# Ultrafine Particle Concentrations in Schoolrooms and Homes

# Final Report: Contract No. 05-305

Prepared for the California Air Resources Board and the California Environmental Protection Agency: California Air Resources Board Research Division 1001 I Street, 5<sup>th</sup> Floor Sacramento, CA 95814

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> > > 7 March 2010

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### Acknowledgments

The authors thank Stephanie Parent, Dane Westerdahl, and Peggy Jenkins of the Research Division of the Air Resources Board for their effective technical management of this project.

This report was submitted in fulfillment of Contract 05-305, "Ultrafine particle concentrations in schoolrooms and homes," by the University of California, Berkeley, under the sponsorship of the California Air Resources Board. Work was completed as of 10 December 2009; final revisions to the final report were made on 7 March 2010.

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#### Abstract

Field monitoring of ultrafine particles and copollutants was undertaken at seven houses and six classrooms in the East Bay region near San Francisco, California. At each site, timeresolved measurements were made indoors and outdoors of particle number (PN) concentration in addition to nitric oxide, ozone, carbon dioxide and carbon monoxide. Data were also acquired through the use of questionnaires (houses), observation (classrooms), and temperature and proximity sensors. The information from each site were analyzed to characterize (1) indoor and outdoor PN concentrations; (2) key factors that influence indoor PN concentrations; and (3) the exposure of building occupants to indoor PN and its determinants.

During the observational monitoring periods, the time-average PN concentrations from the primary indoor monitor varied across the seven house sites from  $3.7 \times 10^3$  particles per cm<sup>3</sup> to  $28 \times 10^3$  particles per cm<sup>3</sup> with a mean of  $14.5 \times 10^3$  particles per cm<sup>3</sup>. The corresponding outdoor concentrations ranged from  $5.5 \times 10^3$  to  $23 \times 10^3$  particles per cm<sup>3</sup> (averaging  $15 \times 10^3$ particles per cm<sup>3</sup>). For five classroom sites (excluding one at which overnight outdoor data were incomplete), the time-average indoor concentrations ranged from  $3.2 \times 10^3$  to  $10.5 \times 10^3$ particles per cm<sup>3</sup> (averaging  $6.9 \times 10^3$  particles per cm<sup>3</sup>); the corresponding outdoor concentrations ranged from  $9.7 \times 10^3$  to  $16 \times 10^3$  particles per cm<sup>3</sup> (averaging  $13 \times 10^3$  particles per cm<sup>3</sup>).

Overall, the results inform the interplay among building factors, human occupancy, and pollutant dynamics as they influence concentrations of and exposures to ultrafine particles in the studied houses and classrooms. Particle levels in classrooms and in houses were much higher when occupied than when vacant. In houses, important contributions to PN levels were attributable to both outdoor particles and indoor emission sources such as cooking (both with natural gas and electricity) and natural gas furnace use. In schools, the dominant PN source was outdoor air and indoor levels were significantly influenced by time-varying ventilation conditions. Daily average PN exposures per person were much higher in houses than in schools.

#### **Executive Summary**

#### Background

Atmospheric ultrafine particles (UFP) are airborne solids or liquids whose individual diameters are smaller than 0.1 micrometers (100 nanometers). This class dominates the number concentration of airborne particles, but makes only a small contribution to mass concentrations. Chemically and physically, UFP are distinct from the currently regulated fine airborne particulate matter, PM<sub>2.5</sub>. Although uncertain, evidence is sufficient to raise concerns about the potential for adverse health effects to result from exposure to UFP in the environment.

A pivotal element in evaluating health risks from air pollutants is to properly characterize exposure. Such characterization requires knowledge about the concentrations of pollutants in the spaces people occupy. Making enough measurements to directly characterize exposure poses great challenges; the effort to understand exposure is facilitated by studying the factors that influence it.

People spend most of their time indoors, and so characterizing conditions in buildings is essential if we are to properly understand exposure to UFP. For particles of outdoor origin, buildings offer some degree of protection. The extent of protection varies with building design and operation and has not been well characterized for UFP. In addition, there are important sources of UFP that can emit directly indoors, thereby substantially influencing exposures.

The main objective of this research is to increase the base of knowledge about the concentrations of UFP in California schoolrooms and residences. A second objective is to advance understanding of the factors that influence UFP levels in these microenvironments.

#### Methods

Field monitoring was conducted in seven houses and six classrooms in the East Bay area near San Francisco, California. The sites were selected to have features that might produce higher-than-average concentrations, but were not intended to be unusual or extreme. At each site, we conducted observational monitoring for several days under normal occupancy and use conditions. We also conducted manipulation experiments designed to characterize ventilation conditions, the indoor penetration and persistence of outdoor particles, and the emissions of particles from potential indoor sources. In all, monitoring at one site or another occurred during ten calendar months between November 2007 and December 2008. (No monitoring was conducted in January, May, July, or August 2008. A supplemental field investigation of emissions from continuous gas pilot lights was undertaken in February 2009.) The seven house sites were occupied, single-family residences built between 1904 and 1996 (average year built = 1945). The six classrooms were from four elementary schools in the urban East Bay.

The primary particle-measuring instruments employed were a set of water-based condensation particle counters (WCPC), which measure the number concentration of airborne particles with excellent time resolution. We monitored and recorded particle number concentrations at one-minute resolution in two indoor locations and also outside of each study site. The WCPC data were augmented with continuous measurements of several copollutants: ozone, nitric oxide, carbon monoxide, and carbon dioxide. We used occupant diaries (in houses) and direct observation (in schools) to record time-dependent information on occupancy, building configuration (e.g., opening of doors and windows), and source-related activities (e.g., cooking). Temperature and proximity sensors provided further information about source activities and building configuration during observational monitoring. Side-by-side sampling was conducted at each site to ensure comparability of indoor and outdoor pollutant concentration measurements.

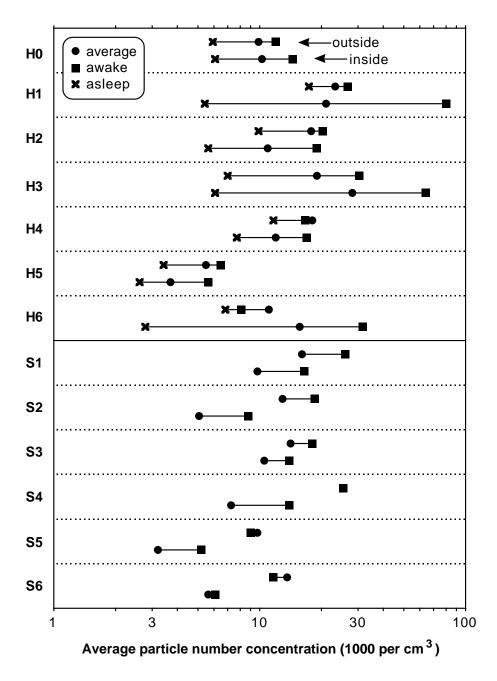
In addition, the flow rates of the WCPCs were checked regularly at each field site and laboratory calibrations of copollutant monitors were conducted at approximately monthly intervals.

The data were analyzed and interpreted to achieve several goals. Time-averaged particle number concentrations were determined for entire periods of observation as well as for periods classified according to the occupancy status of individuals and of the houses and classrooms monitored. Ventilation conditions were characterized using the decay of carbon dioxide as a tracer gas. The contribution of outdoor particles to indoor PN levels was assessed at each site. Contributions of indoor emission sources to indoor particle levels were also evaluated. Indoor exposures to particle number concentrations were assessed for each of the 21 residents of the seven houses studied. We also evaluated exposures for the classroom teacher and for the average student in the six classrooms evaluated. These indoor exposures were apportioned to major source categories: penetration and persistence of particles from outdoor air, emissions from episodic indoor sources, and (at two house sites) continuous emissions from natural-gas pilot lights.

#### Results

A summary of the time-averaged particle number concentrations from observational monitoring is presented in Figure ES.1. For each site, data are presented for outdoor and indoor conditions. The indoor results presented here are based only on the primary indoor monitor, which was placed in a main living space of each house and in the classroom monitored in the case of schools. The overall average monitoring result for the full observational period is represented in the figure by a circle. Averages for parts of the observational monitoring period sorted according to occupancy condition are also presented. For houses, we present the average when all occupants are at home awake (square) and at home asleep  $(\times)$ . For classrooms, we present the average when one or more students are present (square). Note that the horizontal axis is presented on a logarithmic scale spanning a factor of 100. The concentration scale is in units of 1000 particles per cm<sup>3</sup>, so a value of 10 on this scale represents 10,000 particles per cm<sup>3</sup>. On this scale, the overall average indoor concentration in the seven houses ranged from 3.7 (H5) to 28 (H3), with a mean of 14.5. The corresponding outdoor concentrations ranged from 5.5 (H5) to 23 (H1), with a mean of 14.9. For five classroom sites (excluding S4, where we lack a complete record outdoors), the overall average indoor concentration ranged from 3.2 (S5) to 10.5 (S3) with a mean of 6.9, while the corresponding outdoor concentrations ranged from 9.7 (S5) to 16 (S1), with a mean of 13.2.

In houses, the average air-exchange rates varied from 0.1 per hour (H0) to 1.0 per hour (H3) with an overall average of 0.6 per hour, consistent with evidence for overall ventilation conditions for residences in California. In classrooms, air-exchange rates varied strongly with configuration. When all exterior doors were closed and the ventilation system (if any) was off, the average air-exchange rate varied from 0.3 per hour (S2 and S4) to 0.7 per hour (S5) with an overall average of 0.5 per hour. With one or more exterior doors open or with a mechanical ventilation system operating, the air-exchange rates varied from 1.9 per hour (S5) to 4.6 per hour (S3), with an average across all sites of 3.3 per hour. In houses, PN concentration showed little correlation with air-exchange rates. In schools, the higher air-exchange rate condition of the open or mechanically ventilated classroom correlated with higher indoor PN concentrations.



**Figure ES.1.** Average particle number concentrations determined during observational monitoring at seven house sites (H0-H6) and six school sites (S1-S6). For each site, results are reported for outdoor air (upper symbols) and for the primary indoor monitor (lower symbols). The circles represent the overall average for the full observational monitoring period. The squares represent the average for times when the space was occupied with people who were awake. In the case of houses, full occupancy by all residents was required. In the case of schools, the presence of at least one student was required. For houses, the "×" symbols represent average PN concentrations when all of the residents were at home asleep.

In houses, we observed a strong influence of occupant activities on indoor PN levels. In aggregate, for the seven houses monitored for a total of 26 days, we observed 59 emission events

that caused a sudden, usually large rise in the indoor PN level. In most cases, we could correlate the source with a particular activity. For example, in 43 cases showing a sudden particle rise, a single activity was indicated as occurring at the same time, based either on occupant diary information or temperature sensors deployed on suspected sources. For another 14 cases, two activities occurred simultaneously that each might have contributed to the particle concentration rise. Among the single-activity sources, the most common were use of a gas stove or oven (20 cases), use of a gas-fired furnace (9), use of an electric stove or oven (5), and use of a toaster or toaster oven (5).

In schools, episodic sources did not contribute much to indoor particle levels. Instead, the dominant source was outdoor air and indoor levels were influenced by time-varying outdoor levels as well as by time-varying ventilation conditions in the classrooms.

In houses and in classrooms, we evaluated the indoor proportion of outdoor particles. This important parameter (given the symbol  $f_I$  in this report) quantifies the degree of protection afforded by being indoors against UFP pollution in outdoor air. It is a fraction whose theoretical bounds are 0 (implying complete protection) and 1 (implying no protection). For houses, we found that  $f_I$  varied between 0.11 (H1) and 0.51 (H2) with a mean of 0.38. In classrooms, we determined  $f_I$  values in relation to occupancy status and ventilation configuration. Overall, when students were present, the values of  $f_I$  for the six classroom sites varied between 0.48 (S2) and 0.76 (S3) with a mean of 0.57. These results indicate that these indoor environments provide some, but far from complete protection against UFP in outdoor air.

#### Conclusions

Four main qualitative findings summarize the results of this study. First, particle levels in classrooms and in houses are much higher when occupied than when vacant. Second, the indoor proportion of outdoor particles ( $f_1$ ) tends to be higher in classrooms than in homes. Third, indoor emission sources are important contributors to PN levels in houses, but not in classrooms. Fourth, daily average PN exposures per person are much higher in houses than in schools.

In summary, this project was undertaken with the goal of advancing information about ultrafine particles in schoolrooms and in homes in California. This effort is aligned with efforts of the California Air Resources Board to better understand air pollution exposures of California populations. As stated in the Strategic Plan for Research, 2001-2010, "Improved understanding of exposures helps assure that our regulatory activities focus on reducing those that represent the greatest health concerns."

The study design involved detailed investigation, incorporating both observational monitoring and manipulation experiments, at seven houses and six classrooms. With such a small number of sites, we did not intend to collect data that are statistically representative of broader classes of California buildings. In addition, the study is limited to one geographic area in California. Overall, the limits of the study's scope preclude drawing broad conclusions about ultrafine particles in houses and schools throughout California. On the other hand, the intensive monitoring generated rich sets of data that were mined for insights about not only what was observed but also the underlying causes. The study provides information and insight about the interplay among human occupancy, building characteristics, and pollutant dynamic behavior related to the occurrence of ultrafine particles in schoolrooms and homes and the associated exposures. The methods and findings provide an important foundation for future investigations of human exposure to ultrafine particles in California and elsewhere.

#### **1. INTRODUCTION**

Inhalation exposure to airborne particles poses significant adverse health risks (Pope and Dockery, 2006). Federal and California air-quality regulations have been established for ambient mass concentrations of particulate matter smaller than 10 micrometers ( $PM_{10}$ ) and smaller than 2.5 micrometers ( $PM_{2.5}$ ). Evidence indicates that ultrafine particles (UFP), i.e. those smaller than approximately 0.1 micrometers in diameter, may also pose significant health risks (Oberdörster et al., 2005). As noted by Delfino et al. (2005), "High UFP exposures may lead to systemic inflammation through oxidative stress responses to reactive oxygen species and thereby promote the progression of atherosclerosis and precipitate acute cardiovascular responses ranging from increased blood pressure to myocardial infarction."

Ultrafine particles contribute little to the total airborne particle mass. In fact, ambient concentrations of ultrafine particles do not correlate well with ambient  $PM_{10}$  and  $PM_{2.5}$  levels (Woo et al., 2001; Jeong et al., 2004; Watson et al., 2006). Consequently, programs to monitor and control airborne  $PM_{10}$  and  $PM_{2.5}$  levels will not necessarily characterize or reduce health risks associated with exposure to ultrafine particles (UFP).

Because of the significant potential health risks, there is a need to better understand human exposure to ultrafine particles and the factors that influence those exposures. The research described in this report is aimed at adding to the base of knowledge concerning airborne ultrafine particles, particularly as they relate to human exposure in homes and in schools. This research is aligned with goals of the Research Division of the California Air Resources Board as articulated in its strategic plan for research (CARB, 2003). "Another major research priority concerns efforts to advance our understanding of one's total exposure to air pollution. This includes characterizing personal exposure to pollutants from both indoor and outdoor sources. Improved understanding of exposures helps assure that our regulatory activities focus on reducing those that represent the greatest health concerns ..."

Two main sources of ultrafine particles have been substantially studied in ambient air. One source is primary emissions from combustion sources such as motor vehicles. Both diesel and spark-ignition engines are sources of ambient ultrafine particles (Seigneur, 2009). The second source is photochemical production as a nucleation event, whereby new particles are formed in the atmosphere from gaseous precursors (Kulmala et al., 2004). Substantial research efforts have been and are being conducted to characterize the ambient particle levels that result from these processes and how they vary in space and time.

People spend most of their time indoors (Jenkins et al., 1992; Klepeis et al., 2001). Regarding human exposure to air pollution, residences and schools are microenvironments of special concern. Cumulatively, populations spend most of their time in their own homes. Furthermore, sensitive subpopulations such as infants, the elderly, and the infirm spend even higher percentages of their time indoors at home than the population average. The concern with schools arises because of the large proportion of time spent in these microenvironments by children and other juveniles, who may be more susceptible to adverse effects from air pollution than healthy adults. To understand human exposure to ultrafine particles and the potential for adverse health effects from such exposure, we must consider the role of indoor microenvironments as sites of exposure.

Ultrafine particles of outdoor origin may penetrate the building shell along with air provided for ventilation. Moreover, indoor concentrations attributable to outdoor sources are likely to be different from local outdoor levels (Riley et al., 2002). These differences arise because of mechanisms such as penetration losses with infiltrating air, deposition onto indoor

surfaces, and filtration in any mechanical ventilation or air handling system that may be present. As of 2005, when the research reported here was proposed, relatively few studies had investigated the relationship between indoor and outdoor ultrafine particles in houses and none had been conducted in schools. In the past few years, new research on this subject has been reported, as is summarized below.

In addition to the impact of outdoor particles on indoor particle levels and exposures, there are potentially important indoor sources of ultrafine particle emissions. Examples are cooking, use of candles, and ozone-induced oxidation of terpenes. Again, as of 2005, preliminary research had been published that characterized ultrafine particle emissions from these sources. Additional work on these and other indoor ultrafine particle sources has been published in recent years.

#### **1.1 Research Objectives and Goals**

Cumulatively, progress in these areas has been small relative to the complexity of the subject and its importance. The research presented here adds to our collective knowledge and understanding. It presents important new information about (i) the levels of ultrafine particles in a sample of schoolrooms and residences in the East Bay area of Northern California, (ii) the factors that influence those levels, and (iii) the levels of human exposure that result from indoor UFP in these residences and schoolrooms. At the sites studied, the goal of this research has been not just to understand "what" but also "why" with respect to ultrafine particles. Of particular interest is the effect on indoor UFP concentrations of influencing factors: outdoor concentrations; indoor emissions; building factors that influence UFP dynamics; and human activities that affect emissions, dynamic behavior, and human exposure to the resulting UFP levels. These broad objectives have been achieved by pursuing the following specific goals:

- Establish an ensemble of real-time, continuous monitoring instruments for measuring particle number concentrations (which are dominated by UFP) and related copollutants in indoor and adjacent outdoor air.
- Conduct a field monitoring campaign in seven residences and six schoolrooms in the East Bay area of northern California to obtain new experimental data on indoor UFP levels and the factors that influence them. At each of the field sites, data were gathered during a period of observational monitoring, typically of three days duration. The campaign also included manipulation experiments conducted at each field site to further elucidate factors that influence indoor UFP levels.
- Analyze the experimental data, interpret the results, and report the findings to ARB and also to the broader professional community.

#### 1.2 Background

As reviewed by Delfino et al. (2005), recent research has led to a growing concern about potential adverse health effects associated with human inhalation exposure to ultrafine particles (UFP, i.e., particles whose diameter is less than ~ 0.1  $\mu$ m). The diverse sources of UFP and their dynamic behavior raise challenges in characterizing human exposure. Indeed, Sioutas et al. (2005) remarked that, "exposure assessment issues for UFPs are complex.... These issues include high spatial variability, indoor sources, variable infiltration of UFPs from a variety of outdoor sources, and meteorologic factors leading to high seasonal variability in concentration and composition, including volatility."

When viewed through the lens of human exposure, evidence points to several important source classes. Internal combustion engines, both diesel and spark ignition, have been implicated as important sources of urban ambient ultrafine particles, as recently reviewed by Seigneur (2009). In addition, evidence demonstrates compellingly that atmospheric nucleation events are important causes of ambient ultrafine particles (Kulmala et al., 2004). In nucleation events, new particles are formed owing to the oxidation of gaseous precursors - such as sulfur oxides, nitrogen oxides, and certain organic compounds — to lower volatility products. These two source classes — mobile sources and atmospheric nucleation — are clearly important as contributors to ambient UFP. Because particles can penetrate into buildings and other enclosed microenvironments along with ventilation air, important sources of ambient particles are also important sources of human exposure. In addition, many indoor sources of UFP have been identified. Among these are cooking with either gas or electric appliances (Wallace et al., 2008); use of laser printers (Schripp et al., 2008); use of candles (Wright et al., 2007) or incense (See et al., 2007); reaction of ozone with terpenes (Coleman et al., 2008); emissions from heated dust (Pedersen et al., 2003); use of electric motors (Szymczak et al., 2007); and, as reported in one case, a natural-gas clothes dryer (Wallace, 2005). Although these indoor sources may contribute relatively little to ambient concentrations, they may still contribute substantially to human UFP exposure. This apparent paradox is resolved by recognizing that the indoor spaces that people occupy are much smaller and ventilated less intensely per capita than are ambient environments, even in congested cities. Per unit of emissions, the inhalation intake of pollutants emitted indoors can be a few orders of magnitude higher than for pollutants emitted outdoors (Smith, 1993; Bennett et al., 2002; Nazaroff, 2008). Nevertheless, the overall significance of indoor sources in contributing to indoor levels of UFP and to human exposures has not been well characterized.

Since people spend most of their time indoors (Jenkins et al., 1992; Klepeis et al., 2001), understanding the levels of UFP in indoor environments and the factors that control them is an important step for characterizing human exposure. Among various indoor environments, two categories of special concern are schools and residences. Because of the substantial proportion of time spent there by children — on the order of a thousand hours per year — schools are of particular interest. Children may be more vulnerable to air pollution health effects because their bodies are still developing and because they breathe at a higher volumetric rate per body mass than adults. Air quality in schools has not been well studied (Daisey et al., 2003). Residential air quality is also important. In aggregate, residences are the microenvironment where people spend most of their time, so UFP levels in this setting have special significance. Furthermore, other individuals who may have enhanced susceptibility to air pollution — such as infants, the elderly, and the infirm — may spend a higher proportion of their time indoors at home than the population average.

Research on UFP levels in schools and residences, while still small relative to the importance of the subject, has been rapidly expanding. In preparing the proposal for this research in 2005, we made an extensive effort to review the relevant archival literature and were unable to identify any studies that reported UFP levels in occupied schoolrooms and only twelve studies that reported measured UFP levels in residences. Among the studies published as of 2005, only a few were conducted in California (Vette et al., 2001; Suh et al., 2004; Zhu et al., 2005; Kuhn et al., 2005). Highlights are summarized briefly here. Vette et al. monitored indoor and outdoor fine-particle size distributions, down to 0.01  $\mu$ m, over two month-long periods in a single residence in Fresno, California, which was unoccupied except for the researchers. This

study site had no known indoor sources of UFP. The data were interpreted to provide information on particle penetration factors and deposition rates as a function of size. Suh et al. (2004) collected size-resolved particle concentration data inside and outside of 17 homes in Los Angeles. Although the emphasis was on understanding personal exposure to  $PM_{2.5}$  and its constituents, the study includes interesting data on size-integrated particle volume concentrations in the ultrafine size range, 0.02-0.1 µm. The papers by Zhu et al. (2005) and Kuhn et al. (2005) reported UFP levels in four apartments located within 100 m of the I-405 freeway in Southern California. Although these apartments were occupied, an effort was made to minimize the effects of occupancy on measurement results. Instead, the emphasis was on understanding the impact of freeway-generated UFP on indoor levels. Zhu et al. inferred penetration factors and deposition rates as a function of particle size. They observed that penetration factors and deposition rates conformed to model expectations for UFP sizes in the range 20-100 nm, but deviated from expectations for 6-20 nm particles. Kuhn et al. (2005) used a tandem differential mobility analyzer technique to explore the volatility of freeway-generated UFP inside of two of the apartments monitored by Zhu et al.

In preparing this final report, we again surveyed the literature through early 2009, focusing on papers published in peer-reviewed archival journals. A major part of the search was conducted using "Web of Science," with the topic words: "indoor ultrafine." Identified articles were retained if, based on reading the title and abstract, they addressed one or more of these topics: (i) number concentrations (rather than mass or chemical constituents) of particles in either schools or residences; (ii) factors that influence UFP levels in schools or residences; (iii) epidemiological investigations of health effects associated with UFP levels or exposures in schools or residences; and (iv) characterization of exposure to ultrafine particles. These criteria were relaxed somewhat if the study was specific to conditions in California. Table 1.1 presents a brief summary of 60 articles identified through this process. This is a remarkably young literature: a striking feature is that only ten of the 60 articles (< 20%) were published before 2005. A few especially interesting articles in the context of the present study are further discussed in the following paragraphs.

Three recent studies provide interesting information about the potential health risks of inhalation exposure to ultrafine particles in indoor environments. Brauner et al. (2008) conducted an intervention study of a healthy, elderly, nonsmoking population of 21 couples. Using a double blind, crossover study design, the air in each household was either filtered or unfiltered for 48-h periods. Microvascular function (MVF) was measured as the primary end point and several biomarkers of inflammation and oxidative stress were characterized as secondary endpoints. Even with such short intervention periods, filtration significantly improved MVF having reduced both fine-particle mass concentrations (PM<sub>2.5</sub> declined on average from 12.6 to 4.7  $\mu$ g m<sup>-3</sup>) and particle number concentration (from 10,000 per cm<sup>3</sup> to 3,200 per cm<sup>3</sup> for sizes larger than 10 nm). Inflammation and oxidative stress markers were not significantly influenced by the intervention. The authors found that PM<sub>2.5</sub> mass concentration was "more important than the total number concentration for these effects."

Garza et al. (2008) used in vitro cultures of human lung epithelial cells to demonstrate that soot from various combustion sources is cytotoxic and that the "cytotoxicity is not related to [polycyclic aromatic hydrocarbon] content but is related to [reactive oxygen species] generation." Among the sources tested were important indoor combustion sources — candles and natural gas burners — in addition to wood, diesel, and tire smoke. A noteworthy finding was that, in terms of reactive oxygen species generation, "indoor soot production in kitchen

cooking environments may be as efficient in contributing to oxidative stress as outdoor environments where tire burning may occur."

Vinzents et al. (2005) investigated correlations between air pollutant exposures and oxidative DNA damage in 15 healthy nonsmoking subjects. They found that outdoor and indoor exposures to UFP were significant predictors of purine oxidation in DNA. Other air pollutant species were not significant predictors, including PM<sub>10</sub>, NO, NO<sub>2</sub>, CO, and ambient UFP levels. Personal monitoring to UFP was executed using the TSI Model 3007 condensation particle counter, which is designed to be portable and uses isopropanol as its working fluid.

Three studies have been published in the past few years that report particle number concentrations characterized in schools. Fromme et al. (2007) report on an investigation of indoor air quality in 64 schools in and near Munich, Germany. Particle number concentrations (PNC) were determined during a summer sampling period in 36 classrooms, each measured for one 5-h school day, using a scanning mobility particle sizer (SMPS), which responded to particles in the diameter range 10-487 nm. The "median PNC measured in 36 classrooms ranged between 2622 and 12,145 particles cm<sup>-3</sup> (median: 5660 particles cm<sup>-3</sup>)."

Wargocki et al. (2008) reported on an intervention study in which particle filtration conditions were altered in classrooms and children's performance in schoolwork was characterized. These researchers found that the operation of electrostatic air cleaners "considerably reduced the concentration of particles in the classrooms." However, "there were no consistent effects of this reduction on the performance of schoolwork, on the children's perception of the classroom environment, on symptom intensity, or on air quality as perceived by the sensory panel." Particle levels were characterized by means of spot measurements at the end of each week of the study, with the classroom recently vacated, using size-resolved mass determinations as well as number concentration in the size range 20-1000 nm. In this study, outdoor levels were in the range 2800-4250 cm<sup>-3</sup>. Without an active air cleaner, the indoor particle number concentrations were in the range 1300-5600 cm<sup>-3</sup>. With electrostatic air cleaners, the levels declined from the uncontrolled values by 40-80% and varied in the range 700-2300 cm<sup>-3</sup>.

Weichenthal et al. (2008) reported on UFP measurements in 37 classrooms in two schools (one elementary and one secondary) in a rural area of Canada during the wintertime. UFP levels were measured using a TSI model 8525 P-TRAK, which is a particle condensation counter that responds to particles with diameters in the size range 20-1000 nm. Measurements were made continuously indoors throughout the school day for a cumulative total of 60 days in 37 classrooms. Spot measurements of outdoor levels were made at the beginning and end of each school day. (Cold temperatures precluded continuous outdoor monitoring.) For the 30 measurement days at School A, the daily mean indoor PN level was in the range 1160-10,900 cm<sup>-3</sup> with an overall average of 5400; for School B, the corresponding numbers were 1030-11,400 cm<sup>-3</sup> with an overall average of 4600. Two additional schools were subsequently monitored for eight days each and average indoor levels were 2900 cm<sup>-3</sup> in School C, and 7100 cm<sup>-3</sup> in School D. With respect to sources, authors of this study found that "with the exception of an electric kitchen stove in School D, strong indoor UFP sources were not identified in the schools examined."

Wallace et al. (2008) have recently reported on laboratory investigations of UFP emissions from residential cooking appliances, including gas and electric stoves and an electric toaster oven. One particularly strong feature of this study was the advanced battery of aerosol instrumentation used, which allowed detection of particles down to 2 nm diameter. All three

types of cooking appliances generated ultrafine particles at high rates, with or without food being cooked. Aggregated across all 150 experimental tests, reported emission rates were in the range  $(0.06-14) \times 10^{12} \text{ min}^{-1}$ . As an example of significance, a 5-min cooking activity that emits at a rate of  $10^{12}$  particles min<sup>-1</sup> would contribute a peak indoor concentration increase of 17,000 cm<sup>-3</sup> throughout a 300 m<sup>3</sup> house, neglecting losses (e.g., by coagulation or deposition) during the cooking activity.

Fruin et al. (2008) reported on an experimental investigation of the on-road (in vehicle) levels of ultrafine particles and other vehicle-related pollutants in the Los Angeles area. Using their new data along with time-activity information and literature reports of UFP levels in other settings, they estimated that the "time-weighted average population UFP exposure ... was 14 000 particles cm<sup>-3</sup>.... Thirty-six percent of the total exposure ... resulted from drive time, even though drive time only accounts for 6% ... of the day for Californians." The cumulative central estimate for residential exposure in that study (breakfast, dinner, and home) was 147 in units of 1000 particles cm<sup>-3</sup> × h/d, similar in magnitude although somewhat higher than the 119 attributable to on-road exposure. Residential exposure in that paper was estimated based primarily on the work by Cyrys et al. (2004) who studied one hospital building in Erfurt, Germany, and, for cooking, from the detailed investigations conducted by Wallace (2006) in one townhouse in Virginia. A useful contribution of the research reported in the present study would be to improve the empirical basis for estimating residential exposures to ultrafine particles for comparative estimates like those reported by Fruin et al.

### **1.3 Condensation Particle Counters**

A key instrument for measuring UFP is the condensation particle counter (McMurry, 2000). Condensation particle counters (CPCs) provide a means for measuring total particle number concentrations. These instruments detect all particles above a defined threshold. They do not specifically count ultrafine particles. However, ultrafine particles, or at least those particles with diameters below 200 nm, generally dominate the total particle number concentration.

Ultrafine particles have diameters much smaller than the wavelength of visible light, and therefore are too small to be detected directly by means of light scattering. To enable their detection, condensation particle counters first increase the size of the particles through condensation. Most standard, commercially available CPCs use an alcohol, such as butanol or isopropanol, to enlarge particles to enable their detection by optical means. The toxicity of the alcohol working fluid makes them difficult to work with for measurements in occupied indoor environments.

It would be attractive to replace the working fluid in traditional CPCs with a safer substitute. Water would be an ideal candidate. However, it is not possible to use water in a conventional CPC. The design and operation of alcohol-based CPCs specifically require a slowly diffusing species as the condensing vapor. These instruments work by introducing a warm, alcohol-saturated flow into a cold-walled condenser, creating a region of alcohol supersaturation that activates condensational growth of the particles. Key to this process is the slow diffusion of the alcohol relative to the time constant for cooling. The molecular diffusivity for water vapor ( $0.265 \text{ cm}^2/\text{s}$ ) is more than three times higher than that of butanol ( $0.081 \text{ cm}^2/\text{s}$ ). More importantly, it is higher than the thermal diffusivity of air ( $0.215 \text{ cm}^2/\text{s}$ ). Cold-walled condensers cannot work with water vapor because it diffuses too quickly to allow efficient particle activation and growth.

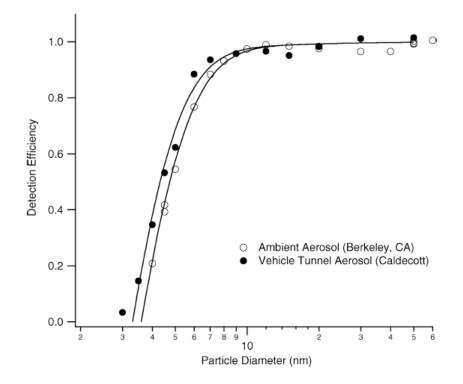
Hering et al. (2005) reported the development of a new, water-based condensational particle counter design that permits the measurement of particle concentrations without alcohol. Their approach centered on a "growth tube" technology that explicitly takes into account the high diffusivity of water vapor, enabling the condensational growth through water condensation in a continuous, laminar flow (Hering and Stolzenburg, 2005). The "growth tube" is a single tube with wetted walls that are warm with respect to the entering flow. The heating of the walls promotes condensation by making a high partial pressure of water vapor available for transport to the center of the flow. The "cooling" necessary for condensation is provided by means of the sample air stream itself.

Several water-based condensation particle counters (WCPC) utilizing this principle have been developed, and are now commercially available. Calibration data showing the lower particle size detected by the first model of this instrument, the TSI 3785, are shown in Figure 1.1 (Hering et al., 2005). Laboratory tests have shown that organic, as well as inorganic salts are activated, grown, and detected (Hering et al, 2005; Petäjä et al., 2006).

Since the 3785 was introduced, two additional models of the WCPC have been designed, an ultrafine WCPC (Model 3786) and a microenvironmental WCPC (ME-WCPC). Each of these outperforms the Model 3785 for direct measurement at the high concentrations encountered in ambient and indoor air. The microenvironmental WCPC (ME-WCPC) is the most recently introduced WCPC. The ME unit is relatively light (2.5 kg) and compact ( $12 \times 18 \times 18$  cm), as depicted in Figure 1.2. It is designed for direct measurement over the concentration ranges found in ambient or indoor air. Like the WCPC described above, the ME units use water as the condensing vapor. The operating temperatures are the same as for the Model 3785 and the lower detectable particle size is similar (Figure 1.3). A difference is that the flow rate has been reduced on the ME-WCPC to eliminate coincidence errors when sampling at the high concentrations found in ambient air.

The prototype ME-WCPC underwent 12 weeks of field-testing in Riverside and Berkeley, CA, with comparison to two butanol-based instruments, under the sponsorship of CARB's Innovative Clean-Air Technologies (ICAT) Program. Good comparability was found between the water-based units, including the ME-WCPC, and butanol-based CPCs. Further details about the development and field-testing of the ME-WCPC unit can be found in Hering (2006).

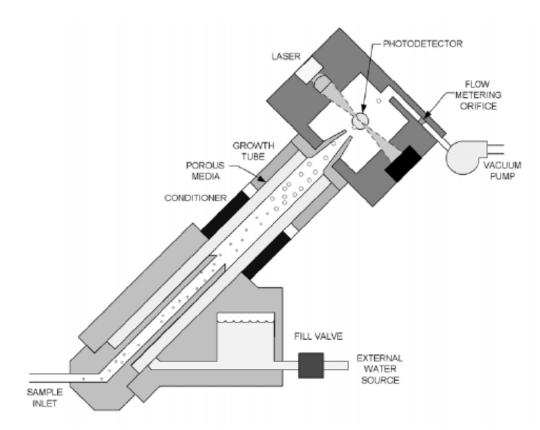
The ME-WCPC is now commercially available as TSI Model 3781. In the present project, we used four instruments, which were designated as QMEa, QMEb, QMEc, and QMEd, respectively. Instrument QMEa was a TSI Model 3781 purchased by ARB from TSI. The other three were similar, early commercial models produced by Quant Technologies and provided for use in the study by Aerosol Dynamics.



**Figure 1.1.** Detection efficiency of the WCPC for atmospheric aerosols from Berkeley, CA and for vehicular particulate sampled from a freeway traffic tunnel. Measurements obtained by reference to a butanol-based ultrafine CPC (TSI Model 3025). Figure reprinted with permission from Hering et al. (2005), copyright American Association for Aerosol Research.



**Figure 1.2.** Photograph of the Micro-Environmental Water-Based Condensation Particle Counter (ME-WCPC) (Hering, 2006).



**Figure 1.3.** Schematic of the microenvironmental water-based condensation particle counter (ME-WCPC), showing conditioner, growth tube, porous media used for wick, and optics head (Hering, 2006).

# 1.4 Research Approach

Field monitoring was conducted at a set of houses and classrooms in the East Bay area of Northern California. At each site, continuous real-time monitoring was conducted for particle number concentration (PN) using WCPCs. Monitoring was also conducted for several copollutants: ozone, nitric oxide, carbon monoxide, and carbon dioxide. Two sets of instruments were assembled into packages, one used for indoor monitoring and the second for outdoor monitoring on site. The purpose of monitoring copollutants was to provide clues about the sources of ultrafine particles and also information about their dynamic behavior. A third WCPC was deployed at a second indoor location to provide information about the spatial variability of indoor PN.

The pollutant concentration measurements were supplemented with data from proximity sensors to monitor the position of a few doors and windows at each site so as to provide ventilation-related information. We also deployed temperature sensors with integral data loggers on suspected PN sources, such as toasters and furnace discharge registers. Questionnaires (in houses) and direct observation (in classrooms) provided supplementary information related to occupant activities and ventilation conditions at the study sites.

At each site, observational monitoring was conducted for approximately three days with the house or classroom in ordinary use. An additional few days of so-called "manipulation experiments" were undertaken under controlled conditions to characterize PN emissions from a few sources, to measure air-exchange rates, and to investigate the penetration and persistence of outdoor particles.

In all, monitoring was undertaken at thirteen field sites, seven houses and six classrooms. The first house site was a pilot for the subsequent investigations. Here, monitoring was undertaken for a longer period, including extensive side-by-side testing of the WCPC instruments both indoors and outdoors. We also tested and refined the protocols used for observational monitoring and manipulation experiments at the remaining sites.

The remainder of the report is organized as follows. Section 2 presents the detailed experimental methods and describes the field sites. Sections 3 and 4 present and discuss results from the house sites and classroom sites, respectively. Sections 5 and 6 present conclusions and recommendations for future research. A list of references and a glossary are respectively presented as §7 and §8. Finally, a series of seven appendices presents supplemental information from the study, as follows: (a) calibration data; (b) forms used in the field; (c) floorplans of the houses and classrooms; (d) time series plots of monitored species at the house sites; (e) probability distribution functions of the minute-by-minute observational data for PN levels at the house sites; (f) time series plots of monitored species at the classroom sites; and (g) probability distribution functions of the minute-by-minute observational data for PN levels at the classrooms.

|                             | To  | pic <sup>a</sup> |          |              |              |              |   |
|-----------------------------|---|------------------|----------|--------------|--------------|--------------|---|
| Reference                   | S   | Н                | Χ        | Ε            | D            | С            | Comment   |
| Afshari et al., 2005        |   |                  |          | ✓            |              |              | Chamber experiments characterizing common potential indoor sources          |
| Alshawa et al., 2007        |   |                  |          | <b>√</b>     |              | $\checkmark$ | Effect of ionization air purifiers on particle removal and generation       |
| Balasubramanian & Lee, 2007 |   | $\checkmark$     |          |              |              |              | Several day investigation of UFP in multistory apartment, Singapore         |
| Berry et al., 2007          |   | ✓                |          |              |              | ✓            | Characterized effect of ionic air cleaner on indoor PM levels including UFP |
| Brauner et al., 2008        |   | ✓                |          |              |              | ✓            | Effects of short-term filtration in households on microvascular function    |
| Coleman et al., 2008        |   |                  |          | <b>√</b>     | 1            |              | Characterize nucleation events involving ozone and terpenes                 |
| Cyrys et al., 2004          |   | ✓                |          |              | 1            |              | I/O ratio of PN monitored for 2-month summer & winter periods, Erfurt       |
| Delfino et al., 2008        |   | ✓                | 1        |              |              |              | PN of outdoor origin correlated with inflammatory markers in elderly        |
| Diapouli et al., 2007       | <ul> <li>Image: A set of the set of the</li></ul> | ✓                |          |              |              |              | Monitored I/O levels of PN in 7 schools & 1 residence in Athens             |
| Evans et al., 2008          |   | <b>√</b>         | <b>\</b> | $\checkmark$ | $\checkmark$ |              | Food frying in residences as a source of exposure to UFP                    |
| Franck et al., 2006         |   | ✓                |          |              |              |              | Reviews residential I/O relationships for PM including UFP                  |
| Fromme et al., 2007         | ✓   |                  |          |              |              |              | Observational monitoring data in many schools in and near Munich            |
| Fruin et al., 2008          |   |                  | <b>\</b> |              |              |              | UFP levels in vehicles on CA roadways; contribution to total exposure       |
| Garza et al., 2008          |   |                  |          | $\checkmark$ |              |              | Cytotoxicity of soot, including emissions from candle & natural gas         |
| Gehin et al., 2008          |   |                  |          | $\checkmark$ |              |              | Emissions from candle, incense, cooking, spray use, cleaning, printing      |
| He et al., 2004             |   | $\checkmark$     |          | $\checkmark$ |              |              | Characterized levels and sources of UFP in 15 houses in Australia           |
| He et al., 2007             |   |                  |          | $\checkmark$ |              |              | UFP emissions from laser printers   |
| Hirsikko et al., 2007       |   |                  |          | 1            |              |              | New secondary particle formation events observed indoors                    |
| Hoek et al., 2008           |   | ✓                |          |              |              |              | I/O levels of PN in households in four European cities                      |
| Hussein et al., 2005        |   | $\checkmark$     |          | $\checkmark$ | $\checkmark$ |              | Monitored home in Finland for 3 weeks; studied sources and dynamics         |
| Hussein et al., 2006        |   | $\checkmark$     |          | 1            | 1            |              | House in Prague monitored; emissions from cooking, smoking, incense         |
| Kagi et al., 2007           |   |                  |          | 1            |              |              | Characterized emissions of UFP from laser & ink-jet printers                |
| Kuhn et al., 2005           |   | ✓                |          |              | 1            |              | Volatility of UFP from freeway in apartments in LA                          |
| Lam et al., 2006            |   |                  |          |              |              |              | Reviews health effects of carbon nanotubes, source includes gas cooking     |
| Langer et al., 2008         |   |                  |          | 1            |              |              | Ozone-limonene reactions as a source of indoor UFP                          |
| Lee et al., 2004            |   |                  |          |              |              | $\checkmark$ | UFP control using unipolar ions discharged into a room                      |
| Lee and Hsu, 2007           |   |                  |          | <b>√</b>     |              |              | Levels of VOC, PM and UFP in 12 photocopy centers in Taiwan                 |
| Liu and Nazaroff, 2001      |   |                  |          |              | $\checkmark$ |              | Particle penetration during infiltration of air across building envelope    |
| Long et al., 2001           |   | $\checkmark$     |          |              | 1            |              | Time and size-resolved PM data in 9 Boston-area homes                       |
| Matson, 2005                |   | $\checkmark$     |          |              | 1            |              | Monitored offices and homes in Sweden and Denmark; I/O ratio                |

**Table 1.1.** Summary of published research in peer-reviewed, archival journals on ultrafine particles in schools and homes

| Mitsakou et al., 2007       |              | $\checkmark$ | 1            |   |   |              | Simulated lung deposition of UFP; considered cooking event                 |
|-----------------------------|--------------|--------------|--------------|---|---|--------------|--|
| Monkkonen et al., 2005      |              | ~            |              |   |   |              | Monitored 11 urban households in India; I/O ratios; cooking as source      |
| Murr et al., 2004           |              |              |              | <ul> <li>Image: A start of the start of</li></ul> |   |              | Carbon nanotubes from natural gas household appliances                     |
| Ogulei et al., 2006         |              | ~            |              | <b>√</b>  |   |              | Source characterization in VA townhouse; positive matrix factorization     |
| Pagels et al., 2009         |              |              |              | <b>√</b>  |   |              | Laboratory characterization of emissions from candles                      |
| Pedersen et al., 2003       |              |              |              | <ul> <li>Image: A start of the start of</li></ul> |   |              | Emissions of VOCs and UFP from heated indoor dust                          |
| Pui et al., 2008            |              |              |              |   |   | 1            | Mitigating exposures using recirculating air filtration                    |
| Schripp et al., 2008        |              |              |              | <ul> <li>Image: A start of the start of</li></ul> |   |              | Characterize emissions of UFP from laser printers                          |
| See & Balasubramanian, 2006 |              |              |              | <b>√</b>  |   |              | Effect of Chinese wok cooking on UFP in Singapore food stall               |
| See et al., 2007            |              |              |              | $\checkmark$  |   |              | Incense smoke particles characterized                                      |
| Sioutas et al., 2005        |              |              | $\checkmark$ |   |   |              | Reviews UFP exposure emphasizing transportation sources                    |
| Sjaastad et al., 2008       |              | <b>\</b>     |              | <b>√</b>  | <b>\</b>  |              | UFP emissions, transport and level from meat cooking indoors               |
| Szymczak et al., 2007       |              |              |              | $\checkmark$  |   |              | UFP and copper emissions from vacuum cleaner motor                         |
| Vinzents et al., 2005       |              |              | $\checkmark$ |   |   |              | Oxidative DNA damage linked to personal exposure to UFP                    |
| Wallace & Howard-Reed, 2002 |              | <b>\</b>     |              | $\checkmark$  |   |              | 18 months of detailed PM and ventilation data in VA townhouse              |
| Wallace et al., 2004a       |              |              |              | $\checkmark$  |   |              | Emissions from cooking on gas stove  |
| Wallace et al., 2004b       |              | ✓            |              |   | $\checkmark$  | $\checkmark$ | Fan and filter effect on UFP in Virginia townhouse                         |
| Wallace et al., 2008        |              |              |              | $\checkmark$  |   |              | Characterize UFP emissions from gas and electric stoves                    |
| Wallace, 2005               |              |              |              | <ul> <li>Image: A start of the start of</li></ul> |   |              | Emissions of UFP from residential, vented clothes dryer                    |
| Wallace, 2006               |              | ~            |              | <b>√</b>  |   |              | Detailed, time-resolved (37 mo.) measurements in townhouse in VA           |
| Wargocki et al., 2008       | $\checkmark$ |              |              |   |   | <b>√</b>     | PN counts (> 20 nm) in schools with different filtration                   |
| Waring et al., 2008         |              |              |              | <ul> <li>Image: A start of the start of</li></ul> |   | 1            | Portable air cleaners can remove UFP but also generate via ozone emissions |
| Weichenthal et al., 2007a   |              | ~            |              | <b>√</b>  |   |              | Cross-sectional survey of 36 Canadian homes; effect of heating on UFP      |
| Weichenthal et al., 2007b   |              |              |              |   |   |              | Reviews link between UFP exposure and childhood asthma                     |
| Weichenthal et al., 2008    | $\checkmark$ |              |              |   |   |              | Measured UFP levels in 37 classrooms in 2 schools; used pTrak              |
| Wright et al., 2007         |              |              |              | ✓   |   |              | Laboratory characterization of UFP emissions from a candle                 |
| Wu et al., 2006             |              |              |              |   | ✓   | $\checkmark$ | Effect of wall surface material on UFP deposition enhanced by air ions     |
| Zhao et al., 2009           |              |              |              |   | <ul> <li>Image: A start of the start of</li></ul> |              | Modeling of transport and deposition of ultrafine particles indoors        |
| Zhou and Levy, 2008         |              |              | 1            |   |   |              | Modeled UFP intake fraction for Manhattan street canyons                   |
| Zhu et al., 2005            |              | ✓            |              |   | 1   |              | Penetration of UFP from freeway into apartments in LA                      |
| Zhu et al., 2006            |              | $\checkmark$ |              | ✓   |   |              | Compared P-Trak vs. CPC for measuring indoor UFP, esp. near freeway        |

<sup>a</sup> Topic code: S = schools, H = homes, X = exposure, E = emissions, D = dynamic behavior, C = control).

# 2. MATERIALS AND METHODS

The research reported here was organized around the central goal of collecting, analyzing, and interpreting new experimental data on ultrafine particle concentrations in schoolrooms and houses. The primary goal is to improve the information that is available about the UFP levels that occur in these microenvironments in California, the factors that influence them, and the resulting exposures associated with these concentrations.

This chapter of the report is divided into two main sections. Section 2.1 focuses on research design, summarizing site selection, describing the instrumentation, and presenting the basic experimental protocols. Section 2.2 presents details about field monitoring at each of the seven house sites and six schoolrooms studied.

# 2.1 Research Design

The basic research scheme embodies these several elements:

- Study sites were selected to span a broad range of expected UFP source conditions.
- Continuous, real-time observational monitoring was conducted in occupied spaces under ordinary use over multiday periods.
- Measurements were made both in school classrooms and in single-family dwellings.
- UFP concentrations were measured along with several copollutants, including nitric oxide and ozone.
- Information was gathered about occupancy, source-related activities, and important building-related factors through questionnaires, direct observation, and the use of supplementary sensors.
- Manipulation experiments were executed to elucidate the factors that influence UFP concentrations and resulting exposures.
- Experimental data were analyzed and interpreted permitting us to glean information about underlying mechanistic causes for what was observed.
- Experimental data were interpreted to provide information about the consequences of indoor UFP levels for exposure of occupants at the studied sites.

Research design inevitably involves tradeoffs. In this study, we have chosen to monitor fairly intensively at a limited number of sites. The intensity of monitoring is designed to elucidate information about underlying physical principles that affect UFP levels in schoolrooms and houses. What is learned from the particulars of the sites studied here can be more generally applied in this case, than would be so with less intensive investigation of each site. On the other hand, because only a fixed number of sites have been investigated (seven houses and six classrooms in this case), the set cannot be considered statistically representative of broader classes of schoolrooms and residences. In particular, the research is not designed to use statistical methods to draw broad inferences about typical or average indoor UFP levels. A second important consideration in the research design involves observational monitoring versus controlled manipulation experiments at field sites. The present project uses a balanced approach with observational and manipulation components. A limitation of purely observational monitoring is the lack of control over the environment, limiting the researchers' ability to draw inferences about causal connections. While well-conceived manipulation experiments overcome this limitation, manipulation raises questions about the relevance of experimental results to understanding real human exposures. By designing a research project that includes both observational monitoring and manipulation experiments, we aim to take advantage of the strengths of each approach.

The field-monitoring phase of the research was conducted from November 2007 through February 2009. During this time, thirteen sites — seven single-family houses and six school classrooms — were studied for approximately one week each. Nominally, at each site, we conducted three days of observational monitoring plus three days of manipulation experiments. The observation period was designed to characterize conditions at study sites under normal occupancy and use. The manipulation experiments were conducted to quantify, under controlled conditions, important factors that affect indoor UFP levels. Specifically, experiments were designed and conducted to characterize at each field site: (a) emissions from one or more potentially important indoor sources; (b) ventilation conditions; and (c) penetration of UFP from outdoors and its persistence in indoor air. In addition to the information gleaned from the manipulation experiments, we found that the intensive observational monitoring periods provided significant information about these three important factors. The first of the field sites, house H0, was used as a pilot to test the performance of the instruments and the experimental protocols.

The research was designed so that approximately equal efforts were made to advance understanding of ultrafine particle concentrations in schools as in houses. The underlying rationale balances several considerations. Overall, people spend more time in houses than in schools, even children. On the other hand, schools are communal spaces, and many of them are in the public trust, so there is a special responsibility of government agencies to ensure their safety and healthfulness. There are likely to be more varied and complex sources of ultrafine particles in residences as compared with schools, which might justify more attention to residential environments. On the other hand, although neither setting has been well studied, houses have been better studied to date than schools, which argues for more attention to be given to school settings. The net effect of these factors suggests that a balanced effort is best.

UFP levels vary temporally in any indoor environment. Furthermore, time-averaged levels would be expected to vary from one location to another. Many factors influence indoor UFP levels. One important set of factors would account for the indoor UFP of outdoor origin. One such factor is the outdoor UFP levels, which would be influenced by subsidiary factors such as the rate of emissions from nearby roadways, the frequency and intensity of atmospheric nucleation events, and meteorological transport and dispersion from localized emissions to the building in question. Indoor UFP levels would also be influenced by the degree to which outdoor UFP penetrate and persist indoors, an idea parameterized as the "indoor proportion of outdoor particles" (IPOP) (Riley et al., 2002). The IPOP is governed by ventilation conditions of the building, particle removal by air filtration (if present), and the rates of particle deposition to indoor surfaces, which would vary across the building stock in a manner dependent on building design and construction and also on building operation.

In addition to variations in the contributions that outdoor particles make to indoor UFP levels, indoor sources can contribute significantly. Expected sources include unvented combustion activities, such as natural gas cooking, candle or incense use, and tobacco smoking. Ozone reactions with terpene-based cleaning products or air fresheners can also generate UFP indoors. The effect of emissions on indoor UFP levels depends on the frequency and intensity of source activities and also on the rates of particle transport and removal from the indoor space, which would in turn vary with ventilation rate, filtration, and deposition to indoor surfaces.

The study reported here did not systematically investigate all of these factors. Site selection satisfied several criteria related to source characteristics as will be described below. However, observational monitoring at each site was conducted over a single period of

approximately three days, so whatever meteorological conditions occurred during that time certainly would have influenced what was observed. Proximity to major ambient sources, building characteristics that affect the IPOP, and indoor source use would be expected to correlate with ethnicity and socioeconomic status of the building occupants and these features were not systematically controlled or characterized here.

It is also stressed that this study is targeted at range-finding with regard to indoor UFP levels, with a bias toward levels that are higher than average, except that there was no smoking at any of the study sites. The study is also targeted at providing information about the cause-and-effect relationships that link sources with indoor concentrations and the consequential exposures. As such, it is not expected that these results could be directly extrapolated to understand indoor UFP exposures for California populations as a whole. On the other hand, the results should provide information that could be invaluable in the design of studies for that purpose.

# 2.1.1 Site selection

All of the experimental monitoring was conducted in Alameda County, located in the East Bay area near San Francisco, California. Among the county's population of 1.4 million (year 2000 data, US Census Bureau, 2009), 94% live in one of the 14 incorporated cities, which range in population from Emeryville's 10,000 to Oakland's 400,000. The overall population density of Alameda County is 760 persons per km<sup>2</sup>. However, considerable portions of the county are hillsides with limited development, preserved parklands, and reservoirs or lakes, and so the cumulative population density of the 14 incorporated cities is much higher than the overall county average, at 1780 persons per km<sup>2</sup> of dry land.

We selected Alameda County for this research because of two main characteristics. First, it is situated amidst one of the larger urban areas of California. As such, there are major primary pollution sources, including heavy vehicular traffic, an important source of atmospheric UFP. Second, it is proximate to the team's base in Berkeley, California, which facilitated field research, allowing us to collect more experimental information using the available resources (money and time) than if we were to sample at a more remote site. On the other hand, one of the limitations of this selection is that ambient ozone levels are lower than in many other urban areas of California.

#### Houses

Seven houses were included in the study. All were occupied, single-family dwellings in Alameda County, California. Recruitment was accomplished through the use of professional and personal contacts of the researchers. Specific sites were selected based on three main criteria. One was in consideration of proximity to major ambient pollution sources. The second was the presence and utilization of potential indoor UFP sources. The third criterion was a practical one, considering the occupants' willingness to provide access and to cooperate with researchers to conduct the observational and manipulation experiments. Compensation was provided at a level of \$100 per research day for occupants' participation and for use of their home. Because the research involved collecting questionnaire data of occupants, its conduct fell under the purview of the Committee for the Protection of Human Subjects at the University of California, Berkeley. The protocols employed in this research were reviewed and approved by that committee.

Overall, the ensemble of houses selected were intended to exhibit diverse characteristics with respect to important factors influencing indoor UFP levels, with a bias in favor of conditions in which elevated but not extreme concentrations are expected. So, for example, we

did not include houses in rural environments where ambient sources are not expected to be large. Likewise, we sought to sample in houses with moderate to high occupancy, as characterized by number of occupants per house volume and by proportion of time spent indoors at home, rather than in houses that are barely occupied. We sought to include houses spanning a range of ages and therefore reflecting different construction styles and degrees of airtightness against infiltration. Because it is the dominant housing type in the US, we only included detached, single-family dwellings and did not study apartments or condominiums. Table 2.1 provides an overview of the houses monitored.

| Table 2.1. Characteristics of house sites studied. |            |  |     |                                |  |  |  |  |  |
|--|------------|--|-----|--------------------------------|--|--|--|--|--|
| ID   | City       | Y built <sup>a</sup> Vol. (m <sup>3</sup> ) <sup>b</sup> Res |     | Residents <sup>d</sup>         |  |  |  |  |  |
| H0   | Oakland    | 1938   | 320 | 2 (M adult and F adult)        |  |  |  |  |  |
| H1   | Oakland    | 1910   | 315 | 4 (M adult, F adult, m 9, m 5) |  |  |  |  |  |
| H2   | Oakland    | 1949   | 328 | 4 (M adult, F adult, m 5, m 3) |  |  |  |  |  |
| H3   | Oakland    | 1928   | 200 | 3 (M adult, F adult, m 11)     |  |  |  |  |  |
| H4   | Oakland    | 1904   | 386 | 4 (M adult, F adult, m 4, m 4) |  |  |  |  |  |
| H5   | Livermore  | 1993   | 420 | 1 (F adult) <sup>c</sup>       |  |  |  |  |  |
| H6   | Emeryville | 1996   | 314 | 3 (M adult, M adult, F adult)  |  |  |  |  |  |

Table 2.1. Characteristics of house sites studied

<sup>a</sup> Estimated year built data from <u>http://www.zillow.com</u> except for H6.

<sup>b</sup> Estimate based on geometric measurement of occupied spaces. Unoccupied spaces such as unfinished basements or garages not included. Not corrected for volume of furniture or cabinets. <sup>c</sup> A second resident was away from the house throughout the study period.

<sup>d</sup> M = male adult; F = female adult; m = male child.

House H0, the first house monitored, was designated as a "pilot" investigation because it was used to test and refine the experimental procedures and instrumentation. The research team deems the quality of the information obtained at H0 sufficient to warrant including it in the analysis as one of seven houses monitored. The seven houses studied do span a broad range of ages, from 10-20 years old (H5 and H6) to more than a century old (H4). Relative to the housing stock of California, a state that has experienced strong population growth on a decadal time scale, the set of houses is older than typical. This outcome resulted from a deliberate choice to emphasize urban core housing, which tends to be older, rather than suburban sites. In general, higher levels of primary pollutants from motor vehicle exhaust are expected in the urban core of cities rather than in the periphery. With respect to occupant density, the occupant-weighted geometric mean household volume per occupant in this set of seven houses was approximately 100 m<sup>3</sup>, lower than the analogous number for all US single-family residences, 160 m<sup>3</sup> (Nazaroff, 2008). Table 2.2 provides additional information about the characteristics of these houses.

Potential sources that could impact indoor UFP levels were important considerations in the selection of field sites. Table 2.3 summarizes the source-oriented criteria to be met in the population of houses to be studied and how these criteria were realized in the seven houses studied.

Figure 2.1 illustrates the spatial relationship of the houses to freeways and major arterial roads. The two sites closest to freeways were H3 and H4. House H3 is sited about 120 m away from freeway I-580, along a stretch where heavy-duty trucks are not permitted and, as a consequence, there is little diesel traffic. The freeway is to the south and west of the house and since the prevailing wind direction is westerly, the house is very often downwind of the freeway.

The average daily traffic count for the nearest milepost (R35.71 106<sup>th</sup> Avenue, Oakland) on that freeway is 167,000 cars per day. House H4 is sited about 30 m away from the edge of I-580 at a section where heavy-duty diesel trucks are allowed. The average daily traffic count for the nearest milepost (45.15, at the junction with routes 24 and 980, Oakland) is 202,000 vehicles per day (Caltrans, 2007).

| Site | Monitoring                 | Location  | Description   |
|------|----------------------------|---|---|
| HO   | Oct-Nov<br>2007            | Urban residential street with<br>light traffic; nearest major<br>roadway is a freeway 500 m to<br>the east; elevation ~ 150 m   | Three-bedroom, two-bath house on split level.<br>Central furnace fired by natural gas and uses<br>natural convection (rather than a fan) for air<br>movement. Electric cooking appliances.<br>Detached garage.  |
| H1   | December<br>2007           | Urban siting on lightly<br>trafficked side street but only<br>five lots from a busy arterial<br>street; closest freeway is 600 m<br>to the southeast; elevation < 50<br>m | Three-bedroom, 2.5-bath house on two levels.<br>Attached garage. Forced air, natural gas<br>furnace; no air conditioning. Gas cooking<br>appliances with continuous pilot lights.   |
| H2   | February<br>2008           | Urban siting on steep low-<br>traffic residential street;<br>moderately busy arterial within<br>50 m of house; nearest freeways<br>are 500 m to NE and 1000 m to<br>SW.   | Three-bedroom, two-bath house on two levels.<br>Attached garage. Cooking appliances are<br>natural gas without pilots.  |
| H3   | March 2008                 | Urban location close to freeway<br>(100-150 m) with annual<br>average daily traffic of ~<br>170,000 vehicles  | Two-bedroom, one-bath house on single level.<br>Forced air natural gas furnace. Natural gas<br>cooking appliances with continuous pilot<br>lights.  |
| H4   | April 2008                 | Urban location very close to<br>freeway (< 50 m) with annual<br>average daily traffic = 202,000<br>vehicles.  | Two-bedroom, two-bath house on two floors.<br>Natural gas appliances for cooking without<br>pilots. Separate central air systems for<br>upstairs and downstairs, both equipped with<br>efficient particle filters. Upstairs system<br>operated continuously; downstairs system<br>under thermostatic control. |
| H5   | June-July<br>2008          | Suburban location on low<br>traffic residential street; 1.25<br>km south of freeway   | Modern two-story, 3-bedroom, 3-bath house.<br>Attached garage. Central, forced air, natural<br>gas furnace with air conditioning. Electric<br>cooking appliances.   |
| H6   | September-<br>October 2008 | Urban location on medium<br>trafficked street; 200 m from<br>heavy urban arterial; 800 m east<br>of freeway; elevation < 50 m.  | Two-bedroom, two-bath house on two floors.<br>Natural gas cooking appliances, without pilot.<br>Central, forced-air heating not used during<br>study.   |

Table 2.2. Additional information about house sites studied.

All of the houses studied were nonsmoking environments and we deliberately avoided peculiarly heavy indoor sources, such as extreme candle use. We explicitly aimed not to investigate environmental tobacco smoke in this project. We did aim to investigate conditions

that are potentially elevated and within the ordinary range encountered by most Californians, but not the most extreme conditions.

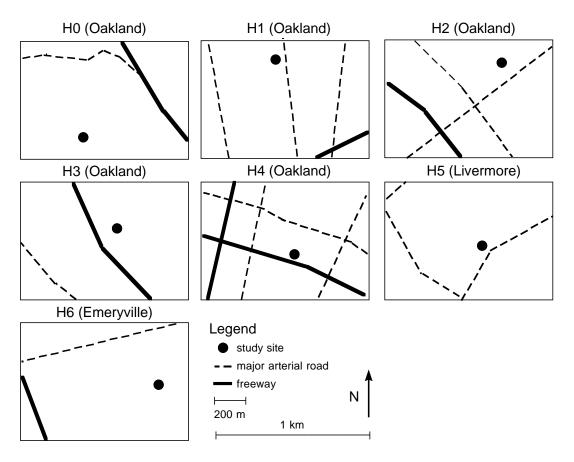
| Attribute             | Criterion                              | Houses w/ attribute  |
|-----------------------|--|----------------------|
| Proximity to heavily  | At least two of the houses will be     | H3, H4               |
| traveled roadway      | selected to be close to freeways with  |                      |
|                       | high traffic density.                  |                      |
| Gas cooking           | At least three of the houses will have | H1, H2, H3, H4, H6   |
|                       | gas-cooking appliances and will use    |                      |
|                       | them to prepare at least one           |                      |
|                       | significant meal during the            |                      |
|                       | observational monitoring period.       |                      |
| Vented natural-gas    | At least three of the houses will have | H0, H1, H2, H3, H4,  |
| appliances            | vented natural-gas-fired appliances    | H5, H6               |
|                       | for one or more of home heating,       |                      |
|                       | water heating, and clothes drying.     |                      |
| Terpene source        | At least two of the houses will        | H4, H5, H6 (terpene- |
|                       | utilize a significant terpene source   | containing cleaning  |
|                       | during the observational monitoring    | products)            |
|                       | period.                                |                      |
| Candle or incense use | At least one of the houses will burn   | H1 and H6 (candles)  |
|                       | candles or incense during the          |                      |
|                       | observational monitoring period.       |                      |

 Table 2.3. Specific source-oriented criteria for selecting study houses.

Significant cooperation was required at all of the field sites. Our ability to secure this cooperation influenced the selection of field sites. However, we do not expect that this cooperation is related to UFP levels or to the factors that influence them, and so this criterion is not expected to bias the research findings.

At each site, both observational monitoring and manipulation experiments were conducted. We aimed to conduct observational monitoring over a contiguous three-day period at each site, including both a weekend day and a weekday. In implementation, some adjustments were necessary to address access limitations and instrument malfunctions. In summary, for the seven house sites, the time monitored was in the range 67 to 140 hours per site (average = 89h/site). At each site, an average of 27 h (minimum = 11 h) of weekend monitoring and an average of 62 h (minimum of 35 h) of weekday monitoring was conducted. At five of the seven sites, the observational monitoring occurred over a single contiguous interval. At one site (H5), monitoring was conducted over two periods with an 8.5-h gap. At H2, three intervals were needed to complete observational monitoring. In this case, we obtained observational monitoring data for almost 4 days (92 h) during a period that extended over eight calendar days. Summarizing the manipulation experiments, a cumulative total of 24 determinations of airexchange rate were made during this phase of research at the seven houses. An outdoor-particle penetration experiment was conducted at each site, but as discussed in §3, we found that the interpretation of observational monitoring data provided a stronger basis for inferring the contribution of outdoor particles to indoor levels. A total of 21 source-activity experiments were also conducted in the seven homes. As planned, these were selected on the basis of relevance to

the particular site. They were also selected so that, in aggregate, at least three different source categories were investigated, including gas-fired cooking, terpene-based cleaning, and the use of candles or incense. In total, the manipulation experiments investigated the following source categories (total number of experiments, number of experiments with quantifiable emissions): gas cooking (5, 5); electric range cooking (3, 3); toaster or toaster oven (3, 2); candles (2, 2); electric iron (1, 1); microwave oven (2, 0); electric kettle (1, 0); vented gas furnace (1, 0); electric clothes dryer (1, 0); and scented cleaning product use (2, 0).



**Figure 2.1**. Schematic showing the spatial relationships between house sites and major nearby roadways. Note that the prevailing wind direction in the study area is from the west.

### Schools

Monitoring was conducted in six classrooms, which were sited at four different public schools. The schools are all located in Alameda County. To preserve anonymity, a condition associated with our receiving approval to monitor at these sites, the school district(s) involved are not named. The protocols employed in this research were reviewed and approved by the Committee for the Protection of Human Subjects at the University of California, Berkeley.

Four broad classes of sources have been identified as potentially strongly influencing indoor UFP levels: ambient nucleation events, emissions from motor vehicles, indoor reactive chemistry between ozone and terpene-emitting products, and indoor unvented combustion activities (for cooking and for other purposes). For schools, it was expected that the first three of these would be the most important in most circumstances, since unvented combustion is not prevalent. Overall, we attempted to select an ensemble of sites whose properties span a range of conditions that might influence indoor UFP levels. Table 2.4 presents a summary of the school sites monitored.

In each monitored schoolroom, our goal was to conduct observational monitoring over at least three normal school days. Our plan was to conduct round-the-clock monitoring rather than monitoring only during hours of occupancy because the additional data may provide important clues about particle sources and dynamic behavior. Mostly, these goals were met. At one site (S4) we did not have access outdoors to electrical power overnight and so we did not operate our outdoor monitoring equipment for overnight periods at this site.

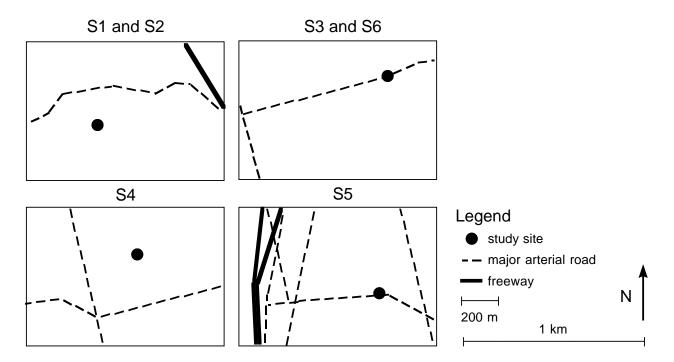
| Site       | Monitoring | Location  | Description   |
|------------|------------|---|---|
| S1         | June 2008  | Quiet residential street;<br>closest freeway is 500<br>m to the east; elevation<br>= 100-150 m  | Third- and fourth-grade students. Older classroom in main building on school site. Natural ventilation using doors and windows to courtyard; no mechanical ventilation. Classroom volume $\sim 290 \text{ m}^3$ .   |
| S2         | Oct. 2008  | Same as S1  | First- and second-grade students. Newly built classroom with mechanical air handling system including particle filter; commonly operated with system fan on and no thermal control. Classroom volume ~ $240 \text{ m}^3$ .  |
| <b>S</b> 3 | Oct. 2008  | Intersection of medium<br>traffic urban street and<br>low-traffic residential<br>street; closest freeway<br>is 1.5-2 km to west;<br>elevation < 50 m      | Second-grade students. Campus is ~ 50 y old, but<br>building containing S3 was built in the 1980s. Building<br>houses three classrooms and a central room;<br>mechanically ventilated room with fixed percentage of<br>outside air and high-efficiency filtration. Classroom<br>volume ~ 205 m <sup>3</sup> . |
| S4         | Nov. 2008  | On residential side<br>street; busy surface<br>streets 100 m to north<br>and 200 m to south;<br>nearest freeway is ~ 1<br>km to west; elevation <<br>50 m | Fifth-grade students. In older building (> 100 y)<br>equipped with radiant heating system. Room ventilation<br>and temperature regulation (cooling) is through use of<br>windows. Classroom volume ~ 230 m <sup>3</sup> .   |
| S5         | Nov. 2008  | Intersection of<br>moderate and heavily-<br>trafficked urban surface<br>streets; nearest freeway<br>is ~ 0.5 km to west;<br>elevation < 50 m              | Fourth-grade students. School campus built in the 1970s. Classroom mechanically ventilated with independent unit equipped with particle filter. Volume $\sim 260 \text{ m}^3$ .   |
| S6         | Dec. 2008  | Same as S3  | Second-grade students. Building containing S6 was<br>constructed in the 1980s. The room is equipped with two<br>wall-mounted ventilation and air treatment units.<br>Classroom volume ~ $300 \text{ m}^3$ .   |

| <b>Table 2.4</b> . | Summarv | of schoolro  | oms studied.  |
|--------------------|---------|--------------|---------------|
|                    | Summury | or senioon o | onio bradica. |

In addition to observational monitoring, manipulation experiments were conducted in each schoolroom. As in residences, the intent of the manipulation experiments was to investigate important attributes that may affect indoor UFP levels in each classroom: (a) ventilation characteristics; (b) dynamic behavior of ambient UFP indoors; and (c) strength and dynamic behavior of emissions from indoor UFP sources. Outdoor emissions from local motor vehicle traffic may impact UFP levels in schoolrooms. The location of schools in relation to major roadways is illustrated in Figure 2.2. Two of the school sites with three of the monitored classrooms were on busy arterial roads. Site S5 was in closest proximity to heavy traffic, being located about 500 m downwind of a major East Bay freeway and also being located directly on a heavily used arterial road.

Recruitment and access to school sites was established through personal contact of research staff with school site directors, such as principals, and with district administrators. The use of an incentive payment of \$100 per day monitored facilitated securing agreement to monitor and also helped ensure strong cooperation of school personnel during monitoring.

It was initially planned that the observational monitoring would proceed with unattended instruments, as in houses. In this approach, information about classroom activities was to be provided by the teacher through filling out daily questionnaires, based on recall. During the more detailed planning in advance of the onset of monitoring, we decided that a preferred approach was to have a member of our research staff continuously in the classroom whenever students were present during observational monitoring. This approach offered two key advantages in relation to the original plan. First, it provided a measure of security and reliability for monitoring to ensure that curious students did not disturb the indoor instrumentation package. Second, the consistency, thoroughness, and reliability of the relevant human observations was undoubtedly enhanced by having a knowledgeable researcher dedicated to making these observations, rather than relying on the recall of a busy teacher. In particular, we took advantage of this arrangement to continuously record (at one-minute resolution) classroom occupancy level and changes in classroom configuration that could influence air-exchange rate, such as the positions of doors and windows.



**Figure 2.2**. Schematic showing spatial relationships between school sites and major nearby roadways.

Cumulatively, classroom monitoring was conducted for a total of 451 h, an average of 75 h per classroom. A total of 18 instructional days were monitored, corresponding to an average of three days per site. At one site (S6) only two instructional days were successfully monitored; at S1, we collected data for four instructional days. At three of the school sites (S1, S4, S5), a single contiguous monitoring period was used. At the other three sites, monitoring was interrupted for one or more of several causes and so the observational monitoring was completed during three or four periods per site. The longest elapsed time from the start to the end of observational monitoring was nine days at site S2. With one exception, all of the monitoring was done on weekdays (i.e., between midnight starting Monday and midnight ending Friday). The exception was at site S6 where one of the overnight periods extended until 0830 AM Saturday morning.

## 2.1.2 Instrumentation

This section of the report describes the instrumentation and other information-acquisition tools used in field monitoring. The central element was instrument packages with real-time monitors for measuring time-resolved particle number concentration plus several other related pollutants and environmental parameters. Three instrument enclosures were assembled for this purpose. Two of these packages were equipped with WCPCs and with monitors for copollutants. These were deployed at all house and school sites during periods of observational monitoring and during manipulation experiments. One enclosure was used to monitor indoor conditions and the second enclosure was used to monitor outdoor conditions on the grounds of the site. A third enclosure, containing only a WCPC, was deployed at a second indoor location to provide information about spatial variability of particle levels within the sites monitored.

Additional information relevant to the interpretation of the experimental data was gathered in three ways. First, during observational monitoring, information was acquired about occupant activities that might be associated with UFP emissions and behavior and also about occupancy status that would be an important consideration in assessing exposure in relation to UFP levels. At the house sites, this information was obtained by having occupants complete activity logs for the observational monitoring days. At the school sites, one member of the research team was present in the classroom throughout the time that students were present to record by direct observation activities and occupancy. Second, temperature sensors and door/window position sensors with integral data loggers were deployed to acquire information related to source activities and to building operation status. For example, common deployments of the temperature sensors were (1) at a supply register on a forced air furnace and (2) on a toaster in the kitchen. The qualitative data from the temperature traces allowed us to detect, for example, when the furnace was operating or when the toaster was used, and this information helped in corroborating or elaborating on evidence gathered from occupant activity logs. Several door and window position sensors were deployed at each site, typically placed on one or two outside doors plus commonly used windows. The information from these sensors helped provide insight into the time variation of building envelope conditions that in turn would influence ventilation rates. The third complementary source of information was acquisition of centralstation data on air pollutant levels and on key meteorological parameters, especially wind speed and direction. The pollutant levels measured at the central station augmented quality assurance information for the outdoor on-site monitoring. In addition, meteorological data provide some insight into the expected impact, for example, of emissions from freeways on UFP levels outside our field sites.

# Instrumentation Packages for Air Quality Monitoring

Air quality monitoring instruments were assembled into packages for deployment in observational monitoring and manipulation experiments. Table 2.5 summarizes the contents of these instrumentation packages.

|                                     |                                   | Enclo | sure <sup>a</sup> |              |
|-------------------------------------|-----------------------------------|-------|-------------------|--------------|
| Parameter                           | Instrument                        | In 1  | In 2              | Out          |
| Particle number concentration (UFP) | ME-WCPC (TSI 3781)                | ✓     | $\checkmark$      | $\checkmark$ |
| CO <sub>2</sub>                     | LI-COR 820                        | ✓     |                   |              |
| $CO_2$                              | TSI Q-Trak Plus 8554              | ✓     |                   | $\checkmark$ |
| СО                                  | TSI Q-Trak Plus 8554              | ✓     |                   | $\checkmark$ |
| Temperature                         | TSI Q-Trak Plus 8554 <sup>b</sup> | ✓     |                   | $\checkmark$ |
| Relative humidity                   | TSI Q-Trak Plus 8554 <sup>b</sup> | ✓     |                   | $\checkmark$ |
| Ozone                               | 2B Technologies Model 202         | ✓     |                   | $\checkmark$ |
| Nitric oxide                        | 2B Technologies Model 400         | ✓     |                   | $\checkmark$ |

Table 2.5. Real-time monitoring instruments.

<sup>a</sup> Three instrument enclosure assemblies were built. The mapping of instruments to enclosures here is associated with how the enclosures were deployed: In1 = primary indoor sampling location; In2 = secondary indoor location; Out = outdoor sampling location.

<sup>b</sup> Temperature and relative humidity measurements were made at some sites using the Onset HOBO U10 T/RH datalogger instead of the Q-Trak Plus instrument.

In addition to monitoring ultrafine particles and basic environmental conditions (temperature and relative humidity), four gaseous pollutants were monitored in this study: carbon dioxide, carbon monoxide, ozone, and nitric oxide. The additional monitoring was done for three purposes. First, we expected that the additional information would help inform us about the relative importance of different UFP sources in contributing to indoor UFP levels. Monitoring ozone was intended to help information about nucleation events. Monitoring NO and CO was intended to provide information about UFP emissions associated with combustion. Simultaneous measurement of these pollutants indoors and outdoors was intended to help distinguish whether the emissions were occurring directly in the indoor environment or whether the source was ambient air.

Second, we expect to obtain information about building ventilation at the monitored sites by means of collecting real-time information on these copollutants. Carbon dioxide proved to be especially useful for this purpose, both as a deliberate tracer in manipulation experiments and also as a tracer of opportunity, especially in classrooms, when the room occupancy state suddenly changed, e.g. at the start of a recess.

A third benefit associated with collecting real-time monitoring data for the copollutants is that these pollutants are of direct interest as indicators of indoor air quality. Extensive data sets do not exist on most of these parameters in indoor air, especially in schools. Toward this end, we calculate and report the time-averaged indoor and outdoor concentrations for ozone, nitric oxide, and carbon dioxide at each site, sorted according to occupancy status of the building. (On average, the carbon monoxide level at each site proved to be lower than accuracy limit of the instrument.) The TSI 8554 Q-Trak Plus with CO provides measurement results for CO<sub>2</sub> and CO, in addition to temperature and relative humidity. The Q-Trak has been used in related studies (e.g., Zhu et al., 2005). Carbon dioxide is measured using a nondispersive infrared technique. According to manufacturer specifications, the instrument has a range of 0-5000 ppm, an accuracy of  $\pm 3\%$  of reading + 50 ppm, a resolution of 1 ppm, and a 20 s response time, making it suitable both for observational monitoring of CO<sub>2</sub> and for using CO<sub>2</sub> as an active tracer for ventilation rate determinations during manipulation experiments. Carbon monoxide is measured using an electrochemical sensor. The instrument range is 0-500 ppm; accuracy is  $\pm 3\%$  or 3 ppm, whichever is greater, and resolution is 0.1 ppm. The response time is < 1 min to reach 90% of final value. This instrument was intended to provide information on pollutant levels influenced by close proximity to roadways or idling vehicles. It was also intended to be useful for monitoring in circumstances where unvented indoor combustion activities produce significant CO.

To ensure high quality data acquisition for carbon dioxide, a second indoor  $CO_2$  instrument was deployed (LI-COR model 820). This instrument also utilizes nondispersive infrared detection and has a manufacturer specified accuracy of  $\pm$  3% plus low drift (http://www.licor.com/env/Products/GasAnalyzers/li820/820\_specs.jsp).

Ozone measurements were made using 2B Technologies Model 202 instrument. This device operates on the basis of UV absorption at 254 nm. According to manufacturer specifications, it has a range of 0-300 ppb with precision and accuracy better than 1.5 ppb or 2%. The instrument was originally developed to be a lightweight, low-power, robust device for measurements of ozone profiles versus altitude on balloon-lifted platforms (Bognar and Birks, 1996). For the current project, the main interest in monitoring ozone is the potential to react with terpenes to generate ultrafine particles (Coleman et al., 2008). Terpene-associated UFP formation events have been detected from indoor monitoring even at ordinary tropospheric ozone levels. We expected to observe a relationship between indoor ozone levels and UFP events associated with terpene use. We found ozone-initiated chemistry to generate UFP in only a few cases, at sites H5 and S1.

Nitric oxide measurements were made using a recently commercialized instrument, 2B Technologies Model 400. The instrument is designed for atmospheric NO measurements and has an analytical range that extends down to 2 ppb, making it suitable for indoor air monitoring. The manufacturer specifies the precision and accuracy to be the greater of 2 ppb or 3%. The specified instrument response time is 20 s, which should have made it well suited for measuring indoor dynamic changes in NO levels. The operating principle is to measure the change in internally generated ozone concentration after exposing an ozone-containing gas stream to the air being sampled. The sensing system is the same as that of the ozone instrument, UV absorption at 254 nm. The virtue of monitoring indoor and outdoor NO is because of the important evidence it may provide in connection with both outdoor and indoor sources of UFP. Nitric oxide is an important copollutant of UFP generated by combustion processes. We anticipated that sites in proximity to roadways would exhibit a significant NO signal that correlates strongly with the UFP signal. However, we did not observe this to be the case. At the sites closest to freeways (H3 and H4) we observed higher-than-average outdoor PN levels, but did not discern a clear time-pattern that would correlate with traffic density. The outdoor NO levels were highest at site H1, which was the site monitored under the most stagnant atmospheric conditions (coldest average temperature and lowest average wind speed. See §3.1).

Nitric oxide was anticipated to be an important copollutant for cooking emissions from natural gas appliances (Traynor et al., 1982). A cooking event would produce an elevated indoor NO level, but not an elevated ambient NO level. We observed elevated indoor NO associated with one or more natural gas cooking events at sites H2 (see Figures 3.11 and D.31), H3 (Figures 3.14 and D.44), H4 (Figures 3.19 and D.57), and H6 (Figures 3.24, 3.26, and D.83). Some cooking-associated PN peaks were not associated with obviously elevated NO or CO levels. For detecting the cause of indoor UFP emissions, we found that the information from occupant diaries and data logging temperature sensors was more consistently reliable.

The instrument enclosures were originally configured with four objectives in mind. First, we sought to provide a convenient unit to support the efforts of research staff in the field. Second, we aimed to provide a secure enclosure that would protect the instruments from wet weather outdoors and from disturbance by building occupants and others who might encounter the instruments. Third, we needed to ensure that the instruments in their enclosures were acceptable for use in occupied houses and classrooms. This consideration required attention to aesthetics, safety, and noise attenuation. Fourth, the instruments were designed to sample air from a consistent height, near a person's breathing zone.

These goals were achieved through assembling the monitoring instruments into commercial polyethylene cases that are designed for housing transportable electronics (1SKB19-10U). Masts were added to the cases to sample air from a height of 1.5-1.6 m, corresponding approximately to the breathing zone of a standing person. Two of the instrument cases are depicted in Figure 2.3.

The instrument cases were designed so that air was sampled near the top of the mast. Gas analyzers (ozone, nitric oxide, and, for one case, the LI-COR  $CO_2$  monitor) were configured so that they sampled air through a Teflon line preceded by a particle filter. The Q-Trak samples air passively to determine  $CO_2$  and CO levels plus temperature and relative humidity. Its sensor probe (and HOBO units, when they were used) was mounted at the top of the mast for each of the two cases so equipped.



Figure 2.3. Instrument cases with sampling masts.

For particle sampling, each of the three cases was also equipped with a sampling line to supply the WCPC. The sampling-line inlet was covered with a wire-mesh screen to protect the WCPC inlet. An auxiliary pump drew air through this line at 8-9 liters per minute and the WCPCs sampled from this line via a tee connection within the box upstream of the pump. External to the case, the line consisted of a length of aluminum tubing (length = 117 cm; inner diameter = 0.79 cm). Within the case, a length of flexible, carbon-impregnated (and therefore electrically conductive) tubing was used (length = 66 cm; inner diameter = 0.64 cm). Particle losses by diffusion to the walls of the sampling line, estimated according to the method of Ingham (1975), were 13%, 6%, 3%, and 2% for 5, 10, 15, and 20 nm diameter particles, respectively.

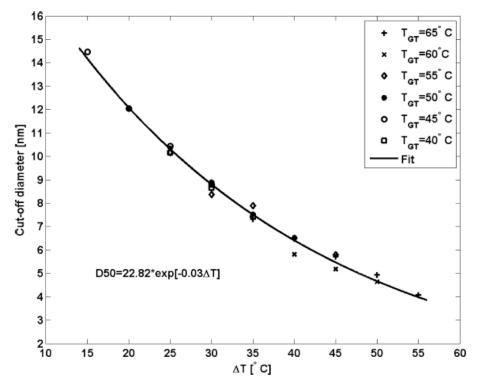
The two main instrument cases were equipped with laptop computers that served as data loggers. The system was programmed to record time-resolved data from each of the instruments. In the enclosure that only housed a WCPC, the internal datalogging function of the instrument was used and the data were transferred to a computer during routine instrument checks. Data were archived at a time resolution of one minute, either by recording data at this time resolution at the field sites or by recording data at a higher time resolution and post-processing to obtain a time series of one-minute-average values.

Initially, the enclosures were only cooled by means of small instrument-cooling fans mounted on the outside of each cabinet. This system, satisfactory at the beginning of the field monitoring campaign in November and December, proved inadequate as weather warmed. Problems were first revealed when, at house site H2, the outdoor enclosure was placed in a location that received direct sun for a portion of the day. The computer datalogger overheated and failed, causing a data lapse for which we compensated by extending the monitoring period. Subsequently, direct solar gain was avoided by more careful siting of the outdoor instrument enclosure. However, at houses H2, H3, H4, and H5, and at school site S1, WCPC instrument status indicators were triggered for a portion of the observational monitoring time. This condition occurred at multiple sites because the indicator that was triggered had multiple possible causes and we did not initially recognize elevated temperatures as the cause.

Once the problem was identified and the cause diagnosed, an intervention was devised that successfully eliminated further occurrences. The intervention involved adding active, thermostat-controlled thermoelectric cooling to the instrument enclosures to offset the internal heat loads and to maintain the enclosure temperature at close to ambient conditions. The absence of further instrument temperature status flags, plus active monitoring of the air temperature within the cabinets at the remaining field sites (H6 and S2-S6) confirm both that the problem was accurately diagnosed and corrected. In addition to this modification of the instrument cases, a review in early August 2008 of the four WCPC instruments deployed in this study by ADI and Quant Technologies personnel led to an upgrading of the heat sink in units QMEb, QMEc, and QMEd to match that in QMEa. Also, at this time, as part of routine maintenance, the nozzles on all units were cleaned and the flow rates recalibrated. A thermoelectric cooling device in unit QMEc was replaced.

It is important to note that the status flag recorded is an indicator of suboptimal instrument conditions, and not an indicator of instrument failure or malfunction. In particular, the flag encountered was a consequence of the temperature of the instrument's saturator (conditioner) rising above its target specification. This condition would result in a reduced temperature difference ( $\Delta$ T) between the saturator and the growth tube. The net effect would be a small shift in the minimum particle size detected by the instrument. The effect is illustrated in

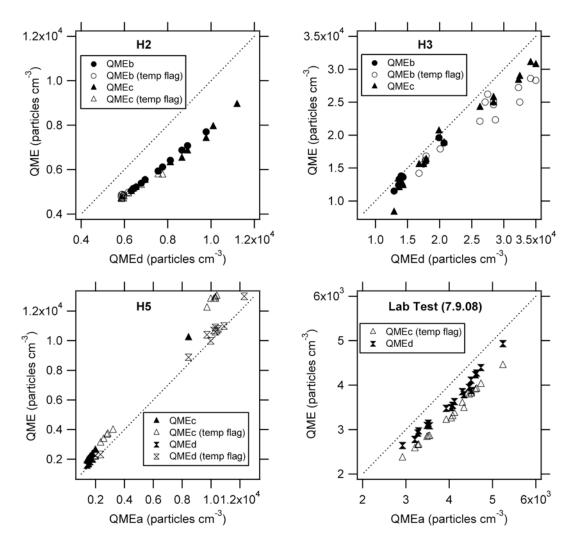
Figure 2.4, which shows the cut-off particle size measured as a function of the temperature difference between the saturator and the growth tube, in this case for silver particles using a TSI Model 3785, which is similar to the 3781 deployed in the present study.



**Figure 2.4.** Minimum particle size detected with 50% efficiency versus temperature difference between saturator (also known as the "conditioner"; see Figure 1.5) and growth tube for the TSI Model 3785 water-based condensation particle counter. These data, for silver particles, illustrate that a higher temperature difference leads to a higher level of supersaturation in the growth tube, which allows smaller particles to serve as nucleation seeds that then grow by water condensation and are subsequently detected. (Figure reproduced from Petäjä et al., 2006.)

The target temperature difference is 40 °C, which produces a cut-off diameter (D<sub>50</sub>) of 7 nm, using the correlation expression displayed in Figure 2.4. The temperature status flag in the WCPC is triggered whenever the conditioner temperature drifts above its target temperature of 20 °C. Laboratory and field tests indicate that the change in temperature difference was 10 °C or less when the status flag was reported. That is, instead of reaching the target  $\Delta T = 40$  °C, the instrument was operating with a  $\Delta T$  in the range 30-40 °C. In the worst of these conditions, the data in Figure 2.4 indicate that the D<sub>50</sub> would have risen from 7 to 9 nm. This shift does not mean that particles between 7 and 9 nm were first counted perfectly and then not counted at all. Instead, it means that the overheating would reduce the measurement efficiency for these particles as well as for particles somewhat smaller and larger. We estimate that the worst-case condition during overheating would have decreased the WCPC is responding to particles that extend to a much larger size range, for which the effect of overheating would be much smaller, we expect that the impact of overheating on overall instrument performance would have been substantially less than the 10-20% maximum indicated.

We have additional direct evidence indicating that the effect of the overheating on data quality is small. As a routine part of our calibration and quality assurance measures, we operated the WCPCs in a side-by-side configuration at each field site. We have some data from these periods when some but not all of the instruments reported an overheating status flag. Sorting the data from these comparisons into periods with and without status flag indicators shows no detectable systematic effect of the overheating condition on instrument comparability. Figure 2.5 illustrates this point.



**Figure 2.5.** A comparison of the particle number concentration (cm<sup>-3</sup>) measured by WCPC units (QMEa, QMEb, QMEc, QMEd) operated side-by-side indoors and outdoors at various residential field sites, and in a laboratory test. Data from monitors designated as reference units (QMEa or QMEd), which did not overheat for the duration of the test are depicted on the x-axis. Each mark represents a 10-min time-weighted-average concentration, and plotted values are raw (uncorrected for calibration). Open symbols are for data from an instrument whose conditioner temperature was out-of-range during the period plotted. A 1:1 dotted line is shown to guide the eye. These frames do not indicate a systematic downward bias in measured particle number concentration attributable to overheating. Although there is a difference between instruments, the difference is consistent per site irrespective of the conditioner temperature status.

In summary, the overheating condition produced a status flag during some of the monitoring periods and for some of the instruments at sites H2-H5 and S1. Indications are that data quality is not substantially degraded by the conditions that triggered this status light signal. In analysis, the data are treated as equivalent, independent of the status flag condition.

The enclosures and each of the sampling instruments was designated with a fixed, unique code. Which instruments were used for monitoring indoors and outside of each house site is summarized in Tables 2.6 and 2.7, respectively.

| Instrument site      | HO    | H1    | H2    | H3    | H4    | H5    | H6    |
|----------------------|-------|-------|-------|-------|-------|-------|-------|
| Main indoor case     | Encl2 | Encl2 | Encl2 | Encl2 | Encl2 | Encl2 | Encl1 |
| Second indoor case   | Encl3 |
| Outdoor case         | Encl1 | Encl1 | Encl1 | Encl1 | Encl1 | Encl1 | Encl2 |
| PN_in1               | QMEb  | QMEb  | QMEd  | QMEd  | QMEd  | QMEc  | QMEa  |
| PN_in2               | QMEc  | QMEc  | QMEb  | QMEa  | QMEa  | (a)   | QMEd  |
| PN_out               | QMEa  | QMEa  | QMEc  | QMEc  | QMEc  | (b)   | QMEc  |
| O <sub>3</sub> _in   | 2B2b  | 2B2b  | 2B2b  | 2B2b  | 2B2a  | 2B2a  | 2B2b  |
| O <sub>3</sub> _out  | 2B2a  | 2B2a  | 2B2a  | 2B2a  | 2B2b  | 2B2b  | 2B2a  |
| NO_in                |       |       | 2B4a  | 2B4b  | 2B4b  | 2B4b  | 2B4b  |
| NO_out               | 2B4a  | 2B4a  | 2B4b  | 2B4a  | 2B4a  | 2B4a  | 2B4a  |
| CO <sub>2</sub> _in1 | LI    |
| CO <sub>2</sub> _in2 | T85b  | T85b  | T85a  | T85b  | T85a  | T85b  | T85a  |
| CO <sub>2</sub> _out | T85a  | T85a  | T85b  | T85a  | T85b  | T85a  | T85b  |
| CO_in                | T85b  | T85b  | T85a  | T85b  | T85a  | T85b  | T85a  |
| CO_out               | T85a  | T85a  | T85b  | T85a  | T85b  | T85a  | T85b  |
| T_in                 | T85b  | T85b  | T85a  | T85b  | T85a  | T85b  | T85a  |
| T_out                | T85a  | T85a  | T85b  | T85a  | T85b  | TRH1  | T85b  |
| RH_in                | T85b  | T85b  | T85a  | T85b  | T85a  | T85b  | T85a  |
| RH_out               | T85a  | TRH1  | T85b  |       | T85b  | TRH1  | T85b  |

Table 2.6. Instruments deployed at house field sites.

<sup>a</sup> QMEd used until 27 Jun 17:59; no data collected thereafter.

<sup>b</sup>QMEa used until 27 Jun 10:00; QMEd used thereafter.

# Instrument Calibration and Performance Checks

This subsection briefly summarizes the calibration and performance checks for the electronic instruments employed in this project. Detailed results of calibrations, side-by-side testing, and comparison of field monitors with central-station monitors for NO and ozone are presented in Appendix A.

Instrument calibrations and performance checks were conducted throughout the period of field monitoring. These steps were designed to ensure, so far as is practicable, that the data collected are accurate. Calibrations were carried out by means of determining instrument response when exposed to known conditions, ideally that span the range expected in the field. Calibrations are not possible for all instruments and are more difficult to execute in the field than in the laboratory. Consequently, another important feature of our quality assurance program was side-by-side testing of the instrument packages, which was conducted for several hours at each of the field sites.

For the WCPCs, the instrument flow rates were checked in the laboratory using an external flow meter (BIOS Defender 510). The measured flow rates were used to adjust instrument response to the target flow of 120 cm<sup>3</sup> min<sup>-1</sup>. Accurate measurement of zero particle concentrations was confirmed by sampling air through a particle filter. All instruments consistently read zero concentration when sampling through the filter. Side-by-side data were collected for all instruments employed at each field site. The side-by-side comparison data were employed by designating one of the instruments as the reference device (QMEa when available or QMEd otherwise) and adjusting the readings of the other instruments to match as closely as possible the results from this reference instrument.

For the ozone monitors, on a few occasions multipoint calibrations were conducted in the laboratory. The aggregate calibration data along with a comparison of our field data to central monitoring results support a finding that the instruments' responses to ozone exposure was accurately recorded in terms of the slope of the response function. Routinely, before monitoring at each field site, the ozone monitors were exposed to ozone-free air by sampling through an ozone scrubber. The resulting readings were used to make zero offset correction.

| Instrument site      | S1    | S2    | <b>S3</b>         | <b>S4</b> | <b>S5</b> | <b>S6</b> |
|----------------------|-------|-------|-------------------|-----------|-----------|-----------|
| Main indoor case     | Encl2 | Encl1 | Encl1             | Encl1     | Encl1     | Encl1     |
| Second indoor case   | Encl3 | Encl3 | Encl3             | Encl3     | Encl3     | Encl3     |
| Outdoor case         | Encl1 | Encl2 | Encl2             | Encl2     | Encl2     | Encl2     |
| PN_in1               | (a)   | QMEa  | QMEa              | QMEa      | QMEa      | QMEa      |
| PN_in2               | QMEd  | QMEd  | QMEd              | QMEd      | QMEd      | QMEd      |
| PN_out               | QMEa  | QMEc  | QMEc              | QMEc      | QMEc      | QMEc      |
| O <sub>3</sub> _in   | 2B2a  | 2B2b  | 2B2b              | 2B2b      | 2B2b      | 2B2b      |
| O <sub>3</sub> _out  | 2B2b  | 2B2a  | 2B2a              | 2B2a      | 2B2a      | 2B2a      |
| NO_in                | 2B4b  | 2B4b  | 2B4b              | 2B4b      | 2B4b      | 2B4b      |
| NO_out               | 2B4a  | 2B4a  | 2B4a              | 2B4a      | 2B4a      | 2B4a      |
| CO <sub>2</sub> _in1 | LI    | LI    | LI                | LI        | LI        | LI        |
| CO <sub>2</sub> _in2 | T85a  |       | T85b <sup>b</sup> | T85b      | T85b      | T85a      |
| CO <sub>2</sub> _out | T85b  | T85b  | T85b <sup>b</sup> | T85a      | T85a      | T85b      |
| CO_in                | T85a  |       | T85b <sup>b</sup> | T85b      | T85b      | T85a      |
| CO_out               | T85b  | T85b  | T85b <sup>b</sup> | T85a      | T85a      | T85b      |
| T_in                 | T85a  | TRH5  | TRH5              | T85b      | T85b      | T85a      |
| T_out                | T85b  | T85b  | T85b              | T85a      | T85a      | T85b      |
| RH_in                | TRH3  | TRH5  | TRH5              | T85b      | T85b      | T85a      |
| RH_out               | T85b  | T85b  | T85b              | T85a      | T85a      | T85b      |

 Table 2.7. Instruments deployed at school field sites.

<sup>a</sup> QMEd used until 3 Jun 8:34; QMEc used thereafter.

<sup>b</sup> At S3, instrument T85b was used during alternating time periods to monitor indoor and outdoor conditions. (Instrument T85a was undergoing repairs at this time.)

For the nitric oxide monitors, laboratory-based calibration was employed to determine instrument response to "zero" air and a span gas (233 ppb). In all, thirteen calibrations were conducted during the monitoring campaign. The most recent laboratory calibration was used as the basis for adjusting the readings of the reference instrument (2B4b). Interpretation of the

response of the other instrument was based on the side-by-side comparison of the two instruments at each field site.

For carbon dioxide measurements, the LI-COR instrument was selected as the reference instrument. It was calibrated on ten occasions in the laboratory using span gases from a compressed gas cylinder and zero air. Carbon dioxide from the Q-Trak instruments were interpreted based on the side-by-side comparison with the LI-COR analyzer.

The manufacturer specification for the Q-Trak carbon monoxide monitor specifies a precision of 0.1 ppm but an accuracy that is the greater of 3 ppm or 3% of the reading. Field data from side-by-side sampling were generally below the 3 ppm accuracy limit, so interpretation of the two Q-Trak instruments was based on laboratory calibrations using zero air and a 40 ppm span gas. Most of the CO data from observational monitoring were of limited value because measurement results were usually not above 3 ppm.

For the temperature and relative humidity measurements, we did not conduct calibrations against primary standards. We did make adjustments in the response of T85a to agree with T85b based on the results of side-by-side testing in the field.

# Questionnaires, Activity Logs and Direct Observation

Information about the field sites and about occupant activities was collected through the use of questionnaires, activity logs, and by direct observation. Full forms are presented in Appendix B. Here, we summarize briefly key aspects.

At house sites, four forms were used, as summarized in Table 2.8. Forms A, B, and C were filled out by research staff based on a combination of inspection and conversation with residents. The occupants themselves filled out occupancy and activity logs (Form D). Form D was devised to be as simple as possible, focusing only on the most important information needed about the occupants. In particular, we sought to know when each of the household occupants was at home, and — for the time at home — to also know when they were awake or asleep. We also sought information from occupants about their use of appliances or products that might be associated with ultrafine particle emissions.

| Form | Purpose   |
|------|---|
| А    | House site characterization: Physical characterization of house and potential sources |
| В    | House site questionnaire: General information on occupants and behaviors              |
| С    | House site observational monitoring: Daily survey                                     |
| D    | House site observational monitoring: Daily activity log                               |

**Table 2.8.** Forms used for acquiring supplementary information in houses.

At the school sites, Form SA, a variant on Form A, was used to guide the collection of information by field research staff on the physical characteristics of the classroom and potential indoor sources. The functions served by the other forms, to acquire information about daily activities of occupants at the field site, were achieved through direct observation by field research staff personnel, one of whom was present in each classroom throughout observational monitoring, whenever a student was in the room. Information was recorded with one-minute resolution on these factors: (a) occupancy (number of students and number of adults present); (b) status of classroom configuration factors that are expected to affect air-exchange rate (position of doors and windows and operating status of any mechanical system); and (c) use of any object that was suspected as a potential source.

# Supplementary Sensors: Temperatures and Door / Window Position

Supplementary dataloggers equipped with open/closed "state" sensors or with temperature sensors were deployed to provide additional real-time monitoring information about potentially important variables at each site. At each site, we used up to four additional open/closed position sensors (Onset HOBO Model U9) and up to six additional temperature sensors (Onset HOBO Model U10). The open/closed position sensors were deployed on windows and doors, mainly to provide information related to occupant activities that would influence the air tightness of the building envelope and consequently might affect the airexchange rate. The temperature sensors were deployed to provide real-time evidence about source-related activities. In this case, the absolute temperature was not of interest as much as time-series behavior indicating a rapid change of conditions. For example, a common deployment of temperature sensors was for cooking appliances, such as a toaster. In this case the sensor was mounted on the outside of the toaster. During operation, a rise in temperature would be registered. Another common deployment was at the position of an air-supply register from the central, forced-air heating or air conditioning system. In this case, operation of the furnace or the air conditioner would be recorded as a sudden change in temperature, respectively, upward or downward. Again, the absolute temperature was not of particular interest, but rather we used the change in temperature as a means of detecting the operation of the device. Tables 2.9 and 2.10 show how these data loggers were deployed at each of the field sites.

| Site       | Sch1               | Sch2                  | Sch3                 | Sch4                   |
|------------|--------------------|-----------------------|----------------------|------------------------|
| H0         | Front door         | Kitchen door          | Kitchen window       | Master bath window     |
| H1         | Front door         | Bathroom window       | Garage door          | Child bedroom window   |
| H2         | Garage door        | Back door, top        | Front door           | Back door, bottom      |
| H3         | Bathroom window    | Door: Kitchen/utility | Front door           | Kitchen window         |
| H4         | —                  | Back door             | Front door           | Kitchen window east    |
| H5         | Door: Garage/house | Front door            | Back door            | Bedroom door (w/ WCPC) |
| H6         | Front door         | Back door             | Upstairs bath window | Middle kitchen window  |
| <b>S</b> 1 | —                  | Back door             | Front door           |                        |
| S2         | Front door         | —                     | Window               |                        |
| <b>S</b> 3 | Front door         | Larger rear door      | Smaller rear door    | Door to central pod    |
| S4         | Front door         | —                     | Window 2             | Window 3               |
| S5         | Back door          | Front door            | Door 1 of hallway    | Door 2 of hallway      |
| S6         | Classroom door     | Cafeteria door        | Window 4             | Window 5               |

Table 2.9. Deployment summary for door and window position sensors at field sites.

### Central Station Monitoring Data

We obtained central-station monitoring data for selected air pollutants and meteorological data, either to support our quality assurance efforts or to provide supplementary information that would assist in interpreting field data. The data we utilized were obtained from California's Bay Area Air Quality Management District, via their website (http://www.baaqmd.gov/). Hourly average results for the duration of observational monitoring at each site were acquired for four ambient pollutant concentrations — carbon monoxide, ozone, nitric oxide, and  $PM_{2.5}$  — and for two meteorological parameters — wind speed and wind direction. At all sites other than H3 and H5, we used pollutant data from the 6<sup>th</sup> Street, Berkeley, monitoring station. For H3, data acquired at 9925 International Boulevard #11, Oakland, were used. For H5, data acquired at 793

Rincon Avenue, Livermore, were used. Meteorological data for all sites was recorded at UC Berkeley. The median distance from a field site to the respective central station monitor was 5 km. All of the field sites were within 11 km (7 miles) of the central station monitors except for the following two cases: (a) site H2 was 10-15 km from the central station pollution monitor; and (b) site H5 was about 50 km from the central meteorological station.

|            |                 |                 | for supprementary temperature sensors at mera sites. |                   |               |                 |
|------------|-----------------|-----------------|--|-------------------|---------------|-----------------|
| Site       | TRH1            | TRH2            | TRH3   | TRH4              | TRH5          | TRH6            |
| H0         | Stove top       | South window    | Toaster  | Supply register — |               | —               |
| H1         | —               | Dishwasher      | Toaster oven   | Supply register   | _             | _               |
| H2         | —               | Clothes dryer   | Enclosure 1  | Supply register   | _             | _               |
| H3         | Enclosure 1     | Supply register | Oven   | Stove             | _             | _               |
| H4         | Supply register | Enclosure 1     | Toaster oven   | Stove top         | _             | —               |
| H5         | —               | Toaster         | Stove  | Enclosure 1       | Oven          | Supply register |
| H6         | Enclosure 1     | Enclosure 2     | Enclosure 3  | Toaster oven      | Stove         | Rice cooker     |
| <b>S</b> 1 | —               | Enclosure 1     |  | _                 |               |                 |
| S2         | Enclosure 1     | Enclosure 2     | Enclosure 3  | Return air vent   |               | —               |
| <b>S</b> 3 | Enclosure 1     | Enclosure 2     | Enclosure 3  | Exhaust vent      | Encl. 1 mast  | Return air vent |
| S4         | Hallway         | Heater          | Printer  | _                 |               |                 |
| S5         | Enclosure 1     | Enclosure 2     | Enclosure 3  | Encl. 3 mast      | Printer       | Supply register |
| S6         | Television      | Enclosure 2     | Enclosure 3  | Enclosure 1       | HAC supply #1 | HAC supply #2   |

Table 2.10. Deployment summary for supplementary temperature sensors at field sites.

# 2.1.3 Experimental Protocols and Procedures

The purpose of this section is to summarize the experimental procedures used to acquire information from the field monitoring sites. The emphasis in this section is on the general protocols and procedures. Details regarding implementation of these general goals at each site are addressed in section 2.2.

# Pilot Study: H0

After the instrumentation packages were assembled and at the beginning of the field monitoring campaign, a pilot-phase monitoring program was undertaken at site H0. In the project proposal, three objectives were established for this task. The first objective was to test the performance of the water-based CPC against a butanol-based instrument in an indoor environment, especially when challenged by common indoor sources. Because of a lengthy delay between project approval and the issuance of funding to start the project, this objective was not pursued. The goal of this objective had already been substantially achieved through the field-testing program described by Hering (2006).

The second objective of the pilot study was to test the side-by-side monitoring performance of the two batteries of real-time monitoring instruments. The third objective was to test the field monitoring protocols both for observational monitoring and for manipulation experiments. Appendix A (§A.1) describes the results of the side-by-side instrument comparison. Overall, we found that the field protocols were suitable for the project with only minor adjustment. Consequently, we decided to treat the observational monitoring and manipulation experiment results at this site in a parallel manner to the other house sites.

### Houses: Observational Monitoring

The first task for observational monitoring was to find appropriate locations for the three instrument enclosures. The primary indoor enclosure (containing WCPC plus copollutant

monitors) was set up in an indoor location centrally located within the house, yet out of the way of the occupants. These goals generally meant that the enclosure was placed along the wall in a frequented gathering room. For five of the houses, the enclosure was placed in a living or family room, and for one house, the enclosure was placed in the dining room. The outdoor enclosure was placed in the backyard at each house, in a location where it could be both secured with a chain and protected from the weather. The supplementary indoor enclosure was placed in a second indoor location of interest. In houses with multiple levels, this second indoor enclosure was placed in a room on the floor less intensely occupied during awake periods. In H3, a smaller one-story house, the ancillary enclosure was placed in the kitchen.

After the enclosures were placed, the instruments were turned on and data were confirmed to be logging to the computer. Starting in September 2008, a check list was added to the protocol, which allowed field research staff to systematically record the following diagnostics after the initial start-up and during each of the field checks: (a) flow to the enclosure was unobstructed; (b) all instruments were logging to the computer; (c) no instrument error signals were being recorded; (d) sufficient water was present in the WCPC reservoir; and (e) cooling fans were operating. These diagnostics were checked at the sites preceding September 2008, but not so systematically.

Before leaving the home, researchers filled out Form A and administered Form B (see Appendix B). To complete Form A, the researchers took stock of all potential sources within the house, made note of the house construction characteristics, and measured the dimensions of all the rooms. Form B was completed by means of questioning one of the residents. The researchers also placed up to six temperature sensors on or near heating/air conditioning supply vents or on surfaces that could become heated and so were suspected of being possible particle sources. Also, up to four state-change sensors were positioned on doors and windows that were reported by occupants to be frequently used.

After these steps were completed, the researchers left the occupants with an occupant journal (Form D), which was formatted for them to both document the times for which they performed potential particle-emitting activities, and the times for which they were home awake, home asleep, or away from the house.

During observational monitoring, the site was visited three times, approximately every 24 hours, to check the diagnostics of the instrument enclosures, to download instrument data from the previous day, and to administer the questions on Form C to the residents. The occupant journal for the previous day was collected. The researchers checked the WCPC data to discern whether any apparent indoor peak events occurred and — if so — that there were corresponding entries for occupant activities during those times.

### Houses: Manipulation Experiments

At house sites, four types of experiments were conducted in the manipulation phase of field research. Respectively, these were designed (a) to measure the air-exchange rate of the house; (b) to measure the penetration and indoor persistence of ultrafine particles of outdoor origin; (c) to characterize emissions characteristics of potentially significant indoor UFP sources; and (d) to test for the presence of combustion emissions directly entering the house on a continuous basis (as would occur, for example, with an unvented pilot light). The following paragraphs summarize the basic experimental protocols. More information is provided about the way in which these experiments were implemented in Section 2.2.

In the original research proposal, it was anticipated that these manipulation experiments would be the primary means by which important factors influencing dynamic behaviors of indoor UFP at the field sites would be assessed. What we learned during the course of the research was that considerable information could be gleaned from the data gathered during observational monitoring for all of these factors. So, in the final analysis, the manipulation experiments provided complementary information rather than primary information about these various factors.

Air-exchange rate. During manipulation experiments, the air-exchange rate was determined by means of tracer-gas decay. In so far as possible, the occupied space of the house was treated as a single, well-mixed volume. Carbon dioxide was used as the tracer gas. A deliberate release was conducted to raise the indoor CO<sub>2</sub> levels, using dry ice (frozen CO<sub>2</sub>) as the source. Dry ice was weighed, and placed in trays of water in several locations throughout the house. Mixing fans were placed immediately in front of the trays, and angled upwards, so as to help with mixing the  $CO_2$  throughout the entire volume of the house as the gaseous  $CO_2$  was released from the sublimating dry ice. Once the indoor concentration reached levels about 2000 ppm, the remaining dry ice was weighed, and then removed from the house. The  $CO_2$  was allowed to mix until concentrations from room to room did not differ by more than 10% (determined using a QTrak). Once this criterion was satisfied, the house was configured to a ventilation mode commonly seen during observational monitoring. The researchers left for a few hours to allow for the concentration decay to be monitored. By fitting an exponential decay curve to the difference between the indoor and outdoor level, the air-exchange rate can be determined. Tracer decay experiments were conducted for key conditions of the building during ordinary occupancy, such as with windows closed or with windows open, or with the mechanical airhandling system on and with it off.

Penetration and indoor persistence of outdoor UFP (Pkd experiment). This procedure is a variant of the "concentration-rebound" method devised by Thatcher et al. (2003). All windows and doors were closed. Three portable high efficiency (HEPA) recirculating fan-filter units were placed around the house and operated at full power. The CO<sub>2</sub> levels were increased to above 2000 ppm using the same procedure described for the air-exchange rate experiment. The particle number concentrations were monitored as they decayed in response to filtration. Once the rate of particle concentration decay within the house began to level off, the researchers removed the HEPA filter units, and exited the house so that it could be monitored without disturbance for a few hours. The data are analyzed using a simple model representation of the dynamic behavior of UFP of outdoor origin in a house. Particles are assumed to fractionally penetrate from outdoors along with ventilation air. Within the house, particles are lost by deposition onto indoor surfaces at a first-order rate and by means of ventilation. The house is modeled as a single, well-mixed space and indoor sources are assumed to contribute negligibly to indoor levels during the experiment. To the extent these conditions are met, then the time-dependent indoor particle number concentration (*C*) can be represented by this differential equation:

$$\frac{dC}{dt} = \lambda P C_o - \lambda C - k_d C \tag{2.1}$$

where  $\lambda$  is the house air-exchange rate,  $C_o$  is the outdoor particle number concentration, P is the fractional penetration from outdoors to indoors and  $k_d$  is the first-order rate constant for deposition of particles onto room surfaces. The goal of the experiment is to determine the

parameters P and  $k_d$  for UFP of outdoor origin by means of fitting the time-dependent indoor UFP concentration to the solution of equation (2.1). As a shorthand notation, this investigation is often referred to as the "Pkd experiment," named after the two key parameters that one seeks to determine from its results.

Indoor source characterization. A few potentially significant indoor UFP emission sources were tested at each house site (see Table 2.11). Selection was based either on the evidence from observational monitoring or because of a priori expectations. When possible, the researchers selected a smaller space within the house for conducting the source experiments, such as the kitchen (when it could be closed off), a bedroom, or a bathroom. The CO<sub>2</sub> within that space was increased to 2000 ppm using the procedure described for the air-exchange rate experiments. After the dry ice had been removed and the CO<sub>2</sub> had mixed throughout the space, the UFP source activity was initiated, and allowed to emit particles for a predetermined period of time. Once the UFP source was terminated, the mixing fans were turned off, and the researchers exited for a few hours while monitoring continued. Data were interpreted using a timedependent material balance model to estimate total emissions (total number of particles emitted), emission rate (particle number emitted per unit time of the activity), and particle decay rate (determined by the extent to which the decay of UFP number concentration exceeded the decay of the outdoor-corrected CO<sub>2</sub> level).

*Persistent combustion emissions test.* This experiment was devised to determine whether a persistent combustion-associated UFP source was present in the house, such as would be associated with a continuously operating, natural-gas pilot light on a cooking appliance. This goal was accomplished using carbon dioxide as a proxy. Since  $CO_2$  is nonreactive, whereas UFP can be lost by deposition on indoor surfaces, a persistent elevation of  $CO_2$  above outdoor levels is a reliable indicator of the presence of an indoor combustion source. In the experiment, the windows and doors of the house were opened to allow for rapid air exchange. Once the  $CO_2$ level within the house dropped to the outdoor value, the doors and windows were closed, and the researchers exited for an hour. If an indoor source was present, it was detectable by observing a time-dependent rise in the indoor  $CO_2$  level.

| Site       | Source(s) tested   |  |  |  |  |
|------------|--|--|--|--|--|
| H0         | Toaster, Electric range (boiling water), Electric range (cooking onion)                |  |  |  |  |
| H1         | Gas range (boiling water), Candles, Toaster oven                                       |  |  |  |  |
| H2         | Gas oven (empty), Gas range (cooking potatoes)   |  |  |  |  |
| H3         | Gas range (cooking onion), Microwave oven (2×), Electric kettle, Gas furnace, Electric |  |  |  |  |
|            | clothes dryer  |  |  |  |  |
| H4         | Toaster oven, Scented cleaning product use   |  |  |  |  |
| H5         | Electric range (cooking rice), Electric iron, Scented cleaning product use             |  |  |  |  |
| H6         | Gas range (cooking onion), Candles   |  |  |  |  |
| <b>S</b> 1 | Mopping with pine-oil based cleaner  |  |  |  |  |
| S2         | Laser printer use  |  |  |  |  |
| <b>S</b> 3 | Candle, Heater   |  |  |  |  |
| S4         | Laser printer use  |  |  |  |  |
| <b>S</b> 5 | Heater   |  |  |  |  |
| S6         | Vacuum cleaner, Laser printer, Scented spray plus ionizing air cleaner                 |  |  |  |  |

**Table 2.11.** Sources tested during manipulation experiments at house and classroom sites.

#### Houses: Pilot-Light Experiments

A few decades ago, it was common for natural gas cooking appliances to have continuously burning pilot lights. These would serve as the ignition source when a stovetop burner or the oven was switched on. Because cooking appliances are typically used only a few hours a day at most whereas the pilot light operates continuously, the amount of gas burned in the pilot could be a significant fraction of the total fuel use. Consequently, in the interest of improving energy efficiency, pilot-free ignition systems were introduced and have become widespread. Nevertheless, some older stoves still have pilot lights. In the present study, it happens that two of the seven houses included as field sites had stoves with continuously burning pilot lights. The presence or absence of pilot lights was not a criterion in house selection. Indeed, we know of no published research reporting that pilot lights might be a source of UFP emissions and did not anticipate that for the present study. However, during observational monitoring at house H1, we observed indoor UFP and  $CO_2$  levels that were persistently elevated relative to our expectations.

An important goal of this project is to apportion the indoor UFP (and consequent exposures) among major source classes, and a dominant division of interest is between particles of outdoor origin and particles emitted from indoor sources. Episodic sources from activities such as cooking are fairly easy to identify and characterize based on their time signatures, as will be demonstrated in Section 3 of the report. Persistent indoor sources that emit at fairly stable rates present a different challenge. Since ventilation rates are uncontrolled and since we are not continuously monitoring this parameter, we do not have enough information to directly determine the contribution of outdoor particles to indoor UFP levels on a time-varying basis. Instead, we must make inferences about that contribution using information gleaned from the cumulative monitoring record (both observational and from manipulation experiments). A persistent indoor source, such as a pilot light, threatens to confound the interpretation.

In light of these considerations, we devised and executed one-day experiments at the two house sites — H1 and H3 — that had continuously operating pilot lights associated with cooking appliances. The goal of the experiments was to quantify the rate of UFP emissions from this source and its consequent impact on indoor particle levels.

The experiment was conducted in an unoccupied and closed house. The real-time instruments used for observational monitoring were operated as usual. Continuous records of indoor and outdoor particle levels and indoor CO<sub>2</sub> levels was generated while the state of the pilot lights was altered, as follows: (a) pilots operating as usual (~ 2 h); (b) pilot lights off (~ 3 h); (c) pilots lights relit and on as usual (~ 2 h). Through analysis of the decline and subsequent rise of UFP levels across these three periods, in combination with the change in the indoor CO<sub>2</sub> levels, we infer best-fit values for the experimental conditions of particle deposition loss rate ( $k_d$ , which considers both outdoor and pilot-light emitted particles), particle penetration efficiency from outdoors (P), and emission rate (E, particles per hour) associated with the continuous pilot lights.

#### Schools: Observational Monitoring

The criteria for siting indoor and outdoor enclosures at schools were identical to those for houses, except that there was the added concern of disruption by curious students or potential vandalism by people who might visit the campus after hours, particularly for the outdoor enclosure. Addressing the security concerns required relaxing our goals for proximity of the outdoor enclosure to the classroom somewhat. For three of the classrooms, the outdoor enclosure was placed in a secure location in close proximity to the classroom. However, for the other three classrooms, finding a secure location for the enclosure was more challenging, and the enclosure had to be located at either an undesirably large distance from the classroom (~100 m), or it had to be unplugged and stored securely indoors overnight.

In addition to instrumental monitoring, questionnaires and data forms were completed. On the first day of observational monitoring, field research staff completed Form SA (a variant on Form A) and supplementary sensors for temperature and door/window position were deployed. Forms B and C were not administered in classrooms, nor was the teacher expected to keep an activity journal during the observational monitoring. Instead, one of the researchers was present within the room during school hours, and documented the number of occupants present, the status of the windows and doors to the classroom, and relevant classroom activities that took place over the course of each monitored school day. The researcher present on these days also conducted a diagnostics check of the instrument enclosures at least once on each of the observational monitoring days.

## Schools: Manipulation Experiments

Air-exchange rate. The same procedure was used as in houses.

*Penetration and indoor persistence of outdoor UFP.* This experiment was conducted using essentially the same procedure as in houses. However, the interpretive model for that procedure depends on the rebound occurring only because of particle penetration directly from outdoors. In isolated classrooms, this assumption is well founded. However, in classrooms contained within larger buildings, particles may enter by several means, including leakage from adjacent indoor spaces (other classrooms or hallways), supply via the mechanical ventilation system, and leakage through infiltration. In this project, we lacked the resources to make a comprehensive evaluation of air flow and particle transport in complex situations such as this. Nevertheless, in some of the classrooms, the QTrak and ancillary WCPC were positioned such that their inlets were immediately below an HVAC supply vent. The intent in doing so was to obtain clues about the extent to which UFP were provided to the room via the HVAC system. And regardless of the limitations of the Pkd experiment, we were able to obtain high quality information about the impact of outdoor particles on indoor UFP levels through analysis and interpretation of observational monitoring.

*Indoor source characterization.* The experiments were conducted using an identical procedure as in houses. Nine experiments were conducted, as summarized in Table 2.11.

*Persistent combustion emissions test.* This experiment was conducted using an identical procedure as in houses.

*Particle concentration decay.* This experiment was conducted in some classrooms to augment the concentration-rebound test. The doors and windows of the room were opened, and the classroom was thereby ventilated rapidly with outdoor air. Once the indoor and outdoor PN levels were approximately equal, the room was sealed, the researchers exited for about two hours, and particle concentration levels were measured as they decayed towards their baseline values.

#### 2.1.4 Data Analysis

The analysis of field data was conducted in three phases, applied to each of the thirteen field sites. The first phase involved generating an archival file of the data acquired. The second phase involved generating time-series plots and other figures depicting key aspects of the results

that could be obtained with limited interpretation. The first two phases were executed using essentially the same procedures for each site. The third phase was a full interpretive analysis of the data. In this third phase, it was necessary to respond to the specific characteristics of each study site in data interpretation, which meant that the procedures varied somewhat. As an example, a result obtained in the second phase of the analysis for each house site was the time-averaged particle number concentrations measured outside and at the two indoor locations, sorted according to the occupancy status of each household resident (away from home, at home awake, and at home asleep). In the third phase of analysis, we sought to apportion the associated at-home exposure estimates for each occupant according to major UFP source categories: outdoors, episodic indoor emissions (with attribution to specific activities), persistent indoor emissions, and unknown. The following subsections provide a summary of the data analysis procedures applied collectively. Details that are specific to each site are addressed in the respective results and discussion sections of the report (Section 3 for houses and Section 4 for schools).

## Analysis Phase 1: Data Archiving

For each field site, data archiving entailed creating a series of files (in MS Word and MS Excel) that contained a structured record of information gathered. Table 2.12 provides a description of the files. These archive files were provided to the Air Resources Board as a term of the research contract. They are also available to other qualified researchers on request.

#### Analysis Phase 2: Time-Series Plots and Other Direct Results

In the second phase, a set of steps was applied to the archived observational monitoring results using basic analysis methods. The primary aims were three: (a) to facilitate communication of findings; (b) to extract interesting and useful information that could be obtained through simple calculations; and (c) to reveal patterns that would help guide subsequent analysis in the third phase. An additional benefit was that producing the time series plots provided a means to audit some aspects of the archive files as a quality assurance measure.

The second phase of analysis produced three primary products. First was a set of timeseries plots for the observational monitoring period, as summarized in Table 2.13. Time-average parameter values were also computed for each of the parameters measured. Second, occupancyassociated probability distribution functions of the one-minute average particle number concentrations were generated. Third, we computed time-averaged particle number concentrations sorted according to occupancy status. The second and third outcomes, described in more detail below, generated figures as well as numerical outcomes.

Before describing the second and third analysis outcomes in this phase, it is important to note differences in the way that houses and classrooms were assessed with regard to occupancy. For houses, each resident's occupancy was characterized independently. Occupancy status was sorted into three states: away from home ("away"), awake at home ("awake"), or asleep at home ("asleep"). Our information about these states derives from self-reports of occupants based on Forms C (administered daily by field staff) and D (log maintained by occupants). For schools, it was deemed impractical to monitor the occupancy status of each student as an individual. Instead, we recorded time series data on the number of students and the number of adults present in the classroom (excluding the field research staff). These data were recorded at one-minute time resolution by means of direct observation during regular school hours, augmented by teachers' reports for the times when students were not in the classroom.

| File name           | Contents   |  |  |
|---------------------|--|--|--|
| Archive_readme.doc  |  |  |  |
|                     | manipulation experiments.  |  |  |
| SiteDescription.doc | Proximity to major roadways and other ambient sources; narrative   |  |  |
|                     | description of site, including ventilation and potential sources; floor plan; occupants and patterns of use.   |  |  |
| Obs.xls             | Primary data record from observational monitoring, organized in three<br>sections: (a) "readme" file that provides documentation for understanding<br>the other sections; (b) "Obs_Adjusted" file that contains time series data<br>incorporating calibration adjustments for all monitored parameters; and<br>(c) "CentralMonit" file that contains time-series data acquired from<br>central station monitors for ambient pollutants and meteorological<br>conditions. |  |  |
| AER.xls             | Primary data record from the manipulation experiment designed to characterize air-exchange rates.  |  |  |
| Pkd.xls             | Primary data record from the manipulation experiment designed to   |  |  |
|                     | characterize the penetration and indoor persistence of outdoor UFP.  |  |  |
| Source.xls          | Primary data record from the manipulation experiment designed to   |  |  |
|                     | characterize emissions from indoor sources.  |  |  |

| Table | 2.12. | Data | archive | files. |
|-------|-------|------|---------|--------|
|-------|-------|------|---------|--------|

**Table 2.13.** Time-series plots generated for the observational monitoring period at each field site.

<sup>a</sup> Although outdoor carbon dioxide levels were measured, the Q-Trak appears to exhibit temperature sensitivity that makes it unreliable for quantifying the fairly narrow range of outdoor concentrations expected. Consequently, outdoor  $CO_2$  levels are not reported based on Q-Trak measurements. Whenever an outdoor value is needed for a calculation result, it is assumed to be the global average background level of 380 ppm.

For the probability distribution function of particle number concentrations, all three of the WCPC monitor results (primary indoor, secondary indoor, and outdoor) were included in the analysis. For houses, we extracted from the minute-by-minute time series data all number concentration results when one of these three conditions was met: (a) all occupants were away from home; (b) all occupants were at home, awake; and (c) all occupants were at home, asleep. When the status of individual occupants differed, the data were not included in this stage of the analysis. For each of the conditions (a) - (c), empirical cumulative probability distributions were computed by sorting the measurements in each group into rank order. Lognormal distribution parameters (geometric mean and geometric standard deviation) were computed for each data subset. The empirical data and the lognormal best fits were then plotted on log-probability coordinates. Thus, for each house site, we have nine probability distribution functions: three particle-monitor locations × three occupancy conditions. At schools, the analysis was similar, except the occupancy sorting criterion was different. The three occupancy states in this case were: (a) one or more students in the classroom; (b) one or more adults in classroom; and (c) classroom vacant. In this case, condition (b) and condition (c) cumulatively occurred 100% of the time, and condition (a) constituted a subset of the condition (b) data.

For the third outcome, again the treatment in houses and in classrooms differed. For houses, we considered each occupant separately. Based on the occupancy status of each occupant, we computed the time-averaged particle number concentration from each monitor separately for each of the three occupancy states (away, awake, asleep). The results are presented as series of bar charts plus the associated numerical values, where for each occupant and each occupancy state there are three bars representing respectively the primary indoor, secondary indoor, and outdoor time-averaged particle number concentration.

### Analysis Phase 3: Interpretation

The broad goal of this phase was to extract whatever meaning and insight could be gleaned from the site-specific data and evidence as a whole to better understand the roles played by various factors in influencing the outcomes of primary concern in this study. In particular, we return to our focus on the ultrafine particle number concentrations indoors, the factors that influence those levels, and the consequences for exposure of building occupants to those particles. We use information gathered both from the observational monitoring phase and also from the manipulation experiments. A key thread throughout is to avoid "forcing" the analysis. Exploration was necessary to allow the data to teach us what it can. It was necessary to maintain a firm, yet flexible sense of direction and goals and at the same time to avoid forcing information to fit into some preconceived notion of what was important. Here are some brief notes about how the analysis was engaged, sorted according to major themes.

For particle concentrations, major influencing factors are (a) sources; (b) ventilation rates; and (c) penetration and persistence of outdoor particles. For (a), we are specifically interested in characterizing the significance of indoor versus outdoor particle sources. Also, when indoor sources are important, can we identify which sources are responsible for which events? The three key tracks investigated were (i) to scrutinize the indoor particle number concentration traces for evidence of episodic sources; (ii) to look in close detail at indoor particle number concentrations in connection with recorded activities that might constitute sources; and (iii) to analyze the "source" manipulation experiments. Characterizing the strength of and decay from indoor episodic sources proved to be a worthwhile effort, both for understanding indoor

particle dynamics and also in carrying out exposure attribution calculations. When relevant, we have tabulated source events and their analysis results.

In the case of ventilation characterization, we were attentive to the potential use of "tracers of opportunity," such as carbon dioxide immediately after a building suddenly becomes unoccupied in addition to the manipulation experiments. We remained mindful of the role that building configuration and weather could play in influencing air-exchange rates. Where possible, we tabulated air-exchange rate determinations and important influencing factors.

A particularly valuable outcome of this study emerged from the coincident collection of time-dependent occupancy information along with indoor particle number concentration data. At the house sites, we are able to explore characterization of exposures for each individual occupant using the microenvironmental approach for estimating exposure. For school sites, we were able to explore the exposure of the main classroom teacher as an individual, and to treat students' exposure statistically.

Because exposure and its associated measures are not fully standardized, it is worthwhile to summarize here the measures that are used and how they are evaluated. Conceptually, exposure refers to the contact between an agent and a target (Zartarian et al., 2005). For the present application, the agent is ultrafine particles, quantified in terms of particle number concentrations, and the targets are the occupants of the residences and classrooms in which monitoring was conducted. The exposure concentration would refer to the UFP concentration in the breathing zone of the exposed individual. In the absence of personal monitoring, we use the stationary microenvironmental PN concentration measurement results as proxy indicators of the breathing zone concentrations for people during the time they are in the buildings being studied. Zartarian et al. also defined a *time-integrated exposure* as the integral of the instantaneous exposure over the exposure duration. In their terminology, the time-integrated exposure would have units of concentration (e.g., 10<sup>3</sup> particles cm<sup>-3</sup>) multiplied by time (e.g., hours) and would be equivalent to the product of the time-averaged exposure concentration multiplied by the exposure duration. Earlier, Zartarian et al. (1997) referred to this same measure as the "temporally-integrated exposure." They further defined the "temporally-averaged exposure" as the temporally-integrated exposure divided by the duration of the time period.

For the purposes of this report, we will use two primary exposure metrics. The ideas are consistent with those presented in Zartarian et al. (1997, 2005), but the naming convention is altered. First, we use the term *exposure* to represent the product of the time-average exposure concentration multiplied by the exposure duration. (This measure is what Zartarian et al., 2005, call the *time-integrated exposure*.) For a household occupant, the PN exposure for any given period is computed by summing the minute-by-minute measures of exposure concentration (in units of  $10^3$  cm<sup>-3</sup>) for each minute that the occupant is inside the house. That result has units of  $10^3$  cm<sup>-3</sup> min and is divided by 60 min h<sup>-1</sup> to convert to units of  $10^3$  cm<sup>-3</sup> h. For students in classrooms, the aggregate exposure for a given period is obtained by summing the minute-by-minute product of student occupancy (number of students in the classroom) multiplied by PN concentration (in  $10^3$  cm<sup>-3</sup> units) for all minutes during the period of interest when at least one student is present in the classroom. The average exposure for each student in the classroom is then estimated by dividing the aggregate exposure by the average occupancy for the minutes when at least one student is present. Dividing that result by 60 min h<sup>-1</sup> produces the average individual student PN exposure in units of  $10^3$  cm<sup>-3</sup> h.

It is illuminating to compare the PN exposures among individuals and from one setting to another. In the present study, the duration of monitoring varied among sites. To facilitate

comparing exposures on a consistent basis, we computed daily exposures, which we refer to as *exposure rates*. For a household occupant, for example, the daily-average exposure rate is determined in two steps: first evaluate that individual's cumulative within-home exposure for the entire monitoring period and second divide by the duration of the monitoring period. For classrooms, the average per-pupil daily exposure rate is determined for each of the school days monitored. One virtue of the exposure-rate measure is that it facilitates the estimation of cumulative exposures for longer time periods. For example, if we can assume that the exposure rates determined in this study are reasonable indicators of average conditions, then the cumulative annual exposure for an individual in their home would be estimated as the product of the daily average classroom exposure rate multiplied by the annual number of school days. We caution that our study is limited in scope and so the exposure rates we determine should be interpreted only as indicators of what the true average conditions might be.

## 2.2 Field Monitoring in Seven Houses and in Six Classrooms

This section of the report describes the thirteen field study sites and the experimental methods used to investigate them. It is not our intent to repeat here the general information described in Section 2.1. Rather, we use this section to present salient information that is specific to each particular field site. With regard to the site itself, we report here on matters such as location, layout, potential UFP sources, ventilation conditions, occupants and their pattern of use of the study site. We also present site-specific information about the observational monitoring effort and the manipulation experiments. Detailed information about the analyses conducted is presented along with the results and discussion in §3 (houses) and §4 (schools).

# 2.2.1 Monitoring in Houses

# 2.2.1.1 Site H0

This split-level house, located in Oakland, was built in 1938. The main level has the kitchen, dining room and large living room. A half flight of steps leads to an upper level toward the back of the house that has three bedrooms and two bathrooms. A utility room is located in a small basement beneath a portion of the upper level. This basement has a vented natural gas dryer. The water heater and furnace are also in the basement area, connected to an unfinished crawlspace that underlies most of the house. The detached garage is north of the house and was actively used. The floor plan is shown in Appendix C (see Figure C.1). The estimated house volume is 350 m<sup>3</sup>; excluding the basement, the remaining house volume is 320 m<sup>3</sup>.

There are hardwood floors in all rooms except the bathrooms, which have tile, and the kitchen, which has a flooring of wood veneer under a plastic laminate. The kitchen has an electric stove, and a separate electric oven. The stove is used every day when the house is occupied. The exhaust hood, which is vented outdoors, is used occasionally.

The house has a central natural-gas furnace with ducted supply registers in each room. However, instead of using a fan to distribute the air, it is configured to use natural convection. The house does not have air conditioning.

These are the identified potential indoor sources of ultrafine particles in the house:

• Electric range top with four burners and equipped with range hood that is vented to the outdoors but is only occasionally used.

- Electric oven.
- Electric kitchen appliances: microwave, toaster oven, espresso machine, dishwasher.
- Vented natural gas appliances (located in basement and crawlspace): furnace, water heater, clothes dryer. The water heater and furnace have continuous pilot lights; these were on during sampling.
- The house has a fireplace, but it was not used during the observational period.

The house is located in the lower part of the Oakland hills (elevation approx. 150 m above sea level) and is therefore somewhat removed from the heart of the urban East Bay area. It is on a residential street with very low traffic volume; we observed a rate averaging less than one car per minute during an observation period on a winter afternoon. The back yard and garage are adjacent to an alley that is closed on the west end and also has very light use.

The closest freeway is Highway 13, which is approximately 500 m to the east. The annual average daily traffic count at the nearest monitoring station on this highway is 74,000 vehicles (Caltrans, 2007).

The main indoor monitoring package (En 2) was placed in the downstairs study. The outdoor enclosure (En 1) was placed in the back yard (to the north of the house), within 6 m of the house. It was protected from rain by being placed under a gazebo installed for that purpose. The second indoor enclosure (En 3, with only a particle monitor) was placed in the bathroom adjoining the hallway on the upper level. The bathroom wasn't used often and the door to the landing was almost always open.

Two adults (M and F) occupied the house. Occupants slept in the master bedroom, but spent the majority of the time that they were indoors and awake on the lower level. Most of the potential sources were in the kitchen (except the furnace, which was in the basement). At this site, the observational monitoring period extended for almost six days (140 h). During this period, each of the occupants was present in the house for four of the six days; their absences did not overlap, so the house was occupied throughout the observational monitoring period.

For a pilot phase of the field experimental work, side-by-side monitoring was conducted with the instrumentation packages for several day periods indoors (21-25 November 2007) and outdoors (25-28 November 2007). The side-by-side monitoring results are described in Appendix A.

The observational monitoring period began on 28 November 2007 (15:45) and ended on 4 December (12:00). There were no interruptions. At this time, the supplementary WCPC had not yet been placed in an enclosure, so the effect of line losses would not be the same for this instrument as for the other two enclosed monitors. Data from this supplementary WCPC are not available at the beginning of the observational monitoring period; data acquisition for this instrument begins on 29 November (16:05). One of the nitric oxide monitors was undergoing repairs with the manufacturer and so there are no indoor NO data at this site. The outdoor relative humidity sensor was also malfunctioning and so there are no outdoor RH data.

Manipulation experiments were conducted on three days: 4, 5, and 7 December. For the air-exchange rate experiment (4 December), the house was configured as follows: exterior door and windows were closed and the interior doors open. The house ( $V \sim 320 \text{ m}^3$  without the basement) was treated as a single, well-mixed space.

An experiment to characterize the impact of outdoor particles on indoor levels was conducted on 7 December. Doors and windows were closed and the indoor particle levels were reduced through the use of two HEPA-filter units. Once particle levels were suitably reduced, the filters were turned off. For a subsequent period of time, the "rebound" in particle number concentration was monitored along with concentration decay of carbon dioxide. At this site, fans were left on and researchers remained in the house for the duration of the particle rebound and  $CO_2$  decay period. Consequently, the data could not be quantitatively analyzed to estimate fractional penetration from outdoors to indoors and the first-order rate constant for deposition of particles onto room surfaces. Instead, we relied upon the interpretation of data from the observational monitoring period.

On 5 December, source investigations were undertaken. We conducted these within the zone proximate to the source – the kitchen – rather than using the entire house. We isolated that zone by closing doors and windows that connect it to the outdoors and to the rest of the house. The net kitchen volume was estimated as  $\sim 37 \text{ m}^3$ . For the remainder of the house (not included in this source zone), we opened windows to the outdoors. A tracer-decay test was conducted for the sealed-off zone, in parallel with the source-investigation experiment.

During cooking, researchers were present in the room. Once cooking was concluded, the room was vacated in some instances. Fans were left on during cooking but turned off during particle decay. Both indoor WCPC instruments were placed in the kitchen zone for these experiments. Three tests were conducted: (a) Toaster. Two pieces of bread were toasted on the <sup>3</sup>/<sub>4</sub> setting, which corresponds to "frozen/pastry." The toast popped after 4 min. (b) Boiling water in a kettle on the electric stove. Data from this test could not be quantitatively analyzed as particle data from outdoors were missing owing to a datalogging failure. (c) Frying an onion on the electric stove. For this test, the range-top burner was set at <sup>3</sup>/<sub>4</sub> power at 17:10 to begin heating pan. After one minute, oil was added. At 17:13, onions were added to the pan and for the next 7 minutes the onions were allowed to cook, with intermittent stirring. At 17:20 the stove was turned off and stirring terminated. The food remained on the hot burner throughout the remainder of the test.

#### 2.2.1.2 Site H1

The house was built in 1910, and has two levels. The top (main) level contains the kitchen, dining and living areas, and the main bedrooms and bathroom. Steps lead to a section slightly elevated above the main floor that was added on to the back of the house. This section has a study, a playroom, and a half bathroom. The lower level consists mainly of the garage, which underlies the main house. The lower level of the add-on has a spare room, and a foyer opening to the backyard and garage. See floor plan in Appendix C (Figure C.2). The volume of the upper level of the house is approximately 315 m<sup>3</sup>.

There are hardwood floors in all rooms except the bathrooms, which are tiled. The kitchen has a Wedgewood gas oven and stove (with continuous pilot lights) with a range hood that vents outdoors. The stove is used daily when the house is occupied and the exhaust hood is used occasionally. There is an exhaust fan in each bathroom. The gas water heater (with pilots) and the gas clothes dryer are located in the garage.

The house is outfitted with a forced air, natural gas furnace with an air filter. There is no air conditioning. The heating system is operated with a programmable thermostat that is designed – in the winter – to keep the house warm during the evening, night, and early morning while occupants are customarily present. The heating system fan operates on thermostat control.

Murphy's oil soap is used about once a month to clean the wood floors, but this cleaning did not occur during the monitoring period. Other surface-cleaning products include Windex, bleach, and dilute Simple Green. There is no known use of terpenes in this household.

Potential indoor UFP sources at this house include the following items:

- Natural gas cooking on the Wedgewood stove and oven, including continuously operating pilot lights.
- Electric kitchen appliances: microwave, toaster oven, espresso machine.
- Vented natural gas appliances: furnace, water heater (with a pilot light, in garage), clothes dryer (garage).
- Candles, which are routinely burned on Friday evening.
- Printer (in study)

The house is located on a residential street with light traffic. During an observation period on an early winter afternoon, the average flow on this street was less than a car per minute. However, only five lots away from this house is a busy street, where, at the same off-peak hour, we counted fifteen cars per minute. At least three other significant arterial roads are within 300-600 m away. The location would be characterized as urban residential.

The closest freeway is Highway 24, which is about 600 m to the southeast. Annual average daily traffic at nearest monitored location is 131,000 vehicles per day (Caltrans, 2007). Other potential nearby ambient sources included smoke from neighboring houses, which could be smelled in the backyard in the morning during the December observational monitoring period. Also, smoke from a nearby restaurant was visible from the house.

The outdoor monitoring enclosure (En 1) was placed in the back yard, to the south of the house, within 6 m of the house. It was protected from rain by being placed under a small wooden playhouse. The main indoor enclosure (En 2) was placed on the east side of the dining room, north of the kitchen. The second indoor particle monitor was placed on a shelf in the study, south of the kitchen.

Two adults (M and F) and two children (m  $\sim 9$  y and m  $\sim 5$  y) occupy the house. Occupants slept in the bedrooms on the top level, and also spent the majority of the time that they were indoors and awake on this main level. They went downstairs to access the backyard or the garage. Most of the potential sources were in the kitchen. Candles were burned in the living room.

Typically, the F adult leaves the house at 7:45 with the two children, and returns with them in the late afternoon (between 15:00 and 16:30). The M adult leaves later in the morning, and returns later in the evening. The children sleep approximately between 20:00 and 7:00; the adults sleep approximately between 11:45 and 6:30. Food is prepared in the morning and in the evening.

Observational monitoring at this site began on 12 December 2007 (16:04) and ended on 15 December (11:13). Observational monitoring data at this site is short of the 72-h target by almost 5 h owing to a data logging error associated with the indoor enclosure at the end of the monitoring period.

Indoor NO and outdoor RH data are missing at this site because the NO instrument was at the manufacturers for repair and the RH sensor was malfunctioning on the Q-Trak. The supplementary indoor WCPC (QMEc) was not in an enclosure. These conditions were the same as at house H0.

Manipulation experiments were conducted at this site on 17-19 and 24 December 2007. An experiment was conducted on 12 February 2009 to characterize emissions from continuous pilot lights on the oven and stove.

We sought to make air-exchange rate measurements while the house was configured according to two common ventilation modes. Since windows were typically closed during the winter (with the exception of the bathroom window), the modes chosen were: thermostat at 69 °F

(measured on 18 December) and thermostat at 64 °F (measured on 24 December) with doors and windows closed. These matched two common settings employed by the occupants. During both experiments, the house was empty of occupants (including researchers).

A CO<sub>2</sub> rebound experiment (18 December 2007) and pilot-light experiment (12 February 2009) conducted at this site indicated the presence of an indoor CO<sub>2</sub> source (pilot lights on the cook stove). Hence, the appropriate "background" level to be employed in air-exchange rate determinations is the indoor steady-state CO<sub>2</sub> level, rather than the outdoor level. Based on data from the rebound experiment, the background CO<sub>2</sub> level is 640-650 ppm when all exterior doors and windows are closed and interior doors are open ( $V \sim 315 \text{ m}^3$ ).

The source experiments were conducted on 19 December in the kitchen ( $V \sim 46 \text{ m}^3$ ), which was isolated from other indoor spaces as much as practical by closing doors and sealing them with tape. The outside door and windows from the kitchen were also closed. Two experiments were conducted: boiling water in a kettle on the stove and using candles.

During active source use, one researcher was in the room. Once the source was extinguished, the room was vacated. In the first experiment, water was boiled in a kettle (half full) on the gas stove. The gas was on medium high for 17 minutes (9:48-10:05); the range hood was off. For the second experiment, two candles (white, from Trader Joe's) were burned (after being ignited with a single match) for 25 minutes (12:13-12:38). The particle concentration and  $CO_2$  concentration decay were monitored for two hours after the candles were extinguished.

#### 2.2.1.3 Site H2

This two-story house was constructed in 1949. The top (main) level has a living room, family room, kitchen, two bedrooms and two bathrooms. (See floor plan in Figure C.3 in Appendix C.) The bottom level has a bedroom and a utility room, a garage, and a large storage closet. The estimated house volume is  $328 \text{ m}^3$  (upstairs  $V \sim 250 \text{ m}^3$ ). There is hardwood flooring in the living room and children's bedroom; tile flooring in the kitchen; carpeting in the master bedroom and bathroom, downstairs bedroom and family room; and an unfinished concrete floor in the utility room and garage. The kitchen has a pilotless gas oven and stove. The fan on the range hood does not function. We believe the water heater is located in the large storage closet downstairs. However, we were unable to confirm this, since the door was obstructed. Consequently, we also were not able to ascertain whether the water heater was gas fired or electric. There is a natural-gas clothes dryer in the utility room.

This house is ventilated through air leakage plus natural ventilation (i.e., through open doors and windows). During observational monitoring, none of the windows were opened. On occasion, the sliding glass doors at the rear end of the house were left open.

Among the identified potential sources of ultrafine particles were cooking with natural gas, a gas-powered clothes dryer, and a wall-mounted gas-fired furnace. The house has a toaster oven in the kitchen; however, it was not used during observational monitoring. No cleaning activities occurred during observational monitoring.

The house is located on a low-traffic residential street that is at a relatively steep incline. Beyond a vacant strip of land, approximately 50 m deep that lies at the rear of the house, there is a moderately busy urban street (approximately 25 cars per minute during a brief period of observation). The house is located approximately 500 m SW of Highway 13 and approximately 1000 m NE of Hwy 580. Note that heavy-duty diesel truck traffic is not allowed on either Hwy 13 or on the nearby portion of Hwy 580. The traffic volume for the nearest monitored location on Hwy 580 is 56,000 vehicles per day (Caltrans, 2007).

Two adults (M and F) and two children (m  $\sim$  5 y and m  $\sim$  3 y) occupy the house. The adult sister of the adult F spent one night at the house during observational monitoring, and the mother of the adult M visited on a few occasions. The sister slept in the bottom floor room during her night at the house; otherwise, it appeared that the family spent little if any time downstairs. Very little cooking occurred in this house; when it did, the cooking activity was simple (fried egg, boiled carrots, etc.). The family spent many evenings away from the home.

Observational monitoring at this site commenced on 7 February 2008 (16:01). The computer in the outdoor enclosure overheated at this site on 8 February at 14:30 and data collection was interrupted then. On 10 February (13:04), the system was restarted and monitoring resumed, but it had to be interrupted on 11 February (9:24) to conduct the scheduled manipulation experiments. Observational monitoring was completed with two additional days after the manipulation experiments were executed, starting 13 February (15:27) and ending on 15 February (16:54). The outdoor instruments (En 1) were positioned on the outer deck. The main indoor instruments (En 2) were placed in the family room upstairs. The supplementary WCPC was placed downstairs just outside the bedroom (En 3). Indoor RH data are missing at this site owing to a sensor malfunction on one of the Q-Traks.

Manipulation experiments were conducted on three days: 11-13 February. For the airexchange rate determinations, we configured the house according to two common ventilation modes. We focused on these two conditions: (a) the entire house was well-mixed and could be considered as one zone, (b) the upstairs and downstairs were isolated from each other, e.g. as a result of a vertical temperature gradient. On 11 February an air-exchange rate measurement was conducted by considering the upstairs and downstairs living areas as one well-mixed zone, and on 12 February the AER experiment was conducted for the upstairs zone only. During both experiments, the house was unoccupied.

For the first experiment, the exterior doors and windows were closed and  $CO_2$  was mixed throughout living areas in both bottom and top floor. The sampling instruments were sited in the same positions as for observational monitoring (outdoors and in family room). The supplemental indoor  $CO_2$  monitor (T85a) was positioned in the upstairs hallway. The  $CO_2$  release occurred over about 90 minutes and the decay was monitored for about 35 minutes. For the second measurement, exterior doors and windows were again closed and  $CO_2$  was only mixed in the upstairs level of the house. Monitoring of  $CO_2$  was done in the family room (LI-COR), in the upstairs hallway (T85a) and in the downstairs bedroom (T85b).

A Pkd experiment was conducted on 11 February using the ordinary procedures. During this experiment, the main instrument enclosures remained as for observational monitoring, outdoors (En 1) and in the family room (En 2). A supplemental indoor  $CO_2$  monitor (Q-Trak) was placed in the upstairs hallway. The third WCPC was placed in the master bedroom.

Two source characterization tests were conducted at this site on 13 February. For the first, the oven was used, but without any food being cooked. For the second, potatoes were cooked in a covered pot on the stove. In these experiments, we isolated the kitchen area (kitchen plus living room plus family room,  $V \sim 140 \text{ m}^3$ ) from the remainder of the house. Bedroom doors were shut and sealed and bedroom windows were opened to the outdoors.

For the first test, the oven was turned on 10:55-11:08 with the temperature slowly increasing. Then, for 30 minutes, the oven was on at the target temperature of  $350 \,^{\circ}$ F. Particle and CO<sub>2</sub> concentration decays were monitored for about 90 minutes after the oven was turned off at 11:38. For the second test, a stainless steel pot with water and two potatoes was placed on the stove. The pot was approximately 30 cm in diameter and had a lid. The stove was turned on to

75% of its maximum setting and operated for thirty minutes (13:30-14:00). Particle and  $CO_2$  concentrations were monitored during this time and for 50 minutes thereafter. During the source tests,  $CO_2$  monitoring occurred in the family room, in the upstairs hallway and in the downstairs bedroom. The WCPCs were positioned in the family room, in the downstairs bedroom, and outdoors.

# 2.2.1.4 Site H3

This single-story house was built in 1928. As illustrated in the floor plan (see Figure C.4, Appendix C), the front door opens into a living room, which adjoins a dining room. On one side of the dining room are the kitchen and utility room. On the other, is a corridor, which opens into the two bedrooms and the bathroom. The airy utility room has a large dog-flap in the door leading from it to the backyard.

There is hardwood flooring in all rooms except the bathroom and kitchen, which have vinyl floors. The utility room has an unfinished wood floor. The kitchen has a Wedgewood gas oven and stove with continuously operating pilot lights and with no range hood. The stove is used every day when the house is occupied. There is a gas water heater (with pilot) and furnace (pilotless) in the storage room beneath the main living area. The utility room houses a clothes washer and electric dryer and the refrigerator. The family has two pets – a dog that travels freely around the house and backyard and a chinchilla that lives in the child's bedroom.

The house is equipped with a central, forced-air, natural-gas-fired furnace. The recirculating fan operates only when heat is actively being supplied. The thermostat does not work, so the system is turned on and off manually by the occupants. There is a wall-unit air conditioner in the child's bedroom.

The house volume is  $200 \text{ m}^3$  (170 m<sup>3</sup> with the "backroom" or utility room excluded). The outdoor monitoring enclosure (En 1) was placed in the back yard (to the southeast of the house), within 10 m of the house. It was protected from rain by being placed under a tarpaulin in a cage also used for weight training. The main indoor enclosure (En 2) was placed on the north corner of the living room, which lies just northwest of the kitchen. The second indoor enclosure (En 3, with the supplementary WCPC) was placed in the kitchen, close to the utility room door.

Among the potential sources identified at this house site are these:

- Gas cooking with the Wedgewood stove and oven with continuous pilot lights.
- Electric appliances: microwave, tea kettle, clothes dryer.
- Vented natural gas appliances: furnace, water heater (with a pilot light) both in underhouse storage room.

The house had no known use of terpenes. Also, there was no fireplace, no use of candles or incense, no air cleaners, and no portable heater use.

The house is located on a street with light to moderate traffic; 5 cars per minute were observed in the early afternoon. The house is situated 120 m east of Hwy 580 along a section where heavy-duty (diesel) truck traffic is not permitted. The annual average daily traffic on the freeway at the nearest monitoring site is 167,000 vehicles per day (Caltrans, 2007). The house also is sited on the edge of a large park or open space.

Three people occupy the house: two adults (M and F) and one child (m  $\sim 11$  y). Typically the adult F cooks in the morning and then leaves the house around 8:30 and returns around 19:00. The boy is home-schooled by the adult M, and the two typically remain in the house during the day, with breaks that take them out of the house for varying lengths of time and without a simple set pattern.

Observational monitoring commenced 13 March 2008 (13:19) and ended 16 March (18:00). At the start of observational monitoring, instrument QMEb was replaced with QMEa; consequently, QMEa was not included in the side-by-side monitoring period at this site. Data for outdoor relative humidity are not available owing to a fault in the RH sensor of a Q-Trak unit.

Manipulation experiments were conducted 19-21 March 2008. The air-exchange rate tests (20 March) were conducted while the house was configured according to these ventilation modes: (i) exterior doors and windows closed, interior door between utility room and kitchen closed; and (ii) utility door open, kitchen window open. During both experiments, the house had no occupants (including researchers). Instrument positioning was as follows: LI-COR in living room, T85a in master bedroom, T85b in kitchen.

The pilot lights experiment conducted at this site indicated the presence of an indoor  $CO_2$  source (pilot lights on the cook stove/oven). Hence, the appropriate "background" level to be employed in air-exchange rate determinations is the indoor steady state  $CO_2$  level (rather than the outdoor level). This steady-state level varies in relation to how much of the interior volume is coupled to the kitchen (e.g., it is lower when all interior doors are open, and higher when the bedrooms are sealed off). Based on data from an unoccupied period, the background  $CO_2$  is approximately 550 ppm when all interior doors are open ( $V \sim 200 \text{ m}^3$ ). When the door to the utility room is closed ( $V \sim 170 \text{ m}^3$ ), the background level in the living room remains the same; but in the kitchen it rises to 650-700 ppm.

We do not have data to assess background levels when doors to bedrooms and the bathroom are sealed off in addition to the door between the kitchen and utility room, which was a configuration employed during a source characterization experiment.

To characterize the influence of outdoor particles on indoor UFP levels (Pkd experiment, 19 March), the basic procedure was followed. Proper interpretation also requires we take into account the emission rate of particles by the pilot lights, which was determined by means of a pilot-light experiment conducted on 8 July 2008. Instrument enclosures were positioned in the same locations as during observational monitoring except that the Q-Trak CO<sub>2</sub> monitors were moved to the kitchen and master bedroom.

Source characterization experiments were conducted on 19 and 21 March 2008. Six tests were implemented to investigate emissions from these activities: (i) heating squash in a plastic bowl in the microwave oven; (ii) heating water in electric kettle; (iii) frying an onion on a gas stove burner; (iv) operation of the central forced-air furnace; (v) use of the electric clothes dryer; and (vi) use of an empty microwave oven.

For tests (i)-(iii), conducted on 21 March, the house was configured to isolate the source kitchen zone, along with the well-connected living room and dining room ( $V \sim 110 \text{ m}^3$ ). In this case, doors to bedrooms, bathroom and utility room were closed and sealed with tape. The exterior windows for the nonsource zone of the house were open. The central heat was turned off. Three fans were used for mixing CO<sub>2</sub> from dry ice, which was released from two locations in the source zone. Fans were turned off once it was determined that CO<sub>2</sub> was well mixed throughout the source zone and prior to the activation of sources. In test (i), squash was heated in a plastic bowl in a microwave oven 11:14-11:16 and monitoring continued thereafter until 12:17. In test (ii), an electric kettle was turned on at 12:19 and monitoring extended until 13:04. In test (iii), the stove was ignited at 13:44 and the burner turned to a medium flame. At 13:45, oil was added to the pan and at 13:46 a half onion, chopped, was added. Cooking continued for five minutes, with occasional stirring. At 13:51 the burner was turned off. Monitoring continued until 15:28, with the pan and onion remaining on the range.

For tests (iv)-(vi), conducted on 19 March 2008, the whole house was treated as one test zone, so that exterior doors and windows were shut, and all interior doors were open. In test (iv), the central heater was operated during 15:06-16:10 and monitoring continued until 16:30, after the heater was turned off. For test (v), the electric clothes dryer was operated 16:49-17:00 and for test (vi) the empty microwave was operated 17:01-17:15.

An experiment to characterize pilot light emissions was conducted on 8 July 2008. Briefly, in this experiment, we monitored indoor and outdoor particle concentrations over the course of a working day. During this time, with house largely vacant, we first had the pilot lights on, then off, and then on again.

#### 2.2.1.5 Site H4

This two-story house with an approximate volume of 386 m<sup>3</sup> was built in 1904. As illustrated in the floor plan (see Figure C.5, Appendix C), the lower level has a living room, family room, kitchen, utility room and bathroom. Upstairs are two more bedrooms, a study, a second bathroom, and a second utility room. There is tile flooring in the bathrooms, study and utility room; synthetic wood flooring in the kitchen; and hardwood flooring in the rest of the house. The kitchen has a pilotless gas oven and stovetop with a range hood that vents outdoors. The occupants reported that the range hood is used infrequently, about once approximately every two weeks. Each bathroom is equipped with an exhaust fan. Each utility room has a water heater and the downstairs utility room contains an electric clothes washer and dryer.

At some point in its history, the house had been broken into two separate flats (one upstairs and one downstairs). The flats were later recombined into a single household. As a result, the house has a separate ducted heating and air distribution system for each floor. The filters within both systems had been changed six months earlier to an Aprilaire Model 2200/2250/2400 filter (advertised as 99% efficient for 5 µm particles). The occupants reported that after the new filters were installed, they would often leave the air systems on continuously in recirculation mode. The upstairs system was continuously on for the duration of observational monitoring in the present study. The fan on the downstairs system only operated when the furnace was on. Occupants reported that generally only bathroom windows are opened during the heating season.

The downstairs water heater is an American Appliance model GBF433TN with a power rating of 33,000 BTU/h. It is a 40-gallon, insulated heater, powered by natural gas, and contains a pilot light. The upstairs water heater is a Reliance Model 501 with a 30-gallon capacity, and is powered electrically.

Potential indoor UFP sources in this house include cooking with the natural gas stove and oven, use of the toaster oven, and surface cleaning with a grapefruit-scented spray ("Method" allpurpose cleaner), which may contain terpenes.

The house is located very close to a freeway, I-580. The rear wall of the house is approximately 30 m away from the freeway edge. On this portion of I-580, heavy-duty diesel truck traffic is permitted. Traffic volume on I-580 at the nearest measurement point (milepost 45.15) averages 202,000 vehicles per day (Caltrans, 2007). The house lot also is sited at the corner of a lightly trafficked residential street and a moderate-use urban street.

Four people occupy the house: two adults (M and F) and twin brothers (~ 4 y). It appears that the majority of time for which all four members of the family are at home and awake is spent in the downstairs level of the house. The living room contained the television and the family room was set up with the children's toys. Meal preparation, eating, and laundry

activities all took place downstairs. Sleeping occurred in the rooms upstairs, and the occupants reported that bedroom doors were closed overnight.

Observational monitoring at this site was initiated on 9 April 2008 (15:55) and completed on 12 April (18:05). There were a few brief periods (10 min or less) of data loss during the observational monitoring period. The outdoor instrument enclosure (En 1) was placed in the backyard. The primary indoor enclosure (En 2) was positioned on the lower floor in the family room. The supplementary indoor enclosure (En 3) was placed in the upstairs study.

Manipulation experiments were conducted at this site during the period 14-17 April. Airexchange rate determinations were made on 15 April. On this day we conducted two air exchange rate experiments while the house was configured according to two common ventilation modes: (a) all doors and windows were closed, the upstairs central system was on, and the downstairs system was off; (b) the two bathroom windows were approximately 50% open, the upstairs central system on, and the upstairs bathroom exhaust fan on. The indoor  $CO_2$ concentration during decay was only monitored in the downstairs living room.

A Pkd experiment was conducted on 14 April. The exterior doors and windows of the house were closed. In this experiment, both upstairs and downstairs central forced-air systems were "off." The monitoring instruments were sited in the same locations as for observational monitoring, with the primary indoor enclosure (En 2) placed in the downstairs living room. Data from this experiment are not available for the supplemental WCPC (upstairs study) owing to an operator error (the data were not internally logged).

Two source characterization experiments were conducted on 17 April 2008. One focused on particle emissions from toasting bread, using the toaster oven. The second investigated the use of a scented cleaning product in the bathroom. For the first experiment, an effort was made to contain the particle emissions to the downstairs area, excluding the bathroom. So, all interior doors on this level were opened except that to the bathroom, which was closed and sealed with tape. The effective volume into which particles were emitted is estimated to be 184 m<sup>3</sup>. In this experiment, two pieces of bread were toasted in the toaster oven and then particle decay was monitored for almost two hours. The instrument location was unchanged from observational monitoring.

For the second source test, "Fantastic" antibacterial heavy-duty spray and "Method" grapefruit scented all-purpose cleaner were applied to surfaces inside the closed bathroom on the lower floor ( $V \sim 15 \text{ m}^3$ ). The main indoor enclosure was moved to inside the bathroom for this experiment to monitor particle and copollutant levels.

#### 2.2.1.6 Site H5

This large  $(420 \text{ m}^3)$ , two-story house was built in 1993. As shown in the house plan (see Figure C.6 in Appendix C), the lower level has the kitchen, dining room, two living rooms, as well as a spare bedroom and bathroom, and a utility room, which contains a washer and dryer. On the upper level are two bedrooms, two bathrooms, and a study.

The floors are carpeted in all rooms except the kitchen, which has vinyl floors, and the bathrooms, which have tile floors. All kitchen appliances are electrically powered. The stove has a range hood vent, which exhausts to the outdoors. The oven is separate from the stove, and does not have exhaust provisions. Bathrooms 1 and 2 have exhaust fans. There is a gas-fired water heater (with continuous pilot) in the garage and a furnace (pilotless) in a closet on the second floor. There is also a fireplace with gas pilot in the second living room.

The house is equipped with a forced air, natural gas furnace and compressor-based airconditioner (installed 2002 or 2003). The recirculating fan is programmed to turn on and off under thermostat control. The thermostat is located on the lower level. It is set to 62 °F (heat) and 82 °F (cooling) and manually adjusted for on-demand thermal conditioning when occupants are present. Supply and return vents are on the ceiling on the first floor and at floor-level on the upper floor. Their locations are marked on the house plan.

The air-handling unit is equipped with an electronic "high-efficiency air filtration system" (Carrier Model EACAAXCC0020). The rated dust-spot efficiency of this unit is 65% and it consists of an aluminum mesh prefilter followed by ionizing wires that produce negative ions and collector plates. Occupants informed research staff that the system has not been serviced during the three years that they have lived in the house, which likely means that the plates have not been cleaned during this time.

Among the potential indoor sources of ultrafine particles at this house were the following:

- Electric cooking appliances: stove and oven.
- Electric kitchen appliances: microwave, toaster oven, coffee pot, "Foreman" grill, electric kettle.
- Vented natural gas appliances: water heater (with a pilot light) in garage, fireplace, furnace (without pilot) in second floor closet. Since monitoring was done in the summer, neither the fireplace nor the furnace was used.
- Electric clothes dryer.
- Air fresheners in bathrooms (used while cleaning).
- Printer in study (use not monitored).
- Electric iron in bedroom.
- Hair dryer in bathrooms.
- Cleaning products containing terpenes.

Cleaning activities were reportedly conducted twice weekly, using the following products (only those suspected to contain terpenes are listed): Pledge lemon-scented furniture cleaner, Scrubble Bubble (lemon scented) bathroom cleaner, spray air fresheners. In addition the occupant routinely used perfumes, and the house consistently had a distinctly pleasant (synthetic) aroma. Candles and incense were not used at this site.

The house is located on a cul-de-sac with very low traffic frequency; no cars passed by during a brief (5 minute) observation period in the late afternoon. The house is 1.2 km south of freeway I-580 in Livermore. At the nearest monitoring point, average daily traffic on this freeway is 173,000 vehicles per day (Caltrans, 2007). Train tracks are located one block north of the house.

Two adults (M and F) live in the house. The M was away during the entire observational monitoring period. The F slept in the master bedroom and usually came in and exited through the door to the garage. On weekdays, she was away at work from ~ 8:00 to 17:00. On the last monitoring day, a Sunday, the F occupant left the house at around 15:00, after which the house was unoccupied overnight.

Observational monitoring was conducted over two periods. The first, about 40 h in duration, began on 25 June 2008 (17:48) and ended on 27 June (09:59). The second, about 60 h in duration, began on 27 June (18:30) and ended on 30 June (06:59). The gap occurred because of the detection of a low-flow reading on one of the WCPC instruments (QMEa, outdoors) during a routine check on 27 June. To ensure that data were reliable, the QMEa instrument was

taken out of service and replaced with the supplementary indoor unit (QMEd). Consequently, for the second period of observational monitoring, data on indoor particle levels are only available from one indoor monitor.

The outdoor NO monitor produced spurious, negative readings after 29 June 12:39; data from that monitor were excluded from analysis from this time to the end of the observational monitoring period.

The outdoor instrument enclosure (En 1) was placed in the backyard, within 10 m of the house. The primary indoor enclosure (En 2) was placed in a living room (LR 1), which is on the front side of the house. The supplementary indoor WCPC (En 3) was operated in the spare bedroom on the second floor for the first 40 h of observational monitoring.

During the observational monitoring period it was warm outside, and the air conditioner was used for cooling in the daytime. Occasionally (on 27 June and during two overnight periods) windows were opened and at that time the air handling system was turned off. During field checks, researchers noted the house was typically cooler indoors than outdoors.

Manipulation experiments were conducted from 30 June through 2 July 2008. Tests were conducted on 30 June and 2 July to measure air-exchange rates while the house was configured according to common ventilation modes from the observational monitoring period: (i) exterior doors and windows closed and ventilation system fan on (cool setting); (ii) same as (i) except ventilation system fan off; (iii) two windows open and ventilation system fan off. During all three experiments, the house was unoccupied. Additional ventilation modes (e.g., house closed and stove range-hood exhaust fan on) were investigated in parallel with the Pkd and source manipulation experiments. Mixing of injected  $CO_2$  was accomplished through the use of the central forced-air system, augmented by three portable fans and five ceiling fans. The  $CO_2$  instruments were sited as follows: T85a was in the kitchen; T85b was in the upstairs hallway; and the LI-COR was in LR 1. In the third test, the kitchen and living room windows were opened.

A CO<sub>2</sub> rebound experiment conducted at this site indicated the presence of a minor persistent indoor combustion source. This is likely one or both of the fireplace and water heater pilot lights. The LI-COR measured a rebound from 384 ppm to 400 ppm, i.e.  $\sim$  15 ppm difference.

A Pkd experiment was conducted with all exterior doors and windows closed and all interior doors open. During this experiment, the WCPC monitors were sited in LR 1 and outdoors. The  $CO_2$  monitors were located in LR 1 (LI-COR), in the kitchen (T85a) and in the upstairs hallway (T85b).

On 1-2 July, three source-characterization experiments were conducted. With variants there were a total of five tests: (i) cooking rice on an electric stove (a) with range hood fan off and (b) with range hood fan on; (ii) using an electric iron; and (iii) using cleaning products that are believed to contain terpenes (a) lightly and (b) heavily. For experiments (i) and (iii), the whole house was used as the test space; for experiment (ii), ironing was done in bedroom 3 ( $V \sim 22 \text{ m}^3$ ), which was closed off from the rest of the house.

For experiment (i), one cup of rice and two cups of water were cooked in a small pot with a lid with the burner on a medium setting. For part (a), cooking occurred from 10:48 to 11:22 and monitoring continued until 12:39. For part (b), cooking was repeated from 12:54 to 13:28 and monitoring continued until 14:25. The kitchen exhaust fan was off for (a) and on continuously throughout (b). Instruments were sited as in the Pkd experiment.

For experiment (ii), the test commenced at 15:04 with the iron turned on to its highest setting, with steam. For three minutes (15:06-15:09), ironing was performed including the use of the water spray feature for steam ironing. Afterward, the hot iron was left on the ironing board with its hot side exposed to air. Monitoring continued with the room vacant until 16:31. Instruments QMEa and T85b monitored particles and CO<sub>2</sub> respectively in the source bedroom; QMEc and the LI-COR monitored in the living room; and T85a monitored in the hallway outside the source bedroom.

For experiment (iii), cleaning took place in the kitchen on 2 July using products that are routinely employed for cleaning in this household. For (a), during 12:44 to 12:51, a glass cleaner was used on two tables, "Scrubbing Bubbles" was used on the sink; and "Pledge" was applied on the granite countertop. The paper towels used for cleaning were left, with residual product, in an open trash receptacle indoors. Monitoring continued until 15:15. Exterior doors and windows remained closed during this test. Test (b) commenced for another six minute period at 15:17 when cleaning was repeated but with a heavy use of terpene-based products. The kitchen and living room windows were opened when the house was vacated at 15:24 and monitoring continued until 16:47. Instrument deployment for this experiment was the same as in the Pkd experiment.

#### 2.2.1.7 Site H6

This two-story house  $(V \sim 314 \text{ m}^3)$  was built in 1996. The configuration has a living room, kitchen, dining room, study and bathroom on the first level, and two bedrooms, a bathroom and a study on the second level (see Figure C.7 in Appendix C). The floor is carpeted in the living room and bedrooms; the kitchen, dining room, bathrooms and downstairs study have tile flooring. The kitchen has a pilotless gas range and an electric oven. The range hood filters air and returns the exhaust back into the kitchen. There is an exhaust fan located in the downstairs bathroom. Two water heaters and furnace are located in a shed behind the house. The water heaters have pilot lights. The gas-fired furnace is pilotless. The house has no clothes washer or dryer.

This house has central heating, but the system was not used during this study. The occupants ventilated the home naturally. The windows opened most frequently included one within each bedroom, one in the upstairs bathroom and one in the kitchen. The windows in the upstairs spaces were often left cracked open.

Potential indoor sources of UFP at this site include cooking (gas range and electric oven), use of an electric toaster, and use of a candle. During observational monitoring, cleaning was done using "Method" grapefruit scented all-purpose cleaner, which may contain terpenes.

The home is located on a medium-trafficked residential street. To the east, 200 m away, is an arterial road with heavy urban traffic. A daycare facility is located next door, so traffic in front of the home increases during the child drop off time in the morning and pick up time in the late afternoon. The home is located approximately ~ 800 m E of I-80 and ~ 1000 m NE of the I-80 and I-580 interchange. Heavy-duty diesel traffic is allowed on both roadways. The nearest monitoring station on I-80 has a reported annual average daily traffic count of 294,000 vehicles per day (Caltrans, 2007).

Three adults (2 M, 1 F) occupy the home. The occupants did not follow a regular schedule, and were often up into the early hours of the morning. A significant amount of cooking occurred in this home and generally involved frying. A rice cooker was also used on a few occasions, but did not appear to be a particle source.

Observational monitoring commenced on 28 September 2008 (12:33) and ended on 1 October (14:11). The primary indoor enclosure (En 1) was positioned on the lower level of the house, in the living room. The primary outdoor instrument package (En 2) was sited in the side yard immediately adjacent to the house. The supplemental WCPC (En 3) was located in the upstairs study.

Manipulation experiments were conducted on 27-28 September and 2 October 2008. Air-exchange rate determinations were made in association with the Pkd experiment on 27 September and then again in a more focused investigation on 28 September. A  $CO_2$  rebound experiment was also conducted on 28 September.

For the CO<sub>2</sub> rebound experiment, the indoor CO<sub>2</sub> levels were measured at 540 ppm at 8:32 on 28 September. The windows and doors were opened for ten minutes, beginning at 8:40, which caused the indoor CO<sub>2</sub> level to drop to 417 ppm. The windows and doors were then closed and the house was vacated to allow for a rebound, with monitoring occurring from 8:55 to 9:45. An observed increase in CO<sub>2</sub> level during the rebound period suggested the possibility of a persistent indoor source of CO<sub>2</sub>. Because such a source might be associated with unvented combustion that could emit ultrafine particles, a follow-up investigation was conducted on 26 February 2009. The two main results from the later investigation were (a) that the cooking appliances in H6 do not have continuous pilot lights; and (b) that no rebound in CO<sub>2</sub> level was observed indoors when both upstairs and downstairs portions of the house are first thoroughly flushed with outdoor air. We suspect that the rebound observed in the first investigation (28 September) resulted from inadequate flushing of the upstairs level, with subsequent transport of CO<sub>2</sub> to the lower level from the residual elevated level upstairs.

For the direct air-exchange rate measurement on 28 September, the house was configured with four open windows, one in each bedroom, a third in the upstairs bathroom, and the fourth in the kitchen. These windows were opened to about 50% of their maximum. Carbon dioxide was released and mixed throughout the whole house. Monitoring was done in the living room (En 1), beginning at 10:40 and continuing for just over an hour.

A Pkd experiment was conducted on 27 September 2008 (19:06-21:40). The house was configured with exterior doors and windows closed and interior doors open. Instruments were in the same locations as used for observational monitoring.

Two source-characterization experiments were conducted on 2 October 2008. For both, the emissions activity was in the kitchen/dining room/living room area. The source zone ( $V \sim 142 \text{ m}^3$ ) was isolated from the remainder of the house by closing interior doors and opening exterior windows for the upstairs bedrooms and bathroom and the downstairs bathroom and study. Carbon dioxide was monitored in the living room and particle concentrations were measured in the living room, the upstairs study and outdoors. During active source use, researchers remained in the room, but with minimal activity. Once the source activity was terminated, the house was vacated.

For the first test, the source was four tea candles, which were burned in the living room for 20 min, beginning at 11:55. Monitoring continued until 14:40. For the second test, a medium-sized onion was fried in a Teflon-coated pan using ~ 30 ml (1/8 cup) of canola oil. The stove burner was set to half of its maximum at 15:02. The onion was added a minute later and cooked with occasional stirring for the next 12 minutes. At 15:15 the burner was turned off. Monitoring continued until 17:48.

# 2.2.2 Monitoring in Schools

# 2.2.2.1 Site S1

This classroom is located in an older building on the school site. The building is single story and the classrooms in the building are arrayed in two rows, each served by a hallway. Ventilation for the building is provided entirely by natural ventilation and infiltration.

For the classroom we monitored, the north side adjoins an enclosed open courtyard and has seven windows opening onto it plus a door that has a window between it and the ceiling. The east and west walls are shared with other classrooms. The south wall adjoins a hallway and the main entrance of the classroom opens into this hallway. The hallway is itself naturally ventilated, by means of an array of windows.

Monitoring at this site took place in June. At this time of the year, the teacher regulates the temperature by opening the door or one or more windows to the outdoor courtyard. Door opening for temperature control occurred during the observational monitoring period; however, window opening did not.

Figure C.8 (Appendix C) shows the layout of the classroom, which has an estimated interior volume of ~ 290 m<sup>3</sup>. The floor is tiled and was partially covered with an area rug of dimensions 2.4 m × 3.2 m. The door dimensions are approximately 1 m × 2 m. Each of the seven single-paned windows to the courtyard measures 2.8 m (H) × 1.1 m. Each of these windows has two panes that can be opened, with each operable pane measuring 0.4 m wide by 0.8 m high. The window closest to instrument enclosure was cracked open during the entire study period to allow passage of the instrument power cord.

During observational monitoring the main indoor instrument enclosure (En 1) was positioned near the northeast corner of the room. The outdoor enclosure (En 2) was placed in the nearby courtyard, within a meter of the building. The supplementary indoor WCPC (En 3) was placed in the hallway, about 2 m from the classroom entrance.

Potential indoor sources of UFP identified at this site include cleaning activities. On occasion, the classrooms and hallway are mopped with a pine-oil based cleaner. The rug in the classroom is occasionally vacuumed. The tile floors are swept daily, after the school day is over, at about 3 PM. Other possible sources include a laser printer, white-board markers, and an overhead projector.

With regard to traffic, the school is located at an elevation of 100-150 m above sea level. As such, it is removed somewhat from the heart of the urban East Bay area, which is mainly concentrated at elevations less than 50 m above sea level. The school site is on a residential street with low traffic overall. The local traffic increases substantially for brief periods at the start and end of the school day when children are dropped off or picked up by parents or other caregivers.

A spot check was conducted one afternoon of the traffic density on the two streets closest to S1. We recorded an average of less than one car per minute between 14:15 and 14:35. At the end of the school day, shortly after 15:00, this number rose to 4-5 cars per minute. By 15:10-15:15 the traffic subsided to 1-2 cars per minute.

The school is approximately 200 m south of a moderately busy street. The nearest freeway is 500 m to the east. Annual average daily traffic on this freeway at the nearest monitoring station is between 60,000 and 80,000 vehicles per day (Caltrans, 2007). We observed no school buses coming to the school.

The students in this classroom are a mix of third- and fourth-graders. On a typical day, the teacher (F, 40-60 y) enters between 7:00 and 8:30. She occasionally uses the printer. The door between the classroom and the hallway is opened when the teacher arrives at the start of the

day and remains open until the teacher leaves. Later, the custodian may reopen the door for cleaning. In the morning before school begins and in the afternoon after school ends the teacher may spend time either inside or outside of the classroom. These changes in occupancy were not monitored. Instead, they are recorded as "occupied by teacher," which is believed to be the dominant state. When children were in the classroom during the observational monitoring period, a researcher in the classroom recorded student and teacher occupancy.

During the afternoon (between 1500 and 1800), the custodian (M, 20-40 y) came into the room to clean (sweep, vacuum, and/or mop the floor). Researchers were not customarily present to observe and record the exact times at which these activities were conducted. He reported to researchers that he did not mop in the classroom during the 4-day observational monitoring period and that he vacuumed occasionally and swept daily. He did mop in the hallway outside the classroom.

One or two assistant teachers (F, 20-50 y) were present intermittently during observational monitoring. At full occupancy, the classroom has 23 students (6-9 y). Additional students were present on one day.

The school day starts at 8:35 and ends at 15:05. There is a ten-minute break between 9:00 and 10:00 and then recess 10:30-10:45. Lunch occurs 12:20-13:05. The transitions are times of high activity as students clean up their work and exit or enter the room. It is also a time when the hallway tends to be occupied. On one of the observational monitoring days, 5 June (Thursday), students were away for much of the day on a field trip.

Observational monitoring was conducted at this site from 2 June 2008 (15:40) until 6 June (15:00). One of the particle monitors (QMEc) became flooded during the first day and consequently was removed from service for repair. Consequently, no supplementary WCPC data are available from the start of observational monitoring until 3 June (8:30). A pump malfunction on this instrument also resulted in the loss of supplementary UFP data after 5 June (19:09).

Three other occurrences during observational monitoring caused a loss of some data. First, data for outdoor  $CO_2$ , CO, air temperature and relative humidity were intermittently lost because of lapses in the performance of the datalogger. Second, a few hours of indoor  $CO_2$ , CO, and air temperature data (4 June 14:27-16:49) were lost. Third, we do not have direct (researcher) observation of occupant activity for the period 08:00-11:20 on 6 June. The researchers were unable to be present at the field site on this day and so instead the activity information was approximated based on discussion with the teacher plus knowledge of habitual activities.

Manipulation experiments were conducted on 6, 10 and 11 June 2008. Instrument positions during the manipulation experiments were substantially the same as during observational monitoring except that the Q-Trak normally in the classroom enclosure was moved to the hallway. The supplemental WCPC did not function properly owing to a flow error.

When needed,  $CO_2$  was released from dry ice at multiple locations. During the release, classroom doors and windows were closed. Doors and windows were also closed during the outdoor particle penetration and source characterization experiments. Hallway windows (from the hallway to outdoors) were closed during all experiments. The classroom was unoccupied during all manipulation experiments.

Air-exchange rate determinations were made on 6 June. For a first experiment, the door to the hallway was opened after the  $CO_2$  release was completed. In response,  $CO_2$  levels were observed to quickly drop (17:14-17:24) in the classroom and become elevated in the hallway until levels between the two were equilibrated. For a second experiment, the decay in  $CO_2$  levels

was monitored (18:20-21:20) with the doors and windows of the classroom closed. There was no detectable indoor  $CO_2$  source at this site.

A Pkd experiment investigating the effect of outdoor particles on indoor levels was conducted on 10 June 2008. For this experiment, doors and windows were closed and the classroom air filtered with two portable HEPA fan-filter units. However, the  $CO_2$  concentration in this experiment behaved in an unexpected manner, confounding analysis. After the room was vacated and closed, even with the  $CO_2$  source removed,  $CO_2$  levels continued to fluctuate rather than decaying smoothly as expected and as observed at previous sites. Particle rebound with door and windows closed was monitored 16:54-17:56.

A source characterization experiment was conducted to investigate UFP generation from use of a pine-oil based cleaner on 11 June 2008. A solution was prepared by diluting a quarter cup of a pine-based cleaner into one gallon of water, which corresponds to a 1:64 dilution, as recommended in the directions on the product container. Approximately 10% of the solution was delivered to the floor during mopping, and the floor area mopped was approximately 60 m<sup>2</sup>. The material safety data sheet for the product indicates that it is 8-10% pine oil. From this information, we estimate that the mopping activity delivered about 0.7 g of pine oil to the floor. Mopping occurred during 16:26-16:43 and monitoring continued until 17:55.

#### 2.2.2.2 Site S2

The school site for classroom S2 was the same as for classroom S1. Classroom S2 is on the second story of a newly built building on the school site. The S2 classroom has a mechanical air handling system (forced air, heat and cooling), which contains a particle filter. Each classroom in the building has its own mechanical unit. Commonly, when temperatures are favorable, the system fan is on even when the thermostat is off so that no heat or cooling is delivered. However, the teacher also reported that she turns it off occasionally when she is disturbed by the sound. Four air supply vents were located on the ceiling, along the midline. A return vent was placed near the thermostat, in the northeast corner.

The floor plan of the classroom is depicted in Figure C.9 (see Appendix C). The classroom volume is  $\sim 240 \text{ m}^3$ , and it is rectangular in shape. The flooring is vinyl. The north and west sides adjoin a basketball court and have windows opening onto it. The east wall is shared with another classroom. The south wall adjoins a courtyard, and the main entrance door opens onto it. The room has five windows, each with a height of 1.4 m.

During observational monitoring, the indoor enclosure (En 1) was placed in the northwest corner of the room. The outdoor enclosure (En 2) was placed in the same location as for S1, i.e. in the outdoor courtyard adjacent to that classroom, which is 30-40 m away from S2. The supplemental indoor WCPC (En 3) was placed in the cafeteria, which is located near the main entrance of the school site. The cafeteria is about 15-20 m away from the S2 classroom entrance, in a separate building. The classroom S2 is about 30 m from the edge of the school property, which is where vehicles pass most closely.

Potential indoor UFP sources are similar to those in classroom S1. With respect to cleaning, there is reported to be occasional mopping of the classroom with a pine-oil-based cleaner. The rugs in the classroom are occasionally vacuumed. There is a daily sweeping of the tiles, which occurs after the end of the students' day, i.e. after 15:00. Other potential indoor sources include a laser printer, white-board markers, and the heated surfaces from an overhead projector. A portable air cleaner was present in the classroom but reported to be rarely used; it was not used during this study.

The typical school day begins with the teacher (F, 30-40 y) entering the room around 7:45. In the morning before school starts and in the afternoon after students have left, she typically leaves the classroom door closed. However, she may frequently open the door to enter or leave to walk between her classroom and the courtyard or primary school building. These high frequency changes in occupancy were not recorded. Instead, the classroom was labeled as being "occupied" by the teacher during these intermittent entry and exit intervals. One or two assistants (F, 20-50 y) may support the teacher and so they are present in the classroom intermittently. At full occupancy, the classroom has 19 students (6-9 y); there are eleven first-graders, and eight second-graders. The second graders are away from the classroom for certain periods of time during the school day.

The classroom schedule is similar to that at S1. The school day starts at 8:35 and ends at 15:05. Recess is held 10:10-10:25 and lunch occurs 12:00-12:45. The level of activity is high during the periods shortly before students leave and shortly after they enter the classroom.

One or more windows and possibly the door to the courtyard are opened when the temperatures are warm; this occurs less often in the winter than during warm weather. The windows and door are typically closed overnight, but were apparently left open during the overnight period on 13-14 October.

Sometime during the afternoon (1500-1800), the custodian (M, 20-40 y) would come into the room to clean (usually sweeping and vacuuming). Our field researchers were not customarily present to observe and record the times at which these activities occurred. The custodian reported that he did not mop the classroom during the observational monitoring period, and that he vacuumed occasionally and swept daily.

The cafeteria is typically empty when classes are in session. It is occupied and heavily used before school and during recesses and lunch and for a short time after school ends. Recesses and lunch periods are staggered for different grade levels, so the cafeteria is occupied for a longer period for recess and lunch than the intervals listed for S2.

Observational monitoring was conducted in three periods. The first began on 6 October 2008 at 0900 and ended that night at 23:30 owing to an electrical short in the indoor enclosure. The instrument package had to be removed from the school site for repairs. The second period began on 9 October at 23:30 and ended on 10 October at 08:30 to allow for completion of the planned manipulation experiments. The third observational monitoring period started on 13 October at 08:20 and was terminated on 15 October at 09:50. During the third observational monitoring period, school personnel inadvertently unplugged the supplemental indoor WCPC on 14 October and again on 15 October, in both instances at about 7:30. In each case, researchers restarted the monitor at about 8:30. Hence, data from this location are missing for around 1 h on two mornings. One of the Q-Trak instruments was at the manufacturer for repair and, because of that, we did not monitor indoor CO levels.

Manipulation experiments were conducted at this site on 9-10 October 2008. The primary indoor (En 1) and outdoor (En 2) instrument enclosure packages were in the same locations for these experiments as for observational monitoring. Data from the supplemental indoor WCPC were not acquired.

As at S1, the classroom was treated as a single, well-mixed space during these experiments. The classroom door and windows were closed during the Pkd and source-characterization experiments. All manipulation experiments were conducted with the classroom unoccupied.

An air-exchange rate measurement was conducted on 9 October 2008. The classroom was configured according to a high airflow configuration commonly noted during observational monitoring: ventilation fan on (heat/cooling off), front door wide open, window opposite front door open about 5 cm.

A Pkd experiment was conducted on 9 October 2008. During the particle rebound and  $CO_2$  decay period (monitored from 16:07 through 23:00) the door and windows were closed and the ventilation system was off.

A source-characterization experiment was conducted on 10 October 2008 to investigate UFP emissions from a laser printer (HP model 1040). Printing occurred over a 20-minute period initiated at 10:08 and entailed printing 350 pages. Carbon dioxide levels were elevated before this time to enable simultaneous determination of the air-exchange rate. During the printing period, researchers were present in the room. Once printing was concluded, the room was vacated. The decay of  $CO_2$  and particle number concentration was monitored for about an hour after printing, terminating at 11:30.

### 2.2.2.3 Site S3

This school campus is approximately 50 years old, but the building containing S3 was built in the 1980s. The building contains three classrooms positioned around a small center room called a "pod" (see Figure C.10 in Appendix C). The pod contains storage space, a small counter and sink, and an electric stove, which teachers use occasionally. The windows within the classrooms are inoperable. Each classroom has two doors to the outside — one single door and one double door — plus one door to the pod. The ceilings of the classrooms are sloped, with a height of approximately 3.3 m in the rear of the room and 2.7 m in the front of the room. The room flooring is a combination of carpet and tile. The approximate dimensions of the room and building are as follows: floor area of the main classroom ~ 68 m<sup>2</sup> and of the entire building ~ 220 m<sup>2</sup>; volume of the main classroom ~ 205 m<sup>3</sup> and of the entire building ~ 640 m<sup>3</sup>.

The classroom has its own HVAC system. The system for S3 was also connected to the center pod, but operated independently of the other two classrooms in the building. The system for S3 is programmed to bring in a fixed percentage of outdoor air, and is equipped with a high-efficiency filter. The building maintenance personnel could not provide specific details on the type of filter within the system (other than to say that it had a "high efficiency" rating), or the percentage of outdoor air mixed in with the supply air. The thermostat allows for the system to be programmed by the teacher. A natural-gas furnace is located in the ceiling plenum.

Among potential indoor sources of UFP are wet cleaning activities and vacuuming. Wet cleaning is done daily for the tile portion of the classroom floor using a "Sundance neutral floor cleaner." The MSDS for this product describes it as a "clear, light orange liquid" with a "fresh odor." The active ingredient is nonylphenol ethoxylate (CAS 9016-45-9). The MSDS for "Sundance nonalkaline floor cleaner" (a concentrated product) indicates that this ingredient constitutes 3-5% of the product. The product "4 Sundance Neutral Floor Cleaner Ready-to-Use" is a 1:256 dilution of the "4 Sundance Neutral Floor Cleaner Concentrate." We do not know whether the product contains terpenes in addition to the nonylphenol ethoxylate.

Mopping also occurred daily in the cafeteria, where the supplementary indoor WCPC was located. In the classrooms, mopping generally occurred in the early evening, whereas in the cafeteria it occurred in the early afternoon. Vacuuming occurred daily on the carpeted section of the classroom. Other potential indoor sources include candles that were burned briefly to

celebrate student birthdays and the electric stove in the center pod that is used occasionally by teachers.

The school is located at the intersection of a medium-trafficked urban street and a light-trafficked residential street. The nearest freeway is 1.7 km to the west and carries annual-average daily traffic of ~ 180,000 vehicles (Caltrans, 2007). We observed no school buses coming to the school.

The students in the S3 classroom are second graders. On a typical school day, the teacher enters the classroom between 7:30 and 8:30 and turns on the heater when the room is cold. Half of the students (early birds) arrive at 8:30 and the other half (late birds) arrive at 9:30. Conversely, the early birds leave for the day at 14:00, while the late birds stay until 15:00. On Wednesdays, all the students arrive at 8:30 and leave at 14:00. The teacher generally leaves between 17:00 and 19:00. She leaves earlier in the afternoon when needed to attend a meeting at the district office.

Observational monitoring extended over three intervals. The first of these began on 20 October at 9:00 and ended at 15:21 the same day. The second interval began on 21 October at 8:30 and ended on 23 October at 7:34. The third interval began on 23 October at 15:17 and ended on 24 October at 8:30. The main indoor instrument enclosure (En 1) was placed in the northwest corner of the classroom. The outdoor enclosure (En 2) was placed within a fenced off and locked area on the north edge of the school campus. This location was about 100 m from the classroom, more distant than desired. That site was chosen because of security concerns. The supplemental WCPC (En 3) was placed at the south end of the cafeteria. The outdoor enclosure was located approximately 10 m from the medium-traffic street, while the classroom was located more than 100 m from the same street.

There was only one Q-Trak available during observational monitoring at this site, because the second had been sent to the manufacturer for repairs of faulty RH and  $CO_2$  sensors. The available Q-Trak was alternated between measuring indoor and outdoor air. Indoor air temperature and relative humidity data were continuously acquired by means of a HOBO temperature sensor, which was attached to the mast of the indoor enclosure.

Manipulation experiments were conducted over a three-day period beginning on 27 October 2008. On the first day, the goal was to conduct an air exchange rate experiment while the classroom was configured according to a common ventilation mode. The entire building was treated as a single zone, with all exterior doors and windows closed and internal doors open. However, with this strategy, we were unsuccessful at raising indoor  $CO_2$  levels above ~ 700 ppm, inadequate for a reliable tracer-gas decay experiment. Eventually, the classroom interior door was closed and the  $CO_2$  level was elevated inside the study classroom only, with subsequent decay monitored at that location. During the experiment, the thermostats were set so that neither heat nor cooling were provided by the ventilation systems, but the fans were on in each case as is commonly done by the respective teachers.

A second effort to measure air-exchange rate was undertaken on 27 October using an integral material balance approach. In this method, the amount of the tracer gas released must be measured along with the time-averaged increase in concentration above the natural background during the rise and fall associated with the release and its removal. For a conserved tracer, such as  $CO_2$ , the total mass released will equal the total mass removed by means of ventilation. Since the rate of removal by ventilation is the product of the unknown airflow rate and the measured net increase in concentration above the background level, the unknown ventilation rate can be determined from the  $CO_2$  release and the measured time-average concentration. In this

experiment, the entire building was again treated as a single, well-mixed zone, with exterior doors and windows all closed and internal doors open. Ventilation fans were on in all classrooms. During the period 18:15-19:40, 5.15 kg of  $CO_2$  was sublimated and mixed into the room air. The  $CO_2$  concentration was monitored during this time and until 20:05.

To investigate the influence of outdoor particles on indoor levels, we used an alternative approach to the Pkd experiment. The high ventilation rate made it impractical for us to significantly reduce the indoor particle levels using portable HEPA-filter units. So, instead, we first flushed the building with unfiltered outdoor air to raise the indoor UFP level. Then, the building was closed and the decay of particles monitored while it was unoccupied. From monitored data on indoor and outdoor particle levels we are able to glean some information about the penetration and decay of outdoor particles. During this experiment, which was carried out on the night of 27-28 October beginning at 20:05, the third WCPC was positioned in the central "pod" of the building. A second brief investigation of outdoor particle influence on indoor levels was executed on 29 October. In this experiment, conducted during 21:05-21:55, researchers simultaneously measured particle levels in outdoor air (instrument QMEc), in the supply air of the center pod (QMEd), and inside the classroom (QMEa).

Source-characterization experiments were conducted on 29 October to investigate emissions from a candle and potential emissions from use of the central heater. For the source experiments, the classroom was closed off from the other three rooms ( $V \sim 205 \text{ m}^3$ ). Three instruments were used for measuring CO<sub>2</sub>: the LI-COR measured levels in the main classroom while the two Q-Traks were deployed in the center pod and in the mirror classroom. Particle number concentrations were monitored in the main classroom, in the center pod, and outdoors. A set of four tests was executed. In the first, the heater was turned on for the period 16:32-17:00. Monitoring continued thereafter until 17:23. For the second test, a candle was lit at 18:00 at the teacher's desk and allowed to burn until 18:10. The decay of particle number concentration and CO<sub>2</sub> level was monitored for this test until 18:38. The third test was a repeat of the first, carried out because the janitor could be heard vacuuming in the building during the first test. In this case, the heat was operated during 18:52-19:21 and then the decay of CO<sub>2</sub> and particle number concentration was monitored until 19:56. The fourth test was a repeat of the candle experiment of the second test, except that all classroom doors to the center pod were closed. The candle was burned 20:21-20:28 and particle concentration decay was monitored until 20:55.

#### 2.2.2.4 Site S4

The site of classroom S4 is more than 100 y old. Additions and remodeling have been completed at intervals since then until the present. The teacher reported that ~ 25 y ago new sprinklers and wiring were installed. Maintenance personnel reported that there was a retrofit in 2002, when the doors and lighting were changed. Since then, there has been trouble with light ballasts failing. Such a failure occurred dramatically in the classroom toward the start of the observational monitoring period, leading to a strong burning smell in the room.

Figure C.11 (Appendix C) presents a floor plan of the classroom, which is carpeted, roughly rectangular in shape and has an interior volume of ~ 240 m<sup>3</sup>. The classroom is on the second floor of the school building. The west side of the classroom adjoins the outdoor school courtyard and has four windows opening onto it. The north wall is shared with another classroom. The south wall adjoins a custodial closet. The east wall adjoins the hallway and includes the entry-door. The four windows are identical, with dimensions 1.6 m × 0.84 m, and with a pane that can be opened measuring 0.43 m high.

Maintenance personnel at the school reported that the classrooms are not mechanically ventilated. Heating is via a radiant hot water system and the boiler room is located below classroom S4. One boiler serves the entire two-story building (18 classrooms). The boiler furnace is vented and has a pilot light. It cycles on and off according to thermostatic control. During our study period at this site, the interior of classroom S4 felt warm (relative to the hallway) whenever the door to the hallway was closed. We presume that this is a result of indirect heating from the warm boiler room below.

Teachers can turn on a fan associated with the radiant heater system at will. Doing so blows air through a filter and over the heated pipes. The fan was not used during the observational period, and the teacher reported that he "almost never" uses it. Radiant heat comes in passively through the vent even when the classroom system is off. There is no thermostat in the classroom.

The teacher opens and closes windows and the door frequently for ventilation and for regulating the air temperature inside the room. Windows are opened when the daytime temperatures rise, usually around 11:00. Windows are used less often in the winter and more often in the summer.

Potential indoor UFP sources include cleaning, a laser printer, an overhead projector, white-board markers, and the boiler under the classroom. Wet mopping was done in the hallways using "Sundance neutral floor cleaner." We observed that the hallways occasionally smelled of terpenes. For restrooms, Pinesol mixed with a "lemon odor" liquid was used on the floor. The reported frequency of vacuuming inside the classroom was once per week; vacuuming did not occur during the observational monitoring period. The room was equipped with a laser printer (HP model 1320). An overhead projector was used in the morning at the start of class, and it is conceivable that its heated surfaces might contribute as a UFP source.

The school is situated on an urban residential street with low traffic, except at the beginning and end of the school day when children are driven to school or picked up in private motor vehicles. The school is less than 100 m south of a moderately busy street and 200 m north of a busy arterial road that feeds to a freeway. The site is approximately 1 km to the east of a heavily used freeway, which has annual average daily traffic of ~ 180,000 vehicles per day (Caltrans, 2007). We observed no school buses coming to the school.

On a typical school day, the teacher (M, 50-60 y) enters around 7:00 and may occasionally use the printer. He initially works in the classroom with the door closed. At ~ 7:45, the classroom door is propped open and it usually remains so until the teacher leaves for the day at 17:00-18:00. An assistant teacher (F, 20-30 y) is present on most days and leaves with the children at the end of the school day. The classroom serves a  $5^{th}$  grade population and has 26 enrolled students.

The school day starts at 8:30 and ends at 15:05, except on Wednesdays when the day ends at 13:45. For this classroom, recesses occur 9:40-9:55 and 13:45-13:55 and lunch occurs 12:00-12:45. During 14:35-15:05, students either have a library period or physical education and so they are outside of the classroom then. The transitions are associated with high levels of activity. Transitions are also a time when the hallway tends to be full of students walking to or away from their respective classrooms.

Observational monitoring began on 4 November 2008 at 6:45 and ended on 7 November at 6:45. Election Day, 4 November, was associated with high levels of activity throughout the school including a greater flux of students in and out of the classroom than is usual.

The primary indoor enclosure (En 1) was placed inside the classroom along the south wall. The supplementary indoor WCPC (En 3) was placed just outside the classroom door, in the second floor hallway (directly across from the elevator). A row of classrooms opened into the same hallway, and it was frequently full of children between classes. The monitoring enclosure for outdoors (En 2) was placed in the courtyard, less than 2 m from the building. We did not have access to a secure outdoor power source at this school site. The custodian turned off the outdoor monitoring equipment at the end of each day, between 23:00 and 01:00. The instruments were restarted when the school opened in the morning,  $\sim 6:45$ .

Manipulation experiments were conducted on 7 November and 10 November 2008. The instrument enclosures were in the same positions as for observational monitoring. The outdoor  $CO_2$  monitor (Q-Trak) was moved to the hallway location so that there was a second indoor  $CO_2$  measure and no measurement of outdoor  $CO_2$  levels.

The rectangular classroom ( $V \sim 240 \text{ m3}$ ) was treated as a single, well-mixed space. The door and windows were also closed during the Pkd and source-characterization experiments. Hallway windows (from the hallway to outdoors) were closed during all experiments. The classroom was unoccupied during all experiments. Further, there was no ventilation system in operation during any of these tests.

Six air-exchange rate determinations were conducted at this site, some independently and others in conjunction with the Pkd or source-characterization experiments. For each experiment, the state (open or closed) of the door and one or more windows was changed.

A rebound experiment conducted at this site indicated the presence of an indoor  $CO_2$  emissions source. So, too, did overnight  $CO_2$  levels. The average indoor  $CO_2$  levels for the period 0200-0600 on the three monitored days were 560, 690, and 500 ppm, respectively. Based on this evidence, we used 530 ppm as the baseline  $CO_2$  level in air-exchange rate determinations.

Three separate determinations of air-exchange rate were made on 7 November. The respective configurations were these (default condition is all windows and the door closed): (1) one window open; (2) three windows open; (3) three windows and the door open. Three additional determinations were made in conjunction with the Pkd (4 and 5) and source-characterization (6) experiments on 10 November. These configurations apply: (4) all windows and the door closed; (5) door open; (6) all windows and door closed.

A Pkd experiment was conducted on 10 November 2008. The period was divided into four intervals. The first interval followed the standard particle rebound experimental protocol. In other intervals, natural-ventilation conditions to the classroom were altered to observe the effect. During the first interval, 11:58-13:20, the rebound of particle concentration and decay of  $CO_2$  was observed with the door and windows closed. During a second interval, 13:20-13:40, the door to the hallway was opened to observe any additional rebound in response to increased air exchange between the classroom and the hallway. The third and fourth periods occurred at the outset of the experiment, before filtration was initiated. For the third period, 9:20-10:00, the classroom door and windows were all opened to allow maximum air-exchange and to raise the indoor particle concentration to a level close to that outdoors. For the fourth period, 10:00-10:30, the door and windows were closed and particle decay observed.

A source-characterization experiment was also conducted on 10 November 2008, investigating the laser printer in the classroom (HP model 1320n). The experiment was conducted with the door and windows closed. During printing, one researcher was present in the room. Once printing was concluded, the room was vacated. A first set of 125 pages was printed

during the period 14:20-14:25. A second set of 11 pages was printed during 14:26-14:35. Particle levels and  $CO_2$  decay continued to be monitored until 16:00.

# 2.2.2.5 Site S5

The school campus for site S5 was built in the 1970s. The classroom we studied opens into a central courtyard and only shares a wall with one other classroom. The wall separating the two classrooms was added some years after the school was built; the two rooms were originally one classroom.

Apart from its ceiling configuration, the classroom has a simple rectangular geometry with an approximate floor area of 90 m<sup>2</sup> and an approximate volume of 258 m<sup>3</sup> (Figure C.12 in Appendix C). There are two external doors in the classroom that, when both opened, provide cross-ventilation for the room; however, the doors were rarely opened at the same time during observational monitoring.

The classroom has its own HVAC system. It is programmed to bring in a fixed percentage of outdoor air, and is equipped with a high-efficiency filter. The building maintenance personnel could not provide specific details on the type of filter within the system (other than to say that it had a "high efficiency" rating), or the percentage of outdoor air mixed in with the supply air. Upon investigation, researchers ascertained that the ventilation fan within the system turns on automatically at 6:45 and turns off at 18:15. The thermostat allows for the temperature to be programmed by the teacher. A natural-gas furnace is located in the ceiling plenum. When the furnace was used for the first time that winter (20 November, during observational monitoring), the room was filled with a "burning hair" smell. This odor did not recur on repeated use.

In addition to the furnace, other potential sources include a rarely used printer, the overhead projector (heated surface) and cleaning activities. Wet mopping was done daily by the janitor using "Sundance neutral floor cleaner" on the tile portion of the classroom floor. Vacuuming was normally done daily on the carpeted section of the classroom, but it only occurred on 19 November during observational monitoring.

The school is situated at the intersection of a heavily traveled urban street that feeds into an interstate highway and a moderately traveled urban residential street. The school is 200 m W of another heavily traveled urban street, and is ~ 500 m E of the nearest freeway, which carries an annual average of ~ 180,000 vehicles per day (Caltrans, 2007). Vehicle traffic immediately in front of the school was heaviest during drop-off and pick-up times. We observed no school buses coming to the school.

On a typical school day, the teacher enters at approximately 7:50, turns on lights, and turns on the heater if the room is cold. Students enter at 8:30. On one day of observational monitoring, there was a choir performance in the morning, and so the students did not enter until 9:37. The observational monitoring at this site occurred during the parent-teacher conference period. Consequently the students left by 13:45 every day, which is earlier than usual. The teacher would have one or two conferences with parents in the early afternoon and would leave by 16:15 every day. A morning recess was held 10:30-10:50 and the lunch period was held 11:50-12:40. The custodian would clean in the classroom on some days during the late afternoon through the evening, 17:00-21:00.

Observational monitoring at this site commenced on 18 November 2008 at 8:57 and ended on 21 November at 8:02. The indoor instrument enclosure (En 1) was positioned at the center of the classroom's west wall, south of the student seating area, and immediately next to

the carpeted area where the children would occasionally sit. The outdoor enclosure (En 2) was placed in an inner courtyard shared by four classrooms. The enclosure was situated in a corner of the courtyard that was diagonally opposite the door to the S5 classroom. The door to another classroom was approximately 0.5 meters from the outdoor enclosure. The supplementary WCPC (En 3) was placed next to double doors in a hallway within a different building on the school campus. The entrance to the school library was located approximately 3 meters from this third enclosure.

Manipulation experiments were conducted at this site on 21, 24 and 25 November 2008. All experiments were conducted using the full volume of the classroom, not including the closet  $(V \sim 258 \text{ m}^3)$ . Experiments were conducted to determine the air-exchange rate for several ventilation configurations. An experiment was devised to explore the HVAC particle filter efficiency and the amount of outdoor air supplied by both the HVAC system and through infiltration. The furnace was tested as a possible UFP source.

Air-exchange rate measurements using tracer-gas decay with carbon dioxide were conducted on 21 November. A series of five decay experiments was conducted on this day, with changes to the classroom configuration, as follows: (1) all doors and windows closed, heat at 61  $^{\circ}F$  (= 16  $^{\circ}C$ ), fan on auto; (2) all doors and windows closed, heating system fan off; (3) windows closed, fan off, door cracked open; (4) windows closed, fan off, door wide open; (5) all doors and windows closed, heat set to 72  $^{\circ}F$  (= 22  $^{\circ}C$ ), fan on "auto." During these experiments, the primary indoor and outdoor instrument enclosures were in the same positions as for observational monitoring; the supplemental WCPC was placed in the adjacent classroom.

On 24 November 2008, an experiment was conducted to investigate the influence of outdoor particles on indoor UFP levels and also to investigate whether there was a persistent indoor source of  $CO_2$ . In this experiment, the indoor Q-Trak and the supplemental WCPC were positioned so that they sampled from the air as supplied at the ventilation system register. During the interval 16:00-17:00, the room was flushed with outside air by opening all doors and windows. Then, for two hours, beginning at 17:00, all doors and windows were closed. The heat was off, but the system fan was on for approximately the first hour (17:00-18:15) until it automatically switched to the "unoccupied" mode.

Later in the evening of 24 November, an air-exchange rate decay experiment was repeated with the same configuration as (1) from 21 November. That is, all doors and windows were closed, the heat was set at 61 °F (= 16 °C) and the fan was on "auto."

Three final manipulation experiments were conducted on 25 November 2008. Two of these were focused air-exchange rate experiments that included an effort to characterize the degree to which the ventilation system brought in outside air rather than simply recirculating room air. To characterize the split, the supplementary WCPC and the indoor Q-Trak were positioned to sample directly from the supply vent. The first tracer-gas decay was conducted 17:30-18:15 with all doors and windows closed, the heat off, and the fan on "auto." The second decay was conducted 21:03-21:23 with the windows closed, the ventilation system fan "on" and the classroom door cracked open.

The third experiment on 25 November followed the Pkd protocol, aiming to characterize the penetration and persistence of outdoor particles in contributing to indoor UFP levels. The room was configured with all doors and windows closed and the ventilation system fan off.

### 2.2.2.6 Site S6

Classroom site S6 was on the same school campus as site S3. The school campus is approximately 50 years old, but the building containing S6 was built in the 1980s. Classroom S6 is contained within a building that has two classrooms and these only share one short wall. The north-facing wall of the classroom has seven tall windows that can be opened. The windows were not opened during observational monitoring, but they have an imperfect seal that should result in a large rate of infiltration. The classroom has a basic rectangular shape with half carpet and half tile flooring; see Figure C.13 (Appendix C) for the floor plan. The classroom floor area is  $\sim 82 \text{ m}^2$  and the volume is  $\sim 297 \text{ m}^3$ .

The classroom is outfitted with two wall-mounted heating and air conditioning units (Fujitsu, Halcyon brand). These units offer the following features: heating, cooling, dehumidification, "air cleaning", fan-on mode (recirculation), and quiet mode (low air flow). Air conditioning is accomplished using a split system design with refrigerant R410A. Heating is accomplished using a reverse refrigeration cycle. Air cleaning is accomplished in effect through electrostatic precipitation. The teacher has the freedom to decide which of these features to apply. During observational monitoring, the system was used for heating, cooling and air cleaning in both quiet and regular air flow modes. Manufacturer information is available at <a href="http://www.fujitsugeneral.com/wallmounted.htm">http://www.fujitsugeneral.com/wallmounted.htm</a>. (This level of information about ventilation systems is not available for the other school sites.)

The tile portion of the classroom floor was wet-mopped daily, using "Sundance neutral floor cleaner." Mopping also occurred daily in the cafeteria, where the 3<sup>rd</sup> WCPC was located. In the classrooms, mopping generally occurred in the early evening, whereas in the cafeteria it occurred in the early afternoon. Vacuuming was done daily on the carpeted section of the classroom. Another potential indoor source is the laser printer. See the description of S3 for information about the proximity of the study site to traffic.

On a typical school day, the teacher enters the classroom between 7:30 and 8:30. The students in this classroom are second graders. Half of the students (early birds) arrive at 8:30 and the other half (late birds) arrive at 9:30. Conversely, the early birds leave for the day at 14:00, while the late birds stay until 15:00. On Wednesdays, all students arrive at 8:30 and leave at 14:00. The teacher generally leaves by 17:00.

Observational monitoring at this site was conducted in four intervals: (i) 2 December 8:15 to 3 December 8:36; (ii) 4 December 14:04 to 5 December 8:30; (iii) 5 December 12:25 to 6 December 8:30; and (iv) 8 December 8:43 to 15:48. The first gap occurred because no research staff was available to be present in the classroom on 3 December owing to illness. The second gap arose because, during morning check on 5 December, the USB cable was inadvertently dislodged from the primary indoor enclosure computer and logging was not restarted until the noon check. The third gap arose because 6-7 December 2008 were weekend days.

During observational monitoring, the primary indoor instrument enclosure (En 1) was placed near the center of the classroom. Because of security concerns, the outdoor enclosure (En 2) was placed within a fenced off and locked area on the north side of the school campus, about 100 m from the classroom. This location was not ideal because it was located approximately 100 m from the classroom, but was chosen because of security concerns. The supplemental WCPC (En 3) was placed in the cafeteria, at the south side.

Manipulation experiments were conducted on 4 December and 6 December 2008. All experiments were conducted within the full volume of the classroom ( $V \sim 297 \text{ m}^3$ ).

Air-exchange rates were determined for three ventilation configurations using CO<sub>2</sub> tracergas decay. In the first configuration (4 December), the door and windows were closed and the wall units were left on the teacher's setting (heat, 64 °F = 18 °C, with fan on "recirculation" mode). The second and third configurations were tested on 6 December. For the second configuration, the wall units were set at higher heating levels (76 °F = 24 °C and 74 °F = 23 °C, respectively). The windows were all closed, but the front door was propped open. For the third configuration, the wall units were shut off and the door closed.

The Pkd experiment was carried out in conjunction with the first air-exchange rate determination (4 December 2008).

We conducted a limited experiment to test the emissions from the classroom printer. In this test, 34 pages were printed. (The small number was to comply with a request from the teacher.) During the Pkd and source characterization experiments on 4 December, the WCPC instruments were in the same positions as for observational monitoring.

Additional experiments were conducted to further characterize possible indoor sources on 6 December. With the doors and windows closed and the wall units off, a researcher vacuumed the carpet 12:13-12:23. Particle monitoring continued until 13:35.

An investigation of possible ozone generation by the wall unit and the potential for such ozone to react with terpenes and thereby produce ultrafine particles was carried out. In this experiment, all windows and doors were closed and the HAC units were set to "heat" (76 °F = 24 °C) with the fan set to "auto" and the "air clean" mode engaged. Monitoring was conducted under these conditions 13:54-14:30. While continuing to monitor, "fresh linen" scented Lysol disinfectant was sprayed five times on a room counter top (14:34-14:35).

For all of the source characterization experiments, the main indoor and outdoor instrument enclosures were situated in the same way as they were for observational monitoring. The supplementary WCPC and the indoor Q-Trak were positioned such that they sampled from directly beneath one of the wall units.

# 3. RESULTS AND DISCUSSION: HOUSES

This section of the report presents the research findings based on monitoring and analysis at the seven house sites investigated in this project. The section is organized in three parts. In the first part, an overview of the observational monitoring results is presented. The aim of this subsection is to provide an overall perspective on what was observed in the seven homes so that the more detailed presentation can be seen in context. The second subsection presents the detailed findings obtained from the seven house sites. This subsection presents the following information: time-series measurement data on indoor and outdoor particle number concentrations; average particle number concentrations indoors and outdoors, sorted according to the occupancy condition of each household resident; air-exchange rate determinations for each household; evaluation of the impact of outdoor particles on indoor UFP levels; characterization of indoor emission sources; analysis of exposure of household occupants to indoor UFP during the observational monitoring period, including an apportionment of those exposures to particles of outdoor origin and major indoor sources.

In the third subsection, we gather and synthesize findings from each of the houses across the full set of study variables, exploring what general insight and understanding of UFP concentrations and exposures in houses can be gleaned from the new data collected and analyzed in this study.

Because of the very large quantity of data gathered in this study, some of the information is presented in appendices. See Appendix D for time-series plots of observational monitoring data from the seven house sites. Cumulative probability distributions of the one-minute resolution particle number concentrations, sorted according to occupancy status of the buildings, are presented and discussed in Appendix E.

### 3.1 Overview of the Research Findings from Observational Monitoring in Houses

Measurement results from observational monitoring at the seven houses are summarized in Table 3.1. The first few rows of this table indicate when (i.e., during which season and year) and for how long monitoring was conducted in each house. The average particle number concentrations presented in the subsequent three lines are of central interest for this research project. Here, time-averaged particle number concentrations from the three WCPCs operated at each site are reported. The primary and supplementary indoor monitors are labeled "PN\_in1" and "PN\_in2," respectively, and the outdoor monitor is labeled "PN\_out." The remaining nine rows report concentrations of other species either measured on-site or at a central monitoring station and basic environmental parameters (temperature and wind speed).

In this table, we have not reported the time-averaged outdoor  $CO_2$  level nor the indoor or outdoor carbon monoxide levels because we deem the measured results for these species to be largely noninformative. The carbon dioxide levels measured by the Q-Trak in ambient air exhibit a much broader range of values than is plausible; we suspect that the instrument has a temperature sensitivity making it unsuitable for ambient concentration measurements. With respect to CO, the manufacturer specifications for the Q-Trak indicate that its accuracy is the larger of 3% of the reading or 3 ppm. At all sites, the average CO monitoring result was less than 3 ppm.

Table 3.2 and Figure 3.1 present summaries of the average particle number concentrations measured at each house site during observational monitoring. These data are sorted according to occupancy conditions in the household. To be included in the "occupants awake" class, all of the occupants of the household must have been at home and awake at that

time. (The one exception to this categorization is for site H5, where one of the two regular occupants was away for the whole observational monitoring period. In this case, we used only the occupancy status of the other occupant to sort the data.) Likewise, to be included in "occupants asleep" and "occupants away" all of the household occupants must have been in that particular status. The percentage of total monitored hours that could be so classified ranged from 40% at H6 to 100% at H5 and averaged 67% across all seven houses.

| Parameter (units)               | HO    | H1    | H2    | H3    | H4     | H5               | H6    |
|---------------------------------|-------|-------|-------|-------|--------|------------------|-------|
| Season/year                     | F '07 | F '07 | W '08 | W '08 | Sp '08 | S '08            | F '08 |
| Duration (h)                    | 140   | 67    | 92    | 77    | 74     | 101              | 74    |
| PN_in1 (1000 cm <sup>-3</sup> ) | 10.2  | 21.0  | 11.0  | 28.1  | 11.9   | 3.7              | 15.6  |
| $PN_in2 (1000 \text{ cm}^{-3})$ | 9.2   | 27.4  | 8.9   | 35.0  | 2.6    | 4.1 <sup>b</sup> | 20.8  |
| $PN_out (1000 \text{ cm}^{-3})$ | 9.9   | 23.2  | 17.9  | 18.9  | 18.0   | 5.5              | 11.0  |
| CO <sub>2</sub> _in (ppm)       | 590   | 1130  | 617   | 800   | 537    | 445              | 549   |
| T_in (°C)                       | 17.4  | 19.4  | 19.7  | 19.0  | 21.3   | 24.3             | 24.7  |
| T_out (°C)                      | 11.8  | 8.3   | 13.2  | 12.8  | 17.4   | 18.8             | 18.6  |
| O <sub>3</sub> _in (ppb)        | 5.1   | 3.1   | 3.2   | 2.4   | 1.4    | 6.1              | 3.3   |
| O <sub>3</sub> _out (ppb)       | 21.7  | 0.4   | 17.4  | 25.8  | 16.8   | 30.4             | 17.3  |
| NO_in (ppb)                     | Na    | na    | 9.8   | 58.8  | 18.6   | 0.9              | 37.3  |
| NO_out (ppb)                    | 3.0   | 51.9  | 20.5  | 7.5   | 13.0   | 0.8              | 22.9  |
| WS (m/s) <sup>a</sup>           | 3.3   | 1.5   | 2.6   | 4.2   | 2.1    | 2.4              | 2.1   |

**Table 3.1.** Time-averaged measurement results inside and outside houses for the observational monitoring periods.

<sup>a</sup> Measurements from central monitoring station.

<sup>b</sup> Average based on the first 40 h of monitoring, only.

House occupancy is important from two complementary perspectives. First, the activities of occupants influence the emissions of UFP from indoor sources. Occupant behaviors also affect building factors that determine the penetration and persistence of particles of outdoor origin as well as the dynamic behavior of particles emitted from indoor sources. A simple example of this latter point is the opening and closing of windows, which influences the air-exchange rate of the building. Second, occupancy and exposure are intimately coupled: there is no exposure to particles indoors when there is no one present indoors. Quantitatively, the exposure of an individual to indoor particles can be evaluated as the time average of the indoor particle concentration *during the time that the individual is indoors*, multiplied by the duration of that presence.

A few key points are made here based on the overall average particle number concentrations. First, considering the total observational periods, the primary indoor monitor (In1) produced an overall average across the seven sites of  $14.5 \times 10^3$  particles cm<sup>-3</sup>, which corresponds to 97% of the overall average of the outdoor monitoring results ( $14.9 \times 10^3$  particles cm<sup>-3</sup>). However, the ratio of In1/Out across sites varied from a low of 61% at H2 to a high of 149% at H3.

The overall comparability of time-averaged indoor and outdoor UFP levels masks a centrally important feature that is relevant for understanding how levels relate to human exposures. The observed indoor levels of ultrafine particles can be decomposed into two primary contributions. One is the penetration and persistence of particles that originated

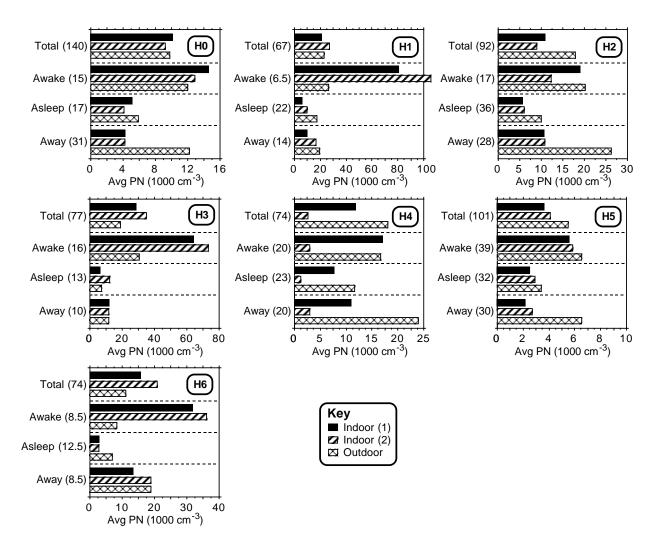
outdoors. The second results from indoor emission sources of UFP. The total observational period represents a composite of contributions from these two sources. However, generally people do not occupy their houses 100% of the time. Many indoor sources of ultrafine particles result from occupant activity. When people are awake at home, the contributions from indoor sources can cause the indoor UFP levels to rise above the outdoor value. Conversely, when people are away or asleep, occupant related source contributions are diminished and, owing to particle loss processes, indoor levels diminish to below the outdoor levels. These points can be observed in the other columns of Table 3.2 and can also be seen graphically in Figure 3.1. For example, the average In1 for the "occupants awake" period is  $33.2 \times 10^3$  particles cm<sup>-3</sup>, which is almost twice the overall average of the outdoor monitoring results during these times  $(17.2 \times 10^3)$ particles cm<sup>-3</sup>). Considering individual sites, the ratio In1/Out during "occupants awake" varied from a low of 86% at H5 to a high of 385% at H6. All of these results are considerably *higher* than the corresponding numbers for the total observational period. Conversely, considering the "occupants away" periods, the average In1 value is  $9.0 \times 10^3$  particles cm<sup>-3</sup>, which is only about half of the outdoor monitoring results during these times  $(17.1 \times 10^3 \text{ particles cm}^{-3})$ . Considering individual sites, the ratio In1/Out during "occupants away" varied from a low of 34% at H5 to a high of 97% at H3. All of these results are considerably *lower* than the corresponding numbers for the total observational period.

|      | <b>Total Observational Period</b> |                  |      |       | Occupants AWAKE |       |      |      |
|------|-----------------------------------|------------------|------|-------|-----------------|-------|------|------|
| Site | In1                               | In2              | Out  | Hrs   | In1             | In2   | Out  | Hrs  |
| H0   | 10.2                              | 9.2              | 9.9  | 140.3 | 14.6            | 12.9  | 12.0 | 15.0 |
| H1   | 21.0                              | 27.4             | 23.2 | 67.2  | 80.6            | 105.9 | 26.7 | 6.5  |
| H2   | 11.0                              | 8.9              | 17.9 | 92.3  | 19.0            | 12.3  | 20.3 | 16.6 |
| H3   | 28.1                              | 35.0             | 18.9 | 76.7  | 63.8            | 73.2  | 30.3 | 15.8 |
| H4   | 11.9                              | 2.6              | 18.0 | 74.2  | 17.0            | 3.0   | 16.6 | 19.8 |
| H5   | 3.7                               | 4.1 <sup>a</sup> | 5.5  | 100.7 | 5.6             | 5.8   | 6.5  | 39.0 |
| H6   | 15.6                              | 20.8             | 11.0 | 73.6  | 31.6            | 36.2  | 8.2  | 8.5  |
| Avg. | 14.5                              | 15.4             | 14.9 | 89.3  | 33.2            | 35.6  | 17.2 | 17.3 |

**Table 3.2**. Summary of particle number concentrations (in units of 1000 particles  $cm^{-3}$ ) measured during observational monitoring at the house sites.

|      | Occupants ASLEEP |      |      |      | Occupants AWAY |      |      |      |  |
|------|------------------|------|------|------|----------------|------|------|------|--|
| Site | In1              | In2  | Out  | Hrs  | In1            | In2  | Out  | Hrs  |  |
| H0   | 5.1              | 4.1  | 5.9  | 17.0 | 4.2            | 4.2  | 12.2 | 30.6 |  |
| H1   | 5.4              | 9.4  | 17.4 | 21.7 | 9.8            | 16.8 | 20.3 | 13.7 |  |
| H2   | 5.6              | 6.0  | 9.9  | 35.8 | 10.9           | 10.9 | 26.1 | 28.4 |  |
| H3   | 6.1              | 12.3 | 7.0  | 13.2 | 11.6           | 11.5 | 12.0 | 9.9  |  |
| H4   | 7.7              | 1.2  | 11.7 | 23.2 | 11.0           | 2.9  | 23.9 | 20.1 |  |
| H5   | 2.6              | 2.9  | 3.4  | 31.5 | 2.2            | 2.7  | 6.5  | 30.2 |  |
| H6   | 2.8              | 2.7  | 6.8  | 12.5 | 13.5           | 19.0 | 19.0 | 8.5  |  |
| Avg. | 5.0              | 5.5  | 8.9  | 22.1 | 9.0            | 9.7  | 17.1 | 20.2 |  |

<sup>a</sup> Average is based on the first 40 h of observational monitoring, only.



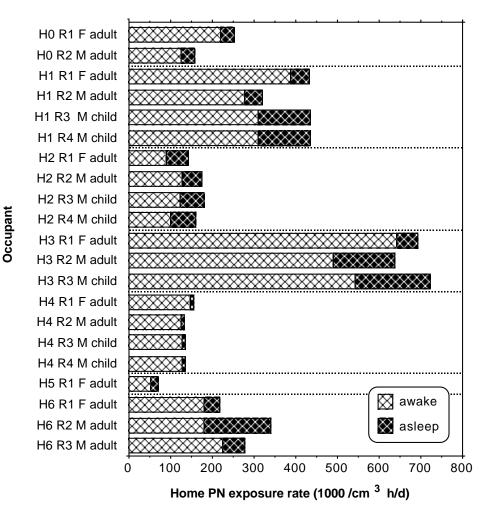
**Figure 3.1.** Time-averaged particle number concentrations measured during observational monitoring at the seven house sites, H0-H6. At each site, the results are presented for each of the three monitors (primary indoor = "Indoor (1)"; supplementary indoor = "Indoor (2)"; and "Outdoor"). The upper set of three bars represent the overall averages for the full observational monitoring period. The other sets of bars represent averages for periods sorted according to the occupancy status of household residents. The number of hours for each condition is indicated in parenthesis. Note that the horizontal axis varies among sites.

In this study, we collected basic time-resolved occupancy information about the household residents, specifically having them record the times when each resident was (a) at home awake, (b) at home asleep, and (c) away from home. Combining this information with the time-resolved measurements of particle number concentration, we can estimate the daily-average household exposure experienced by each of the study subjects. This determination is an estimate because we are not actually measuring the exposure concentrations encountered by the occupants but instead using the information from the stationary microenvironmental indoor monitors as proxies. We have made these estimates, using a fairly simple rubric for assigning measurement results to exposure concentrations. At each site and for each occupant, we used concentrations measured by the primary indoor monitor as the estimate of exposure concentration for the

"awake" period. For the "asleep" period, we selected one of three options, based on judgment about which best represented exposure conditions: (i) primary indoor monitor; (ii) supplementary indoor monitor; or (iii) average of the two indoor monitors. Details about this procedure are presented in §3.2.

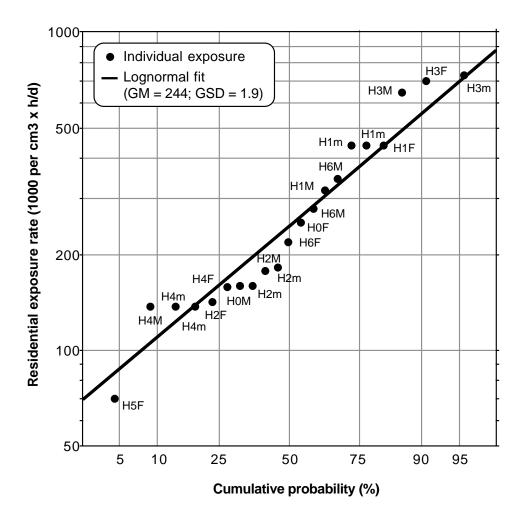
In Figures 3.2-3.3 and Table 3.3, we present a synopsis of the results.

Figure 3.2 is a bar chart showing the estimated household exposure rate for each of the 21 subjects. Each bar is subdivided into a segment representing the "awake" period and a second segment representing the contribution to exposure that occurs during sleep. Figure 3.3 shows the same information presented as a cumulative probability distribution. Table 3.3 presents numerical details of the exposure rate estimates and their components.



**Figure 3.2.** Estimated exposure rate for the 21 occupants in the seven households studied in this project, based on the observational monitoring period. The exposure rate represents the product of the average concentration during occupancy and the daily-average duration of occupancy. So, for example, exposure to an average concentration of  $15 \times 10^3$  particles cm<sup>-3</sup> for an average of 16 h/d while indoors at home would produce an estimated home exposure of 240 in units of (1000 cm<sup>-3</sup> × h/d). Each bar is divided into two sections, one representing the exposure accruing while at home "awake" and the other the exposure while "asleep."

A few key features of these results merit explicit mention. First, most of ultrafine particle exposure occurring in these homes — 79% on average — happens during the "awake" periods rather than during the "asleep" periods. The durations of each of the two states of occupancy are similar in these populations, averaging 8.2 h/d awake and 8.7 h/d asleep. The key difference is that the average indoor PN concentration during the period awake  $(28.0 \times 10^3 \text{ cm}^{-3})$  is 4× as large as that during the asleep period  $(7.0 \times 10^3 \text{ cm}^{-3})$ .



**Figure 3.3.** Cumulative probability distribution of individual average household exposure rates for the 21 occupants of the seven houses studied in this project, based on observational monitoring. These have a GM of 244 (1000 cm<sup>-3</sup> h/d) and a GSD of 1.9. The lognormal distribution with the same GM and GSD is plotted as the straight line. The arithmetic mean of this set of 21 exposure determinations is 297 (1000 cm<sup>-3</sup> h/d) and the standard deviation is 196 (1000 cm<sup>-3</sup> h/d). Each point is designated with an occupant code indicating house number (H0 — H6) and a gender/age indicator (M = male adult; F = female adult; m = male child).

A second noteworthy characteristic of these results is that the variation in exposure across households is much larger than the variation of exposure within households. For the five sites with more than two occupants per household, the relative standard deviation in exposure rate within the households varies from 6% to 22%. Considering the daily average exposure rate for

the occupants in each of the seven houses, the relative standard deviation is much larger, 75%, and the range is an order of magnitude, from  $70 \times 10^3$  cm<sup>-3</sup> h/d in H5 to  $686 \times 10^3$  cm<sup>-3</sup> h/d in H3.

A third interesting feature of the results is that there is not a clear difference among exposure rates by gender and age class (Table 3.4). The difference in averages between any pair of groups (maximum ~  $35 \times 10^3$  cm<sup>-3</sup> h/d) is considerably smaller than the standard error in the mean (~  $65-85 \times 10^3$  cm<sup>-3</sup> h/d); consequently, the differences are not statistically significant.

|           | Awake                    |       |                            |                           | Total |                            |                          |
|-----------|--------------------------|-------|----------------------------|---------------------------|-------|----------------------------|--------------------------|
|           | Exp. Conc.               | Time  | Exp. rate                  | Exp. Conc.                | Time  | Exp. Rate                  | Exp. Rate                |
|           | $(10^3 \text{ cm}^{-3})$ | (h/d) | $(10^3  \mathrm{cm}^{-3})$ | $(10^3 \mathrm{cm}^{-3})$ | (h/d) | $(10^3  \mathrm{cm}^{-3})$ | $(10^3 \text{ cm}^{-3})$ |
| ID        |                          |       | h/d)                       |                           |       | h/d)                       | h/d)                     |
| H0 R1 F   | 18.4                     | 11.8  | 217                        | 3.9                       | 8.7   | 34                         | 251                      |
| H0 R2 M   | 14.9                     | 8.3   | 124                        | 4.1                       | 7.9   | 32                         | 156                      |
| H1 R1 F   | 42.6                     | 9.1   | 389                        | 5.4                       | 8.8   | 47                         | 436                      |
| H1 R2 M   | 38.2                     | 7.2   | 277                        | 5.2                       | 8.5   | 44                         | 321                      |
| H1 r3 m9  | 50.9                     | 6.1   | 310                        | 10.7                      | 11.8  | 126                        | 436                      |
| H1 r4 m5  | 50.9                     | 6.1   | 310                        | 10.7                      | 11.8  | 126                        | 436                      |
| H2 R1 F   | 17.6                     | 5.1   | 90                         | 5.6                       | 9.3   | 53                         | 143                      |
| H2 R2 M   | 18.1                     | 6.9   | 126                        | 5.6                       | 9.3   | 53                         | 178                      |
| H2 r3 m3  | 19.1                     | 6.4   | 122                        | 6.0                       | 10.1  | 60                         | 182                      |
| H2 r4 m4  | 19.5                     | 5.1   | 99                         | 6.0                       | 10.1  | 60                         | 159                      |
| H3 R1 F   | 64.2                     | 10.0  | 644                        | 7.2                       | 6.9   | 50                         | 693                      |
| H3 R2 M   | 36.8                     | 13.3  | 489                        | 19.7                      | 7.6   | 149                        | 639                      |
| H3 r3 m11 | 59.3                     | 9.2   | 543                        | 19.7                      | 9.3   | 183                        | 726                      |
| H4 R1 F   | 15.9                     | 9.3   | 148                        | 1.2                       | 7.5   | 9                          | 157                      |
| H4 R2 M   | 15.7                     | 8.1   | 126                        | 1.2                       | 7.8   | 10                         | 136                      |
| H4 r3 m4  | 16.7                     | 7.6   | 126                        | 1.2                       | 8.6   | 11                         | 137                      |
| H4 r4 m4  | 16.7                     | 7.6   | 126                        | 1.2                       | 8.6   | 11                         | 137                      |
| H5 R1 F   | 5.6                      | 9.3   | 52                         | 2.4                       | 7.5   | 18                         | 70                       |
| H6 R1 F   | 24.7                     | 7.3   | 181                        | 5.3                       | 7.0   | 37                         | 218                      |
| H6 R2 M   | 19.4                     | 9.4   | 182                        | 17.4                      | 9.1   | 159                        | 341                      |
| H6 R3 M   | 23.1                     | 9.7   | 225                        | 7.3                       | 7.3   | 54                         | 278                      |
| Average   | 28.0                     | 8.2   | 233                        | 7.0                       | 8.7   | 63                         | 297                      |
| St. Dev.  | 16.6                     | 2.1   | 162                        | 5.7                       | 1.4   | 53                         | 196                      |

**Table 3.3.** Individual household exposure rates

| Table 3.4. | Average household    | l exposure rates by | gender and   | age class. <sup>a</sup> |
|------------|----------------------|---------------------|--------------|-------------------------|
|            | i i verage nousenone | · onpobulo futob of | Solidor alla | age erabb.              |

|                       |   | Average exposure rate (10 <sup>3</sup> cm <sup>-3</sup> h/d) |            |                  |                  |  |  |
|-----------------------|---|--|------------|------------------|------------------|--|--|
|                       |   | Total  |            |                  |                  |  |  |
| Age/gender group      | Ν | $\mu \pm \sigma$   | Std. Error | $\mu \pm \sigma$ | $\mu \pm \sigma$ |  |  |
| Female adult          | 7 | $281\pm215$  | 81         | $246\pm206$      | $35 \pm 17$      |  |  |
| Male adult            | 7 | $293 \pm 173$  | 65         | $221 \pm 132$    | $71 \pm 58$      |  |  |
| Male child (age 3-11) | 7 | $316 \pm 225$  | 85         | $234\pm164$      | $82 \pm 65$      |  |  |

 $^{a}\mu \pm \sigma =$  arithmetic mean  $\pm$  standard deviation.

# 3.2 Research Findings from Individual Houses

### 3.2.1. Site H0

Observational monitoring was conducted at house site H0 over a period of approximately six days, beginning on 28 November and ending 4 December 2007. The reader is referred to §2.2.1 for detailed background information about the house site. See also Appendix C, Figure C.1, which shows a floor plan. Summarizing briefly, this house has an internal volume of about 320 m<sup>3</sup> and is a conventional single-family, split-level house built in the 1930s with three bedrooms and two bathrooms. It is sited in the lower hills of Oakland, at an elevation of ~ 150 m, in a lightly trafficked neighborhood but in the middle of the metropolitan East Bay area of northern California. The nearest freeway is ~ 500 m to the east. The house has a detached garage, electric cooking appliances, and a central, vented, natural-gas-fired furnace. The house has two adult occupants, a female designated as H0 R1 and a male designated as H0 R2.

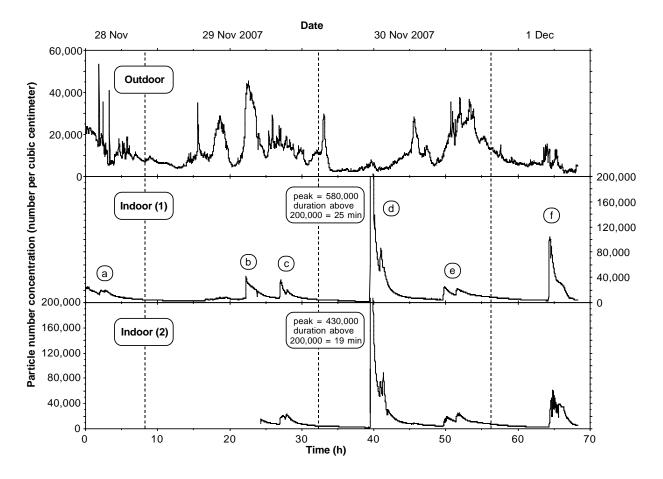
A summary of the time-averaged species concentration and cofactor parameter values was presented in Table 3.1 (§3.1). Of note, the average particle number concentrations measured during the entire observational monitoring period were 9.9 (outdoors), 10.2 (indoors primary), and 9.2 (indoors supplemental) in units of 1000 particles per cm<sup>3</sup>. The primary indoor monitor, also referred to as "PN\_in1" or "Indoor (1)," was sited in the downstairs study. The supplemental indoor monitor, also denoted "PN\_in2" or "Indoor (2)," was positioned in an upstairs bathroom near the central landing at the top of the stairs. The outdoor monitor was sited in the back yard, immediately north of the house.

Figures 3.4a (first half of the period) and 3.4b (second half) show the time-series of particle number concentrations measured by the three monitors. In these figures, the horizontal axis is labeled with elapsed time of monitoring, in hours, with zero corresponding to 15:45 on 28 November. The vertical dashed lines demark midnight and the upper horizontal axis is labeled with the calendar dates. The lower case letters in the middle frames are used to label peaks that correspond to indoor activities with associated episodic emissions of ultrafine particles. These labels are used for identification purposes in later efforts to characterize the indoor sources in four respects: (i) identifying, where possible, the activity or activities that appear responsible for the emissions; (ii) quantifying the source strength of the emissions event (total number of particles emitted; (iii) quantifying the rate of disappearance of indoor particles from indoor air after emissions are complete; and (iv) estimating the contribution of the indoor source event to the exposure of house occupants.

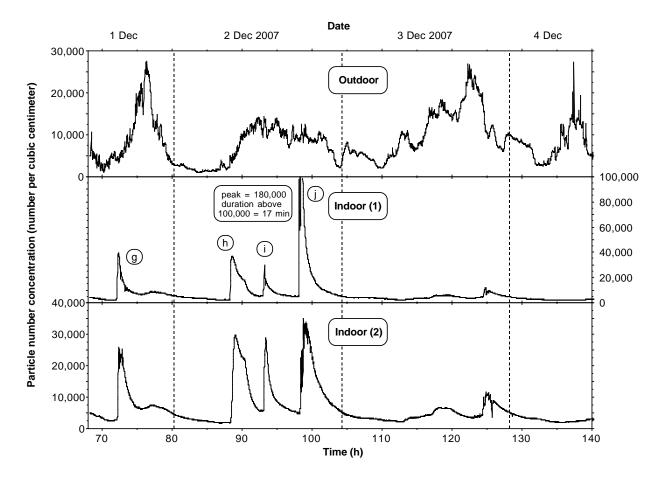
The outdoor concentrations at this site exhibit considerable time variability, spanning a range from ~ 1000 cm<sup>-3</sup> up to ~ 40,000 cm<sup>-3</sup> (with one brief excursion on 28 November to ~ 55,000 cm<sup>-3</sup>). There is a tendency for the levels to be higher in the afternoon and evening as compared with the overnight and early morning hours. This pattern is exhibited for all days monitored, but can be seen most clearly on 2 December. Overall, the average outdoor PN concentration for the hours 10:00-22:00 is  $13.3 \times 10^3$  cm<sup>-3</sup>, about twice the average level for the hours 22:00-10:00 (6.6 ×  $10^3$  cm<sup>-3</sup>).

By contrast, in looking at the traces for the indoor PN concentrations, the dominant impression comes from the labeled peaks — rapid rises and then slower decays — that are seen in both monitors. In total, ten of these episodes occurred during the six-day monitoring period. The highest indoor particle concentrations — by far — occur during these episodes. They are of sufficient magnitude and duration to contribute substantially to the time-averaged indoor particle concentrations (graphically proportional to the area under the traces). Furthermore, because their occurrence tends to coincide with occupancy, they contribute proportionally more to average

indoor *exposures* than they contribute to average indoor *concentrations*. However, even though the particle concentration spikes are associated with emissions from indoor sources, for much of the time, the indoor concentrations are dominated by the penetration into the building and persistence of particles of outdoor origin. These points are explored more thoroughly in the following pages.



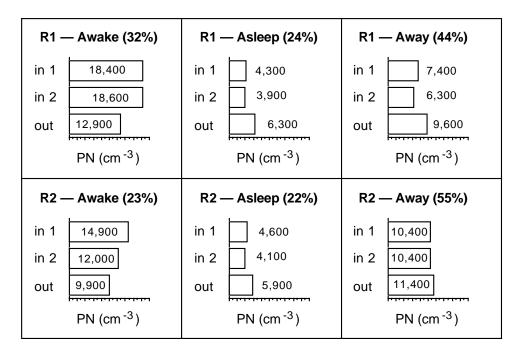
**Figure 3.4a.** Time-series of particle number concentrations during the first half of the observational monitoring period at house site H0. Monitor "Indoor (1)" was located in the downstairs study; monitor "Indoor (2)" sampled from a bathroom off of the upstairs landing. Note that the indoor and outdoor concentration scales are different. The supplementary indoor instrument was not successfully deployed until the second day of the observational monitoring period. Six peaks in indoor particle number concentration are labeled with lower-case letters, a – f. These labels are used to identify the peaks for subsequent discussion.



**Figure 3.4b.** Time-series of particle number concentrations during the second half of the observational monitoring period at house site H0. See caption for Figure 3.4a. Four peaks in indoor particle number concentration are labeled with lower-case letters, g - j. These labels are used to identify the peaks for subsequent discussion.

The occupancy status of the two household residents is an important criterion for investigating the characteristics and consequences of indoor particle number concentration. Figures D.3a and D.3b in Appendix D show the time series of occupancy status for the two residents of H0. The information is based on daily diary entries maintained by the occupants. Figure 3.5 shows the time-averaged particle number concentrations measured over the full observational monitoring period as sorted by the status of each occupant. (Also indicated in the upper portion of each frame is the percentage of the observational monitoring period for which the indicated state applies.) Here are a few noteworthy observations. First, reinforcing the point made in reference to the time-series plots, the outdoor particle concentrations are higher during the daytime periods, when the occupants are commonly either awake at home or away from home, and lower overnight when they are asleep. Second, focusing on the at-home periods, it is noteworthy and significant that the indoor-outdoor (I/O) PN concentration ratios exceed 1.0 for the periods when the occupants are awake, but are less than 1.0 when the occupants are asleep. The episodic indoor emission sources tend to occur when the occupants are awake and at home. Contributions from these sources cause the average indoor levels during this period to exceed the average outdoors. The combined effects of higher outdoor levels during "awake" periods than

"asleep" and also a higher I/O ratio means that the awake-period indoor PN concentrations are considerably higher than the asleep-period indoor concentrations. Focusing on the "in 1" measurement results, the "awake" period average is about 4× higher than the "asleep" period average for R1 and about 3× higher for occupant R2.



**Figure 3.5.** Time-averaged indoor and outdoor particle number concentrations measured during the observational monitoring period at house site H0 and sorted according to the occupancy status of the two inhabitants of the house. Occupant R1 is an adult female and occupant R2 is an adult male. Each bar is labeled with the time-averaged particle number concentration.

A third noteworthy point is that the average concentrations measured at "in 1" and "in 2" agree closely for almost all of the conditions represented in the figure. The ratio "in 2" to "in 1" varies between 81% (R2 "awake") and 101% (R1 "awake"), with an average of 91%. This evidence supports the view that particle number concentrations are reasonably well distributed throughout this house, at least on a time-averaged basis.

Table 3.5 summarizes the average copollutant levels, sorted according to occupancy status of the house. The outdoor NO levels at this site were consistently low. Outdoor ozone levels did not correlate with occupancy status and the indoor to outdoor ratio was in the range 18% to 26% on average. As expected, the CO<sub>2</sub> level is higher when occupants are at home awake as compared to at home asleep and when the house is unoccupied. In Appendix D are presented time-series plots for the copollutants as follows: Figures D.4a and D.4b for ozone; Figures D.5a and D.5b for nitric oxide (outdoor only); Figures D.6a and D.6b for carbon dioxide; and Figures D.7a and D.7b for carbon monoxide.

An important building dynamic factor influencing indoor UFP levels is the air-exchange rate. It has a significant influence on the penetration and persistence of particles originating outdoors. It also is important as a removal process for particles emitted indoors. In general, one expects that the indoor particle concentration will become closer to the outdoor value when the air-exchange rate increases. Conversely, a low air-exchange rate allows for more divergence between the indoor and outdoor particle levels.

At this house site, we were only able to make two determinations of the air-exchange rate using the decay of carbon dioxide concentrations as a tracer. One of these occurred in the middle of the observational monitoring period, when an elevated  $CO_2$  level caused by occupants was followed by a period of vacancy during which the  $CO_2$  decay influenced only by air exchange could be observed. The second determination was from the manipulation experiment conducted on 4 December. The results, summarized in Table 3.6, show a relatively low air-exchange rate of 0.09 and 0.11 per h. During these determinations, all windows and doors in the house were closed. Also, the wind speed (as determined at a central monitoring station) was relatively low and the indoor-outdoor temperature difference (as measured on site) was moderate, as indicated in the table. Overall, for the full observational monitoring period, the average indoor-outdoor temperature difference was 5.6 °C, and the average wind speed was 3.3 m/s.

| Species (units)           | All awake | All asleep | Unoccupied |
|---------------------------|-----------|------------|------------|
| NO_in (ppb)               | n/a       | n/a        | n/a        |
| NO_out (ppb)              | 3.8       | 1.1        | 5.6        |
| O <sub>3</sub> _in (ppb)  | 4.4       | 4.0        | 5.6        |
| O <sub>3</sub> _out (ppb) | 20.7      | 21.9       | 21.8       |
| CO <sub>2</sub> _in (ppm) | 694       | 597        | 521        |

**Table 3.5.** Copollutant levels at site H0 during observational monitoring.<sup>a</sup>

<sup>a</sup> Measurements sorted according to occupancy status of the building and then averaged.

We monitored the open versus closed status of the back door, which is the main entry and exit door for this house, and also the open versus closed position of the window in the master bathroom, which is the window that is most commonly opened during the winter. The back door was open for a total of 1% of the observational monitoring period and the monitored window was open for about 4% of the time. The overall contributions to ventilation from this door and window are expected to be fairly small. An open window might contribute 100-200 m<sup>3</sup> h<sup>-1</sup> to the ventilation rate, so being open 4% of the time indicates an average contribution of ~ 4-8 m<sup>3</sup> h<sup>-1</sup> (Ferro et al., 2009). Published data are lacking on the effect of open doors on air-exchange rates in houses. However, given the relative size of a door and a typical window, one might expect airflow through an open door to be a small integer multiple of that through a window. An air-exchange rate of ~ 0.1 h<sup>-1</sup> in this 320 m<sup>3</sup> house indicates a total ventilation rate of ~ 30-35 m<sup>3</sup> h<sup>-1</sup>. This simple analysis suggests that the average contribution of the open window and door were small but not negligible contributors to the total house ventilation rate, perhaps increasing the average value by ~ 30% over the base value.

**Table 3.6.** Air-exchange rates (AER) determined by carbon dioxide concentration decay in house site H0.<sup>a</sup>

| Time (h)    | $AER(h^{-1})$ | WS (m s <sup>-1</sup> ) | $ \Delta \mathbf{T} $ (°C) | House configuration          |
|-------------|---------------|-------------------------|----------------------------|------------------------------|
| 76.1-77.3   | 0.09          | 2.8                     | 8.4                        | All windows and doors closed |
| A1 (04 Dec) | 0.11          | 1.4                     | 4.5                        | All windows and doors closed |

<sup>a</sup> Uses 424 ppm as the background for observational monitoring; use measured  $CO_2$  outdoors as the baseline for the determination during the manipulation experiment, A1.

The influence of outdoor particles on indoor UFP levels was investigated through two means. One of these was the Pkd manipulation experiment, in which the rebound in indoor particle concentration is measured after the levels are artificially depressed using portable HEPA filter units. However, at this site, the Pkd experiment was not successfully executed, so we had to rely on a second method. In this second method, we compute the ratio of indoor to outdoor particle concentrations, based on time-averaged concentration levels, during times when the house is unoccupied and when there is no clear residual influence of particles from indoor sources. We restricted our attention to times when the dominant configuration of the house applied, namely when all doors and windows were closed.

A total of seven suitable intervals were identified, distributed throughout the observational monitoring period, with a total duration of about 30 h. The results are presented in Table 3.7. For these seven periods, the ratio of average indoor to average outdoor concentrations ranges from 0.22 to 0.92 with a time-weighted average of  $f_1 = 0.36$  for the main indoor monitor (PN\_in1) and, for six periods, from 0.24 to 0.75 with a time-weighted average of  $f_2 = 0.37$  for the supplementary indoor monitor (PN\_in2). Treating each of the intervals as a separate determination, the weighted standard error in these determinations is 0.06-0.07, corresponding to a relative standard error in the mean values of  $f_1$  and  $f_2$  of 15-20%. These  $f_1$  and  $f_2$  parameters are believed to represent the indoor proportion of outdoor particles. That is, on a time-average basis at this site, the evidence suggests that the average indoor concentration of UFP of outdoor origin is about 38-39% of the corresponding average outdoor concentration.

To investigate the significance of indoor sources at site H0, we first compiled information from the activity logs on potential indoor sources. These entries are summarized in Table 3.8. We then examined the time-series of indoor particle concentrations, looking for matches between an entry in the activity log and a sudden increase in the indoor particle concentration. Through this process, each of the peaks in the indoor particle concentration trace was matched to a coincident activity — either cooking (using electric oven, electric range or toaster) or furnace use.

| Time                 | WS           | $ \Delta \mathbf{T} $ | PN_out                   | PN_in1                     | PN_in2                   | $f_{I}$ | $f_2$ |
|----------------------|--------------|-----------------------|--------------------------|----------------------------|--------------------------|---------|-------|
| ( <b>h</b> )         | $(m s^{-1})$ | (°C)                  | $(10^3 \text{ cm}^{-3})$ | $(10^3  \mathrm{cm}^{-3})$ | $(10^3 \text{ cm}^{-3})$ | (—)     | ()    |
| 17.8-20.8            | 1.9          | 3.2                   | 16.2                     | 6.4                        | n/a                      | 0.40    | n/a   |
| 46.8-48.8            | 3.5          | 7.0                   | 9.3                      | 5.3                        | 5.2                      | 0.57    | 0.56  |
| 76.3-77.3            | 2.8          | 8.4                   | 21.5                     | 7.6                        | 6.7                      | 0.35    | 0.31  |
| 87.1-88.3            | 4.2          | 4.3                   | 2.4                      | 2.2                        | 1.8                      | 0.92    | 0.75  |
| 95.1-97.8            | 7.5          | 2.9                   | 9.9                      | 5.5                        | 6.1                      | 0.56    | 0.62  |
| 110.8-125.5          | 4.6          | 2.1                   | 13.4                     | 4.0                        | 4.3                      | 0.30    | 0.32  |
| 134.8-140.3          | 1.5          | 2.9                   | 9.5                      | 2.1                        | 2.3                      | 0.22    | 0.24  |
| Average <sup>b</sup> | 3.9          | 3.0                   | 12.2                     | 4.2                        | 3.7                      | 0.36    | 0.37  |

**Table 3.7.** Analysis of the indoor to outdoor particle concentration ratios at house site H0 for periods when the house is unoccupied and all doors and windows are closed.<sup>a</sup>

<sup>a</sup> The parameter  $f_1$  represents the ratio of the averages for PN\_in1 to PN\_out; the parameter  $f_2$  represents the ratio of the averages for PN\_in2 to PN\_out.

<sup>b</sup> The final row of the table presents time-weighted average values for each entry.

We found that several other recorded activities did not produce a clear change in the indoor particle level. Specifically, these activities were (i) use of a microwave oven; (ii)

vacuuming; (iii) washing clothes; (iv) use of a gas-fired, vented clothes dryer; and (v) use of a dishwasher. The absence of a clear signal does not necessarily mean that these activities emitted *no* ultrafine particles. It does mean that any emissions were insufficient to produce a clear change against the baseline indoor concentration, which averaged ~  $10 \times 10^3$  cm<sup>-3</sup> during daytime awake hours.

Temperature monitors placed on the stove, the toaster, and on a heating-system supply register provided supporting evidence for the activity diary entries for almost all of the peaks. In particular, among the ten peaks, a corresponding clear, sharp increase in one or more appropriate temperature signals occurred for nine, as illustrated in Figures 3.6a and 3.6b. Only for peak i is a corroborating temperature signal missing from the stove.

| Time (h) | Activity                           | PN source? |
|----------|------------------------------------|------------|
| 0.00     | Cooked granola on electric stove.  | ? (Peak a) |
| 0.25     | Baked granola in oven.             | Y (Peak a) |
| 21.25    | Microwaved. Vacuumed bedroom 2.    | Ν          |
| 22.17    | Boiled water on stove.             | Y (Peak b) |
| 26.83    | Boiled rice on stove.              | Y (Peak c) |
| 27.67    | Stir fry                           | Y (Peak c) |
| 39.25    | Furnace on (first use of winter).  | Y (Peak d) |
| 40.75    | Boiled water on stove.             | Y (Peak d) |
| 44.75    | Microwave.                         | Ν          |
| 46.50    | Washed clothes.                    | Ν          |
| 49.50    | Toaster                            | Y (Peak e) |
| 51.25    | Boiled potatoes.                   | Y (Peak e) |
| 52.17    | Dried clothes                      | Ν          |
| 64.08    | Furnace on                         | Y (Peak f) |
| 64.92    | Dishwasher.                        | Ν          |
| 68.17    | Dried clothes.                     | Ν          |
| 72.00    | Boiled water on stove, made toast. | Y (Peak g) |
| 88.25    | Furnace on.                        | Y (Peak h) |
| 92.92    | Ramen noodles on stove.            | Y (Peak i) |
| 98.08    | Toasted 4 slices of bread          | Y (Peak j) |
| 98.25    | Penne pasta boiled on stove.       | Y (Peak j) |

**Table 3.8.** Activity entries in the occupant diary at house site H0 related to potential indoor particle emission sources.<sup>a</sup>

<sup>a</sup> "Peak a" through "peak j" correspond to episodes of elevated indoor particle number concentration as illustrated in Figures 3.4a and 3.4b.

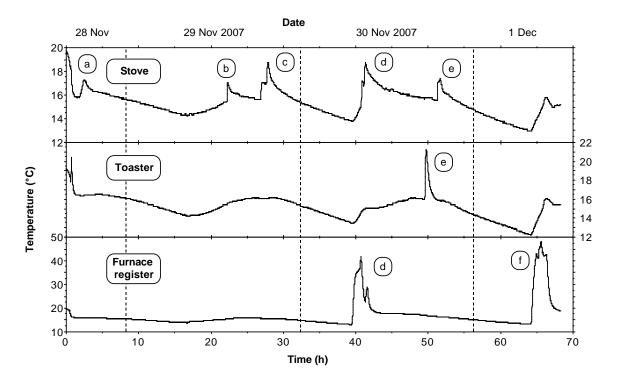
The data from these episodic increases in indoor PN concentrations were analyzed to quantitatively estimate source strength and particle loss rate. For each peak, the analysis proceeds in three stages. First, a net indoor concentration is computed, by subtracting from the measured indoor level a fraction of the outdoor concentration that corresponds to  $f_1$  or  $f_2$ . Second, for the decay period, the natural log of the net indoor concentration is plotted versus time and the slope of the regression line determined as an estimate of the first-order loss-rate coefficient. This coefficient is labeled "k+a," where a is taken to represent the air-exchange rate and k is taken to represent the loss by other processes than air-exchange, such as the deposition

of particles on indoor surfaces. In the third step, the time-integral of the net indoor particle number concentration is computed for the duration of the concentration peak, from the beginning of the rise until the net concentration decays to zero. From the results of the second and third step, the effective particle emission rate per house volume,  $\sigma/V$ , is determined. Finally, the total particle emission rate for the peak event,  $\sigma$ , is computed by multiplying  $\sigma/V$  by the interior house volume, V.

Mathematically, this analysis can be referred to as applying an integral material balance to the net particle number concentration. The starting point is equation 3.1

$$\frac{dN_{net}}{dt} = \frac{E(t)}{V} - \left(k + a\right)N_{net}$$
(3.1)

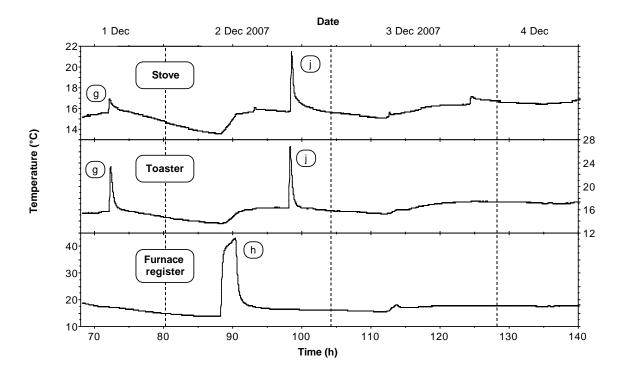
Here,  $N_{net}$  is the increment of indoor particle number concentration associated with the peak event, E(t) is the time-dependent emission rate that causes the increase in particle number concentration, V is the house interior volume, a is the air-exchange rate, and k is the first-order loss-rate coefficient for indoor particles by means other than ventilation.



**Figure 3.6a.** Time-series of sensor temperatures deployed at house site H0 for the first half of observational monitoring. A sudden increase in temperature indicates use of the appliance. The lower-case letters labeling peaks correspond to the rises in indoor particle number concentrations (see Figure 3.4a).

In the analysis, we assume that the emissions (E(t)) are greater than zero for some finite period, after which the following decay expression would apply:

$$\frac{dN_{net}}{dt} = -(k+a)N_{net}$$
(3.2)



**Figure 3.6b.** Time-series of sensor temperatures deployed at house site H0 for the second half of observational monitoring. A sudden increase in temperature indicates use of the appliance. The lower-case letters labeling peaks correspond to the rises in indoor particle number concentrations (see Figure 3.4b).

During the decay time, the particle number concentration will follow an exponential decay curve obtained as the solution to equation (3.2):

$$N_{net}(t) = N_{net}(t^*) \times \exp[-(k+a)(t-t^*)]$$
(3.3)

Here,  $t^*$  denotes the time at the beginning of the decay analysis period. To the extent that this equation is valid, then plotting  $\ln(N_{net})$  versus *t* produces a linear decay whose slope is the negative of (k+a).

Returning to equation (3.1), we can multiply both sides of the equation by dt and then integrate from the beginning to the end of the peak event. The term on the left hand side of the integral equation vanishes because there is no net change in the net number of particles indoors between the beginning of the event and its end. Assuming that (k+a) is time-invariant, we then have

$$\frac{\sigma}{V} = \int \frac{E(t)}{V} dt = (k+a) \int N_{net}(t) dt$$
(3.4)

In this equation,  $\sigma$  represents the total number of particles emitted indoors during the peak event. In practice, it is evaluated by numerically computing the area under the curve of the net indoor particle concentration,  $N_{net}(t)$ , versus time, t, and multiplying that integral by the total indoor particle loss rate, (k+a), which is the other parameter evaluated in this analysis.

Finally, the contribution of the peak event to an occupant's particle exposure is simply the integral of  $N_{net}(t) dt$  over the time that the occupant is present in the household. The integral equals  $(\sigma/V)/(k+a)$  if the occupant is present for the entire peak-event period. If the occupant is present for only a portion of the time, then the contribution to exposure is estimated as the numerically evaluated integral of  $N_{net}(t) dt$  during the overlapping period of occupancy and the peak event.

Table 3.9 presents a summary of the analysis results for the ten identified peak events during observational monitoring at house site H0. For peak b, the decay does not follow a single first-order trajectory, but is better modeled as two separate decay periods, with a loss rate of 1.4  $h^{-1}$  for the first period and 0.2  $h^{-1}$  for the second. For peaks c, d, and e, two activities occurred at slightly offset times, producing overlapping peaks. We have estimated decays and emission characteristics for each subpeak, but these are less reliable than the determinations for the total peak event.

|    | e 3.9. Analysis of m         | Time       | k+a                    | $\sigma/V$                | σ           | $(\sigma/V)/(k+a)$                   |
|----|------------------------------|------------|------------------------|---------------------------|-------------|--------------------------------------|
| ID | Source activity              | (h)        | (h <sup>-1</sup> )     | $(10^3 \mathrm{cm}^{-3})$ | $(10^{12})$ | $(10^3 \mathrm{cm}^{-3} \mathrm{h})$ |
| a  | Stove & oven                 | 0.0-5.8    | 0.6                    | n/a                       | n/a         | 50.0                                 |
| b  | Stove (water)                | 22.2-25.9  | 1.4, 0.24 <sup>b</sup> | 16.3                      | 5.2         | 31.4                                 |
| c1 | Stove (rice)                 | 26.9-27.8  | 1.9                    | (32.6)                    | (10.4)      | (17.0)                               |
| c2 | Stove (stir fry)             | 27.8-30.2  | 0.7                    | (6.6)                     | (2.1)       | (9.2)                                |
| с  | Stove                        | 26.9-30.2  | c                      | 39.3                      | 12.6        | 26.2                                 |
| d1 | Furnace                      | 39.4-44.0  | 2.0                    | (530)                     | (169)       | (270)                                |
| d2 | Stove (water)                | 40.8-45.0  | 0.6                    | (42)                      | (13.5)      | (66)                                 |
| d  | Furnace (& stove)            | 39.4-45.0  | c                      | 572                       | 183         | 336                                  |
| e1 | Toaster                      | 49.5-51.3  | 1.2                    | (21.4)                    | (6.8)       | (17.5)                               |
| e2 | Stove (potatoes)             | 51.3-53.2  | 0.6                    | (7.3)                     | (2.4)       | (12.8)                               |
| e  | Toaster & stove              | 49.5-53.2  | c                      | 28.7                      | 9.2         | 30.3                                 |
| f  | Furnace                      | 64.1-68.0  | 1.2                    | 133                       | 42.6        | 111                                  |
| g  | Toaster & stove              | 72.0-74.8  | 1.0                    | 33.5                      | 10.7        | 33.0                                 |
| h  | Furnace                      | 88.3-92.4  | 2.0                    | 107                       | 34.1        | 53.3                                 |
| i  | Stove (ramen)                | 93.0-94.8  | 1.1                    | 16.0                      | 5.1         | 14.2                                 |
| j  | Toaster & stove <sup>d</sup> | 98.1-102.1 | 1.0                    | 124                       | 39.7        | 130                                  |

**Table 3.9.** Analysis of indoor UFP sources at house site H0 from observational monitoring.<sup>a</sup>

<sup>a</sup> Parameter definitions: k+a = total loss rate coefficient for particles emitted during the event;  $\sigma/V = \text{total particle number per house volume emitted;}$   $\sigma = \text{total particle number emitted;}$  $(\sigma/V)/(k+a) = \text{contribution to exposure for an occupant present for the entire event.}$ 

<sup>b</sup> For this event, the particle concentration decay is better modeled using two separate decay constants, with the larger one applying for the early period (0.5 h) and the smaller one for the later period (3.1 h). The time-weighted average decay rate is 0.4 h<sup>-1</sup>.

<sup>c</sup> The decays of the subpeaks were separately modeled for events c, d, and e.

<sup>d</sup> The range hood exhaust fan was on during this cooking event.

In sum, for these peak events, the observed particle loss rates (k+a) range from 0.4 h<sup>-1</sup> (the time-weighted average for peak b) to 2.0 h<sup>-1</sup> (peak h) with an average value of 1.1 h<sup>-1</sup> and a standard deviation of 0.6 h<sup>-1</sup>. The total particle emissions per event vary over a broader range, from 5 to 183 trillion (10<sup>12</sup>) particles each and an average of  $38 \times 10^{12}$  particles per event. Excluding the one extreme event from the first use of the furnace, the range is narrowed to (5-43)  $\times 10^{12}$  particles per event and the average diminishes to  $20 \times 10^{12}$  particles per event. In terms of potential contributions to exposure (the rightmost column of Table 3.9), the range for these events is from 14 to 336 in units of  $10^3$  cm<sup>-3</sup> h, and the average contribution is  $82 \times 10^3$  cm<sup>-3</sup> h.

Table 3.10 summarizes the results of the three manipulation experiments conducted to investigate source emissions at this study site. Three cooking-related experiments were conducted: (M1) toasting two slices of bread in an electric toaster; (M2) boiling water in a kettle on the electric stove; and (M3) frying an onion in a pan on the electric stove. These experiments were conducted in the kitchen, with it closed off and isolated from the rest of the house. In each case, there was a clear signal of an increase in indoor particle number concentration, so these activities were confirmed as particle sources. For M2, data were not available on outdoor PN concentrations, which precludes quantitative analysis. The results for the other two experiments show decay rates that are similar to those obtained from peak analysis during the observational monitoring period. However, the total particle emission rate,  $\sigma$ , is about an order of magnitude lower than obtained for the observational monitoring peaks (compare entries for  $\sigma$  in Table 3.10 with those for cooking activities in Table 3.9). The reason for this difference is unknown.

|     |                         | Duration | k+a        | $\sigma/V$               | σ           |
|-----|-------------------------|----------|------------|--------------------------|-------------|
| ID  | Source                  | (h)      | $(h^{-1})$ | $(10^3 \text{ cm}^{-3})$ | $(10^{12})$ |
| M1  | Toast 2 slices          | 1.1      | 1.3        | 19.4                     | 0.72        |
| M2  | Boil water <sup>a</sup> |          |            |                          | _           |
| M3  | Fry onion               | 2.9      | 1.0        | 47.2                     | 1.7         |
| 9 0 |                         |          |            |                          |             |

Table 3.10. Analysis of UFP source from manipulation experiments at site H0.

<sup>a</sup> Outdoor particle number concentration data are unavailable for this experiment and so the results cannot be quantitatively analyzed.

An analysis was conducted to estimate residential exposures to particles for the occupants of this household for the observational monitoring period and to apportion those exposures to known indoor sources and to indoor particles of outdoor origin. The results of the analysis are presented in Table 3.11. Cumulative exposures while "awake at home" and "asleep at home" are obtained as the product of the appropriate indoor time-averaged PN concentration and the duration of occupancy awake or asleep. We used PN\_in1 as the best indicator of indoor exposure concentration during awake periods and PN\_in2 as the best indicator when occupants were asleep, since most of their awake time at home was spent downstairs, the level where PN\_in1 was measured, and they slept upstairs, the level where PN\_in2 was sampled. These cumulative exposures were summed to obtain the "total cumulative exposure" for the time spent at home,  $965 \times 10^3$  cm<sup>-3</sup> h for R1 and  $599 \times 10^3$  cm<sup>-3</sup> h for R2. The daily exposure rates were obtained by dividing these figures by the approximate duration of household monitoring during which the occupants were staying at home and therefore accruing exposures, 3.8 days in each case. These measures and the results —  $251 \times 10^3$  cm<sup>-3</sup> h/d and  $156 \times 10^3$  cm<sup>-3</sup> h/d for R1 and R2, respectively — are designed to be independent of the duration of observational monitoring.

The contribution of outdoor particles to exposure was estimated by taking the appropriate fraction of the average measured outdoor concentration during occupancy,  $f_1$  for awake periods and  $f_2$  when the resident was asleep, and computing a cumulative exposure for each occupant. The results were  $286 \times 10^3$  cm<sup>-3</sup> h for R1 and  $179 \times 10^3$  cm<sup>-3</sup> h for R2, corresponding to 30% of the total cumulative exposure for each.

| Parameter  | <b>R1 (F ad)</b> | R2 (M ad) |
|--|------------------|-----------|
| Occupancy status   |                  |           |
| Time at home, awake (h)  | 45.2             | 32.0      |
| Time at home, asleep (h)   | 33.5             | 30.5      |
| Time away from home (h)  | 61.5             | 77.8      |
| Exposure duration (d) <sup>a</sup>   | 3.84             | 3.84      |
| Average exposure concentrations and exposures                                  |                  |           |
| Average concentration (PN_in1), indoor awake $(10^3 \text{ cm}^{-3})$          | 18.4             | 14.9      |
| Average concentration (PN_in2), indoor asleep $(10^3 \text{ cm}^{-3})$         | 3.9              | 4.1       |
| Cumulative exposure, awake at home $(10^3 \text{ cm}^{-3} \text{ h})$          | 833              | 475       |
| Cumulative exposure, asleep at home $(10^3 \text{ cm}^{-3} \text{ h})$         | 131              | 124       |
| Total cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                   | 965              | 599       |
| Cumulative exposure rate $(10^3 \text{ cm}^{-3} \text{ h d}^{-1})$             | 251              | 156       |
| Indoor exposure attributable to particles of outdoor origin                    |                  | •         |
| Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$         | 286              | 179       |
| Percentage attributable to particles of outdoor origin                         | 30%              | 30%       |
| <i>Exposure attributable to indoor source peaks</i> $(10^3 \text{ cm}^{-3} h)$ |                  |           |
| Peak a — bake granola in electric oven   | 50               | 0         |
| Peak b — boil water on electric range  | 31               | 0         |
| Peak c — cook rice and stir-fry vegetables on electric range                   | 26               | 0         |
| Peak d — natural-gas furnace (vented; first use); boil water                   | 336              | 0         |
| Peak e — toaster use and boil potatoes on electric range                       | 30               | 13        |
| Peak f — natural-gas furnace (vented)  | 109              | 111       |
| Peak g — toaster use and boil water on electric range                          | 20               | 33        |
| Peak h — natural-gas furnace (vented)  | 0                | 53        |
| Peak i — cook ramen noodles on electric range                                  | 0                | 12        |
| Peak j — toaster use and cook pasta on electric range (exhaust on)             | 0                | 130       |
| Cumulative exposure attributable to indoor sources                             | 603              | 353       |
| Percentage attributable to quantified indoor sources                           | 63%              | 59%       |
| Indoor exposure of unknown origin  |                  |           |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                         | 73               | 66        |
| Percentage of unknown origin   | 8%               | 11%       |

**Table 3.11.** Exposure analysis for residents of house site H0 during the observational monitoring period.

<sup>a</sup> Exposure duration is computed as the total monitoring period, minus 24 h per day not slept at home, with the result divided by 24 h/d. So, for the two occupants of H0, the result is (140.2-48)/24 = 3.8 d.

The contribution to exposure from indoor peak events was evaluated for each peak separately, as reported in Table 3.11, and summed to obtain the cumulative exposure attributable to indoor sources. The results were  $603 \times 10^3$  cm<sup>-3</sup> h corresponding to 63% of the total for R1 and  $353 \times 10^3$  cm<sup>-3</sup> h corresponding to 59% for R2.

The remainder — 8% for R1 and 11% for R2 — is of unknown origin. The two most likely contributors are (a) persistent low-level indoor sources; and (b) contributions from outdoor sources that are larger than estimated here. Evidence in support of the latter comes from the fact that the outdoor contribution estimate is based on the average indoor to outdoor ratio during times when the house was unoccupied and closed. Overall, though, window and door openings, which did occur to some extent, would likely have produced a higher average indoor to outdoor ratio than obtained for the empty-house conditions.

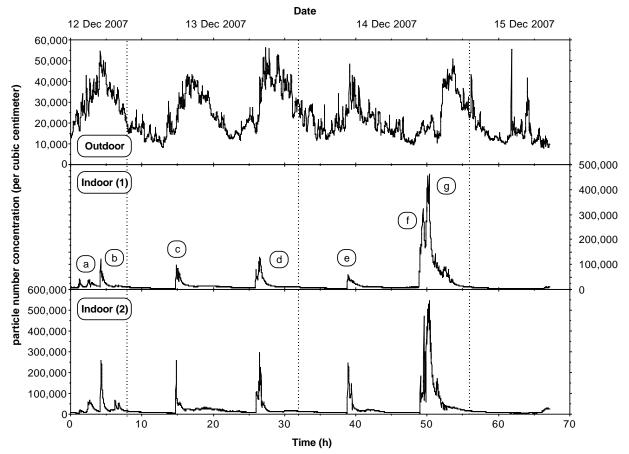
## 3.2.2. Site H1

Observational monitoring was conducted at house site H1 over a period of 67 h, on 12-15 December 2007. The reader is referred to §2.2.1 for detailed background information about the house site. See also Figure C.2 (Appendix C) for a floor plan. Summarizing briefly, this house has two floors, a main upper floor with an internal volume of about 315 m<sup>3</sup> and a lower floor with an attached garage and supplementary rooms. The home is sited in a residential neighborhood within the urban core of Oakland, at an elevation of < 50 m above sea level. The nearest freeway is ~ 600 m to the east; however, the house is < 100 m to the west of a busy arterial road. The house has natural-gas cooking appliances with continuous pilot lights. It has a forced-air central furnace that is vented and gas-fired. The house has four occupants, two adults (R1 = female adult; R2 = male adult) and two male children (R3 = male ~ 9 y old; R4 = male ~ 5 y old).

A summary of the time-averaged species concentration and cofactor values was presented in Table 3.1 (§3.1). The average particle number concentrations measured during the entire observational monitoring period were 23.2 (outdoors), 21.0 (indoors primary), and 27.4 (indoors supplemental) in units of 1000 particles per cm<sup>3</sup>. The outdoor concentrations during observational monitoring were higher at this site than at any other house site in the study and the time-averaged indoor concentrations were the second highest.

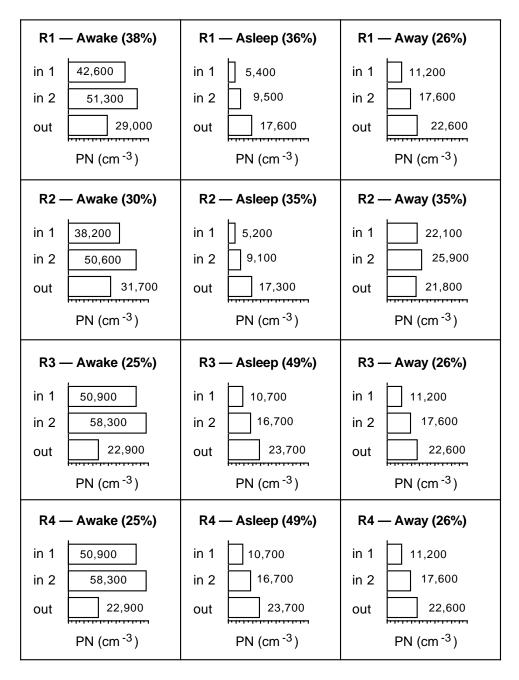
Figure 3.7 shows the time-series of particle number concentrations measured by the three monitors. The horizontal axis is labeled with elapsed time of monitoring referenced to the start time of 16:04 on 12 December 2007. The vertical dashed lines demark midnight. Lower-case letters (a-g) in the middle frame identify peak events believed to be associated with episodic indoor emissions. These are discussed later in the section.

The outdoor concentrations at this site fluctuate between about ~  $10,000 \text{ cm}^{-3}$  and ~  $50,000 \text{ cm}^{-3}$ . There is a suggestion, especially clear during the first half of the observational period, of a diurnal pattern with two peaks, one in the morning (~ 08:00-10:00) and a second one in the evening (~ 19:00-2100). On the linear scale presented in Figure 3.7, the several peaks that are seen in both indoor monitors clearly dominate PN concentrations in the house. Also significant at this site are persistent contributions to indoor UFP levels caused by emissions from the natural-gas pilot lights on the cooking appliance and the penetration and persistence of particles from outdoors.



**Figure 3.7.** Time-series of particle number concentrations during observational monitoring at house site H1. Monitor "Indoor (1)" was located on the main floor (upstairs) in the dining room; monitor "Indoor (2)" sampled from an upstairs study. Note that the indoor and outdoor concentration scales are different. The lower case letters designate peaks in the indoor concentration time series that are discussed subsequently.

The time series of occupancy status of the four residents of H1 is presented in Figure D.16 (see Appendix D). Figure 3.8 shows the time-averaged particle number concentrations measured over the full observational monitoring period as sorted by the status of each occupant. The main feature of this figure is that — for each resident — the average indoor particle levels are much higher when awake at home than when asleep or away. A second, related observation is that the indoor/outdoor ratio is consistently higher than 1.0 for the awake at home periods and consistently less than 1.0 for the asleep and away periods.



**Figure 3.8.** Time-averaged indoor and outdoor particle number concentrations measured during the observational monitoring period at house site H1 and sorted according to the occupancy status of the four inhabitants of the house. Occupant R1 is an adult female, occupant R2 is an adult male, and occupants R3 and R4 are male children. Each bar is labeled with the time-averaged particle number concentration. Also indicated in the upper portion of each frame is the percentage of the observational monitoring period for which the indicated state applies.

Table 3.12 summarizes the average copollutant levels at H1, sorted according to the occupancy status of the house. The outdoor NO levels at this site were moderately high, and higher during the daytime hours (when the house was unoccupied or when all were awake at home) than overnight. Ozone levels were low, both outside and indoors. Carbon dioxide levels

were elevated in this house; likely causes are the low air-exchange rate (discussed below) and the emissions from natural gas cooking and metabolically from occupants. In Appendix D are presented time-series plots for the copollutants as follows: Figure D.17 for ozone; Figure D.18 for NO (outdoor only); Figure D.19 for carbon dioxide; and Figure D.20 for carbon monoxide.

|                           | 1 0       |                |            |  |  |
|---------------------------|-----------|----------------|------------|--|--|
| Species (units)           | All awake | All asleep     | Unoccupied |  |  |
| NO_in (ppb)               | n/a       | n/a            | n/a        |  |  |
| NO_out (ppb)              | 57.3      | 38.2           | 50.9       |  |  |
| O <sub>3</sub> _in (ppb)  | 3.0       | 3.1            | 3.1        |  |  |
| O <sub>3</sub> _out (ppb) | 0.5       | 0 <sup>b</sup> | 5.9        |  |  |
| CO <sub>2</sub> _in (ppm) | 1636      | 1061           | 950        |  |  |

**Table 3.12.** Copollutant levels at site H1 during observational monitoring.<sup>a</sup>

<sup>a</sup> Measurements sorted according to occupancy status of the building and then averaged.

<sup>b</sup> Negative baseline; mean is less than zero.

At this house site, we made five determinations of the air-exchange rate. Three of these occurred during the observational monitoring period, when an elevated  $CO_2$  level caused by occupants was followed by a period of vacancy during which the CO<sub>2</sub> decay influenced only by air exchange could be observed. The other two determinations (A1 and A2) were made during manipulation experiments. The results, summarized in Table 3.13, show a moderately low airexchange rate: average =  $0.23 \text{ h}^{-1}$  (range = 0.1-0.34 per h). For the three determinations during observational monitoring, the wind speed was low (~ 1 m/s) and the temperature difference moderate (8-9 °C). The lowest air-exchange rate occurred during the period when the house was fully closed; somewhat higher values were recorded when the bathroom window was open. In the controlled experiments, the windspeed was higher (3 m/s) but the temperature difference was lower (4-6 °C). In these determinations, the house was fully closed. During one of the two experiments, the home heating system was operating on thermostatic control. During the second experiment, it was off. Central heating systems can influence air-exchange rates either by inducing air leakage through ducts that pass through unconditioned space or by changing the distribution of pressures across the building envelope. At this house, the influence of the central air system on air-exchange rate does not appear to be large.

| Time (h)    | $AER(h^{-1})$ | WS (m s <sup>-1</sup> ) | $ \Delta \mathbf{T} $ (°C) | House configuration                      |  |
|-------------|---------------|-------------------------|----------------------------|--|--|
| 17.4-18.4   | 0.20          | 1.3                     | 9.4                        | Bathroom window open                     |  |
| 42.1-43.1   | 0.10          | 1.3                     | 8.3                        | All windows and doors closed             |  |
| 66.5-67.0   | 0.34          | 1.3                     | 7.8                        | Bathroom window open                     |  |
| A1 (18 Dec) | 0.26          | 3.2                     | 4.3                        | All doors and windows closed,            |  |
|             |               |                         |                            | thermostat at 69 °F (central air cycled) |  |
| A2 (24 Dec) | 0.23          | 3.0                     | 6.2                        | All windows and doors closed,            |  |
|             |               |                         |                            | thermostat at 64 °F (central air off)    |  |

**Table 3.13.** Air-exchange rates (AER) determined by carbon dioxide concentration decay in house site H1.<sup>a</sup>

<sup>a</sup> Uses 650 ppm as the background for observational monitoring and manipulation experiments. The overall average windspeed (WS) and temperature difference ( $\Delta$ T) for the observational monitoring period were 1.5 m/s and 12.4 °C.

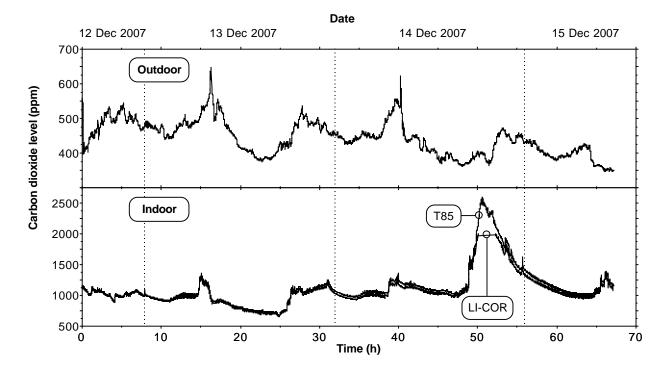
At this site, we monitored the open versus closed status of two doors and two windows. The two doors, the front door and the garage door, were only open 0.3% and 0.6%, respectively, of the observational monitoring period. Monitoring indicated that the children's bedroom window was not opened during the observational period. The bathroom window was open 16% of the time, mainly during a continuous period that began on 13 Dec at 7:43 and ended that same day at 17:15 (monitored time of 15.6-25.2 h). The indications from results reported in Table 3.13 suggest that the open bathroom window might have had a significant influence on the air-exchange rate during this period.

At site H0, we were able to use the indoor to outdoor concentration ratios during unoccupied periods to estimate the contribution of outdoor particles to indoor levels. That approach did not work at site H1, primarily because of persistent emissions of particles from the continuously burning pilot lights on the gas stove and oven. During times when the house is unoccupied, indoor levels are influenced by a combination of the penetration and persistence of particles from outdoors and also the emissions from the pilot lights. Furthermore, the central forced-air system operated a substantial fraction of the time at this site. It contains a particle filter such that when the fan is on there is an additional removal mechanism for indoor particles that must be accounted for to properly characterize the influence of particles of outdoor origin and emissions from indoor sources on indoor levels. The heating system operates for different proportions of time during occupied and unoccupied periods.

An important clue about the potential significance of the pilot lights was revealed in the persistently elevated indoor  $CO_2$  levels (Figure 3.9), even in the absence of source-use activities and during unoccupied periods. This house has an antique Wedgewood stove and oven that is regularly used for cooking. The unit has four pilot lights, two each for the stovetop and the oven. In its normal condition, all four pilots burn continuously. A supplemental experiment was designed and executed on 12 February 2009 to characterize UFP emissions from the pilot lights.

The basic concept in the experiment was to conduct indoor and outdoor monitoring with no indoor sources other than the pilot lights across two changes of state: from pilots on to pilots off and then back to pilots on. Monitoring was conducted according to this schedule:

- 09:29-11:28 four pilots on
- 11:30-14:29 all pilots off
- 14:32-1623 four pilots on
- The house was vacant during these periods and only occupied briefly in the intervening times to change the operational state. Figure 3.10 illustrates the time-dependent particle number concentration during this experiment as measured outdoors (upper frame), indoors on the same floor level as the stove (middle frame), and downstairs (lower frame). The furnace was off during this experiment. The trace in the lower frame indicates that the downstairs zone was not strongly influenced during this experiment by emissions from the pilot lights. The trace in the middle shows two indications of a discernible influence of the particle concentrations on pilot light operation: (i) a downward and sustained break in the slope of the curve when the pilot lights are first shut off; and (ii) a sudden and sustained increase in the indoor particle concentration when the pilot lights are turned on again. The decoupled upstairs and downstairs zones enables focusing on the upstairs volume ( $V \sim 315$  m<sup>3</sup>) in analyzing the results from this experiment.



**Figure 3.9.** Carbon dioxide concentrations measured outside and inside site H1, during observational monitoring. Note that the LI-COR monitor has an upper detection limit of 2000 ppm, which is the source of the divergence between the two indoor monitors at time ~ 50 h.

To characterize emissions from the pilot lights, particle concentration data were analyzed in two stages. To begin, a simultaneously measured decay in the CO<sub>2</sub> concentration yielded an air-exchange rate estimate of a = 0.27 h<sup>-1</sup>. With this information, the time-series indoor PN concentration was modeled for the "pilots-off" period, using this material balance equation:

$$\frac{dN}{dt} = PaN_o - (k_d + a)N \tag{3.5}$$

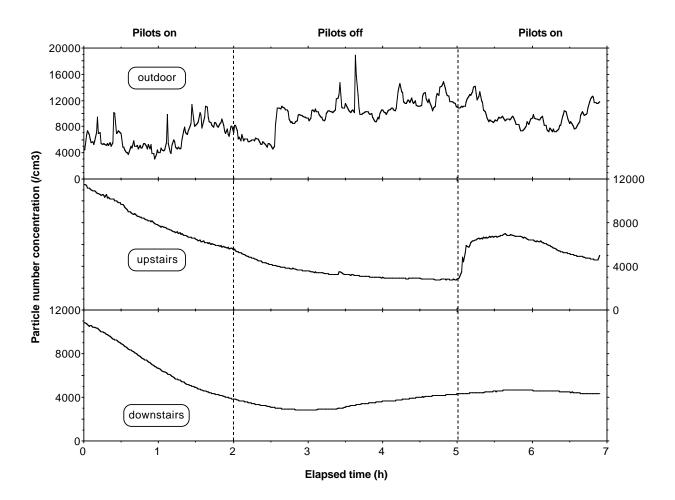
This equation represents the change in indoor concentration as the difference between two terms: penetration from outside (with penetration efficiency, P, and outdoor concentration  $N_o$ ) and removal from indoors, by combined effects of air-exchange (rate constant a) and deposition (rate constant k). The discrete algebraic approximation to this equation was fit to the time-series data for N, given the measured time-dependent outdoor concentration,  $N_o$ , and the tracer-gas determined air-exchange rate, a = 0.27 h<sup>-1</sup> to obtain best-fit values for the penetration efficiency and the deposition rate coefficient, using a least-squares fitting procedure and a time step of 10 minutes. The results were P = 0.61 and k = 0.44 h<sup>-1</sup>.

For the second phase of the analysis, the final period, when pilot lights were restored, is modeled according to this equation:

$$\frac{dN}{dt} = \frac{E}{V} + PaN_o - (k+a)N \tag{3.6}$$

Here, *E* is the particle number emission rate from the pilot lights and *V* is the volume of the upstairs of the house (315 m<sup>3</sup>). Using the parameter results already determined and the measured volume of upstairs, the discrete version of equation (3.6) is fit to the data for the final experimental period to obtain the best estimate value of the particle emission rate, *E*. The result was  $E = 0.58 \times 10^{12}$  particles h<sup>-1</sup>. The influence of this emission rate on indoor particle levels at H1 can be estimated using a steady-state material balance considering particles from this source only:

$$N_{pilot} = \frac{E}{V \times (k+a)}$$
(3.7)



**Figure 3.10.** Particle number concentrations versus time during the pilot light experiment (12 February 2009) at site H1. The upstairs monitor sampled from the dining room. The elapsed time scale begins at 09:29. Pilot lights on the gas-cooking appliance were extinguished at 11:29 (elapsed time = 2 h) and relit at 14:30 (elapsed time = 5 h).

With  $E \sim 0.58 \times 10^{12}$  particles h<sup>-1</sup>,  $V \sim 315$  m<sup>3</sup>,  $k \sim 0.44$  h<sup>-1</sup> and  $a \sim 0.23$  h<sup>-1</sup>, we estimate  $N_{pilot} \sim 2700$  cm<sup>-3</sup> when particle losses are by deposition and air-exchange only. However, different house configurations can have distinct k + a values. Consequently, in assessing and attributing PN exposures for occupants, the analysis of the pilot light contribution to indoor levels is

computed separately for each of three main house states: (i) window closed/central air off, (ii) window closed/central air on, and (iii) window open.

To investigate the effect of outdoor particles on indoor levels in the presence of the pilot lights, we conducted the following analysis using observational monitoring data. We investigated data collected from two extended periods when the house was unoccupied. For one of these (for the period 17.4-24.4 h of elapsed monitoring time), the bathroom window was open; for the other (42.1-46.9 h) all windows and doors were shut. For most of the time during both of these periods, the central heat was off. We fit the following dynamic model to each period separately:

$$\frac{dN}{dt} = \frac{E}{V} + PaN_o - (k+a)N \tag{3.8}$$

In fitting this model, we used the E/V value obtained from the pilot-light experiments, and the measured time-dependent outdoor  $(N_o)$  and primary indoor (N) measured concentrations. The two additional parameters, Pa and (k+a), were treated as constants and evaluated by selecting the combination that produced the least-squares best fit between model and measurement. With these parameters, the ratio  $f_1$  of indoor to outdoor concentration, considering only particles of outdoor origin was estimated as the ratio  $f_1 \sim Pa/(k+a)$ .

We needed to consider one further complication. For these analysis periods, the furnace was off for a large majority of the time. However, during periods of occupancy, the furnace was often on, especially when occupants were awake. The furnace is equipped with a particle filter and so its operation would lead to a net loss of indoor particles, whether originating from indoor or outdoor sources, and that loss should be accounted for in characterizing the dynamic fate of indoor particles.

We modeled this additional loss mechanism as an increase in the first-order loss rate k. In the analysis of PN exposure and its apportionment for occupants at this site, we consider that the house operates in one of three configurations. The baseline state is when all doors and windows are closed and the furnace is off. A second state occurs when the furnace is off, but the bathroom window is open. In the third state, all doors and windows are closed and the furnace is on, the fan cycles on and off under thermostatic control. We do not try to model the on and off periods in these cycles in detail. Instead, we seek to estimate an overall effective first-order loss rate for indoor particles when the furnace is on that combines the filter efficiency, the recirculating flow rate, and the fan duty cycle. There is no extended period when the house is unoccupied and the furnace is on. So, instead, we apply the model in equation (3.7) for occupied periods when no indoor sources other than pilot lights operated.

The results are presented in Table 3.14. In summary, the analysis of the impact of outdoor particles on indoor UFP levels divides the conditions in this house into three configuration states. For each state, we have estimated a value of  $f_I$ , which represents the average indoor concentration of particles of outdoor origin divided by the average outdoor particle concentration during the same period. When the furnace is off and the windows and doors are all closed, the value of  $f_I$  is 0.16. When the bathroom window is open (and the furnace is off), the value of  $f_I$  is 0.25. One expects the opening of a window to increase air-exchange rate and thereby increase the indoor proportion of outdoor particles. When the doors and windows are closed and the furnace is on, the estimated value of  $f_I$  is 0.074. The lower value of  $f_I$  in this

case suggests that the furnace filter at this house is relatively efficient in removing ultrafine particles from the airstream that flows through it.

| Time (h)             | Configuration                                | Pa (h <sup>-1</sup> ) | k+a (h <sup>-1</sup> ) | $f_{I}\left( ight) ^{\mathrm{a}}$ |
|----------------------|--|-----------------------|------------------------|-----------------------------------|
| 17.4-24.4            | Bathroom window open                         | 0.085                 | 0.35                   | 0.25                              |
| 42.1-46.9            | All doors and windows closed                 | 0.055                 | 0.34                   | 0.16                              |
| Various <sup>b</sup> | Forced-air system on, doors & windows closed | 0.055                 | 0.75                   | 0.074                             |

Table 3.14. Results of investigating effect of outdoor particles on indoor levels at site H1.

<sup>a</sup> The average ratio of indoor particles of outdoor origin to outdoor particles estimated as Pa/(k+a).

<sup>b</sup> Analysis was conducted for three periods: 11.4-14.7 h, 35.3-38.8 h, and 54.2-65.9 h. The *Pa* value was assumed to be the same as for the configuration with all doors and windows closed. The k+a value was obtained as the constant parameter providing the best (least-squares) fit between modeled and measured concentrations for the three periods.

To investigate the significance of indoor sources at site H1, we first compiled information from the activity logs on potential indoor sources. These entries are summarized in Table 3.15. We then examined the time-series of indoor particle concentrations, looking for matches between an entry in the activity log and a sudden increase in the indoor particle concentration. Through this process, each of the peaks in the indoor particle concentration trace (see Figure 3.7) was matched to a coincident activity — either cooking or candle use. These labels are used to identify the peaks in subsequent discussion.

We found that several other recorded activities did not produce a clear change in the indoor particle level. Specifically, these activities were (i) use of a blender; (ii) use of a microwave oven; (iii) washing clothes; (iv) use of a toaster oven; (v) use of a dishwasher; (vi) use of a vented natural-gas clothes dryer; (vii) use of a rice cooker; (viii) cleaning with Windex and bleach; (ix) use of a vacuum cleaner; and (x) use of a coffee maker. The absence of a clear signal means that any emissions were insufficient to produce a clear change against the baseline indoor concentration, which was at least ~  $3 \times 10^3$  cm<sup>-3</sup> during daytime awake hours. In addition, in contrast to site H0, at H1 we did not see evidence that the central gas-fired furnace was a source of ultrafine particle emissions.

The data from these episodic increases in indoor PN concentrations were analyzed to quantitatively estimate source strength and particle loss rate. For each peak, the analysis proceeds in three stages. The analysis at this site was similar to that at site H0, with the key differences being in the determination of the net indoor PN concentration caused by the episodic source. At H1, we corrected for outdoor contributions using a configuration-specific factor,  $f_1$ , as detailed in Table 3.14. We also made configuration-specific corrections for the effects of pilot light emissions, using the steady-state material balance estimate.

| Time (h) | Activity   | PN source? |
|----------|--|------------|
| 0.93     | Wiped dining room table with Windex.                               | ? (peak a) |
| 0.93     | Halogen torch light on in living room.                             | ? (peak a) |
| 1.18     | Oven on low (<200 °F) to melt wax from menorahs. Microwave:        | Y (peak a) |
|          | pasta and cabbage soup.  |            |
| 3.93     | Boiled water with gas stove on low.                                | Y (peak b) |
| 5.77     | Made smoothie with blender.  | N          |
| 5.93     | Washed clothes, used toaster oven for pita bread, microwave water. | Ν          |
| 6.60     | Microwave: water.  | Ν          |
| 6.93     | Dishwasher.  | Ν          |
| 14.68    | Stove, microwave, toaster, coffeemaker: boiled water, cooked pasta | Y (peak c) |
|          | with hood fan on low, toasted mini pitas, made coffee              |            |
| 16.93    | Vented natural gas clothes dryer.                                  | N          |
| 25.43    | Rice cooker.   | N          |
| 26.02    | Stir fry on stove. Hood exhaust fan on low.                        | Y (peak d) |
| 27.60    | Toaster oven (pita bread), microwave.                              | N          |
| 30.68    | Cleaned bathroom with bleach and Windex.                           | N          |
| 38.60    | Boiled water on stove.   | Y (peak e) |
| 38.77    | Microwave, toaster (veggie patty sandwiches).                      | ? (peak e) |
| 41.93    | Electric slow cooker.  | N          |
| 47.10    | Vacuumed: bathroom, master bedroom.                                | Ν          |
| 48.93    | Stove, oven: 2 burners used intermittently, range hood fan on low. | Y (peak f) |
| 50.43    | Candles: 2 in dining room.   | Y (peak g) |
| 51.18    | Stove: boiled water, range hood fan on medium.                     | Y (peak g) |
| 51.93    | Dishwasher.  | N          |
| 53.18    | Clothes washer and dryer.  | N          |
| 53.68    | Microwave: water.  | N          |
| 54.43    | Clothes dryer.   | N          |
| 65.93    | Coffeemaker.   | Ν          |

**Table 3.15.** Activity entries in the occupant diary at house site H1 related to potential indoor particle emission sources.<sup>a</sup>

<sup>a</sup> The peak identifying letters (a-g) correspond to the labels on Figure 3.7.

Table 3.16 presents a summary of the analysis results for the seven identified peak events during observational monitoring at house site H1.

In sum, for these peak events, the observed particle loss rates (k+a) range from 0.8 h<sup>-1</sup> to 3.6 h<sup>-1</sup> with an average value of 1.8 h<sup>-1</sup> and a standard deviation of 0.9 h<sup>-1</sup>. The total particle emissions per event vary over a broader range, from 15 to 216 trillion  $(10^{12})$  particles each. The average was  $67 \times 10^{12}$  particles per event. In terms of potential contributions to exposure (the rightmost column of Table 3.16), the range for these events is from 37 to 448 in units of  $10^3$  cm<sup>-3</sup> h, and the average contribution is  $128 \times 10^3$  cm<sup>-3</sup> h.

| _               |                               | Time      | k + a            | $\sigma/V$               | σ                         | $(\sigma/V)/(k+a)$                 |
|-----------------|-------------------------------|-----------|------------------|--------------------------|---------------------------|------------------------------------|
| ID <sup>b</sup> | Source activity               | (h)       | $(h^{-1})$       | $(10^3 \text{ cm}^{-3})$ | $(10^{12} \mathrm{cm}^3)$ | $(10^3 \text{ cm}^{-3} \text{ h})$ |
| a1              | Oven on low &                 | 1.0-1.9   | 2.7 °            | (29.8)                   | (9.4)                     | (11.1)                             |
|                 | microwave                     |           |                  |                          |                           |                                    |
| a2              | Oven on low                   | 2.0-4.0   | 1.0 <sup>c</sup> | (24.6)                   | (7.8)                     | (25.5)                             |
| a               | Oven & micro                  | 1.0-4.0   |                  | 54.4                     | 17.2                      | 36.6                               |
| b               | Stove (water)                 | 4.1-5.9   | 1.6              | 73.8                     | 23.3                      | 46.0                               |
| с               | Stove <sup>d</sup>            | 14.7-16.6 | 3.6              | 187                      | 58.9                      | 51.4                               |
| d               | Stove (stir fry) <sup>d</sup> | 25.9-29.0 | 0.8              | 75.3                     | 23.7                      | 99.2                               |
| e               | Stove (water)                 | 38.8-41.4 | 1.3              | 48.9                     | 15.4                      | 38.0                               |
| f               | Stove <sup>d</sup> & oven     | 48.8-49.8 | 2.1              | 363                      | 114                       | 175                                |
| g               | Stove <sup>d</sup> & candles  | 49.8-54.2 | 1.5              | 686                      | 216                       | 448                                |

Table 3.16. Analysis of indoor UFP sources at house site H1 from observational monitoring.<sup>a</sup>

<sup>a</sup> Parameter definitions: k+a = total loss rate coefficient for particles emitted during the event; $<math>\sigma/V = \text{total particle number per house volume emitted;} \sigma = \text{total particle number emitted;}$ 

 $(\sigma/V)/(k+a) =$  contribution to exposure for an occupant present for the entire event.

<sup>b</sup> The labels correspond to those on peaks displayed in Figure 3.7.

<sup>c</sup> The decays of the subpeaks were separately modeled for events a1 and a2.

<sup>d</sup> The range hood exhaust fan was on during this cooking event.

Table 3.17 summarizes the results of the two manipulation experiments conducted to investigate source emissions at this study site. Three experiments were conducted: (M1) boiling water on the gas range; (M2) using two candles; and (M3) toasting bread in the electric toaster oven. In the first two cases, there was a clear signal of an increase in indoor particle number concentration, so these activities were confirmed as particle sources. The third experiment did not produce a clear increase in indoor particle concentrations against the 5-10 × 10<sup>3</sup> cm<sup>-3</sup> background. The two quantifiable experiments show particle decay rates that are within the range, although higher than average, than those obtained from peak analysis during the observational monitoring period. The total particle emission rate,  $\sigma$ , is also within the range, but less than the average obtained from analysis of the observational monitoring peaks.

|    |                           | Duration | k + a      | $\sigma/V$                 | σ           |
|----|---------------------------|----------|------------|----------------------------|-------------|
| ID | Source                    | (h)      | $(h^{-1})$ | $(10^3  \mathrm{cm}^{-3})$ | $(10^{12})$ |
| M1 | Stove (boil water 17 min) | 2.0      | 2.8        | 475                        | 21.4        |
| M2 | Two candles, 25 min       | 1.9      | 2.7        | 1150                       | 51.6        |

**Table 3.17.** Analysis of UFP source from manipulation experiments at site H1.<sup>a</sup>

<sup>a</sup> A third source experiment (M3) was conducted using an electric toaster oven to prepare toast. It was not seen to be a significant emission source.

An analysis was conducted to estimate residential exposures to particles for the occupants of this household for the observational monitoring period and to apportion those exposures to known indoor sources and to indoor particles of outdoor origin. The results are presented in Table 3.18. Cumulative exposures while "awake at home" and "asleep at home" are obtained as the product of the appropriate indoor time-averaged PN concentration and the duration of occupancy awake or asleep. We used PN\_in1 as the best indicator of indoor exposure

concentration during awake and asleep periods. The cumulative exposures were summed to obtain the "total cumulative exposure" for the time spent at home:  $1,219 \times 10^3$  cm<sup>-3</sup> h for R1, R3, and R4; and 898 × 10<sup>3</sup> cm<sup>-3</sup> h for R2. Daily exposure rates were obtained by dividing these figures by the approximate duration of household monitoring, 2.8 days. These measures and the results —  $436 \times 10^3$  cm<sup>-3</sup> h/d (for R1, R3, and R4) and  $321 \times 10^3$  cm<sup>-3</sup> h/d (for R2) — are designed to be independent of the duration of observational monitoring. The contribution of outdoor particles to exposure was estimated to be the same for each occupant:  $131 \times 10^3$  cm<sup>-3</sup> h, which corresponds to 11-15% of the total cumulative exposure.

| Occupancy status           Time at home, awake (h)         25.5         20.3         17.0         17.0           Time at home, asleep (h)         24.5         23.7         33.0         33.0           Time away from home (h)         17.2         23.2         17.2         17.2           Exposure duration (d) a         2.8         2.8         2.8         2.8         2.8           Average exposure concentrations and exposures   | Parameter   | R1                       | R2       | R3         | R4    |      |
|---|---|--------------------------|----------|------------|-------|------|
| Time at home, asleep (h)24.523.733.033.0Time away from home (h)17.223.217.217.2Exposure duration (d) a2.82.82.82.82.8Average exposure concentrations and exposures2.82.82.82.82.8Average concentration (PN_in1), indoor awake (10 <sup>3</sup> cm <sup>-3</sup> )42.638.250.950.9Average concentration (PN_in1), indoor asleep (10 <sup>3</sup> cm <sup>-3</sup> )5.45.210.710.7Cumulative exposure (10 <sup>3</sup> cm <sup>-3</sup> h)1,2198981,2191,219Cumulative exposure attributable to particles of outdoor origin11%131131131Indoor exposure attributable to particles of outdoor origin11%15%11%11%Indoor exposure attributable to indoor continuous source (gas stove and oven pilot lights)119119119119Cumulative contribution to exposure (10 <sup>3</sup> cm <sup>-3</sup> h)179169179179Percentage attributable to indoor source peaks (10 <sup>3</sup> cm <sup>-3</sup> h)179169179179Percentage attributable to indoor source peaks (10 <sup>3</sup> cm <sup>-3</sup> h)179169179179Peak a — melt wax in gas oven, microwave3703737Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water7382727Peak d — stir fry on gas stove99719999Peak f & g — gas stove and oven use; candle use624 <td>Occupancy status</td> <td></td> <td></td> <td></td> <td></td> | Occupancy status  |                          |          |            |       |      |
| Time away from home (h)17.223.217.217.2Exposure duration (d) a2.82.82.82.82.8Average exposure concentrations and exposuresAverage exposure concentration (PN_in1), indoor awake ( $10^3$ cm <sup>-3</sup> )42.638.250.950.9Average concentration (PN_in1), indoor asleep ( $10^3$ cm <sup>-3</sup> )5.45.210.710.7Cumulative exposure ( $10^3$ cm <sup>-3</sup> h)1,2198981,2191,219Cumulative exposure rate ( $10^3$ cm <sup>-3</sup> h d <sup>-1</sup> )436321436436Indoor exposure attributable to particles of outdoor origin11%131131131Percentage attributable to particles of outdoor origin11%15%11%11%Indoor exposure attributable to indoor continuous source (gas stove and oven pilot lights)Cumulative contribution to exposure ( $10^3$ cm <sup>-3</sup> h)179169179179Percentage attributable to indoor source peaks ( $10^3$ cm <sup>-3</sup> h)179169179179Percentage attributable to indoor source peaks ( $10^3$ cm <sup>-3</sup> h)179169179179Peak a — melt wax in gas oven, microwave3703737Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak e — toaster and microwave use; boil water on gas stove27382727Peaks f & g — gas stov   | Time at home, awake (h)   | 25.5                     | 20.3     | 17.0       | 17.0  |      |
| Exposure duration (d) a2.82.82.82.8Average exposure concentrations and exposuresAverage concentration (PN_in1), indoor awake ( $10^3  {\rm cm}^{-3}$ )42.638.250.950.9Average concentration (PN_in1), indoor asleep ( $10^3  {\rm cm}^{-3}$ )5.45.210.710.7Cumulative exposure ( $10^3  {\rm cm}^{-3}  {\rm h}$ )1,2198981,2191,219Cumulative exposure rate ( $10^3  {\rm cm}^{-3}  {\rm h}  {\rm d}^{-1}$ )436321436436Indoor exposure attributable to particles of outdoor originCumulative contribution to exposure ( $10^3  {\rm cm}^{-3}  {\rm h}$ )131131131131Percentage attributable to particles of outdoor originII with the particles of outdoor origin <td colsp<="" td=""><td>Time at home, asleep (h)</td><td>24.5</td><td>23.7</td><td>33.0</td><td>33.0</td></td>          | <td>Time at home, asleep (h)</td> <td>24.5</td> <td>23.7</td> <td>33.0</td> <td>33.0</td> | Time at home, asleep (h) | 24.5     | 23.7       | 33.0  | 33.0 |
| Exposure duration (d) a2.82.82.82.8Average exposure concentrations and exposuresAverage concentration (PN_in1), indoor awake ( $10^3  {\rm cm}^{-3}$ )42.638.250.950.9Average concentration (PN_in1), indoor asleep ( $10^3  {\rm cm}^{-3}$ )5.45.210.710.7Cumulative exposure ( $10^3  {\rm cm}^{-3}  {\rm h}$ )1,2198981,2191,219Cumulative exposure rate ( $10^3  {\rm cm}^{-3}  {\rm h}  {\rm d}^{-1}$ )436321436436Indoor exposure attributable to particles of outdoor originCumulative contribution to exposure ( $10^3  {\rm cm}^{-3}  {\rm h}$ )131131131131Percentage attributable to particles of outdoor originII with the particles of outdoor origin <td colsp<="" td=""><td>Time away from home (h)</td><td>17.2</td><td>23.2</td><td>17.2</td><td>17.2</td></td>           | <td>Time away from home (h)</td> <td>17.2</td> <td>23.2</td> <td>17.2</td> <td>17.2</td>  | Time away from home (h)  | 17.2     | 23.2       | 17.2  | 17.2 |
| Average concentration (PN_in1), indoor awake $(10^3 \text{ cm}^{-3})$ 42.638.250.950.9Average concentration (PN_in1), indoor asleep $(10^3 \text{ cm}^{-3})$ 5.45.210.710.7Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$ 1,2198981,2191,219Cumulative exposure rate $(10^3 \text{ cm}^{-3} \text{ h} \text{ d}^{-1})$ 436321436436Indoor exposure attributable to particles of outdoor origin131131131131Percentage attributable to particles of outdoor origin11%15%11%11%Indoor exposure attributable to particles of outdoor origin11%15%11%11%Indoor exposure attributable to indoor continuous source (gas stove and oven pilot lights)Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$ 179169179179Percentage attributable to indoor source peaks $(10^3 \text{ cm}^{-3} \text{ h})$ 17916917915%15%Exposure attributable to indoor source peaks $(10^3 \text{ cm}^{-3} \text{ h})$ 19%15%15%15%Peak a — melt wax in gas oven, microwave3703737Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak d — stir fry on gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources8795818798  |   | 2.8                      | 2.8      | 2.8        | 2.8   |      |
| Average concentration (PN_in1), indoor asleep ( $10^3$ cm <sup>-3</sup> )5.45.210.710.7Cumulative exposure ( $10^3$ cm <sup>-3</sup> h)1,2198981,2191,219Cumulative exposure rate ( $10^3$ cm <sup>-3</sup> h) d <sup>-1</sup> )436321436436Indoor exposure attributable to particles of outdoor origin131131131131Percentage attributable to particles of outdoor origin11%15%11%11%Indoor exposure attributable to particles of outdoor origin11%15%11%11%Indoor exposure attributable to indoor continuous source (gas stove and oven pilot lights)Cumulative contribution to exposure ( $10^3$ cm <sup>-3</sup> h)179169179179Percentage attributable to indoor source peaks ( $10^3$ cm <sup>-3</sup> h)17916917915%15%Exposure attributable to indoor source peaks ( $10^3$ cm <sup>-3</sup> h)179169179179Peak a — melt wax in gas oven, microwave3703737Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak d — stir fry on gas stove99719999Peak f & g — gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%  | Average exposure concentrations and exposures   |                          |          |            |       |      |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$ 1,2198981,2191,219Cumulative exposure rate $(10^3 \text{ cm}^{-3} \text{ h} \text{ d}^{-1})$ 436321436436Indoor exposure attributable to particles of outdoor origin131131131131Percentage attributable to particles of outdoor origin11%15%11%11%Indoor exposure attributable to particles of outdoor origin11%15%11%11%Indoor exposure attributable to indoor continuous source (gas stove and oven pilot lights)179169179179Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$ 179169179179Percentage attributable to continuous indoor source15%19%15%15%Exposure attributable to indoor source peaks $(10^3 \text{ cm}^{-3} \text{ h})$ 179169179179Peak a — melt wax in gas oven, microwave3703737Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak d — stir fry on gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin103° m <sup>-3</sup> h)29172929  | Average concentration (PN_in1), indoor awake $(10^3 \text{ cm}^{-3})$                     | 42.6                     | 38.2     | 50.9       | 50.9  |      |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$ 1,2198981,2191,219Cumulative exposure rate $(10^3 \text{ cm}^{-3} \text{ h} \text{ d}^{-1})$ 436321436436Indoor exposure attributable to particles of outdoor origin131131131131Percentage attributable to particles of outdoor origin11%15%11%11%Indoor exposure attributable to particles of outdoor origin11%15%11%11%Indoor exposure attributable to indoor continuous source (gas stove and oven pilot lights)179169179179Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$ 179169179179Percentage attributable to continuous indoor source15%19%15%15%Exposure attributable to indoor source peaks $(10^3 \text{ cm}^{-3} \text{ h})$ 179169179179Peak a — melt wax in gas oven, microwave3703737Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak d — stir fry on gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin103° m <sup>-3</sup> h)29172929  | Average concentration (PN_in1), indoor asleep $(10^3 \text{ cm}^{-3})$                    | 5.4                      | 5.2      | 10.7       | 10.7  |      |
| Indoor exposure attributable to particles of outdoor originCumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$ 131131131131Percentage attributable to particles of outdoor origin11%15%11%11%Indoor exposure attributable to indoor continuous source (gas stove and oven pilot lights)179169179179Percentage attributable to continuous indoor source15%19%15%15%15%Exposure attributable to indoor source peaks ( $10^3 \text{ cm}^{-3} \text{ h}$ )179169179179Peak a — melt wax in gas oven, microwave3703737Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak f & g — gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin1029172929  | Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                                    | 1,219                    | 898      | 1,219      | 1,219 |      |
| Indoor exposure attributable to particles of outdoor originCumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$ 131131131131Percentage attributable to particles of outdoor origin11%15%11%11%Indoor exposure attributable to indoor continuous source (gas stove and oven pilot lights)179169179179Percentage attributable to continuous indoor source15%19%15%15%15%Exposure attributable to indoor source peaks ( $10^3 \text{ cm}^{-3} \text{ h}$ )179169179179Peak a — melt wax in gas oven, microwave3703737Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak f & g — gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin1029172929  | Cumulative exposure rate $(10^3 \text{ cm}^{-3} \text{ h d}^{-1})$                        | 436                      | 321      | 436        | 436   |      |
| Percentage attributable to particles of outdoor origin11%15%11%11%Indoor exposure attributable to indoor continuous source (gas stove and oven pilot lights)Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$ 179169179179Percentage attributable to continuous indoor source15%19%15%15%15%Exposure attributable to indoor source peaks $(10^3 \text{ cm}^{-3} \text{ h})$ 17916917915%15%Peak a — melt wax in gas oven, microwave3703737Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak d — stir fry on gas stove use; boil water on gas stove27382727Peaks f & g — gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin2917292929   | Indoor exposure attributable to particles of outdoor origin                               |                          |          |            |       |      |
| Percentage attributable to particles of outdoor origin11%15%11%11%Indoor exposure attributable to indoor continuous source (gas stove and oven pilot lights)Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$ 179169179179Percentage attributable to continuous indoor source15%19%15%15%15%Exposure attributable to indoor source peaks $(10^3 \text{ cm}^{-3} \text{ h})$ 17916917915%15%Peak a — melt wax in gas oven, microwave3703737Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak d — stir fry on gas stove use; boil water on gas stove27382727Peaks f & g — gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin2917292929   | Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$                    | 131                      | 131      | 131        | 131   |      |
| Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$ 179169179179Percentage attributable to continuous indoor source15%19%15%15%Exposure attributable to indoor source peaks $(10^3 \text{ cm}^{-3} \text{ h})$ 15%15%15%Peak a — melt wax in gas oven, microwave3703737Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak d — stir fry on gas stove99719999Peak f $\&$ g — gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin29172929   |   | 11%                      | 15%      | 11%        | 11%   |      |
| Percentage attributable to continuous indoor source15%19%15%15%Exposure attributable to indoor source peaks $(10^3 \text{ cm}^{-3} h)$ 703737Peak a — melt wax in gas oven, microwave3703737Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak d — stir fry on gas stove99719999Peak e — toaster and microwave use; boil water on gas stove27382727Peaks f & g — gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin29172929  | Indoor exposure attributable to indoor continuous source (gas                             | stove an                 | d oven p | ilot light | s)    |      |
| Exposure attributable to indoor source peaks $(10^3 \text{ cm}^{-3} h)$ Peak a — melt wax in gas oven, microwave3703737Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak d — stir fry on gas stove99719999Peak e — toaster and microwave use; boil water on gas stove27382727Peaks f & g — gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin29172929   | Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$                    | 179                      | 169      | 179        | 179   |      |
| Peak a — melt wax in gas oven, microwave $37$ $0$ $37$ $37$ Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak d — stir fry on gas stove99719999Peak e — toaster and microwave use; boil water on gas stove27382727Peaks f & g — gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin29172929  |   | 15%                      | 19%      | 15%        | 15%   |      |
| Peak a — melt wax in gas oven, microwave $37$ $0$ $37$ $37$ Peak b — boil water on gas stove46104646Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak d — stir fry on gas stove99719999Peak e — toaster and microwave use; boil water on gas stove27382727Peaks f & g — gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin29172929  | Exposure attributable to indoor source peaks $(10^3 \text{ cm}^{-3} h)$                   |                          |          |            |       |      |
| Peak c — electric coffeemaker and toaster use; boil water47514747and cook pasta on gas stove99719999Peak d — stir fry on gas stove99719999Peak e — toaster and microwave use; boil water on gas stove27382727Peaks f & g — gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin29172929  |   | 37                       | 0        | 37         | 37    |      |
| and cook pasta on gas stoveImage: matrix on gas stoveImage: matrix on gas stoveImage: matrix on gas stovePeak d — stir fry on gas stove99719999Peak e — toaster and microwave use; boil water on gas stove27382727Peaks f & g — gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin29172929   |   | 46                       | 10       | 46         | 46    |      |
| Peak d — stir fry on gas stove99719999Peak e — toaster and microwave use; boil water on gas stove27382727Peaks f & g — gas stove and oven use; candle use624410624624Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin29172929  | Peak c — electric coffeemaker and toaster use; boil water                                 | 47                       | 51       | 47         | 47    |      |
| Peak e — toaster and microwave use; boil water on gas stove $27$ $38$ $27$ $27$ Peaks f & g — gas stove and oven use; candle use $624$ $410$ $624$ $624$ Cumulative exposure attributable to episodic indoor sources $879$ $581$ $879$ $879$ Percentage attributable to quantified episodic indoor sources $72\%$ $65\%$ $72\%$ $72\%$ Indoor exposure of unknown origin $29$ $17$ $29$ $29$  | and cook pasta on gas stove   |                          |          |            |       |      |
| Peaks f & g — gas stove and oven use; candle use $624$ $410$ $624$ $624$ Cumulative exposure attributable to episodic indoor sources $879$ $581$ $879$ $879$ Percentage attributable to quantified episodic indoor sources $72\%$ $65\%$ $72\%$ $72\%$ Indoor exposure of unknown origin $29$ $17$ $29$ $29$  | Peak d — stir fry on gas stove  | 99                       | 71       | 99         | 99    |      |
| Cumulative exposure attributable to episodic indoor sources879581879879Percentage attributable to quantified episodic indoor sources72%65%72%72%Indoor exposure of unknown origin29172929   | Peak e — toaster and microwave use; boil water on gas stove                               | 27                       | 38       | 27         | 27    |      |
| Percentage attributable to quantified episodic indoor sources $72\%$ $65\%$ $72\%$ $72\%$ Indoor exposure of unknown originUndoor exposure of unknown originCumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$ $29$ $17$ $29$ $29$  | Peaks f & g — gas stove and oven use; candle use  | 624                      | 410      | 624        | 624   |      |
| Indoor exposure of unknown originCumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$ 291729  | Cumulative exposure attributable to episodic indoor sources                               | 879                      | 581      | 879        | 879   |      |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$ 29       17       29       29  | Percentage attributable to quantified episodic indoor sources                             | 72%                      | 65%      | 72%        | 72%   |      |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$ 29       17       29       29  | Indoor exposure of unknown origin   |                          |          |            |       |      |
|   | Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                                    | 29                       | 17       | 29         | 29    |      |
|   |   | 2%                       | 2%       | 2%         | 2%    |      |

Table 3.18. Exposure analysis for residents of house site H1 during observational monitoring.

<sup>a</sup> Exposure duration is computed as the total monitoring period, minus 24 h per day not slept at home, with the result divided by 24 h/d.

The contribution to exposure from the gas pilot lights was estimated by first estimating the contribution of pilot lights to the indoor concentration as a function of time, using a steady-

state material balance with a removal rate depending on the house configuration, and then summing the concentration increments for each occupant while that person was present at home. The result is  $169 \times 10^3$  cm<sup>-3</sup> h for R2 and  $179 \times 10^3$  cm<sup>-3</sup> h for each of the other three inhabitants. These exposure increments constitute 19% and 15% respectively of the total. The contributions to exposure from the seven peak events were assessed with results of  $581 \times 10^3$  cm<sup>-3</sup> h (65% of total) for R2 and 879  $\times 10^3$  cm<sup>-3</sup> h (72%) for R1, R3, and R4. The small remainders of the estimated exposures that are of unknown origin only amount to  $\sim 2\%$  of the total in each case.

## 3.2.3. Site H2

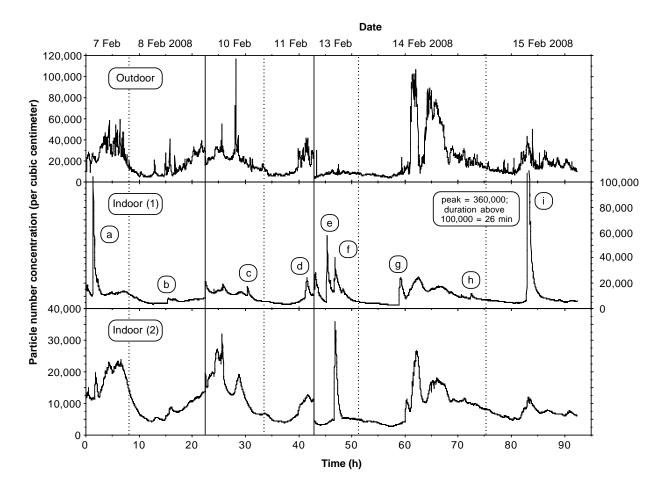
Observational monitoring was initiated at house site H2 at 16:01 on 7 February 2008 and, with two interruptions, was completed at 16:54 on 15 February. The total duration of observational monitoring was 92 h. The reader is referred to \$2.2.1 for detailed background information about the house site. See also Figure C.3 (Appendix C) for a floor plan. Summarizing briefly, this house has two floors, a main upper floor with an internal volume of about 250 m<sup>3</sup> and a lower floor with an attached garage and supplementary rooms. The home is sited in a residential neighborhood within the urban core of Oakland, at an elevation between 50 and 100 m above sea level. A moderately busy arterial road is located about 50 m away and the nearest freeways are 500 m to the NE and 1000 m to the SW. The house has natural-gas cooking appliances without pilot lights. It has a wall furnace that is fired by natural gas and vented. The house has four occupants, two adults (R1 = female adult; R2 = male adult) and two male children (R3 ~ 3 y old; R4 ~ 5 y old).

A summary of the time-averaged species concentration and cofactor values was presented in Table 3.1 (§3.1). The average particle number concentrations measured during the entire observational monitoring period were 17.9 (outdoors), 11.0 (indoors primary), and 8.9 (indoors supplemental) in units of 1000 particles per cm<sup>3</sup>. The outdoor concentrations at this site during observational monitoring were the median among the seven house sites in this study. The timeaveraged indoor concentrations were close to the median, based on the primary indoor monitor.

Figure 3.11 shows the time-series of particle number concentrations measured by the three monitors. The horizontal axis is labeled with elapsed time of monitoring referenced to the start time of 16:01 on 7 February 2008. Vertical solid lines denote the two breaks in the monitoring period. We have adopted a convention such that the time scale measures the elapsed time of active monitoring. This scale is suspended during the breaks. The vertical dashed lines demark midnight and distance from these lines can be used to gauge clock time within a period of continuous monitoring.

The outdoor concentrations at this site vary between about ~  $5,000 \text{ cm}^{-3}$  and ~  $100,000 \text{ cm}^{-3}$ . There is a trend of declining concentrations during the night: the slope of the outdoor PN concentration trace is negative for all four midnights. A strong, short-lived peak on the evening of 10 February is evident. Its short duration suggests that it may have been caused by a local source. A pair of pronounced peaks of several-hour duration is also seen in the outdoor signal on 14 February in the middle of the day.

Indoor PN concentrations exhibit a complex pattern. Focusing on "Indoor (1)," which represents concentrations measured on the main floor of the house, there are a few prominent peaks and several smaller peaks overlaid on time trends that follow the broad outdoor pattern. Because air exchange in houses occurs on a time scale of hours, the high frequency fluctuations observed in outdoor concentrations would not be expected indoors. Instead, the indoor PN concentration caused by the penetration of outdoor particles should follow the time-average trends with an averaging period of a few h and with some decrease in concentration because of penetration and deposition losses indoors. At H0 and H1, the presented PN concentration figures did not reveal this behavior because of the strong dominance of indoor sources. At H2, the indoor sources are less prominent and so the dependence of indoor concentrations on outdoor levels is more clearly seen in the time traces.



**Figure 3.11.** Time-series of particle number concentrations during observational monitoring at house site H2. Monitor "Indoor (1)" was located on the main floor (upstairs) in the family room; monitor "Indoor (2)" sampled from a downstairs utility room. Note that the vertical scales are different among frames. The solid vertical lines demark interruptions in the period of monitoring. Peaks in the indoor PN concentration are labeled with lower case letters (a-i) to facilitate later discussion.

We have labeled nine peaks in the PN concentration data for the primary indoor monitor based on the inference that these indicate episodic emissions from indoor sources. For most of these — notably a, d, e, f, g, and i — the peaks are unambiguous and are clearly not linked to the outdoor concentration trace. However, for three cases — b, c, and h — the identification of peaks related to episodic emissions was made iteratively, incorporating information on activities and utility use in the house. As will be discussed in detail later, we find that cooking is a particle source in this house, being associated with peaks a and i. The operation of the wall heater is associated with each of the remaining peaks. The clothes dryer was operated close to the time of appearance of peak f and may have contributed to it. Some small excursions in the indoor PN concentration signal might also have been labeled as episodic emission events. They were not because (a) the PN signals themselves were ambiguous; and (b) there was no corroborating evidence of any potential indoor sources acting at the same time.

The supplemental indoor monitor ("Indoor (2)") sampled from the downstairs area of the house, which had a guest bedroom, but was otherwise used more as utility space than living

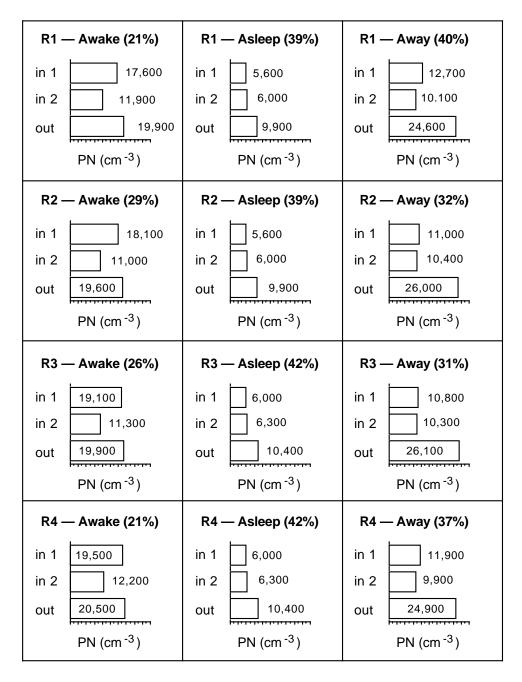
space. The downstairs utility room, in which the supplemental monitor was sited, had a clothes washer and dryer. It is also immediately adjacent to an attached garage. A comparison of the traces for "Indoor (1)" and "Indoor (2)" in Figure 3.11 suggests that the upstairs and downstairs spaces are poorly coupled in an air-exchange sense. Peaks a, i, and j upstairs exhibit weak echoes in the downstairs signal. Peak f has the strongest suggestion of significant mixing between the two zones. However, it is also possible that the upstairs and downstairs peaks at this time are from different causes, with the operation of the wall furnace causing the upstairs peak and the clothes dryer responsible for the downstairs peak. Because of the weak coupling of the zones and because most of the living in this house occurs upstairs, we focus on data from the primary indoor monitor ("Indoor (1)") for characterizing dynamic behavior of indoor particles and in assessing and apportioning indoor exposures for the occupants of this house.

The time series of occupancy data for the four residents of H2 is presented in Appendix D (see Figure D.29). Figure 3.12 shows the time-averaged particle number concentrations measured over the full observational monitoring period as sorted by the status of each occupant. Focusing on the primary indoor monitor, "in 1," we see that the average concentrations when occupants are at home awake,  $18-20 \times 10^3$  cm<sup>-3</sup>, are roughly three times higher than the average concentrations when occupants are at home asleep, ~  $6 \times 10^3$  cm<sup>-3</sup>. We also see that the outdoor concentrations are about twice as high when occupants are awake at home,  $19-21 \times 10^3$  cm<sup>-3</sup>, as when they are asleep, ~  $10 \times 10^3$  cm<sup>-3</sup>. The I/O ratios based on average concentrations "in 1" and "out" are in the range 89-96% when occupants are at home awake, 57-58% when occupants are at home asleep, and 41-51% when occupants are away from home.

Table 3.19 summarizes the average copollutant levels at H2, sorted according to the occupancy status of the house. In Appendix D are presented time-series plots for the copollutants as follows: Figure D.30 for ozone; Figure D.31 for NO; Figure D.32 for carbon dioxide; and Figure D.33 for carbon monoxide. At elapsed time ~ 82 h, the data exhibit significant indoor peaks for particle number concentration (peak i, Figure 3.11), NO (Figure D.31), CO<sub>2</sub> (Figure 3.32), and CO (Figure 3.33). As noted later, this corresponds to one of only two noted cooking events at this house.

At this site, we made five determinations of the air-exchange rate using carbon dioxide as a tracer gas. Three of these occurred during the observational monitoring period, when an elevated  $CO_2$  level caused by occupants was followed by a period of vacancy during which the  $CO_2$  decay influenced only by air exchange could be observed. The other two determinations (A1 and A2) were made from manipulation experiments. The results, summarized in Table 3.20, show moderately high air-exchange rate during observational monitoring, with the three readings of 0.5, 0.7, and 1.3 h<sup>-1</sup>. The highest value occurs at a time when the wind speed was much higher than average. Lower values of the air-exchange rate are obtained during manipulation experiments, 0.2 and 0.3 h<sup>-1</sup>. In all of these experiments, all exterior doors and windows in the house were closed.

To explore the influence of outdoor particles on indoor PN concentrations, we compute the ratio of indoor to outdoor particle concentrations, based on time-averaged concentration levels, during times when the house is unoccupied and when there is no clear residual influence of particles from indoor sources. We restricted our attention to times when the dominant configuration of the house applied, namely when all doors and windows were closed.



**Figure 3.12.** Time-averaged indoor and outdoor particle number concentrations measured during the observational monitoring period at house site H2 and sorted according to the occupancy status of the four inhabitants of the house. Occupant R1 is an adult female, occupant R2 is an adult male, and occupants R3 and R4 are male children. Each bar is labeled with the time-averaged particle number concentration and each frame indicates the percentage of time for that resident for which the occupancy state applies.

| Species (units)           | All awake | All asleep | Unoccupied |
|---------------------------|-----------|------------|------------|
| NO_in (ppb)               | 15.3      | 1.8        | 14.3       |
| NO_out (ppb)              | 21.0      | 12.6       | 29.0       |
| O <sub>3</sub> _in (ppb)  | 3.5       | 2.6        | 3.7        |
| O <sub>3</sub> _out (ppb) | 16.9      | 15.1       | 20.6       |
| CO <sub>2</sub> _in (ppm) | 804       | 532        | 591        |

**Table 3.19.** Copollutant levels at site H2 during observational monitoring.<sup>a</sup>

<sup>a</sup> Measurements sorted according to occupancy status of the building and then averaged.

**Table 3.20.** Air-exchange rates (AER) determined by carbon dioxide concentration decay in house site H2.

| Time (h)    | AER $(h^{-1})$    | WS (m s <sup>-1</sup> ) | $ \Delta \mathbf{T} $ (°C) | House configuration          |
|-------------|-------------------|-------------------------|----------------------------|------------------------------|
| 3.3-4.8     | 0.72              | 1.3                     | 10.1                       | All windows and doors closed |
| 17.0-18.3   | 0.51              | 0.9                     | 8.5                        | All windows and doors closed |
| 61.7-63.2   | 1.27              | 6.7                     | 5.3                        | All windows and doors closed |
| A1 (11 Feb) | 0.20 <sup>a</sup> | 1.8                     | 11.6                       | All windows and doors closed |
| A2 (12 Feb) | 0.30 <sup>a</sup> | 2.6                     | na                         | All doors and windows closed |

<sup>a</sup> For A1, the carbon dioxide tracer gas was released both upstairs and downstairs. The decay is based on a volume-weighted average concentration measured in the upstairs and downstairs zones. For A2, the  $CO_2$  release occurred only upstairs and the air-exchange rate determination is based on decay in this zone only.

Six suitable intervals were identified, distributed throughout the observational monitoring period, with a total duration of 28 h. The results are presented in Table 3.21. For these periods, the ratio of average indoor to average outdoor concentrations ranges from 0.32 to 2.0 with a time-weighted average of  $f_1 = 0.51$  for the main indoor monitor (PN\_in1) and from 0.33 to 0.65 with a time-weighted average of  $f_2 = 0.42$  for the supplementary monitor (PN\_in2). Treating each interval as a separate determination, the weighted standard error for  $f_1$  is 0.18, or 35% of the mean. For  $f_2$ , the standard error is 0.038, 9% of the weighted-average estimate of 0.42.

| Time                 | WS                      | $ \Delta \mathbf{T} $ | PN_out                   | PN_in1                   | PN_in2                   | $f_{I}$ | $f_2$ |
|----------------------|-------------------------|-----------------------|--------------------------|--------------------------|--------------------------|---------|-------|
| ( <b>h</b> )         | $(m s^{-1})$            | (°C)                  | $(10^3 \text{ cm}^{-3})$ | $(10^3 \text{ cm}^{-3})$ | $(10^3 \text{ cm}^{-3})$ | (—)     | (—)   |
| 3.5-4.8              | 1.3                     | 10.1                  | 35.4                     | 11.5                     | 19.3                     | 0.32    | 0.54  |
| 17.1-22.4            | 1.3                     | 5.1                   | 20.4                     | 6.8                      | 9.6                      | 0.33    | 0.47  |
| 28.4-30.2            | 0.7                     | 8.8                   | 21.4                     | 12.1                     | 14.0                     | 0.56    | 0.65  |
| 43.3-45.4            | 6.4                     | 3.6                   | 6.3                      | 12.6                     | 3.1                      | 2.02    | 0.50  |
| 61.4-72.4            | 3.9                     | 5.9                   | 39.9                     | 14.6                     | 13.1                     | 0.36    | 0.33  |
| 85.4-92.3            | 2.5                     | 3.1                   | 15.7                     | 6.7                      | 6.4                      | 0.43    | 0.41  |
| Average <sup>b</sup> | <b>2.9</b> <sup>c</sup> | 5.4 <sup>c</sup>      | 26.5                     | 10.7                     | 10.4                     | 0.51    | 0.42  |

**Table 3.21.** Analysis of the indoor to outdoor particle concentration ratios at house site H2 for periods when the house is unoccupied and all doors and windows are closed.<sup>a</sup>

<sup>a</sup> The parameter  $f_1$  represents the ratio of the averages for PN\_in1 to PN\_out; the parameter  $f_2$  represents the ratio of the averages for PN\_in2 to PN\_out.

<sup>b</sup> The final row of the table presents time-weighted average values for each entry.

<sup>c</sup> Note that the overall average for the observational monitoring for wind speed is 2.6 m/s; for the indoor and outdoor temperatures are 20.0 and 13.2 °C, respectively.

At this site, the occupants only recorded two source-related activities during observational monitoring. As summarized in Table 3.22, both were cooking events on their natural gas stove. Both of these events coincided with and very likely were the cause of indoor PN concentration peaks, labeled as "a" and "i" in Figure 3.11. The occupants reported that the range hood fan was not used during observational monitoring at this site.

We deployed temperature sensing data loggers at this site on the discharge register of the vented natural-gas wall heater and on the natural gas clothes dryer. The wall heater seems clearly to be a source of indoor particles, based on the association of its use with concentration peaks, as illustrated in Figure 3.13. The clothes dryer was used one time during observational monitoring, at a time that coincided with peak f. Note from Figure 3.11 that this is the strongest peak in the downstairs monitor signal. Since the upstairs and downstairs zones of the house appear substantially decoupled, in analyzing indoor emission events, we treat the upstairs peaks b-h as originating from the wall furnace and the downstairs peak f as being caused by the clothes dryer. The emissions characterization analysis is done assuming that the two zones are independent ( $V \sim 250$  m<sup>3</sup> upstairs and  $V \sim 53$  m<sup>3</sup> for the downstairs utility room and bedroom).

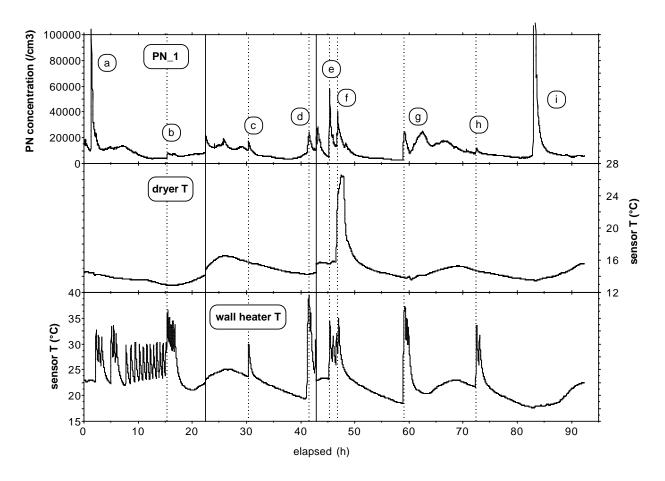
| <b>Tuble 3.22.</b> Relivity notes related to potential Tr emission sources at |                                  |            |  |  |
|---|----------------------------------|------------|--|--|
| Time (h)  | Activity                         | PN source? |  |  |
| 0.98  | Fried egg on gas cook stove      | Y (peak a) |  |  |
| 82.90   | Boiled carrots on gas cook stove | Y (peak i) |  |  |

Table 3.22. Activity notes related to potential PN emission sources at site H2.

For most of the peaks, the analysis is done in the same fashion as for the indoor episodic peaks in H0 and H1. The results are summarized in Table 3.23. A net indoor concentration is computed by subtracting off the contribution from outdoor particles. An exponential curve is fit to the decay period to estimate a total decay rate, k + a, for the particles emitted indoors. The area under the curve of net indoor PN level versus time is integrated for the duration of the peak episode to estimate the emissions per volume,  $\sigma/V$ . And finally, the total emissions ( $\sigma$ ) and total potential exposure [( $\sigma/V$ )/(k+a)] are computed.

For the smaller peaks — b, c, d, and h — this approach does not work because a net indoor concentration cannot be accurately computed. So, in these cases, an emissions strength per unit volume ( $\sigma/V$ ) is estimated as the difference between peak and immediate pre-peak concentration. To estimate the potential contribution to exposure, we use the average k+a of 1.6 per h, based on the other five cases in which a determination can be made.

In sum, using a time-weighted average of  $1.8 \text{ h}^{-1}$  to represent the decay rate for case i, for upstairs peak events a, e, f1, g, and i, the observed particle loss rates (k+a) range from 0.7 h<sup>-1</sup> to 2.3 h<sup>-1</sup> with an average value of  $1.6 \text{ h}^{-1}$  and a standard deviation of  $0.6 \text{ h}^{-1}$ . Across all ten of the events, the total particle emissions per event vary over a broader range, from 1 to 85 trillion  $(10^{12})$  particles. In terms of potential contributions to exposure (the rightmost column of Table 3.23), the range for these events is from 2.4 to 120 in units of  $10^3 \text{ cm}^{-3}$  h. The two cooking events produce the highest emissions and the largest potential contribution to exposure.



**Figure 3.13**. Comparison of indoor particle number concentration measured upstairs with the sensor temperature on the gas dryer and wall heater versus time during observational monitoring at site H2. The vertical dotted lines indicate episodes where operation of the heater is associated with a sudden increase in the indoor PN level.

|    | -                  | Time      | k + a                 | σ/V                      | σ                         | $(\sigma/V)/(k+a)$                   |
|----|--------------------|-----------|-----------------------|--------------------------|---------------------------|--------------------------------------|
| ID | Source activity    | (h)       | $(h^{-1})$            | $(10^3 \text{ cm}^{-3})$ | $(10^{12} \mathrm{cm}^3)$ | $(10^3 \mathrm{cm}^{-3} \mathrm{h})$ |
| а  | Frying egg         | 1.3-2.6   | 2.3                   | 74.9                     | 18.7                      | 32.6                                 |
| b  | Heater – wall unit | 15.1-15.5 | 1.6 <sup>a</sup>      | 4.0                      | 1.0                       | 2.4                                  |
| c  | Heater – wall unit | 30.3-30.9 | 1.6 <sup>a</sup>      | 6.6                      | 1.7                       | 4.0                                  |
| d  | Heater – wall unit | 41.0-42.4 | 1.6 <sup>a</sup>      | 16.0                     | 4.0                       | 9.8                                  |
| e  | Heater – wall unit | 45.2-46.2 | 1.8                   | 54.6                     | 13.6                      | 30.3                                 |
| f1 | Heater – wall unit | 46.7-51.5 | 0.67                  | 19.2                     | 4.8                       | 28.6                                 |
| f2 | Clothes dryer      | 46.5-48.1 | 2.2                   | 40.8                     | 2.2                       | 18.6                                 |
| g  | Heater – wall unit | 58.9-59.9 | 1.6                   | 25.6                     | 6.4                       | 16.0                                 |
| h  | Heater – wall unit | 72.2-72.9 | 1.6 <sup>a</sup>      | 4.0                      | 1.0                       | 2.4                                  |
| i  | Boiling carrots    | 82.9-85.7 | 3.0, 1.1 <sup>b</sup> | 338                      | 85                        | 120                                  |

Table 3.23. Analysis of indoor UFP sources from observational monitoring at site H2.

<sup>a</sup> Estimated as the average of the decay rates for experiments a, e, f1, g, and i.

<sup>b</sup> Decay data are not well fit by a single exponential. Instead, use two separate exponential curves, with decay constant of  $3.0 \text{ h}^{-1}$  for period 83.1-84.0 h and  $1.1 \text{ h}^{-1}$  for period 84.0-85.7 h.

Table 3.24 summarizes the results of the two manipulation experiments conducted to investigate source emissions at this study site: (M1) operating the gas-fired oven (empty); and (M2) cooking potatoes by boiling them in a covered pot on the range top. These experiments were conducted in the kitchen plus family room plus living room ( $V \sim 140 \text{ m}^3$ ), with this area closed off and isolated from the rest of the house. In both cases, there was a clear signal of an increase in indoor particle number concentration, so these activities were confirmed as particle sources. The particle decay rates and total particle emissions in these experiments are comparable to those associated with frying an egg during the observational monitoring period, with overall decay rates of 1.3 and 1.8 per hour and total particle emissions of 18 and 24 trillion particles.

|    |                                 | Duration | k + a      | σ/V                       | σ           |
|----|---------------------------------|----------|------------|---------------------------|-------------|
| ID | Source                          | (h)      | $(h^{-1})$ | $(10^3 \mathrm{cm}^{-3})$ | $(10^{12})$ |
| M1 | Oven (empty)                    | 2.1      | 1.3        | 168                       | 23.6        |
| M2 | Boiling potatoes in covered pot | 1.3      | 1.8        | 129                       | 18.1        |

Table 3.24. Analysis of particle emissions from manipulation experiments at site H2.

As at the other sites, an analysis was conducted to estimate residential exposures to particles for the occupants of this household for the observational monitoring period and to apportion those exposures to known indoor sources and to indoor particles of outdoor origin. The results of the analysis are presented in Table 3.25. We used PN\_in1 as the best indicator of indoor exposure concentration during awake and asleep periods at this house. The "total cumulative exposure" for the time spent at home ranged from  $548 \times 10^3$  cm<sup>-3</sup> h for R1 to  $699 \times 10^3$  cm<sup>-3</sup> h for R3. The daily exposure rates ranged from  $143 \times 10^3$  cm<sup>-3</sup> h/d for R1 to  $182 \times 10^3$  cm<sup>-3</sup> h/d for R3. The cumulative contribution of outdoor particles to indoor exposure ranged from  $379 \times 10^3$  cm<sup>-3</sup> h (R1) to  $451 \times 10^3$  cm<sup>-3</sup> h (R3), which corresponds to 65-69% of the total cumulative residential exposures during observational monitoring at this site.

The contributions to exposure from the nine quantifiable peak events ranged from  $164 \times 10^3$  cm<sup>-3</sup> h (30% of total) for R1 to  $238 \times 10^3$  cm<sup>-3</sup> h (34-35%) for R2 and R3. The small remainders of the estimated exposures that are of unknown origin amount to ~1-2% of the total in each case.

| Parameter   | <b>R1</b> | R2   | <b>R3</b> | R4   |
|---|-----------|------|-----------|------|
| Occupancy status  |           |      |           |      |
| Time at home, awake (h)   | 19.6      | 26.6 | 24.4      | 19.4 |
| Time at home, asleep (h)  | 35.8      | 35.8 | 38.8      | 38.8 |
| Time away from home (h)   | 36.7      | 29.7 | 28.9      | 33.9 |
| Exposure duration (d) <sup>a</sup>                                      | 3.8       | 3.8  | 3.8       | 3.8  |
| Average exposure concentrations and exposures                           |           |      |           |      |
| Average concentration (PN_in1), indoor awake $(10^3 \text{ cm}^{-3})$   | 17.6      | 18.1 | 19.2      | 19.5 |
| Average concentration (PN_in1), indoor asleep $(10^3 \text{ cm}^{-3})$  | 5.6       | 5.6  | 6.0       | 6.0  |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                  | 548       | 684  | 699       | 610  |
| Cumulative exposure rate $(10^3 \text{ cm}^{-3} \text{ h d}^{-1})$      | 143       | 178  | 182       | 159  |
| Indoor exposure attributable to particles of outdoor origin             |           |      |           |      |
| Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$  | 379       | 442  | 451       | 406  |
| Percentage attributable to particles of outdoor origin                  | 69%       | 65%  | 65%       | 67%  |
| Exposure attributable to indoor source peaks $(10^3 \text{ cm}^{-3} h)$ |           |      |           |      |
| Peak a — Frying egg   | 5         | 33   | 33        | 33   |
| Peak b — Heater – wall unit   | 2         | 2    | 2         | 2    |
| Peak c — Heater – wall unit   | 3         | 3    | 3         | 3    |
| Peak d — Heater – wall unit   | 10        | 10   | 10        | 10   |
| Peak e — Heater – wall unit   | 24        | 24   | 24        | 24   |
| Peak f — Heater – wall unit   | 29        | 29   | 29        | 29   |
| Peak g — Heater – wall unit   | 8         | 16   | 16        | 8    |
| Peak h — Heater – wall unit   | 2         | 2    | 2         | 2    |
| Peak i — Cooking carrots  | 80        | 119  | 119       | 80   |
| Cumulative exposure attributable to indoor sources                      | 164       | 238  | 238       | 191  |
| Percentage attributable to quantified indoor sources                    | 30%       | 35%  | 34%       | 31%  |
| Indoor exposure of unknown origin                                       |           |      |           |      |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                  | 25        | 20   | 29        | 32   |
| Percentage of unknown origin  | 1%        | 1%   | 2%        | 2%   |

**Table 3.25.** Exposure analysis for observational monitoring at site H2.

<sup>a</sup> Exposure duration is computed as the total monitoring period, minus 24 h per day not slept at home, with the result divided by 24 h/d.

## 3.2.4. Site H3

Observational monitoring was conducted at house site H3 over a period of 77 h, on 13-16 March 2008. The reader is referred to §2.2.1 for detailed background information about the house site. See also Figure C.4 (Appendix C) for a floor plan. Summarizing briefly, this single-story house has two bedrooms and one bath with an internal volume of about 200 m<sup>3</sup> (170 m<sup>3</sup> excluding a back utility room). The home is sited in a residential neighborhood in the urban core of Oakland at an elevation of < 50 m above sea level. The house is only ~ 120 m east of Hwy 580; however, in this area heavy-duty truck traffic is not permitted on the freeway. The house has natural-gas cooking appliances and a water heater with continuous pilot lights. It has a forced-air central furnace that is vented and gas-fired. The house has three occupants, two adults (R1 = female adult; R2 = male adult) and one male child (R3 = male ~ 11 y old).

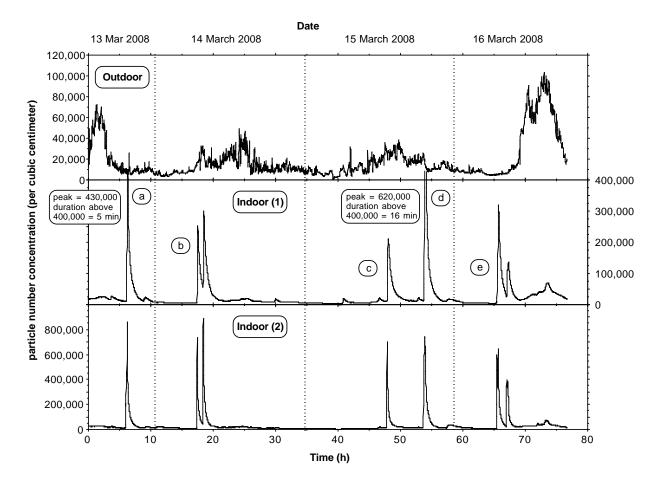
A summary of the time-averaged species concentration and cofactor values was presented in Table 3.1 (§3.1). Average particle number concentrations for the entire observational monitoring period were 18.9 (outdoors), 28.1 (indoors primary), and 35.0 (indoors supplemental) in units of 1000 particles per cm<sup>3</sup>. The outdoor concentrations ranked second highest among the seven sites monitored, about 20% lower than the levels at site H1. However, the indoor concentrations at this site were the highest of any site. Also, based on the primary indoor monitor, this site had the highest I/O ratio based on average concentrations, almost 1.5. It was one of three sites where the I/O ratio exceeded 1.0 (the others were H0 and H6).

Figure 3.14 shows the time-series of particle number concentrations measured by the three monitors. The horizontal axis is labeled with elapsed time of monitoring referenced to the start time of 13:19 on 13 March 2008. The vertical dashed lines demark midnight.

The outdoor PN concentrations at this site span a range of about a factor of 30, from ~ 3,000 cm<sup>-3</sup> to ~ 100,000 cm<sup>-3</sup>. The outdoor levels tend to be higher in the afternoon and lower overnight. Averaging the readings by clock hour over the three-day monitoring period, the maximum occurs at 14:00-14:59 and is  $52 \pm 29 \times 10^3$  cm<sup>-3</sup>; the minimum occurs at 5:00-5:59 and is  $6 \pm 2 \times 10^3$  cm<sup>-3</sup>.

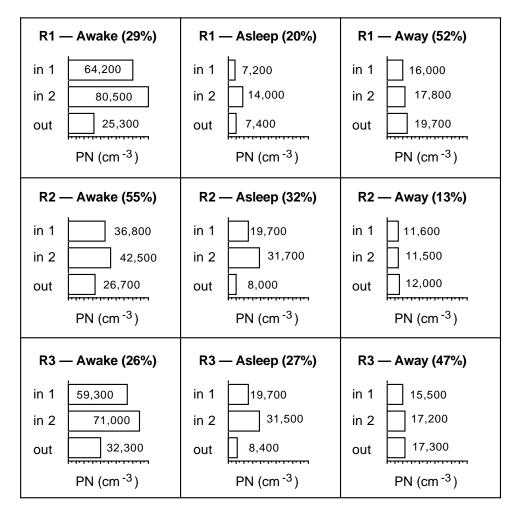
The indoor PN concentration traces in Figure 3.14 are clearly dominated by the several peaks exhibited in the signals from both monitors. In the middle frame, the peak events are labeled with lower-case letters (a-e) to facilitate later discussion. Also significant at this site, but not in evidence from Figure 3.14, are persistent contributions to indoor UFP levels caused by emissions from the natural-gas pilot lights on the cooking appliance and the penetration and persistence of particles from outdoors. The contributions of all three source categories to indoor particle levels and occupant exposure are described in the following paragraphs.

The time-series occupancy status of the three residents of H3, based on daily diary entries maintained by the occupants, is presented in Appendix D (see Figure D.42). Figure 3.15 shows the time-averaged particle number concentrations measured over the full observational monitoring period as sorted by the status of each occupant. (Also indicated in the upper portion of each frame is the percentage of the observational monitoring period for which the indicated state applies.) The main feature of this figure is that — for each resident — the average indoor particle levels are much higher when awake at home than when asleep or away. When occupants are at home — whether awake or asleep — the average indoor PN level generally exceeds the average outdoor level. (The one minor exception to this pattern is for R1 when asleep, where the "in 1" to "out" ratio is 0.97.) When occupants are away from home, the I/O ratios are less than 1.0, ranging from 0.81 for the primary indoor monitor for R1 to 0.99 for the supplementary indoor monitor for R3.



**Figure 3.14.** Time-series of particle number concentrations during observational monitoring at house site H3. Monitor "Indoor (1)" was located in the living room; monitor "Indoor (2)" sampled from the kitchen. Note that the concentration scales are different on the three frames. Lower-case letters designate peaks in the indoor PN concentration time series.

Table 3.26 summarizes the average copollutant levels at H3, sorted according to the occupancy status of the house. In Appendix D are presented time-series plots for the copollutants as follows: Figure D.43 for ozone; Figure D.44 for NO; Figure D.45 for carbon dioxide; and Figure D.46 for carbon monoxide. Given the close proximity to the freeway, the outdoor NO levels are surprisingly low. Indoor NO levels are very much higher. As seen in Figure D.44, the indoor NO level has a high baseline, suggesting the potential contribution from pilot lights. Cooking-related peaks are seen in both the NO and CO time-series data (Figure D.46).



**Figure 3.15.** Time-averaged indoor and outdoor particle number concentrations measured during the observational monitoring period at house site H3 and sorted according to the occupancy status of the three inhabitants of the house. Occupant R1 is an adult female, occupant R2 is an adult male, and occupant R3 is a male child. Each bar is labeled with the time-averaged particle number concentration.

| Tuble 5.201 Copolitical for the at site 115 during observational h |           |            |            |  |  |
|--|-----------|------------|------------|--|--|
| Species (units)  | All awake | All asleep | Unoccupied |  |  |
| NO_in (ppb)  | 73.2      | 45.8       | 43.4       |  |  |
| NO_out (ppb)   | 6.0       | 7.6        | 8.3        |  |  |
| O <sub>3</sub> _in (ppb)   | 2.4       | 2.1        | 2.4        |  |  |
| O <sub>3</sub> _out (ppb)  | 30.2      | 17.4       | 27.6       |  |  |
| CO <sub>2</sub> _in (ppm)  | 930       | 794        | 564        |  |  |

Table 3.26. Copollutant levels at site H3 during observational monitoring.<sup>a</sup>

<sup>a</sup> Measurements sorted according to occupancy status of the building and then averaged.

At this house site, we made six determinations of the air-exchange rate using carbon dioxide as a tracer gas. Two of these occurred during the observational monitoring period, when an elevated  $CO_2$  level caused by occupants was followed by a period of vacancy during which the  $CO_2$  decay influenced only by air exchange could be observed. Three additional

determinations were made during the March manipulation experiments (A1-A3). The sixth determination (A4) was made in connection with a supplemental pilot-light source characterization experiment. The results, summarized in Table 3.27, show a moderately high air-exchange rate, typical of older and smaller single-family dwellings (Chan et al., 2005). The six determinations have an average of  $1.0 \text{ h}^{-1}$ , a median of  $0.7 \text{ h}^{-1}$  and a range of  $0.5-2.2 \text{ h}^{-1}$ .

**Table 3.27.** Air-exchange rates (AER) determined by carbon dioxide concentration decay in house site H3. <sup>a</sup>

| Time (h)    | AER $(h^{-1})$ | WS $(m s^{-1})$ | $ \Delta \mathbf{T} $ (°C) | House configuration                     |
|-------------|----------------|-----------------|----------------------------|---|
| 3.3-3.6     | 2.2            | 6.7             | 4.5                        | All doors & windows closed <sup>b</sup> |
| 26.7-28.6   | 0.63           | 6.5             | 4.9                        | All doors and windows closed            |
| A1 (19 Mar) | 0.79           | 3.2             | 8.9                        | All doors and windows closed            |
| A2 (20 Mar) | 0.45           | 3.8             | 6.1                        | All doors and windows closed            |
| A3 (20 Mar) | 1.3            | 4.8             | 6.5                        | Kitchen window open. Door between       |
|             |                |                 |                            | kitchen and utility room closed.        |
| A4 (8 July) | 0.58           | 2.8             | 9.5                        | All doors and windows closed            |

<sup>a</sup> The CO<sub>2</sub> decay determinations are referenced to a baseline value of 550 ppm for observational monitoring and manipulation experiments, with the elevated baseline accounting for the effect of emissions from the pilot lights on the gas cookstove. However, A4 (8 July) was conducted with pilot lights extinguished. Hence to interpret results from this test, a fixed nominal outdoor CO<sub>2</sub> value of 380 ppm is used as the background.

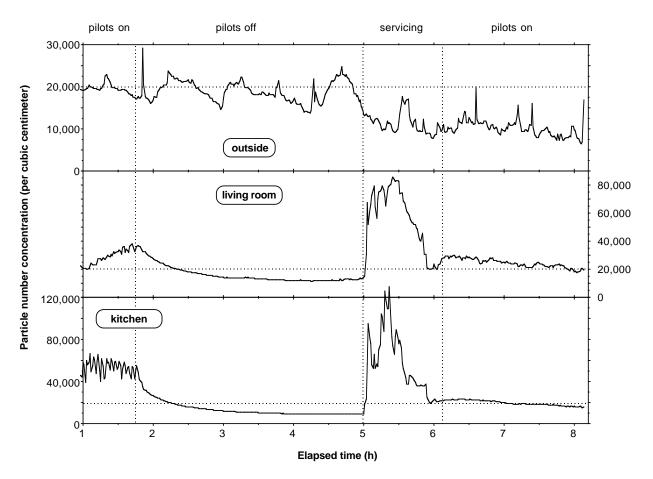
<sup>b</sup> The AER from this experiment was quite high and the steady-state indoor CO<sub>2</sub> level low. It seems possible that a window or door was open during this period but not recorded.

In characterizing the sources of indoor particles at this site, we focused on three issues: (a) continuous emissions of particles from pilot lights on the gas cooking appliances; (b) penetration and persistence of outdoor particles in the house; and (c) episodic indoor emissions producing the spikes in the indoor PN concentration time series. The effects of the episodic emissions are easy to isolate from the continuous contributions based on the time-series data. As is evident in Figure 3.14, the strong emissions from the labeled peaks produce a signal that is clearly based on the episodic events. Analysis of the time traces associated with these peaks provides information about the particle emissions and dynamic behavior associated with these episodic events.

More challenging is to separate the influence of the continuous pilot-light emissions from the penetration and persistence of outdoor particles. Both of these source categories are believed to significantly influence indoor particle levels at all times.

To investigate the levels of indoor particles from pilot lights and from outdoor sources, we considered several sources of information. We studied the indoor and outdoor particle traces during the two periods when the house was unoccupied. We conducted and analyzed a particle rebound experiment during the manipulation phase. Here, we artificially decreased the indoor particle level by means of recirculating filtration. Then, with the house vacant, we monitored the rebound of particle concentrations over time. Both of these data sources provide some information about the behavior of the continuous particle sources, but it is difficult to differentiate between pilot light emissions and particle penetration from outdoors on this basis.

We also conducted a supplemental pilot-light characterization experiment at the house during a return visit in July 2008. Figure 3.16 displays the time series of particle number concentrations during this experiment.



**Figure 3.16.** Particle number concentrations versus time during the supplemental pilot-light experiment conducted on 8 July 2008 at H3. The time scale is referenced to the beginning of monitoring at 9:32. The vertical dashed lines separate different operational states of the cook stove. During "servicing" a technician was adjusting the gas burners, so these data are not included in the analysis of particle emissions and dynamic behavior from pilot lights. The horizontal dashed lines at 20,000 particles cm<sup>-3</sup> are intended to guide the eye. During this experiment, the utility room was isolated from the main part of the house so that the indoor volume impacted by the pilot light emissions was ~ 170 m<sup>3</sup>.

Qualitatively, it is clear from Figure 3.16 that the pilot lights are a significant source of particles in this house. When the pilots are extinguished at t = 1.75 h, the indoor concentrations in both the kitchen and the living room exhibit a clear and pronounced decay, decreasing to roughly half of the outdoor level. When the lights are relit (and after servicing of the burners), levels indoors stabilize at ~ 20,000 cm<sup>-3</sup>, considerably higher than the outdoor level.

We analyzed the data from this experiment to estimate quantitative characteristics of the particle emissions from pilot lights and also the penetration and persistence of particles from outdoors. In brief, simultaneous monitoring of the decay of deliberately injected  $CO_2$  indicated

an air-exchange rate of a = 0.58 h<sup>-1</sup> during this experiment. The time-scale for the decay of particles in the living room during the "pilots off" period indicated a loss rate (k + a) of 1.89 h<sup>-1</sup>. Application of an integral material balance for the period t = 1.75-4 h indicated that the indoor proportion of outdoor particles (Pa/(k+a)) was 0.66. This result can be interpreted as the steady state indoor concentration of particles when the outdoor level is constant and there are no indoor sources. However, substituting parameter values of a and k+a into this last result would suggest that the penetration efficiency of outdoor particles to indoors exceeds one (P = 2.2), a physically implausible result. We believe that the discrepancy in this outcome arises because we have determined the decay constant, k, from the disappearance of particles originating from pilot lights, but then apply it for estimating deposition loss of particles of outdoor origin. It is well understood that particle size influences the deposition loss rate (Lai and Nazaroff, 2000; Riley et al., 2002). For ultrafine particles, larger particles will deposit at slower rates than smaller particles. It is likely that the fresh emissions from the pilot lights on a gas range have a smaller size and therefore a higher k value than the particles penetrating from outdoors. Despite the discrepancy in estimating k for outdoor particles, we believe that the (Pa/(k+a)) estimate of 0.66 is a valid determination of the proportion of outdoor particles that penetrate and persist indoors.

An emissions estimate from the pilot light experiment was made by applying an integral material balance to the measured PN concentration data for the period t = 6.13-8.13 h. We used this governing equation for the indoor particle number concentration:

$$\frac{dN}{dt} = \frac{E}{V} + PaN_o(t) - (k+a)N(t)$$
(3.9)

Here, *E* represents the particle emissions rate from the pilot lights. Both sides of the equation are multiplied by dt and then the terms are integrated over the two-hour period. We assume, in doing so, that *Pa* and (*k*+*a*) can be treated as constants and that the values for these parameters described in the previous paragraphs also apply for this final test period. The equation that is evaluated to estimate emissions is then:

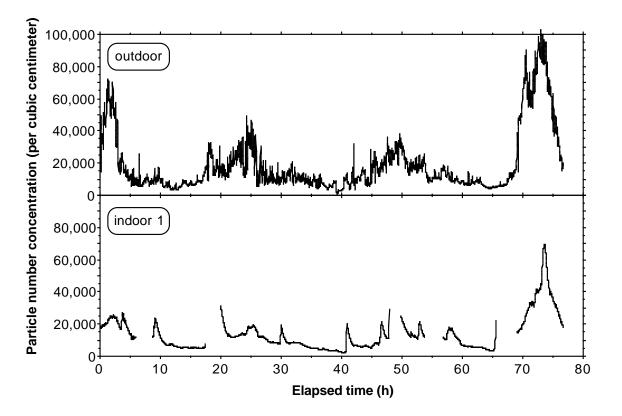
$$\frac{\sigma}{V} = \frac{t_s}{V} = \left[N(t_e) - N(t_s)\right] + \left(k + a\right) \int_{t_s}^{t_e} N(t)dt - Pa \int_{t_s}^{t_e} N_o(t)dt \qquad (3.10)$$

Here,  $\sigma$  represents the cumulative pilot light emissions for the duration of the analysis period, beginning at  $t_s = 6.13$  h and ending at  $t_e = 8.13$  h. All of the terms on the right-hand side can be evaluated. From this approach, we estimate  $\sigma = 9.8 \times 10^{12}$  particles for the two-hour period, so that the time-averaged emissions rate from the pilot lights are estimated to be  $E = 4.9 \times 10^{12}$ particles per hour. The estimated average contribution of pilot light emissions to the indoor PN number concentration is then estimated as

$$N_{pilot} = \frac{E}{V(k+a)} \tag{3.11}$$

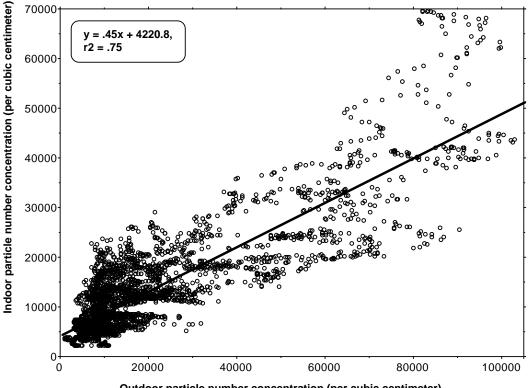
Applying  $V \sim 200 \text{ m}^3$  and  $(k+a) \sim 1.89 \text{ h}^{-1}$ , we arrive at  $N_{pilot} \sim 13,000$  particles cm<sup>-3</sup>. Comparing this result with the data presented in Figure E.4b and E.4c (see Appendix E), we find that this determination of the impact of pilot lights on indoor PN concentrations must overstate substantially the time-averaged contribution during observational monitoring. When occupants are asleep (Figure E.4b), the PN concentration in the living room is almost always less than this estimated contribution from pilot lights. And when the occupants are away, the median level indoors is less than this estimated contribution of pilot lights alone, even without accounting for the contribution from outdoor particles. For reasons we cannot explain, then, this supplemental pilot-lights experiment produces an emissions estimate that is too high to represent the average contribution during observational monitoring.

Finally, then, we adopted the following alternative approach to assess both the impact of pilot light emissions and the penetration and persistence of outdoor particles indoors during the observational monitoring period. We considered the indoor PN concentration measured in the living room as representative of household average conditions. We removed from the time series of indoor and outdoor PN concentrations the five identified (and labeled in Figure 3.14) peak events. The resulting time series are displayed in Figure 3.17. In this case, with the peaks removed, we believe that the indoor concentrations are a result of the superposition of contributions from outdoor particles and emissions from the pilot lights. (A few small peaks are evident in the indoor trace, which probably represent effects of minor episodic emissions. These contribute little to the overall time average and so are not removed in this analysis.)



**Figure 3.17.** Time series of outdoor and indoor particle number concentrations measured at site H3 during observational monitoring with the data for peaks a-e removed.

On average, we expect that the contributions from the continuous pilot lights to indoor concentrations are independent of the outdoor PN level, whereas the indoor contribution caused by outdoor particles should scale in proportion to the outdoor PN concentration. To investigate, we carried out a regression analysis, plotting the minute-by-minute indoor PN levels against the corresponding outdoor levels. Plotting minute-by-minute results preserves all of the high-frequency fluctuations in the outdoor signal, which are physically averaged by the building, and so introduces some scatter in the regression. Nevertheless, as illustrated in Figure 3.18, the data are well correlated. We interpret the slope as a best estimate of the average penetration and persistence of outdoor particles: (Pa)/(k+a) = 0.45. We interpret the intercept, 4221 cm<sup>-3</sup>, as the best estimate of the time-average indoor concentration caused by pilot light emissions alone. Applying the steady-state material balance (equation 3.11), with  $V \sim 200 \text{ m}^3$  and  $(k+a) \sim 1.89 \text{ h}^{-1}$  (based on the pilot-light experiment), we estimate  $E \sim 1.6 \times 10^{12}$  particles per hour as the emission rate from the pilot lights. This result is similar in magnitude, but about 2.8× the value  $(0.58 \times 10^{12} \text{ particles/h})$  determined for the pilot lights at site H1.



Outdoor particle number concentration (per cubic centimeter)

**Figure 3.18.** Regression of minute-by-minute measurements of indoor PN concentration (measured in the living room) against simultaneous outdoor PN concentration measurements obtained during observational monitoring at site H3. The data were first edited to remove the periods when episodic peak events a-e affected the indoor concentration.

Next, we direct the reader's attention to emissions from episodic indoor sources. At this site, cooking on the natural gas stove was a clear source of indoor particles. Cooking was

recorded on the stove by the occupants on four occasions, which correspond to the peaks a, b, d, and e. These were all of the recorded uses of the stove. For peak c (elapsed time ~ 48 h), the occupants did not record a cooking activity, but a small increase in the temperature of the sensor deployed on the stove (see Figure D.49 in Appendix D) suggests that it may have been used at this time. A sharp increase in the indoor NO level at this time (see Figure D.44 in Appendix D) supports the inference.

Several other potential source activities were noted in the activity logs. These are presented in Table 3.28. None of the activities other than cooking produce associated indoor particle concentration peaks. (Note that the minor peaks seen in Figure 3.17 do not correspond in time with the entries in Table 3.28.)

| Time (h) | Activity  | PN source? |
|----------|---|------------|
| 0.00     | Electric dryer  | Ν          |
| 1.68     | Sweeping  | Ν          |
| 6.10     | Cooking on stove (2 burners on, fried meat and noodles).            | Y (peak a) |
| 6.35     | Microwave and electric kettle                                       | Ν          |
| 17.35    | Cooking on stove, electric kettle                                   | Y (peak b) |
| 18.27    | Cooking on stove.   | Y (peak b) |
| 19.68    | Washed dishes with dish soap  | Ν          |
| 20.33    | Electric tea kettle   | Ν          |
| 21.28    | Electric tea kettle   | Ν          |
| 22.30    | Electric tea kettle   | Ν          |
| 47.82    | Electric tea kettle   | ?          |
| 47.83    | Stove (use not reported; inferred through sensor & indoor NO level) | Y (peak c) |
| 47.87    | Electric tea kettle   | ?          |
| 53.52    | Electric tea kettle   | ?          |
|          | Cooking on stove (onions and meat on one burner, cooked noodles     | Y (peak d) |
| 53.68    | on a second burner), electric tea kettle, microwave                 |            |
| 56.18    | Electric tea kettle   | Ν          |
| 65.43    | Cooking on stove (oatmeal on one burner, meat on second)            | Y (peak e) |
| 66.98    | Cooking on stove  | Y (peak e) |
| 70.05    | Electric tea kettle   | Ν          |
| 72.47    | Clothes washer  | Ν          |
| 73.35    | Electric tea kettle   | Ν          |
| 73.90    | Cleaning in bathroom with "Holy Cow"                                | Ν          |
| 74.20    | Electric clothes dryer  | Ν          |
| 76.27    | Microwave   | Ν          |

Table 3.28. Activity notes related to potential PN emission sources at house site H3.

We quantitatively analyzed the emissions and dynamic behavior of particles associated with peaks a-e. The analysis followed the same approach used at the earlier sites. The results are summarized in Table 3.29. The particle decay rate (k+a) determinations are contained within a narrow range, 1.7-2.2 h<sup>-1</sup>. The average value, 1.9 h<sup>-1</sup>, agrees with the value obtained in the pilot-light experiment. The particle emissions per episode ranges from 49 to 110 trillion ( $10^{12}$ ) particles; the average is  $87 \times 10^{12}$  particles per event.

|    |                               | Time      | k + a          | $\sigma/V$                | σ                         | $(\sigma/V)/(k+a)$                   |
|----|-------------------------------|-----------|----------------|---------------------------|---------------------------|--------------------------------------|
| ID | Source activity               | (h)       | $(h^{-1})$     | $(10^3 \mathrm{cm}^{-3})$ | $(10^{12} \mathrm{cm}^3)$ | $(10^3 \mathrm{cm}^{-3} \mathrm{h})$ |
| a  | Stove (meat & noodles)        | 6.0-8.7   | 1.7            | 352                       | 70                        | 209                                  |
| b1 | Stove                         | 17.4-18.4 | 2.2            | (265)                     | (53)                      | (122)                                |
| b2 | Stove                         | 18.4-19.9 | 1.9            | (259)                     | (52)                      | (139)                                |
| b  | Stove                         | 17.4-19.9 | <sup>b</sup>   | 523                       | 105                       | 261                                  |
| с  | Stove                         | 47.9-49.8 | 2.0            | 246                       | 49                        | 123                                  |
| d  | Stove (onions, meat, noodles) | 53.7-56.8 | 1.7            | 551                       | 110                       | 321                                  |
| e1 | Stove (oatmeal, meat)         | 65.5-67.0 | 2.0            | (370)                     | (74)                      | (181)                                |
| e2 | Stove                         | 67.0-69.0 | 1.9            | (146)                     | (29)                      | (77)                                 |
| e  | Stove                         | 65.5-69.0 | — <sup>b</sup> | 516                       | 103                       | 258                                  |

Table 3.29. Analysis of indoor UFP sources from observational monitoring at site H3.<sup>a</sup>

<sup>a</sup> Parameter definitions: k+a = total loss rate coefficient for particles emitted during the event; $\sigma/V = \text{total particle number per house volume emitted;}$   $\sigma = \text{total particle number emitted;}$  $(\sigma/V)/(k+a) = \text{contribution to exposure for an occupant present for the entire event.}$ 

<sup>b</sup> The decays of the subpeaks were separately modeled for events b and e.

Table 3.30 summarizes the results of one manipulation experiment conducted to investigate source emissions at this study site. In total, six experiments were attempted. Five of these did not produce a detectable increase in the indoor PN concentration trace: microwave oven use (M1 and M6), electric kettle use (M2), vented natural-gas furnace use (M4), and electric clothes dryer use (M5). Experiment M3 confirms the evidence from observational monitoring that cooking on the gas range is a source of ultrafine particles in this house. The loss rate of particles (k+a) during this experiment was comparable (1.4 h<sup>-1</sup> compared with 1.7-2.2 h<sup>-1</sup>) to the results obtained from analysis of observational monitoring. The particle emission rate is several times less than for the observational monitoring peaks (22.5 versus 49-110 in units of  $10^{12}$  particles per event).

|    |                       | Duration | k + a      | $\sigma/V$                | $\sigma$    |
|----|-----------------------|----------|------------|---------------------------|-------------|
| ID | Source                | (h)      | $(h^{-1})$ | $(10^3 \mathrm{cm}^{-3})$ | $(10^{12})$ |
| M3 | Stove (frying onions) | 1.7      | 1.4        | 205                       | 22.5        |

Table 3.30. Analysis of UFP source from manipulation experiments at site H3.

We estimated residential exposures to particles for the occupants of this household for the observational monitoring period and to apportion those exposures to known indoor sources and to indoor particles of outdoor origin. The results of the analysis are presented in Table 3.31. We used PN\_in1 as the best indicator of indoor exposure concentration during awake and asleep periods at this house. The "total cumulative exposure" for the time spent at home was  $1523 \times 10^3$  h cm<sup>-3</sup> for R1,  $2041 \times 10^3$  h cm<sup>-3</sup> for R2 and  $1593 \times 10^3$  h cm<sup>-3</sup> for R3. Cumulative exposure rates are computed by dividing cumulative at-home exposures by the number of monitored days when the occupants were at home. At this house, two of the occupants (R1 and R3) were away for one day during the observational monitoring period, so their rates are based on 2.2 days of monitored occupancy, while R2's rate is based on 3.2 days. The results are 693 ×  $10^3$  cm<sup>-3</sup> h/d for R1,  $639 \times 10^3$  cm<sup>-3</sup> h/d for R2, and  $726 \times 10^3$  cm<sup>-3</sup> h/d for R3.

The contribution of outdoor particles to exposure was computed by taking the appropriate fraction,  $f_1$ , of the average measured outdoor concentration during occupancy and computing a cumulative exposure for each occupant. The result,  $314-598 \times 10^3$  cm<sup>-3</sup> h, corresponded to 21-29% of the total cumulative exposure for the three occupants. Potential contributions of the nearby freeway to UFP levels at this house are explored in §D.3.

The contribution to exposure from the gas pilot lights was estimated by multiplying the estimated increment in particle concentration caused by the pilots, 4220 cm<sup>-3</sup>, times the duration of occupancy, awake plus asleep, for each occupant. The results are in the range 157-282 × 10<sup>3</sup> cm<sup>-3</sup> h, which corresponds to 10-14% of the total exposures. The contributions to exposure from the five quantified peak events collectively made the highest contributions to exposure:  $1031 \times 10^3$  cm<sup>-3</sup> h (68% of the total) for R1,  $1173 \times 10^3$  cm<sup>-3</sup> h (58% of the total) for R2 and  $1031 \times 10^3$  cm<sup>-3</sup> h (65% of the total) for R3. These three source categories accounted for all of the exposure to within 1%, as indicated by the small residual in each case.

| Parameter  | <b>R</b> 1  | R2            | R3   |
|--|-------------|---------------|------|
| Occupancy status   |             |               |      |
| Time at home, awake (h)  | 22.0        | 42.5          | 20.1 |
| Time at home, asleep (h)   | 15.2        | 24.3          | 20.3 |
| Time away from home (h)  | 39.5        | 9.9           | 36.3 |
| Exposure duration (d) <sup>a</sup>   | 2.2         | 3.2           | 2.2  |
| Average exposure concentrations and exposures                                  |             |               |      |
| Average concentration (PN_in1), indoor awake $(10^3 \text{ cm}^{-3})$          | 64.2        | 36.8          | 59.3 |
| Average concentration (PN_in1), indoor asleep $(10^3 \text{ cm}^{-3})$         | 7.2         | 19.7          | 19.7 |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                         | 1523        | 2041          | 1593 |
| Cumulative exposure rate $(10^3 \text{ cm}^{-3} \text{ h d}^{-1})$             | 693         | 639           | 726  |
| Indoor exposure attributable to particles of outdoor origin                    |             |               |      |
| Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$         | 314         | 598           | 369  |
| Percentage attributable to particles of outdoor origin                         | 21%         | 29%           | 23%  |
| Indoor exposure attributable to indoor continuous source (gas s                | tove and ov | en pilot ligl | hts) |
| Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$         | 157         | 282           | 171  |
| Percentage attributable to continuous indoor source                            | 10%         | 14%           | 11%  |
| <i>Exposure attributable to indoor source peaks</i> $(10^3 \text{ cm}^{-3} h)$ |             |               |      |
| Peak a — meat and noodles on gas stove   | 209         | 209           | 209  |
| Peak b — gas stove   | 261         | 261           | 261  |
| Peak c — gas stove   | 0           | 123           | 0    |
| Peak d — onions, meat, and noodles on gas stove                                | 321         | 321           | 321  |
| Peak e — oatmeal and meat on gas stove   | 240         | 258           | 240  |
| Cumulative exposure attributable to episodic indoor sources                    | 1031        | 1173          | 1031 |
| Percentage attributable to quantified episodic indoor sources                  | 68%         | 58%           | 65%  |
| Indoor exposure of unknown origin  |             |               |      |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                         | 20          | -12           | 22   |
| Percentage of unknown origin   | 1%          | -1%           | 1%   |

Table 3.31. Exposure analysis for residents of house site H3 during observational monitoring.

<sup>a</sup> Exposure duration is computed as the total monitoring period, minus 24 h per day not slept at home, with the result divided by 24 h/d.

## 3.2.5. Site H4

Observational monitoring was conducted at house site H4 for a continuous 74-h period commencing at 15:55 on 9 April 2008 and ending at 18:05 on 12 April. The reader is referred to  $\S2.2.1$  for detailed background information about the house site. See also Figure C.5 (Appendix C) for a floor plan. Summarizing briefly, this house has two full floors with an internal volume of about 386 m<sup>3</sup>. The home is sited very close to — about 30 m separate the back wall of the house from the edge of the freeway — interstate freeway 580 in Oakland, along a stretch of the freeway that is open to all traffic (including heavy duty trucks, which are restricted along some portions of 580 in Oakland). The house has natural-gas cooking appliances without pilot lights. The house has two separate central forced-air systems, one serving each floor. The upstairs system is equipped with a high quality particle filter and operates continuously. The fan for the downstairs system, which also has a high efficiency filter, is under thermostatic control. The house has four occupants, two adults (R1 = female adult; R2 = male adult) and two twin boys (R3 and R4 = male ~ 4 y old).

A summary of the time-averaged species concentration and cofactor values was presented in Table 3.1 (§3.1). The average particle number concentrations measured during the entire observational monitoring period were 18.0 (outdoors), 11.9 (indoors downstairs), and 2.6 (indoors upstairs) in units of 1000 particles per cm<sup>3</sup>. It is noteworthy that even though this site is immediately adjacent to a heavily used freeway, the average outdoor concentration during observational monitoring was the third highest of the seven home sites. The indoor concentrations on the first floor also represent the median across all seven houses. The average upstairs level was the lowest recorded value at any site. That these levels were so low is almost certainly attributable to the continuously operating recirculating air filter system.

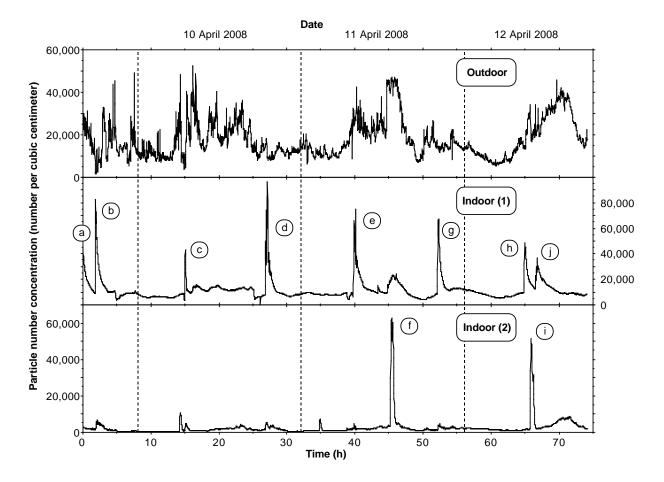
Figure 3.19 shows the time-series of particle number concentrations measured by the three monitors. The horizontal axis is labeled with elapsed time of monitoring referenced to the start time of 15:55 on 9 April 2008. The vertical dashed lines demark midnight and distance from these lines can be used to gauge clock time.

The outdoor concentrations at this site vary between ~ 2,000 cm<sup>-3</sup> and ~ 50,000 cm<sup>-3</sup>. In Figure 3.19, average levels appear to be higher in the midday period and lower overnight. Averaging according to clock hour, the highest levels are seen during 13:00-13:59 at  $33.6 \times 10^3$  cm<sup>-3</sup> and the lowest diurnal levels occur 4:00-4:59 at  $8.5 \times 10^3$  cm<sup>-3</sup>.

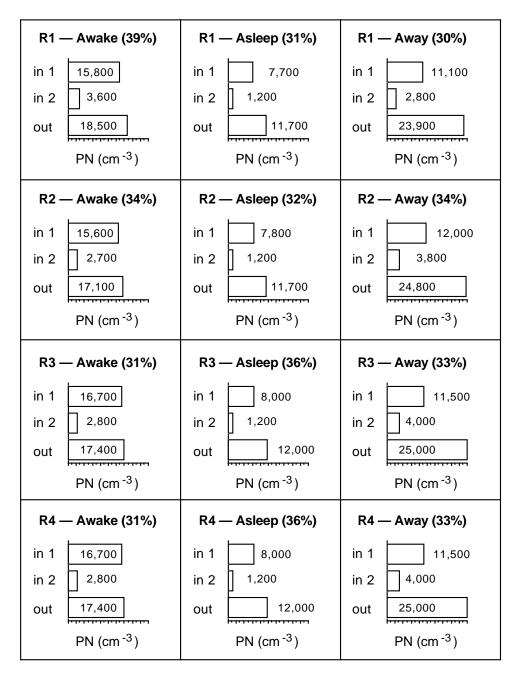
As we have seen at other sites, the indoor particle levels on the first floor of this house appear to reflect a superposition of a baseline influence of outdoor particles with episodic indoor emission events. In this case, the episodic emissions produce concentration peaks that are not much higher than the peak concentrations observed outdoors. Overall, levels on the ground floor vary from ~ 4,000 to ~ 90,000 cm<sup>-3</sup>. Upstairs, where the levels are influenced by the continuous operation of the air filtration system, the baseline levels are substantially lower than downstairs. There are a few prominent peaks that do not persist long. The upstairs and downstairs zones appear to be substantially decoupled, in the sense that the peaks upstairs are barely detectable downstairs and vice versa.

The occupancy status of the four residents, based on daily diary entries, is presented in time-series form in Appendix D (see Figure D.55). Figure 3.20 presents the time-averaged particle number concentrations measured over the full observational monitoring period as sorted by the status of each occupant. Focusing initially on the primary indoor monitor, "in 1," we see that the average concentrations when occupants are at home awake,  $15-17 \times 10^3$  cm<sup>-3</sup>, are similar to the outdoor average concentrations during these times,  $17-19 \times 10^3$  cm<sup>-3</sup>. When occupants are

away from home, the indoor levels are on the first floor are lower,  $11-12 \times 10^3$  cm<sup>-3</sup>, whereas the outdoor levels are higher,  $24-25 \times 10^3$  cm<sup>-3</sup>. The levels upstairs ("in 2") are consistently lower than levels recorded by other monitors. For example, when occupants are asleep (in the upstairs bedrooms), the average upstairs level is  $1.2 \times 10^3$  cm<sup>-3</sup>, compared with the downstairs average of  $\sim 8 \times 10^3$  cm<sup>-3</sup> and the outdoor average of  $\sim 12 \times 10^3$  cm<sup>-3</sup>.



**Figure 3.19.** Time-series of particle number concentrations during observational monitoring at house site H4. Monitor "Indoor (1)" was located on the main floor (downstairs) in the family room; monitor "Indoor (2)" sampled from an upstairs study. Note that the vertical scales are different among frames. Lower case letters label peak concentrations in the indoor PN concentration signal to facilitate later discussion.



**Figure 3.20.** Time-averaged indoor and outdoor particle number concentrations measured during the observational monitoring period at house site H4 and sorted according to the occupancy status of the four inhabitants of the house. Occupant R1 is an adult female, occupant R2 is an adult male, and occupants R3 and R4 are male children. Each bar is labeled with the time-averaged particle number concentration. In the upper portion of each frame is the percentage of the observational monitoring period for which the indicated state applies.

Table 3.32 summarizes the average copollutant levels at H4, sorted according to the occupancy status of the house. In Appendix D are presented time-series plots for the copollutants as follows: Figure D.56 for ozone; Figure D.57 for NO; Figure D.58 for carbon dioxide; and Figure D.59 for carbon monoxide. Average outdoor NO levels are not particularly elevated at

this site, which is surprising given the close proximity to the freeway. Referring back to Table 3.1, we can see that the overall average outdoor NO level here, 13 ppb, ranks as the median outdoor level across the seven house sites. Table 3.32 shows that indoor levels are only moderately higher than the outdoor level. Ozone levels are quite low indoors relative to the outside level.

| Species (units)           | All awake | All asleep | Unoccupied |
|---------------------------|-----------|------------|------------|
| NO_in (ppb)               | 15.3      | 20.1       | 14.3       |
| NO_out (ppb)              | 9.8       | 19.0       | 7.2        |
| O <sub>3</sub> _in (ppb)  | 1.3       | 1.2        | 1.8        |
| O <sub>3</sub> _out (ppb) | 14.7      | 6.1        | 31.1       |
| CO <sub>2</sub> _in (ppm) | 636       | 482        | 499        |

**Table 3.32.** Copollutant levels at site H4 during observational monitoring.<sup>a</sup>

<sup>a</sup> Measurements sorted according to occupancy status of the building and then averaged.

At this house site, we made eight determinations of the air-exchange rate using carbon dioxide as a tracer gas (see Table 3.33). Four of these occurred during the observational monitoring period. The other four determinations were made from various manipulation experiments. The average air-exchange across the eight determinations was  $0.83 \text{ h}^{-1}$  with a standard deviation of  $0.34 \text{ h}^{-1}$ .

| <b>Table 3.33.</b> Air-exchange rates (AER) determined by carbon dioxide concentration decay in |
|---|
| house site H4. <sup>a</sup>   |

| Time (h)  | $AER(h^{-1})$ | WS (m s <sup>-1</sup> ) | $ \Delta \mathbf{T} $ (°C) | House conditions                          |
|-----------|---------------|-------------------------|----------------------------|---|
| 16.0-18.0 | 0.88          | 1.5                     | 6.9                        | Downstairs bathroom window open and       |
|           |               |                         |                            | exhaust fan on. Upstairs central fan on.  |
| 22.5-24.5 | 0.98          | 3.6                     | 0.4                        | Downstairs bathroom window open and       |
|           |               |                         |                            | exhaust fan on. Upstairs central fan on.  |
| 41.3-42.3 | 0.42          | 1.3                     | 2.9                        | Doors/windows closed. Exhaust fans off.   |
|           |               |                         |                            | Upstairs central fan on.                  |
| 67.5-70   | 0.37          | 2.2                     | 4.8                        | Doors/windows closed. Exhaust fans off.   |
|           |               |                         |                            | Upstairs central fan on.                  |
| Pkd (14   | 1.27          | 8.0                     | na                         | Windows/doors closed. Central systems     |
| Apr)      |               |                         |                            | off.                                      |
| A1 (15    | 0.76          | 5.4                     | na                         | Windows/doors closed. Upstairs central    |
| Apr)      |               |                         |                            | system on (no heat).                      |
| A2 (15    | 1.26          | 5.8                     | na                         | Bathroom windows open. Bathroom           |
| Apr)      |               |                         |                            | exhaust fans on. Upstairs central fan on. |
| M1 (16    | 0.67          | 3.6                     | na                         | Tracer downstairs only. Windows/doors     |
| Apr)      |               |                         |                            | closed. Central systems off.              |

<sup>a</sup> Uses 411 ppm as the background  $CO_2$  level for AER determinations during observational monitoring; uses measured  $CO_2$  outdoors for manipulation experiments. The first four entries are from the observational monitoring period. The remaining four are from manipulation experiments: (a) the Pkd experiment for measuring particle penetration and persistence from outdoors; (b) two experiments targeted at measuring air-exchange rate (A1 and A2); and (c) a source characterization experiment (M1).

To explore the influence of outdoor particles on indoor PN concentrations at site H4, we computed the ratio of indoor to outdoor particle concentrations, based on time-averaged concentration levels, during times when the house is unoccupied. We restricted our attention to times when the dominant configuration of the house applied, namely when all doors and windows were closed. We assessed the upstairs and downstairs zones of the house separately.

Five suitable intervals were identified, distributed throughout the observational monitoring period, with a total duration of 20.5 h. The analysis results are presented in Table 3.34. For these five periods, the ratio of average indoor downstairs PN concentration to the average outdoor PN concentration varied over a narrow range, from 0.35 to 0.58, with a time-weighted average value of  $f_1 = 0.47$ . Likewise, for the average indoor upstairs PN concentration to the average outdoor PN concentration, the ratio varies from 0.06 to 0.17, with a time-weighted average of  $f_2 = 0.11$ . Treating each of the intervals as a separate determination, the weighted standard error in the determination of  $f_1$  is 0.04, or 8% of the mean. For  $f_2$ , the standard error is 0.017, 15% of the weighted average.

**Table 3.34.** Analysis of the indoor to outdoor particle concentration ratios at house site H4 for periods when the house is unoccupied and all doors and windows are closed.<sup>a</sup>

| Time                 | WS           | $ \Delta \mathbf{T} $ | PN_out                     | PN_in1                   | PN_in2                   | $f_1$ | $f_2$ |
|----------------------|--------------|-----------------------|----------------------------|--------------------------|--------------------------|-------|-------|
| ( <b>h</b> )         | $(m s^{-1})$ | (°C)                  | $(10^3  \mathrm{cm}^{-3})$ | $(10^3 \text{ cm}^{-3})$ | $(10^3 \text{ cm}^{-3})$ | (—)   | (—)   |
| 16.1-21.1            | 2.1          | 3.9                   | 22.5                       | 13.0                     | 1.3                      | 0.58  | 0.06  |
| 22.1-25.1            | 3.4          | 0.5                   | 23.3                       | 12.6                     | 2.5                      | 0.54  | 0.11  |
| 41.6-43.3            | 1.8          | 1.7                   | 21.5                       | 11.0                     | 2.0                      | 0.51  | 0.09  |
| 46.1-50.6            | 4.2          | 4.7                   | 19.2                       | 8.9                      | 2.1                      | 0.46  | 0.11  |
| 67.9-74.2            | 2.6          | 4.4                   | 29.4                       | 10.2                     | 5.0                      | 0.35  | 0.17  |
| Average <sup>b</sup> | 2.9          | 3.5                   | 23.9                       | 11.0                     | 2.9                      | 0.47  | 0.11  |

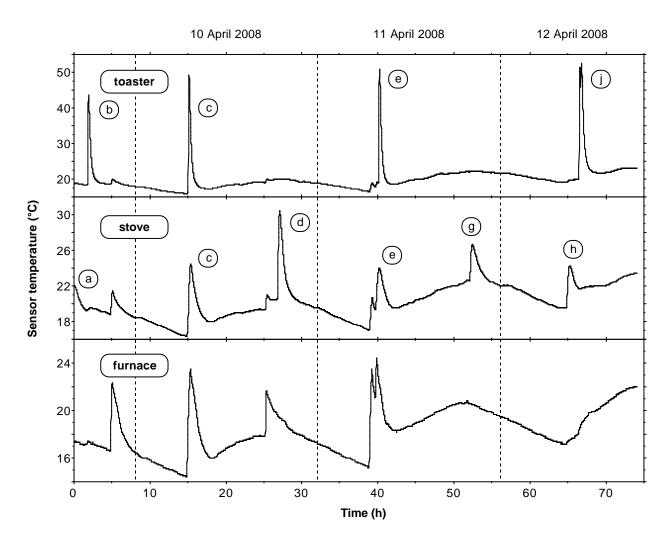
<sup>a</sup> The parameter  $f_1$  represents the ratio of the averages for PN\_in1 to PN\_out; the parameter  $f_2$  represents the ratio of the averages for PN\_in2 to PN\_out.

<sup>b</sup> The final row of the table presents time-weighted average values for each entry. Note that the overall average meteorological results for the observational monitoring period are indoor temperature =  $21.3 \,^{\circ}$ C, outdoor temperature =  $17.4 \,^{\circ}$ C, and wind speed (WS) =  $4.7 \,\text{m s}^{-1}$ .

At this site, there do not appear to be significant persistent indoor sources of PN emissions. The presence of episodic emission sources is evident from Figure 3.19. To investigate the causes of these sources, we utilized information from occupant activity diaries and temperature sensors. Information from the diaries that relates to potential PN sources is reproduced in Table 3.34. Figure 3.21 displays the time-series data for temperature sensors deployed on the toaster oven (upper frame), gas stove (middle frame) and a furnace discharge register on the lower floor (lower frame). In the upper two frames of this figure, the sharp, pronounced peaks in the temperature signals indicate use of the appliance. There is complete concordance for the toaster and the stove with information recorded in the occupant diaries. Furthermore, these sensor temperature signals and occupant use records correspond with all of the downstairs peak events in PN concentration: peaks a-e, g, h, and j. On the other hand, the source of the two sharp peaks (f and i) in the upstairs PN concentration signal do not correspond to any information from the sensor temperatures or from the activity diaries, so we cannot determine their cause. At some sites (H0 and H2) the furnace was identified as a particle source. Here, the furnace does not appear to be a PN source and furthermore evidence indicates that the operation of the furnace fan reduces indoor PN concentrations, in all likelihood because of the high-efficiency filter. Figure 3.21 shows four spikes in the furnace register temperature signal, indicating furnace use. The first of these, which occurs at  $t \sim 5$  h, coincides with a sudden decrease in indoor particle levels, as can be discerned in Figure 3.19, middle frame. The second use of the furnace, at  $t \sim 15$  h, coincides with stove and toaster oven use and so there is a PN concentration spike at this time. Note, though, the spike in concentration is quite short-lived, which is expected if there is a high removal rate associated with recirculating air filtration. The third use of the furnace, at  $t \sim 25$  h, again produces a sharp dip in the indoor PN concentration trace, a few hours ahead of peak d. Finally, the fourth use of the furnace, at  $t \sim 39$  h, again produces a dip in the indoor PN concentration signal immediately ahead of the stove and toaster use that generate peak e.

| Time (h) | Activity  | PN source? |
|----------|---|------------|
| 0.00     | Water boiled on stove                                     | Y (peak a) |
| 1.58     | Toaster oven and microwave used                           | Y (peak b) |
| 2.08     | Clothes washing machine                                   | Ν          |
| 3.08     | Clothes dryer   | Ν          |
| 4.08     | Turned on downstairs heater                               | Ν          |
| 15.08    | Water boiled on stove top; microwave & toaster oven used  | Y (peak c) |
| 15.58    | Microwave used  | Ν          |
| 25.05    | Clothes dryer used; heater turned on                      | Ν          |
| 26.08    | Burned tortilla on stove when making quesadillas; exhaust | Y (peak d) |
|          | fan turned on; microwave used                             |            |
| 27.58    | Used Fantastik spray in downstairs bathroom               | Ν          |
| 28.58    | Used Method spray in downstairs bathroom                  | Ν          |
| 31.08    | Turned off downstairs bathroom exhaust fan                | Ν          |
| 39.73    | Boiled water on stove; used microwave                     | Y (peak e) |
| 40.08    | Used toaster oven   | Y (peak e) |
| 44.08    | Used microwave  | Ν          |
| 45.10    | Turned on upstairs bathroom vent; turned on dryer         | ? (peak f) |
| 46.10    | Turned on dryer   | Ν          |
| 51.75    | Turned on dryer   | Ν          |
| 52.10    | Boiled water on stove, and turned on dryer                | Y (peak g) |
| 54.60    | Turned on dryer   | Ν          |
| 64.10    | Turned on dryer, boiled water on stove                    | Y (peak h) |
| 65.08    | Microwaved milk and coffee                                | Ν          |
| 66.10    | Used toaster oven   | Y (peak j) |

| Table 3.35. Activity notes from occupan | t diaries at H4 related to | potential PN sources |
|---|----------------------------|----------------------|
|---|----------------------------|----------------------|



**Figure 3.21.** Time series of sensor temperature readings deployed on the toaster oven, the gas cook stove, and a furnace discharge register on the lower floor of the house site H4. The labeling with lower case letters corresponds to the identification of peaks in Figure 3.19.

We quantitatively analyzed the peak events to determine first-order loss rate coefficients (k + a) and source strengths ( $\sigma$ ) using the integral material balance approach described in connection with site H0 (§3.2.1). The results are reported in Table 3.36. We treated the two floors of the house as independently well-mixed zones. In particular, this means that for upstairs peak events (f and i), we used  $V = 186 \text{ m}^3$  to estimate total emissions ( $\sigma$ ) from the emissions per volume determination. Conversely, for the downstairs events, we only used the downstairs house volume,  $V = 199 \text{ m}^3$  in estimating  $\sigma$ . Since the emissions for peak a occurred before the onset of monitoring, we can only determine a decay term for this event, not the total emissions.

For all ten episodic emissions events, we are able to characterize an effective indoor particle loss rate, k+a. For the two upstairs peaks (f and i) and for one of the downstairs events, peak c, the loss-rate coefficients are very high, ~ 10 h<sup>-1</sup>. It is probable that for all three cases the high rates incorporate important contributions from the high removal rates in the recirculating, forced-air heating system. Recall that the fan on the upstairs system operated continuously. Evidence indicates that the downstairs system was on during event c. For the remaining seven

events, the loss rates are more moderate, with values similar to what we have observed at other sites. The range is  $1.3-3.6 \text{ h}^{-1}$  with an average of  $1.9 \text{ h}^{-1}$  (standard deviation =  $0.8 \text{ h}^{-1}$ ).

Emissions were quantified for seven downstairs events, all associated with cooking either on the gas range or with the toaster oven (or both). The total particle emissions ( $\sigma$ ) for these events range from 3 to 15 trillion (10<sup>12</sup>) particles; the average is 11 × 10<sup>12</sup> particles per event. The two upstairs events have higher estimated emissions, 53 × 10<sup>12</sup> for f and 36 × 10<sup>12</sup> for i.

|    |                              | Time      | k + a      | $\sigma / V$             | σ                         | $(\sigma/V)/(k+a)$                   |
|----|------------------------------|-----------|------------|--------------------------|---------------------------|--------------------------------------|
| ID | Source activity              | (h)       | $(h^{-1})$ | $(10^3 \text{ cm}^{-3})$ | $(10^{12} \mathrm{cm}^3)$ | $(10^3 \mathrm{cm}^{-3} \mathrm{h})$ |
| a  | Stove (water)                | <0.0-1.8  | 1.8        | na                       | na                        | na                                   |
| b  | Toaster oven                 | 1.8-3.8   | 1.8        | 52.0                     | 10.4                      | 29.6                                 |
| с  | Stove (water) & toaster oven | 15.0-15.3 | 10.3       | 54.0                     | 10.8                      | 5.3                                  |
| d  | Stove (burned tortilla)      | 26.8-29.0 | 1.6        | 70.2                     | 14.0                      | 42.8                                 |
| e  | Stove (water) & toaster oven | 39.8-42.4 | 1.7        | 50.4                     | 10.0                      | 30.3                                 |
| f  | Unknown (upstairs)           | 45.2-46.1 | 11.1       | 283                      | 52.5                      | 25.5                                 |
| g  | Stove (water)                | 52.1-54.0 | 1.8        | 65.0                     | 12.9                      | 35.6                                 |
| h  | Stove (water)                | 64.8-66.5 | 3.6        | 73.5                     | 14.6                      | 20.6                                 |
| i  | Unknown (upstairs)           | 65.7-66.9 | 9.5        | 192                      | 35.8                      | 20.2                                 |
| j  | Toaster oven                 | 66.6-68.5 | 1.3        | 16.6                     | 3.3                       | 13.2                                 |

Table 3.36. Analysis of indoor UFP sources at H4 from observational monitoring.

Two manipulation experiments were conducted at this site to further investigate PN emission sources. Experiment M1 involved toasting two pieces of bread in the toaster oven. Experiment M2 entailed use of a grapefruit-scented cleaning product believed to contain terpenes. Experiment M2 did not produce a detectable increase in indoor PN concentrations, and so it was not quantitatively analyzed. Experiment M1 did produce a clear increase in indoor particle levels, which constitutes corroborating evidence from observational monitoring that the toaster oven is a particle source at this site. A quantitative analysis of the experimental results, following the same procedure used for observational monitoring, yielded the results summarized in Table 3.37. Both the particle decay rate (k + a) and the source strength ( $\sigma$ ) agree well with results from the analysis of peaks detected during observational monitoring.

 Table 3.37. Analysis of UFP source from manipulation experiment M1 at site H4.

|    |   | Duration | k + a      | $\sigma/V$               | σ           |
|----|---|----------|------------|--------------------------|-------------|
| ID | Activity                                    | (h)      | $(h^{-1})$ | $(10^3 \text{ cm}^{-3})$ | $(10^{12})$ |
| M1 | Two slices of bread toasted in toaster oven | 1.4      | 1.9        | 66.8                     | 13.3        |

The exposure analysis results for H4 occupants are summarized in Table 3.38. For the "awake at home" time, we used the downstairs PN concentration (PN\_in1) as the best estimate of the exposure concentration, taking into account information from the residents that they spent most of their awake time on the lower level of the house. For the "asleep" time, we used the upstairs PN concentration (PN\_in2) as the best estimate of exposure concentration. The "total cumulative exposure" for the time spent at home ranged from  $419 \times 10^3$  cm<sup>-3</sup> h for R2 to  $485 \times 10^3$  cm<sup>-3</sup> h for R1. The daily exposure rates are estimated to range from  $136 \times 10^3$  cm<sup>-3</sup> h/d for R2 to  $157 \times 10^3$  cm<sup>-3</sup> h/d for R1.

| Parameter   | <b>R1</b> | R2   | R3   | <b>R4</b> |  |
|---|-----------|------|------|-----------|--|
| Occupancy status  |           |      |      |           |  |
| Time at home, awake (h)   | 28.8      | 24.9 | 23.3 | 23.3      |  |
| Time at home, asleep (h)  | 23.3      | 24.0 | 26.5 | 26.5      |  |
| Time away from home (h)   | 22.2      | 25.3 | 24.3 | 24.3      |  |
| Exposure duration (d) <sup>a</sup>                                      | 3.1       | 3.1  | 3.1  | 3.1       |  |
| Average exposure concentrations and exposures                           |           |      |      |           |  |
| Average concentration (PN_in1), indoor awake $(10^3 \text{ cm}^{-3})$   | 15.9      | 15.7 | 16.7 | 16.7      |  |
| Average concentration (PN_in2), indoor asleep $(10^3 \text{ cm}^{-3})$  | 1.2       | 1.2  | 1.2  | 1.2       |  |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                  | 485       | 419  | 422  | 422       |  |
| Cumulative exposure rate $(10^3 \text{ cm}^{-3} \text{ h d}^{-1})$      | 157       | 136  | 137  | 137       |  |
| Indoor exposure attributable to particles of outdoor origin             |           |      |      |           |  |
| Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$  | 279       | 230  | 223  | 223       |  |
| Percentage attributable to particles of outdoor origin                  | 58%       | 55%  | 53%  | 53%       |  |
| Exposure attributable to indoor source peaks $(10^3 \text{ cm}^{-3} h)$ |           |      |      |           |  |
| Peak a — boiled water on stove  | 17.5      | 17.5 | 17.5 | 17.5      |  |
| Peak b — toaster oven   | 29.6      | 29.6 | 29.6 | 29.6      |  |
| Peak c — boiled water on stove & toaster oven                           | 3.1       | 2.5  | 2.5  | 2.5       |  |
| Peak d — burned tortilla on stove                                       | 42.8      | 42.8 | 42.8 | 42.8      |  |
| Peak e — boiled water on stove and used toaster oven                    | 27.0      | 11.0 | 27.0 | 27.0      |  |
| Peak g — boiled water on stove  | 35.6      | 35.6 | 35.6 | 35.6      |  |
| Peak h — boiled water on stove  | 20.6      | 20.6 | 20.6 | 20.6      |  |
| Peak j — toaster oven   | 13.2      | 13.2 | 13.2 | 13.2      |  |
| Cumulative exposure attributable to indoor sources                      | 189       | 173  | 187  | 187       |  |
| Percentage attributable to quantified indoor sources                    | 39%       | 41%  | 44%  | 44%       |  |
| Indoor exposure of unknown origin                                       |           |      |      |           |  |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                  | 16.4      | 16.8 | 12.4 | 12.4      |  |
| Percentage of unknown origin  | 3%        | 4%   | 3%   | 3%        |  |

Table 3.38. Exposure analysis for residents of house site H4 during observational monitoring.

<sup>a</sup> Exposure duration is computed as the total monitoring period, minus 24 h per day not slept at home, with the result divided by 24 h/d.

The contribution of outdoor particles to exposure was computed by taking the appropriate average fraction,  $f_I = 0.47$  for "awake" and  $f_2 = 0.11$  for "asleep," of the average measured outdoor concentration during respective occupancy states for each resident and computing an exposure increment for each occupant. The cumulative contribution of outdoor particles to indoor exposure ranged from  $223 \times 10^3$  cm<sup>-3</sup> h (R3 and R4) to  $279 \times 10^3$  cm<sup>-3</sup> h (R1), which corresponds to 53-58% of the total cumulative exposure.

The contributions to exposure from the eight quantifiable peak events were assessed with cumulative totals ranging from  $173 \times 10^3$  cm<sup>-3</sup> h (44% of total) for R2 to  $189 \times 10^3$  cm<sup>-3</sup> h (39%) for R1. The small remainders of the estimated exposures that are of unknown origin amount to ~3-4% of the total in each case. Because we do not know that occupants were upstairs during the two peak events of unknown origin that occurred there (peaks f and i), we did not include any contributions from these events to the exposure estimates.

## 3.2.6. Site H5

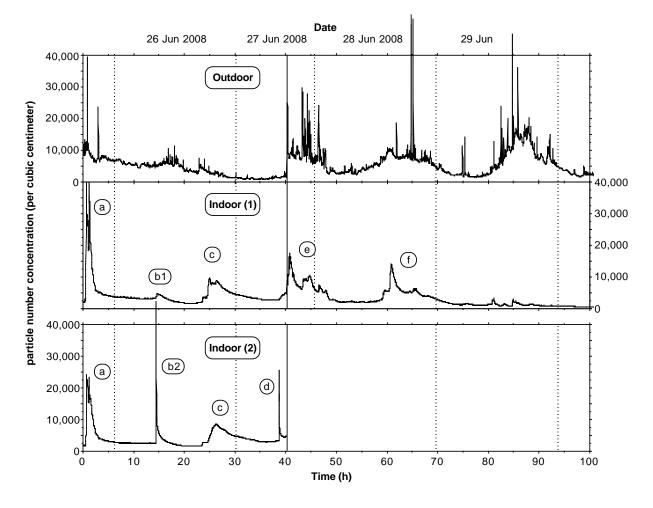
Observational monitoring commenced at house site H5 at 17:48 on 25 June 2008. There was an interruption for 8.5 h beginning at 10:00 on 27 June. Monitoring was terminated at 6:59 on 30 June, so that the cumulative duration of observational monitoring was 100 h. The interruption was caused by an instrument malfunction in the outdoor WCPC. To allow monitoring to continue, we removed the outdoor monitor from service and used in its place the supplementary indoor monitor. Consequently, for the period 40-100 h of elapsed monitoring time, we acquired PN concentration data with only one outdoor and one indoor monitor.

For detailed background information about the house site, the reader is referred to \$2.2.1. See also Figure C.6 (Appendix C) for a floor plan. Summarizing briefly, this large, modern house has two floors with an internal living volume of about 420 m<sup>3</sup>. The house is in Livermore, about 30 miles east of Oakland. It is situated on a quiet residential street, about 1.2 km from interstate freeway 580. The house has all electric cooking appliances. It is equipped with a central forced-air heating and air conditioning system. During the summer study period, the air conditioner was used; heating was not. The system operates under thermostatic control with a manual override. The fan only operates when heating or cooling is being used. In the recirculating line is an electrostatic particle filter. This filter would be expected to reduce indoor particle concentrations during times that the system fan is on. Electrostatic precipitators have also been associated with ozone production (Waring et al., 2008). The house has two permanent occupants, both adults (R1 = female adult; R2 = male adult). During the observational monitoring period, R2 was away and so only R1 was considered as a resident for analysis.

A summary of the time-averaged species concentration and cofactor values was presented in Table 3.1 (§3.1). The average particle number concentrations measured during the entire observational monitoring period were 5.5 (outdoors), 3.7 (indoors downstairs), and 4.1 (indoors upstairs, for the first 40 h of monitoring) in units of 1000 particles per cm<sup>3</sup>. The average outdoor PN levels measured at this site were substantially lower than at any other site in this study. The indoor levels were also low, well below the average levels measured at any other site except for the upstairs floor of house H4.

Figure 3.22 shows the time-series of particle number concentrations measured at this site. The horizontal axis is labeled with elapsed time of monitoring referenced to the start time of 17:48 on 25 June 2008. The vertical dashed lines demark midnight and distance from these lines can be used to gauge clock time. The break in monitoring is denoted by a solid vertical line.

The outdoor concentrations at this site exhibit spiky upward excursions above a baseline that oscillates from < 1,000 cm<sup>-3</sup> to ~ 10,000-15,000 cm<sup>-3</sup>. The spikes are especially pronounced from the evening of 28 June through mid afternoon of 29 June and suggest intermittent influence of significant local sources. Winds at this time (based on central station monitoring data) were steady from the west and moderately strong, varying from ~ 5 m/s at the beginning and end of the interval to ~ 2 m/s in the middle. During the first 40 h of monitoring, the main trend of PN levels shows a steady decline. After the break, there is more evidence of a diurnal pattern, with higher concentrations in the late afternoon and evening, and lower concentrations in the morning. Overall, this monitoring period coincided with a time of intense forest fires throughout northern California, which had substantial impacts on regional air quality. Central station PM<sub>2.5</sub> levels (in Livermore) reached 100  $\mu$ g/m<sup>3</sup> late on 26 June and remained elevated, in the range 15-35  $\mu$ g/m<sup>3</sup>, for most of 28-29 June. In studies of urban air quality, it has been noted that UFP levels and PM<sub>2.5</sub> mass concentrations are not correlated (e.g., Jeong et al., 2004). Not only are the sources potentially different, but the coagulation of UFP onto accumulation mode particles, which



typically make up most of the  $PM_{2.5}$  mass, can serve as an important sink for atmospheric UFP, which would contribute to an anticorrelation between these two air-quality parameters.

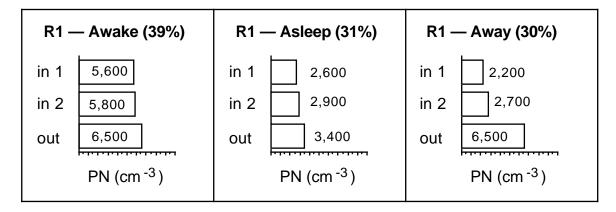
**Figure 3.22.** Time-series of particle number concentrations during observational monitoring at house site H5. Monitor "Indoor (1)" was located on the main floor (downstairs) in the living room; monitor "Indoor (2)" sampled from an upstairs bedroom. The solid vertical line marks a break in observational monitoring beginning at 10:00 on 27 June caused by a malfunction of the outdoor WCPC. The indoor (2) unit was moved outdoors and monitoring was resumed at 18:30. Lower case letters label peak concentrations in the indoor PN concentration signal to facilitate later discussion.

As at other sites, the baseline indoor PN concentrations in this house exhibit an influence of the time varying outdoor concentrations, with the high-frequency fluctuations filtered out. Added to this baseline is an apparent influence of several episodes, almost certainly from indoor emission sources. Compared with other sites, the episodic peaks in indoor concentrations are mostly of lower intensity.

Comparing the upstairs ("indoor (2)") and downstairs ("indoor (2)") monitoring traces for the first 40 h of monitoring, we see similar profiles for peaks a and c. But peaks b and d show very different profiles. Furthermore upstairs, the b2 and d2 peaks exhibit a very sharp rise and fall before, at least in the case of b2, returning to a more common form of gradual decay back to

the baseline. As will be discussed later in this subsection, we believe that the peaks b and d are associated with the use of an electric steam iron upstairs. Peak a is associated with cooking downstairs. Peak c, which exhibits a long persistence both upstairs and downstairs, appears to be significantly influenced by particle generation from ozone-terpene chemistry, related to the use of cleaning products.

The occupancy status of the adult female who resided in H5 during observational monitoring is presented in Figure D.68 (see Appendix D). Figure 3.23 shows the time-averaged particle number concentrations measured during observational monitoring as sorted by the occupancy status of R1. Focusing initially on the primary indoor monitor, "in 1," we see that the average indoor concentration when the occupant is at home awake,  $5.6 \times 10^3$  cm<sup>-3</sup>, is roughly 15% less than the outdoor average concentration during these times. When the occupant is away from home, the indoor level is much lower,  $2.2 \times 10^3$  cm<sup>-3</sup>, whereas the outdoor level is unchanged. The level upstairs ("in 2") is comparable to that measured downstairs. Average PN levels for the first 40 h of monitoring only are presented in Table 3.39. With these more directly comparable data, we see that the indoor level downstairs averages about 10% higher than the level upstairs when the house is occupied and are equal when the house is unoccupied.



**Figure 3.23.** Time-averaged indoor and outdoor particle number concentrations measured during the observational monitoring period at house site H5 and sorted according to the occupancy status of the one resident inhabitant of the house, an adult female. Each bar is labeled with the time-averaged particle number concentration. Note that the average values for "in 2" (measured indoors upstairs) reflect only the first 40 h of the observational monitoring period. For "in 1" and "out" the average for the full 100 h of observational monitoring is reported. The percentage of the observational monitoring period for which the indicated state applies is noted at the top of each frame.

**Table 3.39.** Average particle number concentrations (cm<sup>-3</sup>) for the first 40 h of observational monitoring, sorted according to occupancy status of R1 at site H5.

|              | Awake | Asleep | Away  |
|--------------|-------|--------|-------|
| Inside 1     | 6,400 | 3,300  | 2,700 |
| Inside 2     | 5,800 | 2,900  | 2,700 |
| Outside      | 4,700 | 3,600  | 4,600 |
| Duration (h) | 17.4  | 13.0   | 9.8   |

Table 3.40 summarizes the average copollutant levels at H5, sorted according to the occupancy status of the house. In Appendix D are presented time-series plots for the copollutants as follows: Figure D.69 for ozone; Figure D.70 for NO; Figure D.71 for carbon dioxide; and Figure D.72 for carbon monoxide. Average outdoor NO levels are very low at this site, both outdoors and indoors. Ozone levels outdoors are moderate, with afternoon peak values reaching 60-80 ppb. Indoor levels average about 20% of the outdoor value.

| Species (units)           | Awake | Asleep           | Away |
|---------------------------|-------|------------------|------|
| NO_in (ppb)               | 0.6   | 0.3              | 2.1  |
| NO_out (ppb)              | 0.7   | 0.0 <sup>b</sup> | 4.3  |
| O <sub>3</sub> _in (ppb)  | 8.2   | 4.3              | 5.3  |
| O <sub>3</sub> _out (ppb) | 38.5  | 22.0             | 28.7 |
| CO <sub>2</sub> _in (ppm) | 458   | 442              | 430  |

**Table 3.40.** Copollutant levels at site H5 during observational monitoring.<sup>a</sup>

<sup>a</sup> Measurements sorted according to occupancy status of R1 and then averaged.

<sup>b</sup> Owing to a negative baseline, the mean value is less than zero

At this house site, we made twelve determinations of the air-exchange rate using carbon dioxide as a tracer gas (see Table 3.41). Three of these occurred during the observational monitoring period; the other nine (denoted A1-A9 in the table) were made during manipulation experiments. For the five determinations under baseline conditions (exterior doors and windows closed; air-handling system off), the air-exchange rates were in the range  $0.08-0.20 \text{ h}^{-1}$ , with a mean value of  $0.17 \text{ h}^{-1}$ . During three determinations, the exterior doors and windows were closed, and the air conditioner was operating (cycling on and off). The air-exchange rates for these cases were somewhat higher,  $0.32-0.36 \text{ h}^{-1}$ . An even higher value,  $0.59 \text{ h}^{-1}$ , was determined for a case with the stove range hood fan continuously operating. Much higher air exchange rates — 2.0 and  $3.1 \text{ h}^{-1}$  — were obtained during manipulation experiments when two windows were deliberately opened. These values are similar to the AER obtained during the second observational monitoring interval (62.8-63.3 h). We believe it likely that a window was open during this time because of the high air-exchange rate, but have no confirming evidence.

To explore the influence of outdoor particles on indoor PN concentrations at site H5, we computed the ratio of indoor to outdoor particle concentrations, based on time-averaged concentration levels, during times when the house is unoccupied. Four suitable intervals were identified, with a total duration of 29 h. The results are presented in Table 3.42. For three of these periods, the air conditioner was off. In these cases,  $f_I$  ranges from 0.24 to 0.60 with a time-weighted average value of 0.43. When the air-conditioner is on, the value of  $f_I$  is considerably lower, 0.11. Because of this observation, combined with the expectation that recirculating the air through the air handling system filter reduces indoor particle concentrations, we use these different  $f_I$  values combined with sensor-based information on air conditioner operation to estimate the indoor PN concentrations of outdoor origin in apportioning PN exposure to its causes.

Information about the indoor concentration measured upstairs in relation to the outdoor level is only available for one interval, the first. During this time, the average indoor PN level upstairs matches the value downstairs, and the ratio of indoor (upstairs) to outdoor is  $f_2 = 0.49$ .

| Time (h)    | <b>AER</b> $(h^{-1})$ | <b>WS</b> (m $s^{-1}$ ) | $ \Delta \mathbf{T} $ (°C) | House configuration                  |
|-------------|-----------------------|-------------------------|----------------------------|--------------------------------------|
| 16.2-18.2   | 0.08                  | 1.8                     | 1.3                        |                                      |
| 62.8-63.3   | 2.3                   | 4.5                     | 1.1                        | Open window? <sup>c</sup>            |
| 85.2-87.2   | 0.36                  | 4.5                     | 6.5                        | Air conditioner cycled on and off.   |
| A1 (30 Jun) | 0.20                  | 4.0                     | 1.9                        |                                      |
| A2 (30 Jun) | 0.32                  | 4.5                     | 4.2                        | Air conditioner cycled on and off.   |
| A3 (30 Jun) | 0.20                  | 4.5                     | 3.3                        |                                      |
| A4 (30 Jun) | 0.34                  | 4.9                     | 4.2                        | Air conditioner cycled on and off.   |
| A5 (1 July) | 0.17                  | 2.7                     | 2.6                        |                                      |
| A6 (1 July) | 0.59                  | 3.1                     | 6.0                        | Stove range hood exhaust fan on.     |
| A7 (2 July) | 2.0                   | 3.1                     | 0.5                        | Kitchen and living room window open. |
| A8 (2 July) | 0.19                  | 3.6                     | 9.1                        |                                      |
| A9 (2 July) | 3.1                   | 3.6                     | 8.3                        | Kitchen and living room window open. |

**Table 3.41.** Air-exchange rates (AER) determined by carbon dioxide concentration decay in house site H5.<sup>a, b</sup>

<sup>a</sup> Unless otherwise noted, the configuration of the house during these measurements was this: all exterior doors and windows closed; all interior doors open; central air system off.

<sup>b</sup> The net indoor CO<sub>2</sub> levels are referenced to a baseline value of 400 ppm, based on the result of a CO<sub>2</sub> rebound experiment.

<sup>c</sup> The high air-exchange rate during this time makes it improbable that the house was entirely closed; however, we have no confirmation of open windows.

**Table 3.42.** Analysis of the indoor to outdoor particle concentration ratios at house site H5 for periods when the house is unoccupied and all doors and windows are closed.<sup>a</sup>

|             |     | WS           | $ \Delta \mathbf{T} $ | PN_out                     | PN_in1                   | PN_in2                   | $f_1$ | $f_2$ |
|-------------|-----|--------------|-----------------------|----------------------------|--------------------------|--------------------------|-------|-------|
| Time (h)    | AC? | $(m s^{-1})$ | (°C)                  | $(10^3  \mathrm{cm}^{-3})$ | $(10^3 \text{ cm}^{-3})$ | $(10^3 \text{ cm}^{-3})$ | (—)   | (—)   |
| 14.8-23.3   | Ν   | 2.1          | 3.5                   | 5.0                        | 2.4                      | 2.4                      | 0.49  | 0.49  |
| 62.8-67.7   | Ν   | 3.8          | 6.6                   | 8.5                        | 5.1                      |                          | 0.60  |       |
| 85.2-93.7   | Y   | 3.7          | 5.5                   | 10.6                       | 1.1                      | _                        | 0.11  |       |
| 93.7-100.7  | Ν   | 1.5          | 9.6                   | 2.9                        | 0.7                      |                          | 0.24  |       |
| Avg. AC off |     | 2.3          | 6.4                   | 5.1                        | 2.5                      | 2.4                      | 0.43  |       |

At this site, evidence suggests that the electric cook stove, a clothes iron, cleaning product and, possibly, microwave oven use were associated with episodic indoor particle emissions. This evidence is based primarily on a comparison of information from occupant diaries (Table 3.43) with the time series of measured indoor and outdoor PN concentrations.

Recorded activities that were not associated with clear increase in indoor PN concentrations include these: use of electric kettle (seven recorded uses with only one potentially contributing, to peak d); electric clothes washer (two uses); electric clothes dryer (one use); vacuuming (two occasions); use of the dishwasher (one occasion); use of Lysol spray (one time); and mopping with "Armstrong" floor cleaner (one occasion).

We quantitatively analyzed the peak events using an integral material balance approach. Where possible, we quantified first-order loss rate coefficients (k + a) for individual peak events. We also estimated source strengths ( $\sigma$ ). The results of this analysis are reported in Table 3.44. For the first ironing event, we separately analyzed the impact upstairs (b2) and downstairs (b1) because of the distinct, short-lived concentration peak that only appeared in the upstairs monitor. For the second ironing event, a peak again appeared in the upstairs monitor but there was no clear effect on downstairs PN concentrations. For these cases, we treated the two floors of the house as independent (upstairs  $V = 144 \text{ m}^3$ ; downstairs  $V = 276 \text{ m}^3$ ). The emissions from peak *e* began during the interval when we were not monitoring, so we could not estimate total particle emissions associated with this event.

| Time (h) | Activity   | PN source?     |
|----------|--|----------------|
| 0.37     | Electric stove, 2 burners  | Y (Peak a)     |
| 2.87     | Electric kettle  | Ν              |
| 13.95    | Iron, blow dryer, perfume  | Y (Peak b)     |
| 14.70    | Electric kettle  | Ν              |
| 23.45    | Electric stove. Boiled rice, fried onions, used pressure cooker. Kitchen exhaust fan on. | Y (Peak c)     |
| 24.20    | Cleaned downstairs (some products had terpenes)  | Y (Peak c)     |
| 24.20    | Electric washer  | Ν              |
| 25.70    | Electric kettle  | Ν              |
| 38.20    | Iron, electric kettle  | Y (Peak d)     |
| 40.20    | Electric stove. Cooked dosa, 3 burners, exhaust fan on                                   | Y (Peak e)     |
| 54.45    | Electric kettle  | Ν              |
| 57.95    | Cleaning upstairs (window opened)  | Y (Peak f)     |
| 60.45    | Microwave, warmed food   | Y (Peak f)     |
| 62.37    | Iron, perfume  | ? <sup>a</sup> |
| 68.03    | Electric kettle  | Ν              |
| 80.20    | Electric kettle  | Ν              |
| 80.70    | Vacuumed upstairs, Lysol spray   | Ν              |
| 81.20    | Electric washer  | Ν              |
| 81.45    | Vacuumed downstairs  | Ν              |
| 81.62    | Mopped downstairs, "Armstrong" floor cleaner   | Ν              |
| 82.28    | Electric dryer   | Ν              |
| 83.03    | Iron   | ? <sup>a</sup> |
| 84.45    | Dishwasher   | Ν              |

Table 3.43. Activity notes from occupant diary at H5 related to potential PN sources

<sup>a</sup> PN concentration data from upstairs are not available after 40 h elapsed time. A peak from ironing was previously detected in the upstairs PN data on two occasions, but not so clearly in the downstairs data.

The two fully quantifiable events associated with cooking on the electric stove (peaks a and c) had the highest emissions,  $26 \times 10^{12}$  and  $25 \times 10^{12}$  particles per event. The impact of cleaning product use (peaks c and f) is somewhat ambiguous, because we were unable to isolate these effects from those of cooking activities. However, in the case of peak f, the confounding activity is use of a microwave oven, which was not observed to be a source of UFP at other sites in this study. Consequently, we believe that peak f is mainly attributable to cleaning.

Three manipulation experiments were conducted at this site to further investigate PN emission sources. Experiment M1a and M1b involved cooking rice on the electric stove.

Experiment M2 involved ironing clothes. Experiment M3a and M3b involved use of several cleaning products (glass cleaner, "Scrubbing Bubbles," and "Pledge"). Experiments M1 and M2 produced clear peaks in the indoor PN concentration and so were confirmed as emission sources. Experiment M3 produced ambiguous results and is not analyzed further. (M3a was conducted with closed windows, which led to very low indoor ozone levels. For M3b, the windows were opened, which increased indoor ozone and indoor particle levels. We are unable to discern whether the increase in indoor particle levels is attributable to ozone + terpene chemistry or is simply a consequence of the higher rate of introduction from outdoors owing to the higher air-exchange rate.) A quantitative analysis of the experimental results for M1 and M2, following the same procedure used for observational monitoring, yielded the results summarized in Table 3.45. Particle decay rates ( $k + a = 0.9-2.1 \text{ h}^{-1}$ ) agree with findings from observational monitoring. The emission rates are also comparable for ironing ( $\sigma \sim 2 \times 10^{12}$  particles) and of similar magnitude from the two cooking activities combined ( $\sigma \sim 15 \times 10^{12}$  particles total for M1a and M1b).

|    |  | Time      | k+a              | $\sigma/V$                | σ                         | $(\sigma/V)/(k+a)$                 |
|----|--|-----------|------------------|---------------------------|---------------------------|------------------------------------|
| ID | Source activity                        | (h)       | $(h^{-1})$       | $(10^3 \mathrm{cm}^{-3})$ | $(10^{12} \mathrm{cm}^3)$ | $(10^3 \text{ cm}^{-3} \text{ h})$ |
| а  | Electric stove (2 burners)             | 0.4-3.0   | 1.6 <sup>a</sup> | 61.0                      | 25.6                      | 37.9                               |
| b1 | Iron (downstairs)                      | 14.4-16.0 | 0.59             | 1.3                       | 0.4                       | 2.2                                |
| b2 | Iron (upstairs)                        | 14.4-16.0 | 1.9              | 13.6                      | 2.0 <sup>b</sup>          | 7.1                                |
| с  | Stove & cleaning products <sup>c</sup> | 23.5-38.6 | 1.0 <sup>d</sup> | 59.2                      | 24.8                      | 59.2                               |
| d  | Iron (upstairs)                        | 38.6-39.5 | 1.8              | 10.4                      | 1.5 <sup>b</sup>          | 5.7                                |
| e  | Stove (3 burners) <sup>c</sup>         | 40.2-42.3 | 1.0              | n/a                       | n/a                       | 14.2                               |
| f  | Cleaning & microwave                   | 59.2-62.7 | 0.65             | 11.2                      | 4.7                       | 17.2                               |

**Table 3.44.** Analysis of indoor UFP sources at H5 from observational monitoring.

k+a estimate includes decay due to removal by air treatment device in air handling system.

<sup>b</sup> The upstairs house volume is used to obtain this estimate (144 m<sup>3</sup>)

<sup>c</sup> Range hood exhaust fan on during cooking.

<sup>d</sup> No clear decay period, so k+a could not be calculated. Value recorded  $(1.0 \text{ h}^{-1})$  is an estimate, was used to calculate  $\sigma/V$  and  $\sigma$  using the integral material balance approach.

| Table 3.45. Analysis of PN emission source charac | teristics from manipulation experiments at |
|---|--|
| site H5.  |  |

|     |  | Duration | <i>k</i> + <i>a</i> | $\sigma/V$                | σ           |
|-----|--|----------|---------------------|---------------------------|-------------|
| ID  | Source   | (h)      | $(h^{-1})$          | $(10^3 \mathrm{cm}^{-3})$ | $(10^{12})$ |
| M1a | Electric stove, boiled rice                    | 1.9      | 1.2                 | 21.2                      | 8.9         |
| M1b | Same as M1a but with range hood exhaust fan on | 1.2      | 2.1                 | 14.2                      | 6.0         |
| M2  | Ironing  | 1.5      | 0.9                 | 91.0                      | 2.0         |

The PN exposure analysis for occupant R1 at site H5 is summarized in Table 3.46. For the "awake at home" time, we used the downstairs PN concentration (PN\_in1) as the best estimate of the exposure concentration. For the "asleep" time, we used the upstairs PN concentration (PN\_in2) as the best estimate of exposure concentration. Those data were only available for the first two nights. The ratio of the average PN\_in2 to average PN\_in1 for those two nights was 0.89. So, to estimate exposure concentrations while sleeping for nights three and four, we used 89% of the average PN\_in1 concentration. The "total cumulative exposure" for the

time spent at home,  $294 \times 10^3$  cm<sup>-3</sup> h, corresponds to a daily exposure rate of  $70 \times 10^3$  cm<sup>-3</sup> h/d. This is the lowest exposure rate for any of the house sites we studied. Among the contributing factors are (a) low concentrations in outdoor air; (b) large house volume within which to dilute indoor emissions; and (c) minimum number of occupants in the house. With respect to (c), it is noteworthy that emissions for certain occupant activities, such as cooking, would be expected to increase with an increasing number of occupants. The result would be higher indoor concentrations to which all household occupants would be exposed.

| Parameter   | R1 (F)           |  |  |
|---|------------------|--|--|
| Occupancy status  |                  |  |  |
| Time at home, awake (h)   | 39.0             |  |  |
| Time at home, asleep (h)  | 31.5             |  |  |
| Time away from home (h)   | 30.2             |  |  |
| Exposure duration (d) <sup>a</sup>                                      | 4.2              |  |  |
| Average exposure concentrations and exposures                           |                  |  |  |
| Average concentration (PN_in1), indoor awake $(10^3 \text{ cm}^{-3})$   | 5.6              |  |  |
| Average concentration (PN_in2), indoor asleep $(10^3 \text{ cm}^{-3})$  | 2.4 <sup>b</sup> |  |  |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                  | 294              |  |  |
| Cumulative exposure rate $(10^3 \text{ cm}^{-3} \text{ h d}^{-1})$      | 70               |  |  |
| Indoor exposure attributable to particles of outdoor origin             |                  |  |  |
| Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$  | 133              |  |  |
| Percentage attributable to particles of outdoor origin                  | 45%              |  |  |
| Exposure attributable to indoor source peaks $(10^3 \text{ cm}^{-3} h)$ |                  |  |  |
| Peak a — stove  | 38               |  |  |
| Peak b — iron (average of upstairs and downstairs)                      | 3                |  |  |
| Peak c — stove and cleaning products                                    | 598              |  |  |
| Peak d — iron (average of upstairs and downstairs)                      | 2                |  |  |
| Peak e — stove  | 14               |  |  |
| Peak f — cleaning products and microwave                                | 17               |  |  |
| Cumulative exposure attributable to indoor sources 13.                  |                  |  |  |
| Percentage attributable to quantified indoor sources                    | 45%              |  |  |
| Indoor exposure of unknown origin                                       |                  |  |  |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                  | 29               |  |  |
| Percentage of unknown origin  | 10%              |  |  |

Table 3.46. Exposure analysis for resident of house site H5 during observational monitoring.

<sup>a</sup> At all other sites, exposure duration is computed as the total monitoring period, minus 24 h per day not slept at home. Here, the observational monitoring period extended over a fifth night, even though the total period, 100 h, was only slightly over four days. So, even though the occupant was away the final night, we treated the exposure duration as the full monitoring period divided by 24 h/d.

<sup>b</sup> The average PN concentration indoors during "asleep" periods is estimated as the upstairs average concentration. For the third and fourth nights of sleeping, only downstairs data are available. These were used in computing the average with a 0.89 correction factor, based on the average upstairs to downstairs ratio during the sleeping periods on the first and second nights.

The contribution of outdoor particles to exposure was estimated by taking the appropriate average fraction of the appropriate averaged outdoor concentration. We used  $f_1 = 0.43$  for "awake" periods with the air conditioning off;  $f_1 = 0.11$  for "awake" periods with the air conditioning on; and  $f_2 = 0.49$  for periods "asleep". The cumulative contribution of outdoor particles to indoor exposure was estimated to be  $133 \times 10^3$  cm<sup>-3</sup> h, corresponding to 45% of the total cumulative exposure.

The contributions to exposure from the six quantifiable peak events were assessed with a cumulative total contribution to exposure of  $133 \times 10^3$  cm<sup>-3</sup> h, which is 45% of total for R1 during the observational monitoring period. The remainder — 10% of the total — is of unknown origin.

## 3.2.7. Site H6

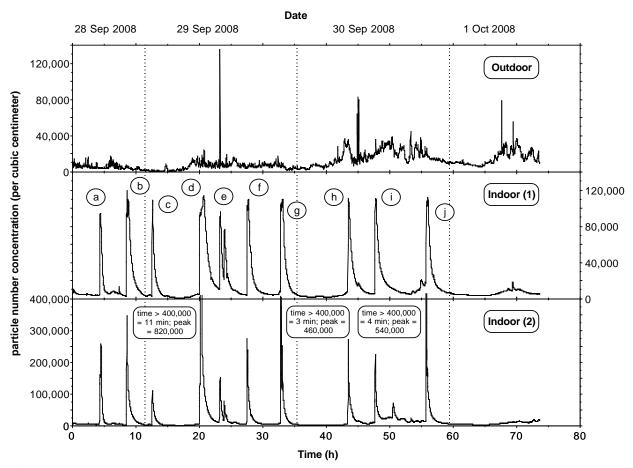
Observational monitoring was conducted at house site H6 for a continuous 73-h period commencing at 12:33 on 28 September and ending at 14:11 on 1 October 2008. For detailed background information about this house site, the reader is referred to 22.1. See also Figure C.7 (Appendix C) for a floor plan. Summarizing briefly, this recently constructed (1996) two-story house has an internal living volume of about  $314 \text{ m}^3$ . The house is in Emeryville, a small city that neighbors Oakland on the northwest. It is situated on a medium-trafficked residential street, about 200 m west of a heavy arterial road and about 800 m east of I-80, a very heavily used freeway. The house has a pilotless gas cooking range and an electric oven. It is equipped with central forced-air heating, but that system did not operate during the course of this study owing to the comfortable ambient temperatures. The house has three occupants, all adults (R1 = female adult; R2 = male adult; R3 = male adult).

A summary of the time-averaged particle number (PN) concentrations was presented in Table 3.1 (§3.1). The average levels measured during the entire observational monitoring period were 11.0 (outdoors), 15.6 (indoors downstairs), and 20.8 (indoors upstairs) in units of 1000 particles per cm<sup>3</sup>. The average outdoor PN levels measured at this site were on the lower side of the range observed in this study (the average for all seven sites was  $14.9 \times 10^3$  particles cm<sup>-3</sup>). The indoor levels, on the other hand, were on the high side of the central tendency. Focusing on the main indoor monitor, the average for all seven sites was  $14.5 \times 10^3$  particles cm<sup>-3</sup> about 7% lower than the average at this house site. Another interesting feature of the overall data comes from a comparison of the PN concentrations measured by the supplemental and primary monitors. H6 was one of three sites (H1 and H3 were the others) where the supplemental monitor recorded a substantially higher average PN concentration than the primary monitor. These are the three sites with the highest overall average concentrations recorded by the primary indoor monitor. The ratios of supplemental to primary concentrations are similar in all three cases, in the range 1.2-1.4. These are also the three sites where cooking made the largest contributions to indoor PN concentrations. An interesting and surprising feature of the elevated concentrations in the supplemental monitor at H6 is that this monitor was placed on the second floor of the house, whereas cooking occurred on the first floor. As we will see, the peak concentrations associated with cooking events are also higher in the supplemental monitor than exhibited by the primary monitor. It is plausible that the buoyancy of cooking emissions induced more efficient transport into the upper story of the house than throughout the main floor.

Figure 3.24 shows the time-series of particle number concentrations measured at this site. The horizontal axis is labeled with elapsed time of monitoring referenced to the start time of 12:33 on 28 September 2008. The vertical dashed lines demark midnight and distance from these lines can be used to gauge clock time.

The outdoor concentrations at this site display an overall increase between the lower values during the first half of the monitoring period and higher values during the second half. To elaborate, the average outdoor PN concentration from the start of the monitoring period until the end of the day on 29 September (35.5 h) was  $5.9 \times 10^3$  particles cm<sup>-3</sup>. From that point until the end of the monitoring period, the average level rose by almost  $3 \times to 15.7 \times 10^3$  particles cm<sup>-3</sup>. In addition to this overall trend, the levels exhibit a diurnal pattern that is broadly similar to those observed at other sites, with higher daytime levels and lower levels overnight. However, at this site, the peak in the diurnal cycle occurs earlier than elsewhere, with hourly average levels of ~  $18.2 \times 10^3$  particles cm<sup>-3</sup> for the 7:00-8:00 and 8:00-9:00 hours, perhaps reflecting the influence

of morning traffic and the generally weaker vertical dispersion in the morning than at night. The lowest levels in the diurnal cycle are seen 4:00-5:00 at  $4.6 \times 10^3$  particles cm<sup>-3</sup>.

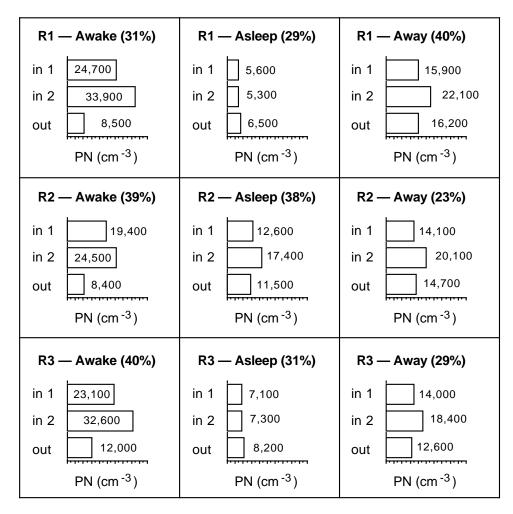


**Figure 3.24.** Time-series of particle number concentrations during observational monitoring at house site H6. Monitor "Indoor (1)" was located on the main floor (downstairs) in the living room; monitor "Indoor (2)" sampled from an upstairs study. On three instances, corresponding to approximate monitoring times of 20 h, 33 h, and 56 h, the "Indoor (2)" monitor recorded levels above 400,000 cm<sup>-3</sup>. The maximum values and the duration of these excursions are noted in boxes. Note that the vertical scales vary among these frames. Indoor concentration peaks are labeled with lower case letters to facilitate later discussion.

Scanning the time-series trace of the indoor PN concentrations, the clearly dominant feature is the sequence of pronounced peaks, characterized by rapid rises and slower decays. There are ten clear peaks that are remarkably similar to one another in the "indoor (1)" trace, representing concentrations measured in the downstairs living room. Each of these peaks rises to roughly 100,000 particles per cm<sup>3</sup>, much higher than the average outdoor level. Upstairs ("indoor (2)") the traces are more variable, but most of the peaks rise to even higher levels than in the downstairs data: eight peaks rise above 200,000 particles per cm<sup>3</sup> and three exceed 400,000 particles cm<sup>-3</sup>. As we will see, all of these ten peaks are associated with cooking events.

The occupancy status of the three household residents of H6 is presented in time-series form in Appendix D (see Figure D.81). Figure 3.25 shows the time-averaged PN concentrations

during observational monitoring sorted by occupancy status. Focusing on the primary indoor monitor, "in 1," we see that the average concentrations when the occupants are at home awake,  $19-25 \times 10^3$  cm<sup>-3</sup>, are considerably higher than the corresponding outdoor concentrations, 8-12 ×  $10^3$  cm<sup>-3</sup>. Compared to levels when they are awake at home, when the occupants are individually away from the house, the indoor levels are lower,  $14-16 \times 10^3$  cm<sup>-3</sup>, even though the outdoor levels are relatively higher during these times,  $13-16 \times 10^3$  cm<sup>-3</sup>. The lowest average indoor PN concentrations occur for each occupant when they are asleep,  $6-13 \times 10^3$  cm<sup>-3</sup>, whereas the outdoor levels are unchanged. The levels for resident R2 are relatively high during the "asleep" time compared with residents R1 and R3. The explanation is that R1 did most of the cooking and R2 reported being asleep during some of the cooking times. The elevated concentrations when occupants were away can also be understood as a residual effect related to cooking.



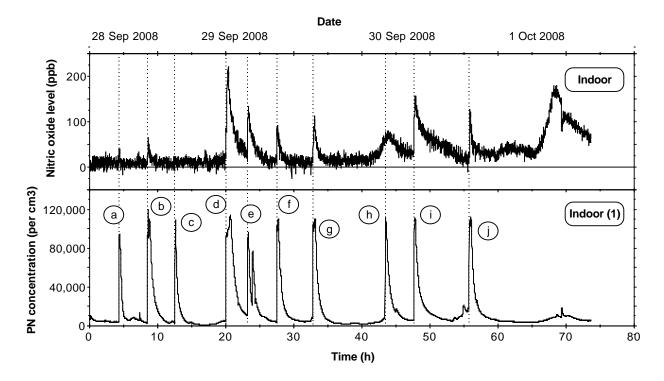
**Figure 3.25.** Time-averaged indoor and outdoor particle number concentrations measured during the observational monitoring period at house site H6 and sorted according to the occupancy status of the three residents of the house. R1 is an adult female; R2 and R3 are adult males. Each bar is labeled with the time-averaged particle number concentration. Indicated in each frame is the percentage of the observational period for which the indicated state applies.

Table 3.47 summarizes the average copollutant levels at H6, sorted according to the occupancy status of the house. In Appendix D are presented time-series plots for the copollutants as follows: Figure D.82 for ozone; Figure D.83 for NO; Figure D.84 for carbon dioxide; and Figure D.85 for carbon monoxide. Outdoor NO levels are low except for a pronounced peak episode on the morning of 1 October when the level rose above 300 ppb. Indoor NO levels exhibit several peaks that correspond to some, but not all, of the PN concentration peaks, as illustrated in Figure 3.26. For seven of the ten identified indoor PN peaks (all except a, c, and h), sudden increases in the NO levels coincide with increases in the PN levels, indicating common sources.

| Species (units)           | All awake | All asleep | Unoccupied |
|---------------------------|-----------|------------|------------|
| NO_in (ppb)               | 32.9      | 21.4       | 44.2       |
| NO_out (ppb)              | 14.1      | 19.9       | 13.3       |
| O <sub>3</sub> _in (ppb)  | 3.3       | 2.8        | 3.6        |
| O <sub>3</sub> _out (ppb) | 17.8      | 10.6       | 24.4       |
| CO <sub>2</sub> _in (ppm) | 600       | 530        | 542        |

**Table 3.47.** Copollutant levels at site H6 during observational monitoring.<sup>a</sup>

<sup>a</sup> Measurements sorted according to occupancy status of the building and then averaged.



**Figure 3.26.** Time-series plots showing the correspondence between the indoor NO and indoor particle number concentrations. The individual frames shown here are reproduced from Figure D.83 (NO) and Figure 3.24 (PN). The vertical dashed lines correspond to peaks in the indoor PN level and are presented to guide the viewer's eye. The elevated indoor NO levels at peak h and at time ~ 70 h are probably caused by penetration from outdoors.

At this house site, we made three determinations of the air-exchange rate using carbon dioxide as a tracer gas (see Table 3.48). One of these occurred during the observational monitoring period; the other two (denoted A1-A2) were made during manipulation experiments. For one of the determinations, the doors and windows were closed. For the other two cases, either two or four windows were open. For each of the three determinations, the estimated air-exchange rate was ~0.8-0.9 h<sup>-1</sup>. It is surprising that the AER value is as high with the windows closed as with the windows open. Note, though, that the indoor-outdoor temperature difference was much larger for the determination with the windows closed. Air infiltration rates depend on wind speed and indoor-outdoor temperature differences as the driving forces.

**Table 3.48.** Air-exchange rates (AER) determined by carbon dioxide concentration decay in house site H6. <sup>a, b</sup>

|             |                       |                         |                            | · · · · · · · · · · · · · · · · · · · |
|-------------|-----------------------|-------------------------|----------------------------|---------------------------------------|
| Time (h)    | <b>AER</b> $(h^{-1})$ | <b>WS</b> (m $s^{-1}$ ) | $ \Delta \mathbf{T} $ (°C) | House configuration                   |
| 24.5-26.0   | 0.78                  | 3.6                     | 3.0                        | Two windows open: upstairs bathroom   |
|             |                       |                         |                            | & rear bedroom                        |
| A1 (27 Sep) | 0.93                  | 2.7                     | 9.4                        |                                       |
| A2 (28 Sep) | 0.84                  | 2.2                     | 1.7                        | Four windows open: upstairs bathroom, |
|             |                       |                         |                            | both bedrooms, kitchen                |

<sup>a</sup> Unless otherwise noted, the configuration of the house during these measurements was this: all exterior doors and windows closed; all interior doors open; central air system off.

<sup>b</sup> The net indoor  $CO_2$  levels are referenced to a baseline value of 418 ppm.

To investigate the influence of outdoor particles on indoor PN concentrations, we computed the ratio of indoor to outdoor particle concentrations, based on time-averaged concentration levels, during times when we believe there is negligible influence of indoor sources on indoor particle levels. At most of the other house sites we could conduct this analysis using unoccupied periods. At this house, however, the periods when the house was vacant were brief and there is also evidence during some of these times of the influence of indoor emission sources. Consequently, supplemented the periods of vacancy with analysis of the times when all occupants were asleep. Discussion of Figures E.7b and E.7c (see Appendix E) substantiates the appropriateness of these decisions. Table 3.49 summarizes the results of the five monitoring periods (duration = 16 h) identified in this manner. The ratio  $f_1$  of average indoor PN concentration (downstairs) to average outdoor concentration ranges in these five periods from 0.30-0.81. The mean is 0.44 with a standard error of 0.06 (13%). Considering the average upstairs concentration to average outdoor concentration ratio, the parameter  $f_2$  varies in these five periods from 0.28-0.87, with a mean value of 0.51 and a standard error of 0.08 (16%).

At this site, evidence indicates that the gas cook stove, electric toaster oven, and candle use are associated with episodic indoor particle emissions. This evidence is based primarily on a comparison of information from occupant diaries with the time series of measured indoor and outdoor PN concentrations. Table 3.50 labels occupant activity information with peaks in a manner that corresponds to the information presented in Figure 3.24.

We quantitatively analyzed the peak events using an integral material balance approach. We evaluated first-order loss rate coefficients (k + a) for individual peak events by regressing the natural logarithm of the slope of the net downstairs PN concentration (PN\_in1 minus the product of  $f_1$  and PN\_out) against time. We also estimated source strengths ( $\sigma$ ) by applying the integral material-balance equation. The results are reported in Table 3.51. Peak e is divided into two

subpeaks for the source characterization analysis. Because the peaks overlap, they are combined in exposure apportionment.

**Table 3.49.** Analysis of the indoor to outdoor particle concentration ratios at house site H6 for periods when the house is either unoccupied or all occupants are sleeping and there is no evidence of the influence of indoor sources on indoor PN levels.

| Time (h)               | WS           | $ \Delta \mathbf{T} $ | PN_out                   | PN_in1                     | PN_in2                   | $f_1$ | $f_2$ |
|------------------------|--------------|-----------------------|--------------------------|----------------------------|--------------------------|-------|-------|
|                        | $(m s^{-1})$ | (°C)                  | $(10^3 \text{ cm}^{-3})$ | $(10^{3} \text{ cm}^{-3})$ | $(10^3 \text{ cm}^{-3})$ | (—)   | (—)   |
| 14.5-18.9 <sup>a</sup> | 1.8          | 7.8                   | 3.6                      | 2.0                        | 1.8                      | 0.54  | 0.50  |
| 25.3-26.4 <sup>b</sup> | 3.7          | 3.5                   | 10.3                     | 8.3                        | 8.9                      | 0.81  | 0.87  |
| 38.5-42.4 <sup>a</sup> | 0.9          | 9.4                   | 8.9                      | 2.7                        | 2.5                      | 0.30  | 0.28  |
| 51.5-54.6 <sup>b</sup> | 2.8          | 5.0                   | 21.7                     | 7.1                        | 16.5                     | 0.33  | 0.74  |
| 62.5-66.4 <sup>a</sup> | 1.0          | 10.2                  | 8.3                      | 3.9                        | 4.0                      | 0.47  | 0.48  |
| Average                | 1.7          | 7.9                   | 9.9                      | 4.0                        | 5.7                      | 0.44  | 0.51  |

<sup>a</sup> Period when all three occupants were at home asleep and there was no evident influence of indoor sources on indoor PN levels.

<sup>b</sup> Period when the house was vacant and there was no evident influence of indoor sources on indoor PN levels.

| Time (h) | Activity   | PN source? |
|----------|--|------------|
| 4.28     | Used rice cooker. Grilled fish and broccoli on stove.          | Y (peak a) |
| 8.45     | Fried egg on gas stove.  | Y (peak b) |
| 11.45    | Lit a candle.  | Y (peak c) |
| 12.45    | Used popcorn maker.  | ? (peak c) |
| 19.95    | Fried potstickers on stove.                                    | Y (peak d) |
| 23.45    | Cooked ramen noodles on stove.                                 | Y (peak e) |
| 23.78    | Used toaster oven.   | Y (peak e) |
| 27.45    | Reheated potstickers and broccoli on stove.                    | Y (peak f) |
| 32.30    | Made stew on stove. Used microwave.                            | Y (peak g) |
| 43.28    | .28 Used toaster oven.   |            |
| 47.45    | Cooked noodles and potstickers on stove.                       | Y (peak i) |
| 55.45    | Cooked on stove and in George Foreman grill.                   | Y (peak j) |
| 56.95    | Cleaning using Zep glass cleaner & Method all-purpose cleaner. | Ν          |
| 68.45    | Used toaster oven, made coffee.                                | ?          |

Table 3.50. Activity notes from occupant diary at H6 related to potential PN sources

The quantified emission events are fairly consistent in their key attributes. Across the eleven events (counting e1 and e2 as separate events), the effective particle decay rate, k+a, ranges from 1.2 to 3.6 h<sup>-1</sup>, with a mean value of 1.9 h<sup>-1</sup> and a relative standard error of 11%. Combining e1 and e2 for this second analysis (summing to  $41 \times 10^{12}$  particles emitted), the total estimated particle emissions per event varies over a narrow range,  $26-56 \times 10^{12}$  particles per event, with a relative standard error of 6%.

Two manipulation experiments were conducted at this site to further investigate PN emission sources. Experiment M1 entailed frying onions on the gas range. Experiment M2 involved lighting four tea candles and allowing them to burn for 20 minutes. A quantitative

analysis of the experimental results for M1 and M2, following the same procedure used for observational monitoring, yielded the results summarized in Table 3.52. Particle decay rates  $(k + a = 1.5 \text{ and } 1.9 \text{ h}^{-1})$  agree with findings from observational monitoring. The estimated emission rates are of similar magnitude but several times lower than inferred for the emission episodes during observational monitoring, 11 and  $16 \times 10^{12}$  particles per event for the manipulation experiments, as compared with  $26-56 \times 10^{12}$  particles per event during observational monitoring.

|    |                           | Time      | k+a        | σ/V                      | σ           | $(\sigma/V)/(k+a)$                 |
|----|---------------------------|-----------|------------|--------------------------|-------------|------------------------------------|
| ID | Source activity           | (h)       | $(h^{-1})$ | $(10^3 \text{ cm}^{-3})$ | $(10^{12})$ | $(10^3 \text{ cm}^{-3} \text{ h})$ |
| a  | Stove & rice cooker       | 4.2-5.4   | 3.6        | 141                      | 44          | 39                                 |
| b  | Stove (frying)            | 8.5-10.8  | 1.5        | 133                      | 42          | 89                                 |
| с  | Candle                    | 12.5-13.9 | 1.9        | 84                       | 26          | 44                                 |
| d  | Stove (frying)            | 20.0-23.1 | 1.2        | 179                      | 56          | 152                                |
| e1 | Stove (water)             | 23.1-23.9 | 2.8        | 116                      | 36          | 42                                 |
| e2 | Toaster oven              | 23.9-26.2 | 1.9        | 17                       | 5           | 9                                  |
| f  | Stove (frying)            | 27.4-29.8 | 1.6        | 125                      | 39          | 79                                 |
| g  | Stove (water) & microwave | 32.8-34.8 | 1.8        | 146                      | 46          | 81                                 |
| h  | Toaster oven              | 43.4-44.9 | 1.7        | 112                      | 35          | 65                                 |
| i  | Stove (water & frying)    | 47.6-50.3 | 1.5        | 110                      | 35          | 72                                 |
| j  | Stove & GF grill          | 55.6-59.1 | 1.5        | 127                      | 40          | 83                                 |

Table 3.51. Analysis of indoor UFP sources at H6 from observational monitoring.

**Table 3.52.** PN emission source characteristics from manipulation experiments at site H6.

|    |                          | Duration | k + a      | $\sigma/V$                | σ           |
|----|--------------------------|----------|------------|---------------------------|-------------|
| ID | Source                   | (h)      | $(h^{-1})$ | $(10^3 \mathrm{cm}^{-3})$ | $(10^{12})$ |
| M1 | Gas stove, frying onions | 2.8      | 1.9        | 114                       | 16          |
| M2 | Candles                  | 2.8      | 1.5        | 79                        | 11          |

Exposure analysis for H6 occupants is summarized in Table 3.53. For the "awake at home" time, we used the downstairs PN concentration (PN\_in1) and for the "asleep" time, we used the upstairs PN concentration (PN\_in2) as the best estimates of exposure concentrations. The total cumulative exposure for the time spent at home ranged from  $669 \times 10^3$  cm<sup>-3</sup> h for R1 to  $1045 \times 10^3$  cm<sup>-3</sup> h for R2. The daily exposure rates were obtained by dividing these figures by the duration of household monitoring when the occupants were at home (judged by where they slept), 3.1 days in each case. The results range from  $218 \times 10^3$  cm<sup>-3</sup> h/d for R1 to  $341 \times 10^3$  cm<sup>-3</sup> h/d for R2.

The contribution of outdoor particles to exposure was computed by taking the appropriate average fraction,  $f_1 = 0.44$  for "awake" and  $f_2 = 0.51$  for "asleep" of the average measured outdoor concentration during respective occupancy states for each resident and computing an exposure increment for each occupant. The cumulative contribution of outdoor particles to indoor exposure ranged from  $155 \times 10^3$  cm<sup>-3</sup> h (R1) to  $270 \times 10^3$  cm<sup>-3</sup> h (R2). Outdoor particles that penetrate and persist indoors are estimated to contribute 23-29% of the in-house exposure of these occupants during the observational monitoring period.

The contributions to exposure from the ten quantifiable peak events were assessed with cumulative totals ranging from  $506 \times 10^3$  cm<sup>-3</sup> h for R1 to  $791 \times 10^3$  cm<sup>-3</sup> h for R2. For the three occupants of H6, the estimated contribution of indoor emission sources to PN exposure indoors is in the range 72-76%. The small remainders of the estimated exposures that are of unknown origin amount to only ~1% of the total in each case.

| Parameter  | <b>R1</b> ( <b>F</b> ) | <b>R2</b> (M) | R3 (M) |
|--|------------------------|---------------|--------|
| Occupancy status   |                        |               |        |
| Time at home, awake (h)  | 22.5                   | 28.8          | 29.8   |
| Time at home, asleep (h)   | 21.5                   | 28.0          | 22.5   |
| Time away from home (h)  | 29.7                   | 16.9          | 21.4   |
| Exposure duration (d) <sup>a</sup>   | 3.1                    | 3.1           | 3.1    |
| Average exposure concentrations and exposures                                  |                        |               |        |
| Average concentration (PN_in1), indoor awake $(10^3 \text{ cm}^{-3})$          | 24.7                   | 19.4          | 23.1   |
| Average concentration (PN_in2), indoor asleep $(10^3 \text{ cm}^{-3})$         | 5.3                    | 17.4          | 7.3    |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                         | 669                    | 1045          | 854    |
| Cumulative exposure rate $(10^3 \text{ cm}^{-3} \text{ h d}^{-1})$             | 218                    | 341           | 278    |
| Indoor exposure attributable to particles of outdoor origin                    |                        |               |        |
| Cumulative contribution to exposure $(10^3 \text{ cm}^{-3} \text{ h})$         | 155                    | 270           | 251    |
| Percentage attributable to particles of outdoor origin                         | 23%                    | 26%           | 29%    |
| <i>Exposure attributable to indoor source peaks</i> $(10^3 \text{ cm}^{-3} h)$ |                        |               |        |
| Peak a — Stove and rice cooker   | 0                      | 0             | 39     |
| Peak b — Stove (frying)  | 89                     | 89            | 89     |
| Peak c — Candle  | 38                     | 44            | 38     |
| Peak d — Stove (frying)  | 119                    | 281           | 152    |
| Peak e — Stove (water) and toaster oven  | 37                     | 64            | 64     |
| Peak f — Stove (frying)  | 76                     | 76            | 44     |
| Peak g — Stove (water) and microwave   | 81                     | 81            | 0      |
| Peak h — Toaster oven  | 1                      | 70            | 73     |
| Peak i — Stove (water & frying)  | 0                      | 3             | 28     |
| Peak j — Stove and grill   | 64                     | 83            | 83     |
| Cumulative exposure attributable to episodic indoor sources                    | 506                    | 791           | 611    |
| Percentage attributable to quantified episodic indoor sources                  | 76%                    | 76%           | 72%    |
| Indoor exposure of unknown origin  |                        |               |        |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                         | 8                      | -16           | -9     |
| Percentage of unknown origin   | 1%                     | -2%           | -1%    |

**Table 3.53.** Exposure analysis for resident of house site H6 during observational monitoring

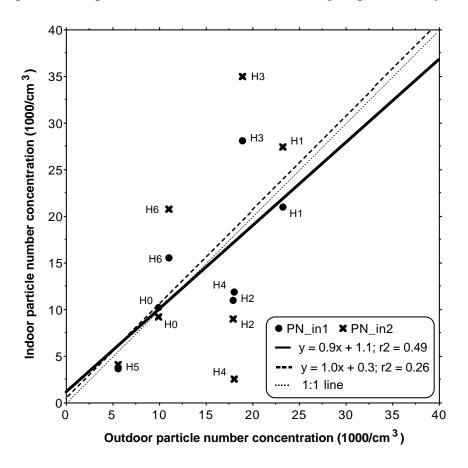
<sup>a</sup> Exposure duration is computed as the total monitoring period, minus 24 h per day not slept at home, with the result divided by 24 h/d.

## 3.3 Synthesis of the Research Findings from the Seven House Sites

In this subsection, we explore the study results from the seven house sites in aggregate. The presentation addresses the main themes discussed for each of the sites individually:

- Particle number PN concentrations
- House air-exchange rates
- Influence of outdoor particles on indoor levels
- Indoor particle emission sources
- Exposure of occupants to PN and its attribution to sources

Table 3.1 (see §3.1) presented a summary of parameter values measured during observational monitoring at the seven house sites. We have seen that outdoor particles are attenuated in their influence on indoor PN levels. Three main effects can cause indoor PN levels to be less than outdoor levels in the absence of indoor sources: particle loss that may occur as air leaks into the building envelope; particles deposit on interior house surfaces; and particles may be removed from indoor air by means of active filtration, e.g. in a house's central air handling system. An interesting and important question is this: to what degree can indoor PN levels be predicted by outdoor concentrations? Figure 3.27 displays a scatter plot with the main (in1) and the supplemental (in2) PN concentrations as the dependent variables and the outdoor level as the independent variable. Each point represents a time-average value for the entire monitoring period (except for in2 at H5, for which the average represents only the first 40 h of monitoring).



**Figure 3.27.** Scattergram of time-averaged particle number concentrations measured indoors (primary =  $PN_in1$ ; supplemental =  $PN_in2$ ) versus outdoors at seven home sites.

The data do show the expected trend, that higher indoor PN levels are correlated with higher outdoor levels. However, the scatter about the trend lines is large, as reflected in the low correlation coefficients ( $r^2 = 0.49$  for the primary indoor site and  $r^2 = 0.26$  for the supplementary indoor site). This poor correlation can be understood mechanistically. Houses vary in two important ways that affect the outcome observed here. First, the indoor proportion of outdoor particles can vary moderately from one house to another. These differences can arise because of variability among houses in factors such as tightness of the building envelope (which affects air-exchange rates and can influence particle penetration), indoor air motion and degree of furnishing (which can affect indoor particle deposition rates), and the extent of operation and associated effectiveness of filtration in residential air-handling systems. Second, the indoor source characteristics can vary substantially from one house to another. These features are explored later in this subsection.

An interesting and unexpected outcome is that the indoor and outdoor concentrations, averaged across the seven sites, are approximately the same. Table 3.54 presents the average and standard error for the time-averaged PN concentration measurements across all seven sites. The averages are in agreement within the uncertainty indicated by the standard error.

**Table 3.54.** Overall summary statistics of particle number concentrations ( $\times 10^3$  cm<sup>-3</sup>) measured at the seven house sites.

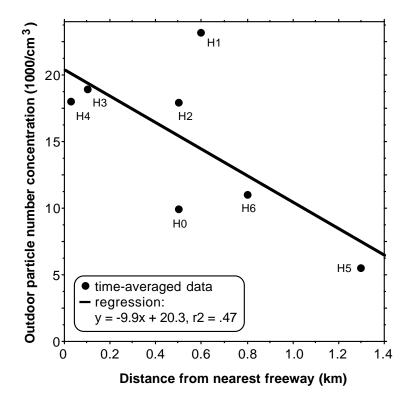
| Parameter                 | Average <sup>a</sup> | Std. Error | Range    |  |
|---------------------------|----------------------|------------|----------|--|
| Outdoor (PN_out)          | 14.9                 | 2.3        | 5.5-23.2 |  |
| Primary indoor (PN_in1)   | 14.5                 | 3.0        | 3.7-28.1 |  |
| Secondary indoor (PN_in2) | 15.4                 | 4.7        | 2.6-35.0 |  |

<sup>a</sup> The time-weighted average is first computed for each site over the full observational monitoring period; the results for the seven sites are linearly averaged to obtain the entries in this column.

The proximity to major roadways was correlated to some extent with outdoor PN levels. Table 3.55 summarizes the data and Figure 3.28 presents a simple regression analysis, which exhibits a moderate  $r^2$  value of 0.47. A few important caveats must be noted. First, H5 in Livermore is quite different than the other six sites, which are in the more highly urbanized cities of Oakland (H0-H4) and Emeryville (H6). If H5 is removed from the analysis, the resulting  $r^2$ declines to 0.12. Second, each of these house sites was measured over a different 3+-day period. Clearly, PN levels at any site vary with time according to factors such as meteorological conditions. At the level of comparing one site to another, our study design does not allow us to separately consider the effects of time and space on outdoor PN levels. We note specifically that the highest time-averaged wind speed during the monitoring period occurred at H3 (4.2 m/s, compared with an average of 2.3 m/s for the other six sites), which is one of two sites within a few hundred m of the nearest freeway. The high winds during our monitoring period could have reduced the local effect of PN emissions from the freeway. Also, monitoring at H5 coincided with a period of intense forest fire activity throughout northern and central California. Ambient PM<sub>2.5</sub> levels were especially high at that time and high levels of fine particles can suppress PN concentrations by serving as a condensational sink for vapors and as a coagulation sink for ultrafine particles.

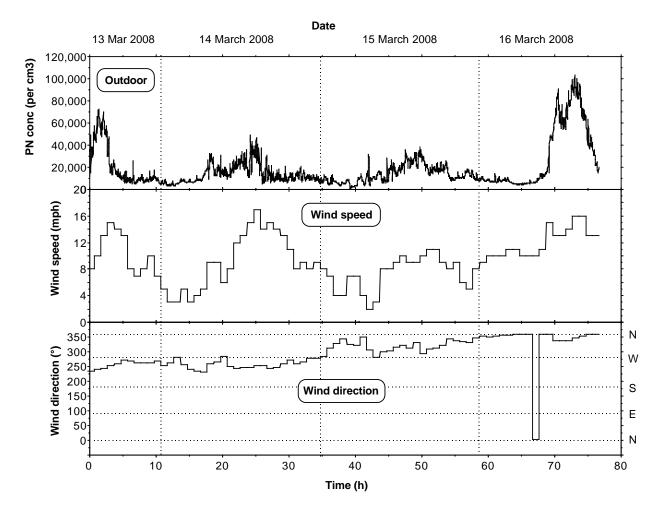
| Site | Avg. PN_out | Fwy location | Note   |
|------|-------------|--------------|--|
| H0   | 9.9         | 0.5 km E     | Second freeway is 1-2 km NW of house           |
| H1   | 23.2        | 0.6 km SE    | House is in the urban core of north Oakland    |
| H2   | 17.9        | 0.5 km NE    | Second freeway is 1 km SW of house             |
| H3   | 18.9        | 0.1 km W     | No heavy-duty truck traffic on nearest freeway |
| H4   | 18.0        | 0.03 km S    | House lot adjacent to freeway                  |
| H5   | 5.5         | 1.3 km N     | House in Livermore, less urban than Oakland    |
| H6   | 11.0        | 0.8 km W     | Second freeway is about 1 km S of house        |

**Table 3.55.** Summary of average outdoor PN concentrations ( $\times 10^3$  cm<sup>-3</sup>) in relation to distance from closest freeway at the seven house sites.



**Figure 3.28.** Correlation between time-averaged outdoor PN level measured at seven house sites and the distance of each site from the nearest freeway.

As a further exploration of these data, we now consider the diurnal pattern of outdoor PN levels at the two sites closest to freeways in relation to wind speed and wind direction. Figures 3.29 and 3.30 present stacked time series plots for H3 and H4, respectively. In these figures, the upper frame displays the onsite measurement results for outdoor particle number concentration. The second and third frames show the wind speed and wind direction, as measured at the UC Berkeley meteorological station. An unavailable piece of important evidence is the time-dependent emissions profile of UFP from the freeway. One expects that the highest emissions would occur during periods when the traffic density is the highest, i.e. early to mid morning and then again mid to late afternoon.

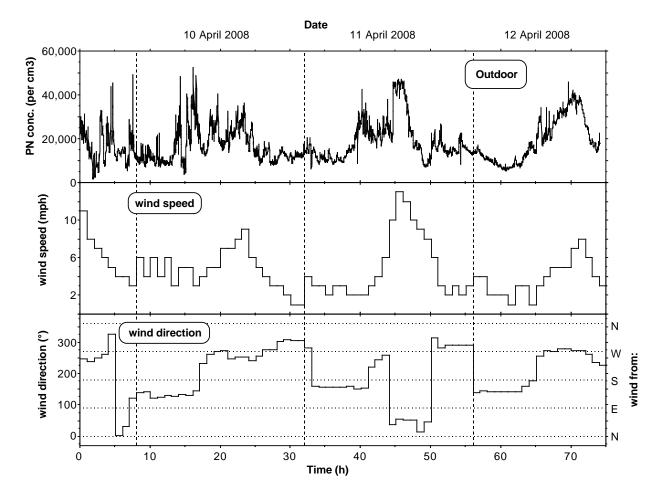


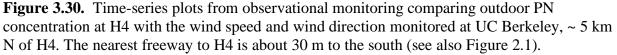
**Figure 3.29.** Time-series plots from observational monitoring comparing outdoor PN concentration at H3 with the wind speed and wind direction monitored at UC Berkeley, 15-20 km NW of H3. The nearest freeway is about 120 m to the west of H3 (see also Figure 2.1).

The plots do not provide clear evidence that the local outdoor PN levels are strongly influenced by emissions from the nearby freeway. At H3, for example, on 14 and 15 March, the outdoor PN concentration peaks midday when the wind speed is highest. There is no pronounced signal that one might associate with heavier rush-hour traffic. Furthermore, the outdoor particle levels are much higher on 16 March than on 14-15 March. However, on this last day, the wind direction has shifted to be from the north, which is roughly parallel to the freeway direction, rather than from the west, for which the freeway would be immediately upwind.

At H4, the higher PN levels on 11 and 12 April correspond in time to when the wind speed is higher, even though one might expect higher wind speeds to cause higher rates of dilution and therefore lower particle levels near strong sources. Furthermore, comparing the two days, the wind directions are different during the high concentration periods, being from the north on 11 April and from the west on 12 April. Nevertheless, the peak PN concentrations on these two days are roughly the same.

The main point from this brief exploration is this. Vehicles traveling on freeways may be important sources of ultrafine particles. But vehicles operate on surface streets, too, and so urban emissions of UFP from vehicles are certainly not restricted to freeways. Furthermore, vehicles are not the only source of ambient UFP. In this research project, we explore the influence of proximity to roadways as one of many possible factors that can influence indoor UFP levels. Relative to other variables, we did not find the influence to be strong.





Air-exchange rate might be expected to influence the indoor PN levels. All else being equal, a higher air-exchange rate should increase the indoor proportion of particles of outdoor origin. On the other hand, a higher air-exchange rate would also decrease the indoor concentration of particles of indoor origin. Combine the effects in a situation where indoor and outdoor sources are of comparable significance, as they are here, and we might expect the relationship between indoor PN levels and air-exchange rate to be small.

In aggregate, we made 41 determinations of the air-exchange rate at the 7 house sites. Table 3.56 summarizes these measurement results. The (arithmetic) average air-exchange rate determined at these seven home sites has a GM of  $0.50 \text{ h}^{-1}$  and a GSD of 2.4. These statistics are

similar to those of a large (although not statistically representative) data set from across the US (N = 2844, GM = 0.5 h<sup>-1</sup>, GSD = 2.1) (Murray and Burmaster, 1995; Nazaroff, 2004).

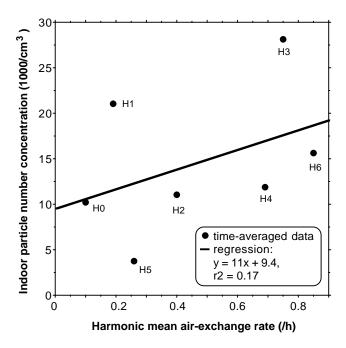
| Site | Ν   | HM $(h^{-1})^{a}$ | $Avg(h^{-1})$ | Range (h <sup>-1</sup> ) | Avg. PN_in1 |
|------|-----|-------------------|---------------|--------------------------|-------------|
| H0   | 2   | 0.10              | 0.10          | 0.09-0.11                | 10.2        |
| H1   | 5   | 0.19              | 0.23          | 0.10-0.34                | 21.0        |
| H2   | 5   | 0.40              | 0.60          | 0.20-1.3                 | 11.0        |
| H3   | 6   | 0.75              | 0.99          | 0.45-2.2                 | 28.1        |
| H4   | 8   | 0.69              | 0.83          | 0.37-1.3                 | 11.9        |
| H5   | 12  | 0.26              | 0.82          | 0.08-3.1                 | 3.7         |
| H6   | 3   | 0.85              | 0.85          | 0.78-0.93                | 15.6        |
| Avg. | 5.9 | 0.46              | 0.63          | 0.3-1.3                  | 14.5        |

**Table 3.56.** Air-exchange rate and primary indoor PN (PN\_in1) concentration ( $\times 10^3$  cm<sup>-3</sup>), determined at seven house sites.

<sup>a</sup> The harmonic mean is the reciprocal of the mean of the reciprocals. So, for example, where the arithmetic mean of 1 and 3 is 2, the harmonic mean of 1 and 3 is 1.5. The harmonic mean is relevant for assessing the relationship of average indoor pollutant levels to average indoor emission sources because the steady-state material balance for concentration depends on the reciprocal of the air-exchange rate.

Figure 3.31 presents a regression analysis between the average primary indoor PN concentration measurements and the harmonic mean air-exchange rate, which shows a weak association ( $r^2 = 0.17$ ). (Regressing the indoor PN concentration against the arithmetic mean air-exchange rate produces an even lower correlation coefficient,  $r^2 = 0.02$ .) In the presence of the other influencing factors, the overall average air-exchange rate is not seen to have a prominent influence on average indoor PN levels at these house sites.

Occupancy was an important factor correlating with indoor PN concentrations. The underlying reasons for the influence are two: (a) the significance of indoor sources contributing to indoor PN concentrations, and (b) the dependence of indoor sources on occupant activities. This finding also matters because occupancy is a necessary component of indoor exposures. Significantly, we found that for each one of the 21 subjects in this study, the time-averaged indoor PN concentration when they were at home exceeded the time-averaged indoor PN concentration for the entire monitoring period. Table 3.57 illustrates the influence, by presenting lognormal distribution parameters for the primary indoor monitor at each site. The minute-byminute measurements at each site were first sorted according to occupancy, with "awake" requiring that all occupants report being home awake, "asleep" requiring that all occupants report being at home asleep, and "away" requiring that the house be vacant. As a measure of the importance of occupant activities on indoor PN levels, compare the GMs when occupants are all awake to the GMs when they are all asleep. These awake/asleep GM ratio varies from 1.8 to 8.3, with an average value across the seven sites of 4.4. To some extent, this ratio is influenced by the fact that outdoor concentrations are lower at night than during the daytime. Perhaps a more fair comparison, then, is to examine indoor concentrations during "awake" and "away" periods. The ratio of GMs for "awake" to "away" is again consistently above 1.0, ranging from 1.3 to 4.3 with an average value of 2.5.



**Figure 3.31.** Regression analysis of average indoor PN concentration (primary site) at each of the seven house sites plotted against the harmonic mean air-exchange rate of the site.

|      | Awake |      |     | Asleep |      |     | Away |      |     |
|------|-------|------|-----|--------|------|-----|------|------|-----|
| Site | GM    | GSD  | %   | GM     | GSD  | %   | GM   | GSD  | %   |
| H0   | 9.6   | 2.44 | 11% | 4.8    | 1.47 | 12% | 3.8  | 1.59 | 22% |
| H1   | 41.7  | 3.76 | 10% | 5.0    | 1.46 | 32% | 9.6  | 1.25 | 20% |
| H2   | 13.9  | 1.86 | 18% | 5.3    | 1.43 | 39% | 9.8  | 1.56 | 31% |
| H3   | 39.9  | 2.41 | 21% | 5.8    | 1.30 | 17% | 10.9 | 1.39 | 13% |
| H4   | 13.6  | 1.90 | 27% | 7.6    | 1.25 | 31% | 10.4 | 1.42 | 27% |
| H5   | 4.3   | 2.09 | 39% | 2.4    | 1.52 | 31% | 1.6  | 2.11 | 30% |
| H6   | 19.1  | 2.77 | 12% | 2.6    | 1.48 | 17% | 10.7 | 1.81 | 12% |

**Table 3.57.** Summary statistics of the lognormal parameters for indoor PN concentrations measured at the seven home sites, with the data first sorted according to occupancy status.<sup>a</sup>

<sup>a</sup> GM = geometric mean, with units of  $10^3$  cm<sup>-3</sup>; GSD = geometric standard deviation; % = percentage of the observational monitoring period when the indicated state applied.

We next turn attention to the sources of particles indoors, first considering the role of outdoor particles and then indoor emission sources. For outdoor particles, at each site we sought to assess parameters we refer to as  $f_1$  and  $f_2$ . These parameters represent the time-averaged concentrations of indoor particles divided by the time-averaged outdoor particle concentration, under conditions in which the indoor sources have negligible influence. We can also refer to these parameters as the "indoor proportion of outdoor particles (IPOP)" (Riley et al., 2002). The primary indoor monitor results (PN\_in1) were represented in  $f_1$  while the supplemental monitor results (PN\_in2) are incorporated in  $f_2$ .

Table 3.58 presents a synopsis of the determinations of  $f_1$  and  $f_2$  at the seven house sites. Note that the values of  $f_2$  are reported only for sites where the supplementary indoor monitor was used in exposure assessment. Focusing on the  $f_1$  values, across the seven sites, the values range from 0.11 to 0.51 with a mean of 0.38 and a standard deviation of 0.14 (RSD = 36%). Considering the broad range of house conditions, the varying times of the year when monitoring was conducted, and the different techniques used in the evaluation, this important parameter demonstrates remarkable coherence across sites. The values of  $f_2$ , for the four sites where we have made a determination, are similar: average = 0.37 and standard deviation = 0.19. To be clear, an average of  $f_1 = 0.38$  means that the expected average indoor PN concentration is 38% of the outdoor level when the only source of particles is outdoor air. This result indicates an average level of protection of 63% by being indoors in a residence against ultrafine particles of outdoor origin, as determined by the particle number concentration measured with a WCPC.

|      | sites. <sup>a, b</sup> |       |       |  |  |  |  |  |  |
|------|------------------------|-------|-------|--|--|--|--|--|--|
| Site | Dur.                   | $f_1$ | $f_2$ | Note   |  |  |  |  |  |
| H0   | 30 h                   | 0.36  | 0.37  |  |  |  |  |  |  |
| H1   | 30 h                   | 0.11  |       | Time-weighted average of conditions with bathroom window open $(f_1$ |  |  |  |  |  |

| <b>Table 3.58.</b>     | Evaluation results for the indoor proportion of outdoor particles at the seven house |
|------------------------|--|
| sites. <sup>a, b</sup> |  |

| пі               | 50 fi  | 0.11 |      | = 0.25); windows closed and air handler off ( $f_1 = 0.16$ ); and windows    |  |  |  |
|------------------|--|------|------|--|--|--|--|
|                  |  |      |      | ,, , , , , , , , , , , , , , , , , , ,                                       |  |  |  |
|                  |  |      |      | closed with air on ( $f_1 = 0.074$ ). Model fit developed using integral     |  |  |  |
|                  |  |      |      | material balance approach accounting for emissions from pilot lights.        |  |  |  |
| H2               | 28 h   | 0.51 |      |  |  |  |  |
| H3               | 63 h   | 0.45 | —    | Based on regression analysis of indoor vs. outdoor concentrations for        |  |  |  |
|                  |  |      |      | full monitoring period after first removing from record times when           |  |  |  |
|                  |  |      |      | indoor sources had an evident influence on PN levels.                        |  |  |  |
| H4               | 22 h   | 0.47 | 0.11 | Upstairs $(f_2)$ floor has continuously operating air cleaner.               |  |  |  |
| H5               | 29 h   | 0.29 | 0.49 | Activity-weighted average of AC off ( $f_1 = 0.43$ ) and AC on ( $f_1 =$     |  |  |  |
|                  |  |      |      | 0.11) yields $f_1$ value. The value for $f_2$ is based on 8.5 h with AC off. |  |  |  |
| H6               | 16 h   | 0.44 | 0.51 | Includes periods when all occupants are asleep (12 h) in addition to         |  |  |  |
|                  |  |      |      | period when house is vacant and there is no evident influence of             |  |  |  |
|                  |  |      |      | indoor sources in indoor PN levels (4 h).                                    |  |  |  |
| <sup>a</sup> Dur | <sup>a</sup> Dur $-$ duration of partial for which data are analyzed to produce average f and f values: f is |      |      |  |  |  |  |

Dur. = duration of period for which data are analyzed to produce average  $f_1$  and  $f_2$  values;  $f_1$  is the ratio of the average indoor concentration measured with the primary monitor to the average outdoor concentration;  $f_2$  is the same as  $f_1$  except that the indoor concentration is based on the supplementary monitor results.

<sup>b</sup> Except where otherwise noted, the  $f_1$  and  $f_2$  results are obtained by analyzing indoor and outdoor concentrations when the house is vacant and there is no evident influence of indoor sources on indoor PN levels.

Indoor emissions were detected and quantified at each of the seven house sites. We separately considered sources that emitted continuously and those that emitted episodically. The only continuous sources we detected were pilot lights associated with the gas cooking appliances in houses H1 and H3. Inferred emission rates and steady-state indoor concentration increments from pilot lights are presented in Table 3.59.

Episodic PN emissions were observed several times at each of the seven house sites. In total, 59 peak events were investigated, which corresponds to an average of 8 peaks per site or about 2.4 peaks per day monitored. Based on a combination of occupant diary information and judiciously deployed temperature sensors, we have identified one or two likely causes of the

episodic peaks in 57 of the 59 events. For 42 of these events, we have identified a single likely source factor. For the other 15 events, two sources were identified. Table 3.60 summarizes the sources that were associated with these peak events.

For the peak events, we were able to evaluate a first-order decay constant (k+a) for 54 episodes and quantify emission source strength ( $\sigma$ ) for 56. Table 3.61 presents a summary of the statistical attributes of the decay constants (39 instances) and source strengths (41 instances) for the peak events that were associated with identifiable single activities.

**Table 3.59.** Summary of emission estimates and steady-state increments to indoor PN concentrations from continuously burning natural-gas pilot lights.

| Site | Emission rate (10 <sup>12</sup> particles/h) | PN increment (1000 cm <sup>-3</sup> ) |
|------|--|---------------------------------------|
| H1   | 0.58   | 2.7                                   |
| H3   | 1.6  | 4.2                                   |

**Table 3.60.** Compilation of activities or events identified as associated with episodic particle emissions during observational monitoring at the seven house sites. <sup>a</sup>

| Source                                      | N1 | N2 |
|---|----|----|
| Gas stove or oven                           | 20 | 9  |
| Furnace (gas fired, central or wall)        | 9  | 1  |
| Electric stove (range) or oven              | 5  | 7  |
| Toaster or toaster oven                     | 4  | 5  |
| Candles                                     | 1  | 1  |
| Ironing clothes                             | 2  | 0  |
| Gas clothes dryer                           | 1  | 0  |
| Microwave oven                              | 0  | 3  |
| Cleaning product use                        | 0  | 2  |
| Miscellaneous (electric grill, rice cooker) | 0  | 2  |

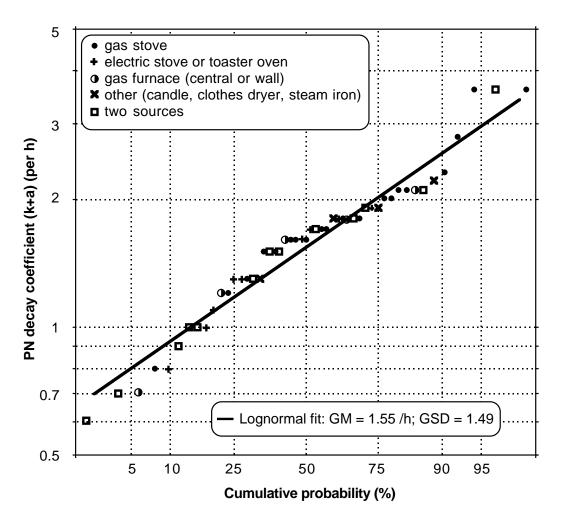
<sup>a</sup> N1 indicates that only one indicated potential source was identified with a particular peak (total = 42 such peaks); N2 indicates that two sources were each potential contributors to a given peak event (15 such peaks).

**Table 3.61.** First-order decay constants and source strengths for peak events at the seven houses associated with identified single activities. <sup>a</sup>

|                      | Decay constant $(k+a)$ $(h^{-1})$ |           |               | Source strength ( $\times 10^{12}$ particles) |           |               |  |
|----------------------|-----------------------------------|-----------|---------------|---|-----------|---------------|--|
| Emission source      | Ν                                 | GM (GSD)  | $AM \pm SD$   | Ν   | GM (GSD)  | $AM \pm SD$   |  |
| Gas stove            | 20                                | 1.8 (1.4) | $1.9 \pm 0.7$ | 19  | 38 (2.1)  | $48 \pm 34$   |  |
| Furnace, central air | 2                                 | 1.6 (1.5) | $1.7 \pm 0.6$ | 2   | 41 (1.1)  | $41 \pm 5$    |  |
| Candle               | 1                                 | 1.9       | 1.9           | 1   | 26        | 26            |  |
| Toaster oven         | 4                                 | 1.7 (1.2) | $1.7 \pm 0.3$ | 4   | 9 (2.8)   | $13 \pm 15$   |  |
| Electric stove       | 5                                 | 1.1 (1.3) | $1.2 \pm 0.3$ | 4   | 10 (2.1)  | $12 \pm 10$   |  |
| Furnace, wall        | 3                                 | 1.3 (1.7) | $1.4 \pm 0.6$ | 7   | 3.1 (2.7) | $4.6 \pm 4.5$ |  |
| Clothes dryer        | 1                                 | 2.2       | 2.2           | 1   | 2.2       | 2.2           |  |
| Steam iron           | 2                                 | 1.5 (1.2) | $1.6 \pm 0.6$ | 2   | 1.9 (1.4) | $2.0 \pm 0.6$ |  |

<sup>a</sup> GM = geometric mean; GSD = geometric standard deviation; AM = arithmetic mean; SD = arithmetic standard deviation.

Figure 3.32 presents a cumulative distribution function for the decay constants (k+a) for 51 of the 54 episodes quantified. These episodes include those summarized in Table 3.61 and also the two-source events and the few cases for which the source is unknown. Overall, the decay constants in these events conform well to a lognormal distribution with a GM of 1.55 h<sup>-1</sup> and a GSD of 1.49. The GSD from the ensemble distribution is similar to the GSDs for events that are clustered by source (see Table 3.61). The implication is that the decay rate constant for UFP emitted in these events does not vary markedly among source types. The three decay constants excluded from the distribution in Figure 3.32 all occurred at H4 and are dominated by the high rate of removal in the recirculating air-filtration systems at that site. The three excluded values were 9.5, 10, and 11 h<sup>-1</sup>. Excluding these three high-decay events, the arithmetic mean value of k+a across all events was 1.7 h<sup>-1</sup>.

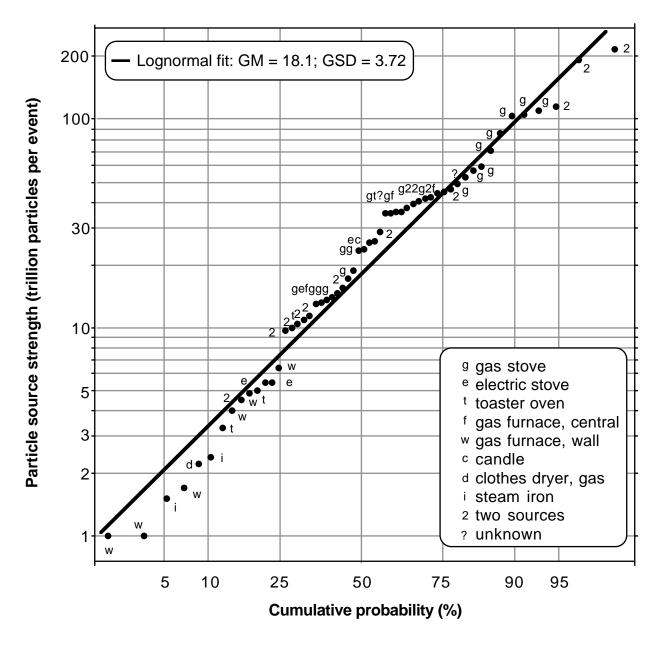


**Figure 3.32.** Cumulative distribution function of 51 determinations of the first-order particle decay rate coefficient obtained from observational monitoring at the seven home sites. For five peaks, the decay coefficient could not be determined (typically because the peaks were too small). For three peaks at H4, the decay coefficients were much higher than the values in this distribution (9-11  $h^{-1}$ ).

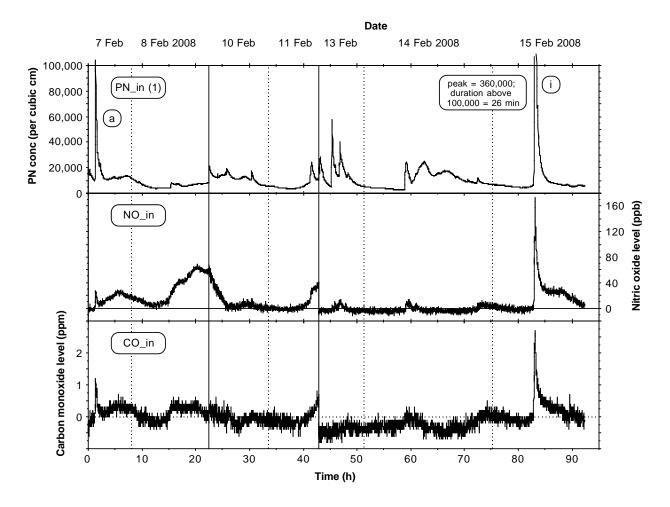
We were able to quantify the source strength of PN emissions for 56 of the 59 peak events. (When we could not quantify emissions, it was because the emissions began at a time when we were not monitoring.) Table 3.61 summarizes the emissions source strength ( $\sigma$ , in units of 10<sup>12</sup> particles per event) for those cases associated with a single activity. Figure 3.33 presents a cumulative distribution function of emissions source strength for all quantifiable events. The straight line in this figure corresponds to a lognormal distribution with GM = 18.1 × 10<sup>12</sup> particles per event and a GSD of 3.72. The overall arithmetic mean is 36 × 10<sup>12</sup> particles per event. The much higher arithmetic mean than GM corresponds to the relatively high dispersion in the distribution, as reflected in the high GSD. Note the high variability in GM or AM values among sources, varying by more than an order of magnitude between the lowemissions events (steam iron, clothes dryer, and wall furnace) and the high-emissions sources (use of a gas stove, central forced-air furnace, or candle). Much of the variance revealed in the high GSD for the aggregate distribution reflects differences in the mean values among these sources.

To put these numbers in perspective, note that an emissions event that releases  $36 \times 10^{12}$  particles suddenly into a house of volume 300 m<sup>3</sup> will increase the PN concentration by  $\sigma/V = 120 \times 10^3$  cm<sup>-3</sup>. This is a substantial increase given the typical average measured indoor concentrations (e.g., see Table 3.57). With regard to an exposure caused by indoor emissions episodes, consider an occupant who is continuously present from the start of the event until the peak has fully decayed. The resulting exposure increment would be  $(\sigma/V)/(k+a)$ . With a typical source strength of  $\sigma/V \sim 120 \times 10^3$  cm<sup>-3</sup> and a typical decay constant of  $(k+a) \sim 1.7$  h<sup>-1</sup>, the exposure increment from this one indoor release event would be  $\sim 70 \times 10^3$  h/cm<sup>3</sup>. Toward the end of this section, we will present information on total PN exposure levels occurring in houses that will permit the reader to see this result in the broader context of overall residential exposures.

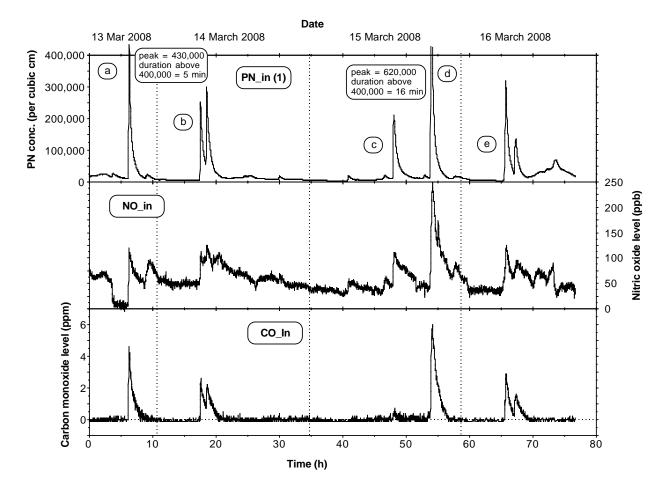
For most of the PN concentration peaks associated with natural gas cooking, copollutant monitoring results provides reinforcing evidence. At five of the seven house sites (all except H0 and H5), natural-gas appliances were used. At four of these sites, we have data from indoor NO and CO monitoring. (At H1, no indoor NO data were acquired.) Figures 3.34-3.37 present stacked time-series plots of observational monitoring data at H2, H3, H4, and H6, respectively, showing (a) the indoor PN concentration at the primary monitor; (b) the indoor NO level; and (c) the indoor CO level. Labels (lower case letters) have been retained on only those peaks when gas-cooking activities on a range top burner occurred. Among the 22 peaks associated with gas cooking in these figures, in all but one case (site H6, peak a) is there *not* a corresponding peak in the NO signal. For carbon monoxide, the evidence is more mixed. At H2 and H3, there are CO peaks for each of the gas cooking-related PN except for peak c at H3. Conversely, at H4 and H6, there are no CO peaks associated with gas cooking, except for peak d at H6. It is possible that significant nitric oxide emissions occur more consistently from gas combustion appliances whereas carbon monoxide, being a product of incomplete combustion, is not significantly emitted from well-tuned gas appliances.



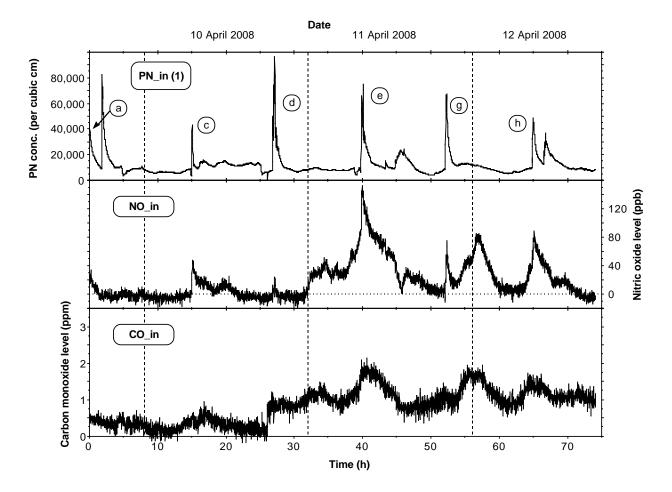
**Figure 3.33.** Cumulative distribution of particle source strength for 56 episodic events that were quantified from all observational monitoring at the seven home sites.



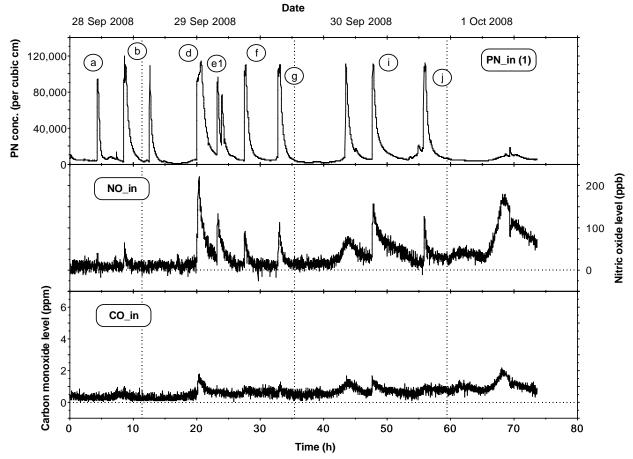
**Figure 3.34.** Time-series plot of the indoor particle number concentration, nitric oxide level, and carbon monoxide level at site H2. The labeled peaks a and i correspond to cooking activities using a natural-gas stove. (See Table 3.23.)



**Figure 3.35.** Time-series plot of the indoor particle number concentration, nitric oxide level, and carbon monoxide level at site H3. The labeled peaks a, b, c, d, and e correspond to cooking activities using a natural-gas stove. (See Table 3.29.)



**Figure 3.36.** Time-series plot of the indoor particle number concentration, nitric oxide level, and carbon monoxide level at site H4. The labeled peaks a, c, d, e, g, and h correspond to cooking activities using a natural-gas stove. (Peaks c and e also corresponded to the use of a toaster oven.) (See Table 3.36.)



**Figure 3.37.** Time-series plot of the indoor particle number concentration, nitric oxide level, and carbon monoxide level at site H6. The labeled peaks a, b, d, e1, f, g, i, and j correspond to cooking activities using a natural-gas stove. (Peak a also corresponded to the use of a rice cooker; peak j also corresponded to the use of a George Foreman grill. (See Table 3.50.)

We conducted manipulation experiments at each site to investigate the properties of emission sources. These experiments are helpful for confirming that a particular activity was (or was not) a source at a given house site. We also quantified emissions and decay constants from the manipulation experiments in which a clear increase in particle number concentration was detected. In total, we ran 20 tests, 13 of which produced an analyzable peak. Table 3.62 summarizes the results. Note that a negative finding does not demonstrate that the particular activity or process is never a UFP source. With our protocol, it is difficult to quantify sources that emit far fewer than ~  $10^{12}$  particles. Furthermore, we observed with some source types that there were clear emissions in some cases but not in others. One example is the toaster (or toaster oven), which was unambiguously a PN source in H0 and H4, probably a source in H6 (based on observational monitoring), and clearly not a significant source in H1. A second example is the (vented) gas furnace, which, based on observational monitoring evidence, was certainly a source in H0, very probably a source in H2, and very likely not a source in H1.

The loss rate coefficients (k+a) obtained from manipulation experiments were similar to those inferred from observational monitoring. The lognormal parameters from the 13 quantified manipulation experiments were GM = 1.6 h<sup>-1</sup> and GSD = 1.4. (Recall that the 51 of 59 quantified observation-period experiments produced a GM = 1.55 h<sup>-1</sup> and GSD = 1.5.) However,

the source strengths from the manipulation experiments were somewhat lower than inferred from observational monitoring events. For the manipulation experiments, the lognormal statistics were  $GM = 9.2 \times 10^{12}$  particles per event with a GSD of 3.4. In the observational monitoring case, the GM was about twice as large, at  $18.1 \times 10^{12}$  particles per event and a GSD of 3.7.

| Site-Test | Source          | k+a (h <sup>-1</sup> ) | $\sigma$ (10 <sup>12</sup> particles) |
|-----------|-----------------|------------------------|---------------------------------------|
| H0-M1     | Toaster         | 1.3                    | 0.7                                   |
| H0-M3     | Electric range  | 1.0                    | 1.7                                   |
| H1-M1     | Gas range       | 2.8                    | 21                                    |
| H1-M2     | Candles         | 2.7                    | 52                                    |
| H2-M1     | Gas oven        | 1.3                    | 24                                    |
| H2-M2     | Gas range       | 1.8                    | 18                                    |
| H3-M3     | Gas range       | 1.4                    | 23                                    |
| H4-M1     | Toaster oven    | 1.9                    | 13                                    |
| H5-M1a    | Electric range  | 1.2                    | 9                                     |
| H5-M1b    | Electric range  | 2.1                    | 6                                     |
| H5-M2     | Ironing clothes | 0.9                    | 2                                     |
| H6-M1     | Gas range       | 1.9                    | 16                                    |
| H6-M2     | Candles         | 1.5                    | 11                                    |

Table 3.62. Summary of results from the source-characterization manipulation experiments.<sup>a</sup>

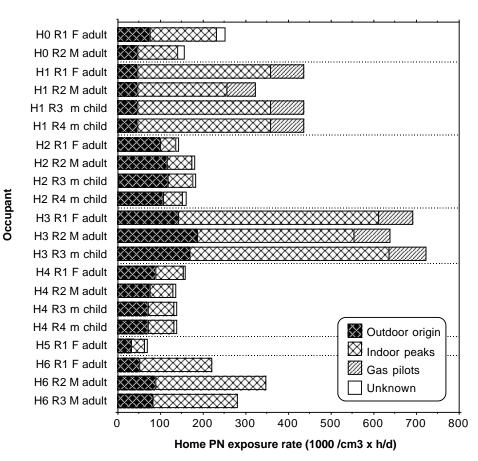
<sup>a</sup>Negative test results: toaster oven (H1); microwave oven (twice, H3); electric kettle (H3); gas furnace (H3); electric clothes dryer (H3); cleaning products (H4); cleaning products (H5).

To complete this section, we now turn to the issue of the residential PN exposure of the 21 occupants of the seven houses investigated in this study, and its apportionment among sources. In §3.1, we presented a summary of the daily residential PN exposure rate for the occupants of the seven houses. Recall, the units of measure are  $10^3 \text{ cm}^{-3}$  h/d, where a value of 1 implies that the average exposure condition is equivalent to one hour per day at  $10^3$  particles per cm<sup>3</sup> or any other combination of concentration and daily duration that yields the same multiplicative product. Figure 3.38 shows how the daily average exposure rate for each occupant is apportioned into different major source categories. The categories considered here are (i) particles of outdoor origin; (ii) particles originating from episodic indoor emissions; (iii) particles originating from gas pilot lights; and (iv) particles of unknown origin.

The main story revealed by this information has these key elements. First, at all sites there are substantial contributions to residential exposure both from the penetration of outdoor particles and from episodic indoor emissions. In fact, at every site and for all occupants, these two source categories dominate. Second, at the three sites with the lowest exposure rates (H2, H4, and H5), the outdoor sources contribute as much or more than the indoor emissions. However, at the other four sites, where the overall exposure rates are the highest, emissions from indoor sources are more important than outdoor particles in contributing to the overall rate of residential PN exposure.

To quantify these last points, let's consider the percentage contribution to exposure rate that is associated with outdoor particles and with indoor sources. Further, let's consider the houses in two groups: Group A is H2, H4, and H5 where outdoor sources are most important; and Group B is H0, H1, H3, and H6, where indoor sources dominate. For the 9 occupants in the Group A houses, their overall average exposure rate is  $144 \times 10^3$  cm<sup>-3</sup> h/d (range = 70-182) and

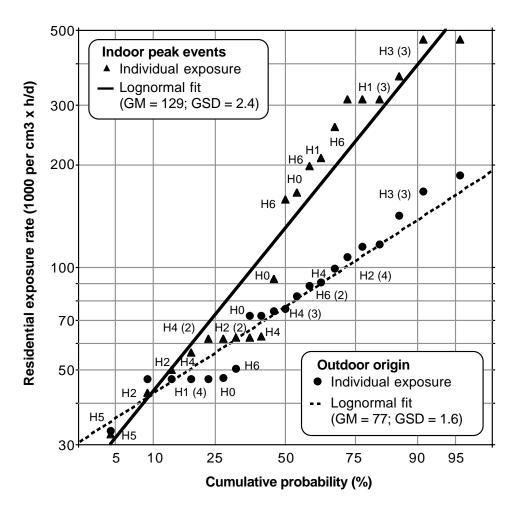
the average contribution to this exposure from outdoor sources is 59% (range: 45-69%). For the 12 occupants in Group B, the average overall exposure rate is almost  $3 \times$  higher,  $411 \times 10^3$  cm<sup>-3</sup> h/d (range = 156-726). Note that the distributions of overall exposures for these two groups barely overlap, as the highest exposure rates at H2 are only slightly above the lower exposure rate at H0. For the Group B residents, the average contribution to exposure from outdoor sources averages only 22% (range 11-30%), whereas, conversely, the average contribution from indoor PN sources is 76% (range 59-87%).



**Figure 3.38.** Apportionment of indoor exposure rate for the 21 occupants of the seven houses studied according to PN source category. The length of the bars in each case only considers exposure that occurs within the homes of the individuals and can be understood as the product of the average indoor PN concentration while at home  $(1000/\text{cm}^3)$  multiplied by the daily average time spent at home (h/d).

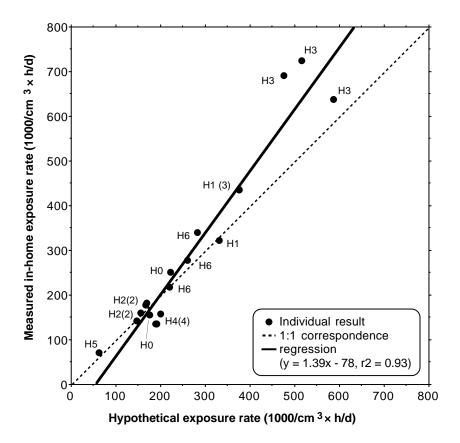
Figure 3.39 explores the distribution of residential PN exposure contributions caused by particles of outdoor origin and by particles from episodic indoor sources. The distributions each conform reasonably well to lognormals (as reflected in this plot by the degree to which the data points follow the straight lines). The parameter values indicate a lower overall average contribution to residential exposure caused by outdoor sources as compared to indoor episodic emissions as well as less variability.

As a conclusion to this section, we consider an important issue in exposure assessment. To what extent can exposures to ultrafine particles (as measured by particle number concentration) be predicted based on outdoor concentration measurements? In exploring this important question, we consider only the component of exposure that occurs when one is indoors at home, since this is the only component we have measured. We also consider the inferred exposure based on outdoor concentrations in the following manner. For each occupant, we compute the average outdoor PN concentration, as measured at their house site, during the hours that they were indoors at home. The product of the average outdoor concentration multiplied by the duration of occupancy (hours per day) yields an exposure indicator that could be derived using only the outdoor concentration measurements and information on occupancy. Let us call this parameter the hypothetical exposure rate.



**Figure 3.39.** Cumulative distribution functions of residential PN exposure rates caused by particles of outdoor origin (circles, dashed line) and by particles emitted from indoor episodic sources (triangles, solid line) for the 21 occupants of the seven houses studied in this project. The points are labeled with the house site (and, to minimize clutter, parenthetical numbers indicate how many nearby points are to be associated with each label).

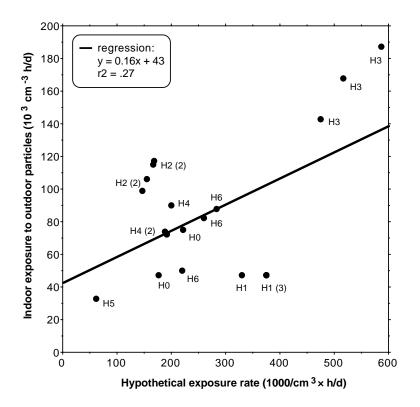
In Figures 3.40 and 3.41, the exposure rates we have determined in this study are compared with the hypothetical exposure rates for the 21 occupants. In Figure 3.40, the dependent variable is the total residential exposure rate. In Figure 3.41, the variable plotted on the y-axis is the estimated residential exposure only to particles of outdoor origin.



**Figure 3.40.** Scatterplot and regression of measured residential exposure rate against the hypothetical exposure rate based solely on outdoor PN concentration measurements. The measured residential exposure rate represents the average indoor concentration during occupancy multiplied by the duration of occupancy (h/d). For the hypothetical exposure rate, the average outdoor concentration during occupancy is multiplied by the duration of occupancy.

Figure 3.40 shows that the hypothetical exposure rate can predict well the measured inhome exposure, with an  $r^2$  value of 0.93. Note, though, that the overall exposure rate is not the same as the hypothetical exposure rate: the slope of the regression line is 1.4, and the data exhibit a strong tendency to lie above the 1:1 line for higher overall exposure rates.

One of the limitations of measuring particle number concentration with a WCPC is that it does not distinguish among particle sources. There are reasons to be concerned with distinguishing between exposures to particles of outdoor and indoor origin. Figure 3.41 shows the relationship between the estimated residential exposure to particles of outdoor origin and the hypothetical PN exposure rate. The regression is only fair, with an  $r^2$  value of 0.27.



**Figure 3.41.** Scatterplot and regression of inferred residential exposure rate to particles of outdoor origin against the hypothetical exposure rate based solely on outdoor PN concentration measurements. The measured residential exposure rate represents the average indoor concentration during occupancy that is attributable to outdoor particle sources multiplied by the duration of occupancy (h/d). For the hypothetical exposure rate, the average outdoor concentration during occupancy is multiplied by the duration of occupancy.

# 4. RESULTS AND DISCUSSION: SCHOOLS

This section of the report presents research findings based on monitoring and analysis at six classroom sites. As for houses, this section is organized in three parts. In the first part, an overview of the observational monitoring results is presented. The aim of this subsection is to provide an overall perspective on what was observed in the six classrooms so that the more detailed presentation can be read in context. The second subsection presents the detailed findings obtained from the six sites. In the third subsection, we gather and synthesize findings from the monitored classrooms across the full set of study variables. We seek general insight and understanding of UFP concentrations and exposures in classrooms that can be gleaned from the new data collected and analyzed in this study.

Because of the very large quantity of information gathered in this study, time-series plots of data gathered but not discussed in detail are presented in an appendix (Appendix F). Also, cumulative distribution functions of particle number concentrations, sorted according to occupancy conditions of the classrooms, are presented and discussed in Appendix G.

## 4.1 Overview of Research Findings from Observational Monitoring in Schools

Measurement results are summarized in Table 4.1. The first few rows indicate when (i.e., during which season and year) and for how long monitoring was conducted in each classroom. The average particle number concentrations in the subsequent three lines are of central interest for this project. Here, time-averaged particle number concentrations from the three WCPCs operated at each site are reported. The primary and supplementary indoor monitors are labeled "PN\_in1" and "PN\_in2," respectively, and the outdoor monitor is labeled "PN\_out." The remaining rows report concentrations of other species (copollutants) either measured on-site or at a central monitoring station and basic environmental parameters (temperature and wind speed) that can influence ventilation, the rate of transport and dispersion of pollutants from ambient sources, and potentially other factors that could have affected what was observed.

In this table, we have not reported the time-averaged outdoor  $CO_2$  level nor the indoor or outdoor carbon monoxide levels because we deem the measured results for these species to be largely noninformative. The carbon dioxide levels measured by the Q-Trak in ambient air exhibit a much broader range of values that is plausible; we suspect that the instrument has a temperature sensitivity making it unsuitable for ambient concentration measurements. With respect to CO, the manufacturer specifications for the Q-Trak indicate that its accuracy is the larger of 3% of the reading or 3 ppm. At all sites, the average CO monitoring result was less than 3 ppm.

Figure 4.1 and Table 4.2 present summaries of the average particle number concentrations measured at each school site during observational monitoring. These data are sorted according to occupancy conditions in the classroom.

A few key points are made here based on the average particle number concentrations. First, considering the *total* observational periods, the primary indoor monitor (In1) produced an overall average across the six sites of  $6.9 \times 10^3$  particles cm<sup>-3</sup>, which corresponds to 47% of the overall average of the outdoor monitoring results ( $14.6 \times 10^3$  particles cm<sup>-3</sup>). The I/O ratio based on overall average concentrations was consistently less than 1.0, varying from a low of 33% at S5 to a high of 74% at S3.

| Parameter (units)               | <b>S1</b>         | S2    | <b>S3</b> | S4                | <b>S</b> 5 | <b>S6</b> |
|---------------------------------|-------------------|-------|-----------|-------------------|------------|-----------|
| Season/year                     | Sp '08            | F '08 | F '08     | F '08             | F '08      | F '08     |
| Duration (h)                    | 95                | 73    | 71        | 72                | 71         | 70        |
| PN_in1 (1000 cm <sup>-3</sup> ) | 9.8               | 5.1   | 10.5      | 7.3               | 3.2        | 5.6       |
| PN_in2 (1000 cm <sup>-3</sup> ) | 16.9 <sup>a</sup> | 10.5  | 12.6      | 9.8               | 7.1        | 8.0       |
| $PN_out (1000 \text{ cm}^{-3})$ | 16.0              | 12.9  | 14.2      | 21.3 <sup>b</sup> | 9.7        | 13.4      |
| CO <sub>2</sub> _in (ppm)       | 464               | 504   | 473       | 802               | 596        | 634       |
| T_in (°C)                       | 24.0              | 26.6  | 21.8      | 23.6              | 21.5       | 21.8      |
| T_out (°C)                      | 17.7              | 17.5  | 17.4      | 17.4              | 15.5       | 11.7      |
| O <sub>3</sub> _in (ppb)        | 8.6               | 4.1   | 7.9       | 6.2               | 3.7        | 1.1       |
| O <sub>3</sub> _out (ppb)       | 25.2              | 19.4  | 14.8      | 13.4              | 18.4       | 5.8       |
| NO_in (ppb)                     | 0.8               | 3.0   | 8.2       | 13.1              | 14.1       | 17.5      |
| NO_out (ppb)                    | 3.8               | 12.1  | 16.3      | 13.9              | 3.5        | 46.4      |
| WS (m/s) <sup>c</sup>           | 2.8               | 2.0   | 1.9       | 1.9               | 2.1        | 2.1       |

**Table 4.1.** Time-averaged measurement results inside and outside school classrooms for the observational monitoring periods.

<sup>a</sup> The "PN\_in2" monitor only sampled for 59 h ( $t \sim 17-76$  h of elapsed time in the monitoring period). The average reported here is based on the monitored interval, only.

<sup>b</sup> At site S4, we were unable to monitor the outdoor PN concentrations overnight (typically from about midnight until sometime between 0600 and 0700). The reported average is based only on the available data, which is likely biased high because outdoor PN levels were commonly observed to be lower overnight than during the day.

<sup>c</sup> Measurements from central monitoring station.

**Table 4.2.** Time-average particle number concentrations (in units of 1000 particles cm<sup>-3</sup>) measured during observational monitoring at the school sites.

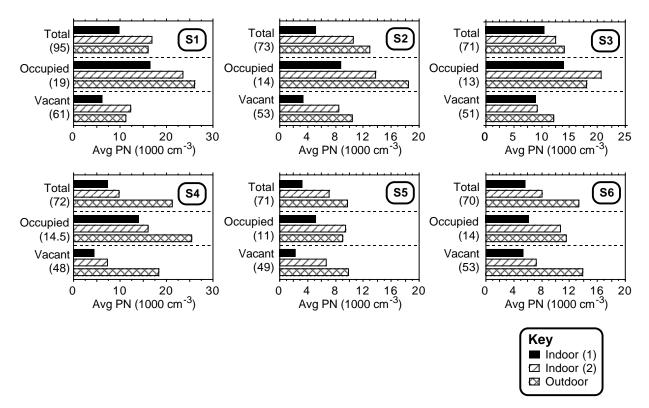
|            | Total Obs. Period |                   |                   |      | Occupied by students |      |      | Unoccupied |     |      |                   |      |
|------------|-------------------|-------------------|-------------------|------|----------------------|------|------|------------|-----|------|-------------------|------|
| Site       | In1               | In2               | Out               | Hrs  | In1                  | In2  | Out  | Hrs        | In1 | In2  | Out               | Hrs  |
| S1         | 9.8               | 16.9 <sup>a</sup> | 16.0              | 95.3 | 16.5                 | 23.6 | 26.0 | 18.9       | 6.1 | 12.2 | 11.2              | 61.0 |
| S2         | 5.1               | 10.5              | 12.9              | 73.0 | 8.8                  | 13.8 | 18.5 | 13.9       | 3.3 | 8.5  | 10.4              | 52.9 |
| <b>S</b> 3 | 10.5              | 12.6              | 14.2              | 70.7 | 14.0                 | 20.8 | 18.1 | 13.0       | 8.8 | 9.2  | 12.1              | 50.8 |
| S4         | 7.3               | 9.8               | 21.3 <sup>b</sup> | 72.4 | 14.0                 | 16.1 | 25.5 | 14.5       | 4.5 | 7.3  | 18.3 <sup>a</sup> | 48.2 |
| S5         | 3.2               | 7.1               | 9.7               | 71.1 | 5.2                  | 9.5  | 9.0  | 11.3       | 2.4 | 6.6  | 9.8               | 49.3 |
| S6         | 5.6               | 8.0               | 13.4              | 70.0 | 6.1                  | 10.7 | 11.6 | 13.9       | 5.3 | 7.3  | 13.9              | 52.5 |
| Avg.       | 6.9               | 10.8              | 14.6              | 75.4 | 10.8                 | 15.8 | 18.1 | 14.3       | 5.1 | 8.5  | 12.6              | 52.5 |

<sup>a</sup> The "PN\_in2" monitor only sampled for 59 h ( $t \sim 17-76$  h of elapsed time in the monitoring period). The average reported here is based on the monitored interval, only.

<sup>b</sup> The outdoor WCPC at S4 was not operated overnight (approximately midnight to 6 AM). The entries represent averages during the periods monitored. The results for the total observation period and for the unoccupied period are likely biased high compared to the true time averages because overnight levels tend to be lower than 24-h averages.

However, more relevant for exposure considerations are PN concentrations when students occupy classrooms. During occupancy hours, indoor levels are consistently higher than when classrooms are unoccupied. This outcome is a consequence of two independent factors: the outdoor concentrations tend to be higher during school hours than when school is not in session and the I/O ratio tends to be higher when classrooms are occupied compared to when they are unoccupied. To elaborate, during periods when classrooms are occupied, the average outdoor particle concentration across the six sites is  $18.1 \times 10^3$  particles cm<sup>-3</sup>, 24% higher than the average for all periods of observational monitoring. The average I/O ratio is 59%, which is 25% higher than the overall average. The overall average indoor concentration, then, during periods when classrooms are occupied was  $10.8 \times 10^3$  particles cm<sup>-3</sup>, which is 56% above the overall average indoors ( $1.22 \times 1.27 = 1.55$ , which accounts for the 56% increase).

This pattern is fairly consistent across all sites. Each of the six classrooms exhibits a higher I/O ratio during periods that the classroom is occupied compared to when it is vacant. Four of the six sites exhibit higher outdoor PN concentrations during periods of occupancy than when the classrooms are vacant; the exceptions to this trend occur at S5 and S6 where the outdoor concentrations are higher during periods when the classroom is vacant.



**Figure 4.1.** Time-averaged particle number concentrations measured during observational monitoring at the six school sites, S1-S6. At each site, the results are presented for each of the three monitors (primary indoor = "Indoor (1)"; supplementary indoor = "Indoor (2)"; and "Outdoor"). The upper set of three bars represent the overall averages for the full observational monitoring period. The other two sets of bars represent averages for periods sorted according to the occupancy status of the classroom. "Occupied" implies that one or more students is present in the classroom; "vacant" indicates that the classroom is unoccupied. The number of hours for each condition is indicated in parenthesis. Because teachers can be present when students are not, the sum of "occupied" and "vacant" hours is always somewhat less than the "total." Note that the horizontal axis scale varies among sites.

In the houses studied, indoor particle sources made a major contribution to average indoor particle levels and especially to exposures. In the classrooms studied, indoor emission sources contributed relatively little to concentrations and exposures. Considering the six classrooms as a whole, outdoor particles were the predominant source of indoor UFP. The key features, then, governing indoor particle concentrations and exposures involved the penetration and persistence of outdoor particles into the classroom environment. Again in contrast to the case in houses, where all study sites were ventilated by a combination of natural ventilation and infiltration, at some of the school sites classrooms were equipped with mechanical ventilation systems, which could also incorporate active particle filtration devices into the air supply. Four of the classrooms (all except S3 and S5) also had windows that could be opened to provide natural ventilation. And, air could leak into the classroom as infiltration through openings in the envelope. The penetration of ultrafine particles would have been influenced by the quality of a filtration device, if any, in the mechanical ventilation system, if present, and by the relative degree to which ventilation air was supplied mechanically versus entering via natural ventilation or infiltration. The persistence of UFP in the classrooms would be influenced by the overall airexchange rate, by the tendency for UFP to deposit onto room surfaces (including the clothing and skin and in the lungs of classroom occupants) and by filtration, if any, in mechanically recirculated airflows, if any. An important feature of the changing I/O ratio between occupied and unoccupied periods in the classroom appears to be the extent to which natural ventilation is used in the classroom. We observed in some classrooms the use of open windows to regulate classroom temperature, especially when it became too warm. In these cases the I/O ratio tended to be higher when more windows were open.

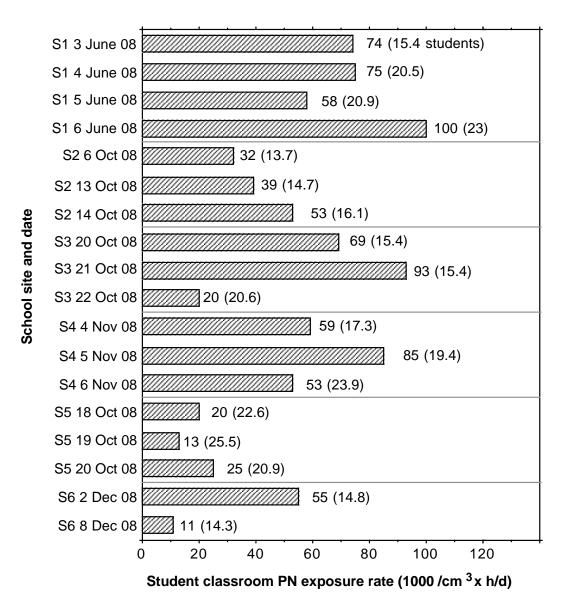
For each school day monitored in each classroom, we determined an average indoor exposure for the students who occupied the classroom that day. These results were computed by combining the time-series data on indoor PN concentration, as measured by the primary WCPC, with time-resolved occupancy data as observed and recorded by a member of our research team. This latter data set reported the number of students in the classroom as a function of time, with one-minute resolution.

The total exposure summed over all students was computed by multiplying the measured PN concentration by the number of students present, summing over all time for a school day. We computed an average student occupancy level by first excluding minutes from the time series when no students were present and then computing the average number of students present during the remaining times. In other words, this occupancy indicator represents the average number of students in the classroom whenever there is at least one student present. The average exposure rate per student was approximated as the ratio of the cumulative exposure to the average student occupancy.

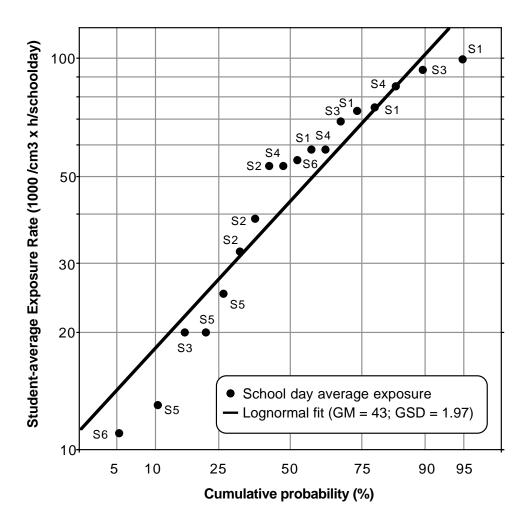
The daily average classroom exposure to PN concentrations was determined for each of the eighteen days of observational monitoring in schools. The results are presented in Figures 4.2 and 4.3. In Figure 4.2, the results are presented as a bar chart, with one bar per school day. Figure 4.3 presents the same results, but in the form of a cumulative probability distribution plotted on log-probability coordinates.

Considering each monitored day individually, the average per-student classroom exposure rate varies from  $11 \times 10^3$  cm<sup>-3</sup> h/d to  $100 \times 10^3$  cm<sup>-3</sup> h/d, with an overall average of 52  $\times 10^3$  cm<sup>-3</sup> h/d. Variation within sites and across sites contribute to the overall range observed. Averaging the exposures for the monitored days at each site, the range of averages is  $19 \times 10^3$  cm<sup>-3</sup> h/d at S5 to  $77 \times 10^3$  cm<sup>-3</sup> h/d at S1. A key reason for the differences among exposures at

the different sites is the difference in outdoor PN concentration during observational monitoring. The nature of this study does not allow us to discern why the outdoor PN concentrations might be higher or lower at one site versus another. Since monitoring was done on different dates for each site and because four different school sites were studied, we cannot discriminate between space (e.g., proximity to sources) and time (e.g., meteorology affecting transport and dispersion) as the underlying causes of different outdoor PN concentrations.



**Figure 4.2.** Daily average classroom exposure to particle number concentration for each of the eighteen observational monitoring days in the six classrooms. The left-hand axis indicates the classroom school site (S1-S6) and the monitored date. The numbers adjacent to the bars indicate the average exposure per student, in units of  $1000 \text{ cm}^{-3} \times \text{h/d}$  (and the average number of students in the classroom when occupied by students).



**Figure 4.3.** Cumulative probability distribution of daily average classroom PN exposure rates for the 18 days of monitoring at the six classrooms studied in this project. These eighteen data points have a GM of 43 (1000 cm<sup>-3</sup> h/d) and a GSD of 1.97. The lognormal distribution with the same GM and GSD is plotted as the straight line. The arithmetic mean of this set of 18 exposure determinations is 52 (1000 cm<sup>-3</sup> h/d) and the standard deviation is 28 (1000 cm<sup>-3</sup> h/d).

## 4.2 Research Findings from Individual Classrooms

#### 4.2.1 Site S1

Observational monitoring was conducted at classroom site S1 for a continuous 95-h period commencing at 15:40 on 2 June and ending at 15:00 on 6 June 2008. For detailed background information about this school site, the reader is referred to §2.2.2. See also Figure C.8 (Appendix C) for a floor plan. Summarizing briefly, the school site is on a residential street with low traffic overall at an elevation of 100-150 m above sea level. The nearest freeway is ~ 500 m to the east. The classroom is located in an older, single-story building (perhaps 50 y old) on the school site. The classroom we monitored is situated such that two of its walls are shared with neighboring classrooms, a third (S) wall adjoins the corridor that provides access to the classroom, and beyond the fourth (N) wall is an outdoor courtyard. Ventilation is provided entirely by natural ventilation (through open windows and doors) and infiltration (air leakage). During the June monitoring period, the teacher was observed to regulate the classroom temperature by opening the door to the courtyard. At this time of year, heating was not needed.

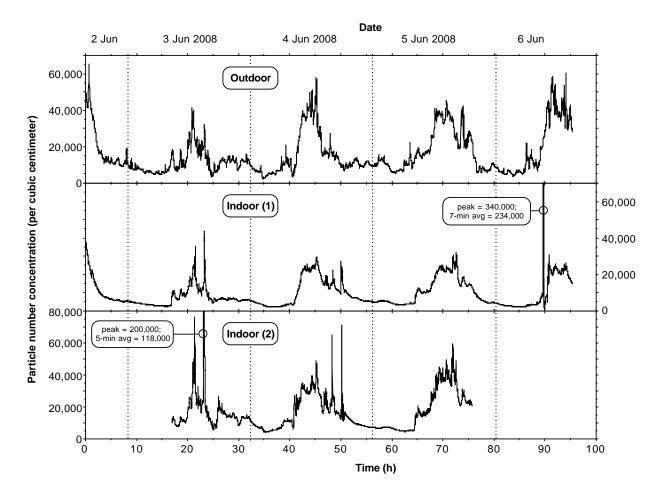
The primary indoor monitoring package was deployed inside the classroom, in the NE corner of the room. The outdoor package was set up to monitor in the courtyard immediately adjacent to the classroom. The supplementary indoor monitor was placed in the corridor across from the door to the classroom.

A summary of the time-averaged species concentration and cofactor values was presented in Table 4.1 (§4.1). The average particle number concentrations measured during the entire observational monitoring period were 16.0 (outdoors), 9.8 (indoors classroom), and 16.9 (indoors corridor) in units of 1000 particles per cm<sup>3</sup>. The average outdoor PN levels measured at this site were on the high end of the range observed in this study. Excluding S4, the average outdoor PN level at the other five sites was in the range 9.7-16  $\times 10^3$  particles cm<sup>-3</sup>, with an average of 13.2  $\times$  $10^3$ . (At S4, we were unable to monitor outdoor PN levels overnight, which biases upward the average monitored result.) The indoor levels were also on the high side of the central tendency. Focusing on the main indoor monitor, the average for all six sites was  $6.9 \times 10^3$  particles cm<sup>-3</sup>, about 30% lower than the average at this classroom. We observed at all six classroom sites that the supplementary indoor monitor produced higher time-averaged PN concentrations than were obtained from the classroom monitor. Here, the supplementary indoor monitor, sampling from the corridor, produced a time-averaged PN concentration that was ~ 70% higher than the classroom concentration, and within 10% of the outdoor level. Note: the supplementary monitor was only available for a portion of the observational monitoring period, from  $t \sim 17-76$  h. Restricting the comparison to this period only does not change the result markedly: the average outdoor PN level was  $16.2 \times 10^3$  cm<sup>-3</sup> and the average PN level recorded by the classroom monitor was  $10.4 \times 10^3$  cm<sup>-3</sup>. We don't know the reason for the higher levels in the corridor. The two monitors compared well in side-by-side monitoring on site (Appendix A), so we believe that the difference in measurement results reflects differences in concentrations in the two microenvironments.

Figure 4.4 shows the time-series of particle number concentrations measured at this site. The horizontal axis is labeled with elapsed time of monitoring referenced to the start time of 15:40 on 2 June 2008. The vertical dashed lines demark midnight and distance from these lines can be used to gauge clock time.

The outdoor concentrations at this site display a diurnal pattern that is similar to that observed at several other sites, with markedly higher PN concentrations during midday periods than overnight. To quantify the diurnal pattern, we computed hourly average concentrations for

each clock hour of the monitoring period and then averaged the results for all like hours over the four-day period. The resulting maximum was  $37.0 \times 10^3$  cm<sup>-3</sup> at 12:00-13:00, more than  $6\times$  the diurnal minimum,  $5.8 \times 10^3$  cm<sup>-3</sup>, at 3:00-4:00. The dominant temporal trend in the classroom PN level tracks the outdoor behavior, with higher concentrations during the daytime hours and lower levels overnight. There is one prominent spike in the classroom PN level, which occurs at elapsed time  $t \sim 90$  h. These traces suggest, and analysis supports, that the main source of classroom PN levels most of the time is the penetration and persistence of particles that originate in outdoor air. There are contributions from indoor emission sources. However, compared with what we observed in houses, the relative influence of indoor sources compared to outdoor particles is less pronounced. That is true not only at this site but as a consistent pattern across the classrooms monitored.

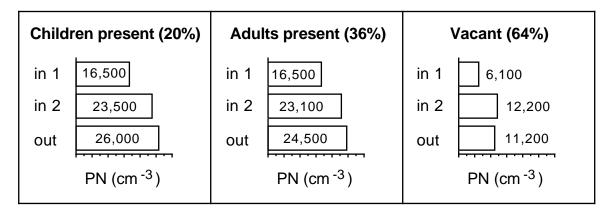


**Figure 4.4.** Time-series of particle number concentrations during observational monitoring at classroom site S1. The "Indoor (2)" monitor only sampled between 3 June at 8:37 and 5 June at 19:19 (corresponding to elapsed times of 17.0-75.7 h). On one occasion, corresponding to approximate monitoring time 23 h, the "Indoor (2)" monitor recorded levels above 80,000 cm<sup>-3</sup>. An analogous excursion was recorded on the "Indoor (1)" monitor at  $t \sim 90$  h. The maximum values and the duration of these excursions are noted in boxes.

At each of the classroom sites, we had a researcher present throughout the school day to observe and record information about factors that could influence indoor PN levels and also that are related to exposures. One category of information was occupancy — the numbers of adults and children in the classroom at any given time — recorded with a time-resolution of one minute. The occupancy time series for site S1 is presented in Appendix F (Figure F.3).

Figure 4.5 displays the time-averaged particle number concentrations measured during observational monitoring as sorted by occupancy status of the classroom. Focusing on the primary indoor monitor, "in 1," we see that the average concentrations when the classroom is occupied,  $16.5 \times 10^3$  cm<sup>-3</sup>, are considerably higher than the indoor average concentrations when the classroom is vacant,  $6.1 \times 10^3$  cm<sup>-3</sup>. There are three reasons for the difference. First, and most important, the outdoor PN levels are much higher when the classroom is occupied (25-26 ×  $10^3$  cm<sup>-3</sup>) than when it is vacant ( $11 \times 10^3$  cm<sup>-3</sup>). This difference follows from the fact that the hours of the school day overlap with the higher outdoor PN levels in the diurnal cycle. A second influencing factor is that the average ventilation rate of the classroom is higher during periods of occupancy than during vacant times. A higher ventilation rate would tend to produce a higher indoor:outdoor ratio, when outdoor sources dominate. The third factor is that — as in houses — episodic indoor emissions tend to coincide with occupancy. This last factor, which had a prominent influence in houses, appears to be not so important in the classroom sites monitored in this study.

Note that the time-averaged concentration recorded by the supplementary monitor matches fairly closely to the outdoor air concentration, agreeing to within 10% for each of the three conditions. Conditions in the corridor appear to be well coupled to those outdoors. That finding is reinforced by the time-series of the ratio of the corridor to outdoor concentration (see Appendix F, Figure F.2), which fluctuates in the vicinity of 1.0 for most of the time. This behavior would be expected if the air-exchange rate of the corridor were particularly high.



**Figure 4.5.** Time-averaged indoor and outdoor particle number concentrations measured during the observational monitoring period at classroom site S1 and sorted according to the classroom occupancy status. The period with children present is a subset of the period when adults are present. (Note that the average for the supplementary indoor monitor ("in 2") is based on sampling for only a portion of the period, from t ~ 17.0-75.7 h.) Indicated in the upper portion of each frame is the percentage of the observational monitoring period for which the indicated state applies.

Table 4.3 summarizes the average copollutant levels at S1, sorted according to the occupancy status of the classroom. In Appendix F are presented time-series plots for the copollutants as follows: Figure F.4 for ozone; Figure F.5 for NO; Figure F.6 for carbon dioxide; and Figure F.7 for carbon monoxide. Outdoor and indoor NO levels are consistently low at this site. Ozone levels average approximately 30 ppb outdoors when school is occupied and the indoor concentration is roughly 40% of the value outdoors.

| Species (units)           | Students | Adults | Unoccupied |  |
|---------------------------|----------|--------|------------|--|
| NO_in (ppb)               | 0.8      | 0.6    | 0.9        |  |
| NO_out (ppb)              | 4.3      | 3.8    | 3.8        |  |
| O <sub>3</sub> _in (ppb)  | 10.4     | 12.4   | 6.4        |  |
| O <sub>3</sub> _out (ppb) | 27.7     | 29.8   | 22.6       |  |
| CO <sub>2</sub> _in (ppm) | 650      | 568    | 405        |  |

Table 4.3. Copollutant levels at site S1 during observational monitoring.<sup>a</sup>

<sup>a</sup> Measurements sorted according to occupancy status of the building and then averaged.

We made five determinations of the effective air-exchange rate (AER) based on an analysis of tracer gas decay using carbon dioxide. The results are summarized in Table 4.4. Four of these determinations were made during observational monitoring with doors to the classroom open, as they typically were when the classroom was occupied. The fifth determination was made during a manipulation experiment with the doors closed. Windows were closed for all determinations except that one window was cracked open to allow for the passage of electrical cords to power the outdoor monitoring equipment. (That condition was maintained throughout the field-monitoring period.)

Before discussing the results, two aspects of the AER determination are discussed. First, this classroom — like some of the others in our monitoring campaign, and as is common for many classrooms — is potentially coupled by means of airflow to other indoor spaces. The coupling is unlikely to be strong enough to produce a larger well-mixed space of which the classroom is a part. On the other hand, it could well be too large to be ignored. In the case of the S1 classroom, the room air is almost certainly coupled to some extent to the air in the corridor, especially when the classroom is occupied and the door to the corridor is open, as it often is. The classroom might also experience some airflow coupling with adjacent rooms. What does an airexchange rate mean in this case? In essence, by using carbon dioxide generated by occupants as a tracer, as we do in the observational monitoring period, we are determining an effective airexchange rate that takes account of the degree to which the air flowing into the room is contaminated with CO<sub>2</sub> from sources in other interior spaces. Consider, for example, that outdoor air has a  $CO_2$  level of 380 ppm. Let us refer to the net  $CO_2$  level as the increment of  $CO_2$  above this outdoor baseline. So, if the air in the classroom contains 780 ppm of  $CO_2$  the net  $CO_2$  level would be 400 ppm. Now, let's assume that the net  $CO_2$  level in the corridor is exactly half of the net CO<sub>2</sub> level in the classroom and that all of the air entering the classroom comes from the corridor. In this highly idealized case, the air-exchange rate determined by tracer gas decay would be exactly half of the true airflow rate from the corridor.

The second consideration is that completely unoccupied periods suitable for inferring airexchange rates from  $CO_2$  decay were rare — in fact, there was only one during observational monitoring at S1. A few other periods exhibited well-behaved  $CO_2$  decay curves that coincided with light levels of occupancy. We analyzed these to determine air-exchange rates by fitting a theoretical solution to the  $CO_2$  level versus time, taking into account estimated metabolic emissions of  $CO_2$  from occupants. Uncertainty introduced because we are estimating the rate of emissions of  $CO_2$  from occupants is generally small because we pick times for analysis in which there is a large change in occupancy, i.e. from a fully occupied classroom (~ 20 persons) to a lightly occupied room (only a few occupants). A clear decrease in  $CO_2$  occurs over time in these cases, which provides the basis for analysis. The sensitivity of the determined air-exchange rates to the assumed  $CO_2$  emission rates from the few occupants remaining is small.

For the four observational monitoring periods, we found the effective air-exchange rate to be in the range 1.3-2.6 h<sup>-1</sup>, with a mean  $\pm$  standard deviation of  $2.1 \pm 0.6$  h<sup>-1</sup>. With the room closed during the manipulation experiment, we found the air-exchange rate to be 0.5 h<sup>-1</sup>. In this case, because school was not in session, we were able to measure the air-exchange rate of the classroom itself, effectively in isolation from potentially coupled spaces. However, although the experimental conditions are much better defined, the manipulation experiment is less relevant than the inferences from observational monitoring for describing conditions in the classroom as occupied.

**Table 4.4.** Air-exchange rates (AER) determined by carbon dioxide concentration decay in classroom site S1.

| Time (h)   | <b>AER</b> $(h^{-1})$ | <b>WS</b> (m $s^{-1}$ ) | $ \Delta \mathbf{T} $ (°C) | <b>Classroom configuration &amp; occupancy</b> <sup>a</sup> |
|------------|-----------------------|-------------------------|----------------------------|---|
| 44.9-45.9  | 2.3                   | 5.4                     | 4.0                        | Three adults plus researcher present. For                   |
|            |                       |                         |                            | first 9 min, 4 children also present.                       |
| 67.3-68.3  | 2.4                   | 3.6                     | 2.0                        |   |
| 71.4-72.4  | 2.6                   | 3.6                     | 0.4                        | One teacher plus researcher present. For                    |
|            |                       |                         |                            | first 24 min, 2 children also present.                      |
| 92.7-93.7  | 1.3                   | 3.6                     | 3.1                        | One teacher plus researcher present.                        |
| A1 (6 Jun) | 0.5                   | 3.6                     |                            | Doors closed.   |

<sup>a</sup> Unless otherwise noted, the configuration of the classroom during these measurements was this: both doors (an exterior door and an interior door to a hallway) open; windows closed except for one that was cracked open to allow passage of a power cord; room vacant. All air-exchange rate determinations are based on the assumption that the outdoor  $CO_2$  level is 380 ppm. When occupants are present, we account for their metabolic emissions of  $CO_2$  in the analysis, using a  $CO_2$  emission rate of 0.013 mol/min (= 830 g  $CO_2/d$ ) for teachers and 0.009 mol/min (= 580 g  $CO_2/d$ ) for students (see also Bartlett et al., 2004). The estimate for teachers is derived from ASHRAE Standard 62-1989, assuming a metabolic level of 1.2 met units, corresponding to "office (seated)" and a respiratory quotient of 0.83, corresponding to a "normal mix" diet. For students, we have assumed that their rate of metabolic product of  $CO_2$  is 70% of the teacher's rate.

We next explore the sources of UFP in this classroom. Evidence presented thus far suggests that outdoor air is the dominant source at almost all times during the observational monitoring period. Furthermore, the air-exchange rate of the class is sensitive to the position of doors. Air-exchange rate is expected to influence the indoor proportion of outdoor particles. Occupancy is also a potentially significant factor, certainly with regard to quantifying exposure and possibly because occupancy alters the rate of deposition of particles onto indoor surfaces.

To investigate, we removed from the data set the two periods when there was evident influence of indoor sources on indoor PN levels. We then sorted the remaining minute-by-

minute PN concentration into groups based on occupancy and door position and computed average PN concentrations determined by the three monitors for each of the groups. Finally, we computed the ratio of classroom average to outdoor average concentrations  $(=f_1)$  and the ratio of classroom average to corridor average concentrations  $(=f_{12})$ . The results show some effect, although small, of door position on the concentration ratios (see Table 4.5). Focusing on parameter  $f_1$ , we see that when students are in the classroom, the ratio of average indoor to outdoor concentrations is 65 when both doors are open (10 h) as compared with 0.55 when only the interior door is open (7 h). When the room is vacant and both doors are closed, the ratio is 0.52 (54 h).

| State                          | <b>Duration</b> (h) | <b>WS</b> (m $s^{-1}$ ) | $ \Delta \mathbf{T} $ (°C) | $f_{I}(-)$ | $f_{12}()$ |  |  |
|--------------------------------|---------------------|-------------------------|----------------------------|------------|------------|--|--|
| At least one student present   |                     |                         |                            |            |            |  |  |
| Exterior door (only) open      | 0.8                 | 3.1                     | 3.1                        | 0.63       |            |  |  |
| Interior door (only) open      | 7.0                 | 3.5                     | 5.1                        | 0.55       | 0.68       |  |  |
| Both doors open                | 9.7                 | 3.4                     | 3.4                        | 0.65       | 0.59       |  |  |
| Both doors closed <sup>b</sup> | 0.5                 | 2.7                     | 5.2                        | 0.39       |            |  |  |
| Average <sup>b</sup>           | 18.0                | 3.4                     | 4.1                        | 0.58       | 0.63       |  |  |
| At least one teacher present   |                     |                         |                            |            |            |  |  |
| Exterior door (only) open      | 0.8                 | 3.1                     | 3.1                        | 0.48       |            |  |  |
| Interior door (only) open      | 13.9                | 3.5                     | 4.4                        | 0.63       | 0.69       |  |  |
| Both doors open                | 16.2                | 3.5                     | 3.3                        | 0.69       | 0.61       |  |  |
| Both doors closed <sup>b</sup> | 2.1                 | 3.8                     | 4.2                        | 0.55       | 0.78       |  |  |
| Average <sup>b</sup>           | 33.0                | 3.5                     | 3.8                        | 0.64       | 0.65       |  |  |
| Unoccupied                     |                     |                         |                            |            |            |  |  |
| Exterior door (only) open      | 3.0                 | 2.8                     | 5.2                        | 0.61       | 0.63       |  |  |
| Both doors open                | 3.7                 | 3.1                     | 2.2                        | 0.62       | 0.60       |  |  |
| Both doors closed              | 54.1                | 2.5                     | 9.3                        | 0.52       | 0.55       |  |  |
| Average                        | 61.0                | 2.7                     | 8.6                        | 0.54       | 0.57       |  |  |

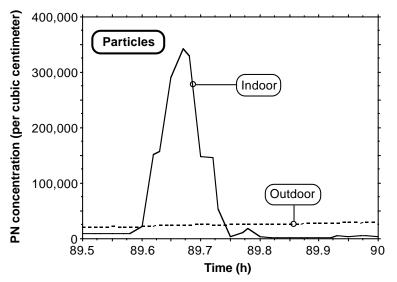
**Table 4.5.** Analysis of the indoor to outdoor and indoor to hallway particle concentration ratios at classroom site S1 for various door configurations.<sup>a</sup>

 $^{a}$  All windows were closed at all times except for one window which was cracked open to allow the passage of a power cord.

<sup>b</sup> Analysis excludes 6-min period when indoor levels in the classroom were strongly elevated.

At this site, there was only one clear source of particle emissions that occurred within the classroom. An electric griddle was used to make pancakes as part of a classroom activity one morning. Figure 4.6 displays the particle traces indoors and outdoors in association with this event. Two features merit explicit mention: the very high peak concentration (> 300,000 particles cm<sup>-3</sup>) and the very short duration of the event (< 10 min). The rapid decay of particles after the peak corresponds to a first order loss rate of ~ 30 h<sup>-1</sup>, which greatly exceeds the rate that can be explained by deposition and ventilation. One possible explanation is incomplete mixing; however, the smoothness of the rise and fall suggests that the room air was probably reasonably well mixed. Another possible alternative explanation for the rapid decay is a removal mechanism such as evaporation that can act on a much faster time scale than deposition or ventilation. Our data are inadequate to explore the cause of the rapid decay further. However, we can estimate the contribution to exposure of occupants caused by this peak. It is represented

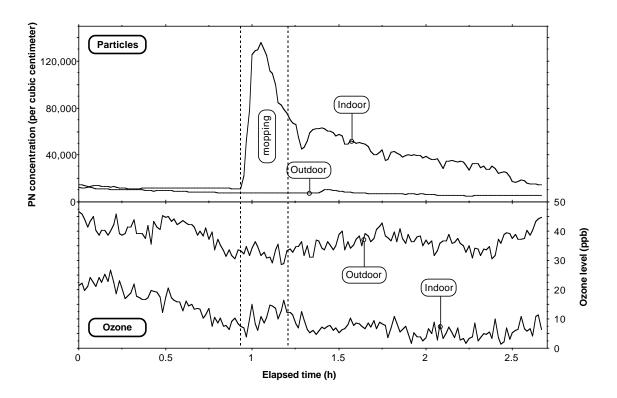
by the area under the "indoor" curve in Figure 4.6, minus the expected contribution from outdoor particles, which is estimated as the area under a trace given by  $f_1 \times$  outdoor PN concentration. The result of that calculation is  $27 \times 10^3$  particles/cm<sup>3</sup> h. This is the estimated increment in exposure that would have been experienced by each occupant in the classroom during this event associated with emissions from the use of the griddle.



**Figure 4.6.** Particle number concentration versus time inside the classroom ("indoor") and outside at site S1. The time scale is referenced to the beginning of monitoring (15:40 on 2 June 2008). The peak is associated with an event that involved cooking pancakes in the classroom on an electric griddle.

There was one cleaning activity that might also have contributed to indoor PN levels. This event occurred on the afternoon of 5 June beginning at elapsed time 71.4 h and involved the use of a pine-scented cleaning product someplace in the school building, but outside of the classroom. Approximately 20 min after the smell of terpenes reached the classroom, the classroom particle trace exhibited a small peak, increasing from ~ 21,000 particles cm<sup>-3</sup> to ~ 29,000 particles cm<sup>-3</sup>. Concurrently, outdoor levels were experiencing a downward trend. Hallway levels increased from ~33,000 to ~59,000 particles cm<sup>-3</sup>. Since this peak was not attributable to an in-classroom source, because it was relatively small, and because school had already been dismissed for the day, the peak was not quantitatively analyzed.

The potential for the use of terpene-containing cleaning products to contribute to indoor PN levels was investigated further in a manipulation experiment at this site. The classroom floor was mopped with a pine-oil based cleaner. Figure 4.7 shows that particle formation occurred as a result of this activity, with peak concentrations reaching a level of ~ 120,000 cm<sup>-3</sup>. The PN concentration decay curve is reasonably approximated as a first-order loss process with a rate constant of  $k+a \sim 0.7$  h<sup>-1</sup>. Applying the integral mass balance approach to the indoor concentration peak, we estimate that ~  $20 \times 10^{12}$  particles were emitted in the classroom from this activity. The relatively slow decay (compare with the k+a values of ~ 1.5-2 h<sup>-1</sup> observed in houses) suggests that particle formation was continuing throughout the duration of the experiment and potentially beyond, which suggests that the estimated cumulative emissions should be regarded as an approximate lower bound.



**Figure 4.7.** Time traces of particle concentrations and ozone levels during the source-investigation manipulation experiment at site S1. Mopping entailed use of a pine-oil based cleaner during the period 0.93-1.20 h.

Our field research staff maintained a log of activities that might possibly be associated with UFP emissions during observational monitoring. Table 4.6 summarizes the entries from that log. Among the activities recorded, only one was unambiguously an emission source of particles, the cooking of pancakes on an electric griddle. We reiterate an important point: an entry of "N" in the "PN source?" column of Table 4.6 does not indicate that *no* particles were emitted from the activity. Rather, it indicates that the activity did not produce a clear episodic peak in the indoor PN concentration, measured against the background concentration of ~  $10^4$  particles cm<sup>-3</sup>. For a source to have been clearly detected, it would have had to be responsible for an increase in PN concentration of at least ~ 3,000 cm<sup>-3</sup>. Given the room volume of ~ 290 m<sup>3</sup>, the minimum emissions scale that could be clearly detected would have been ~  $10^{12}$  particles.

An exposure analysis based on observational monitoring at site S1 is presented in Table 4.7. In comparison with the house sites, where we tracked individuals, here we are only able to treat students as a group and assess average exposures of the individuals in that group. For the four school days of monitoring, the cumulative time that at least one student was present in the classroom was 18.9 h or 4.7 h/d. The average number of students present in the classroom during the time when any student was present was 19.5. For each minute when one or more student was present in the classroom, an increment of exposure is computed as the student occupancy (number of students) multiplied by classroom PN level ( $10^3 \text{ cm}^{-3}$ ) and multiplied by the duration of the observation interval (one minute). These increments, summed over all periods of student occupancy during observational monitoring, totaled 6214 ×  $10^3 \text{ cm}^{-3}$  h.

Dividing this cumulative exposure by the average occupancy yields an average individual exposure of  $319 \times 10^3$  cm<sup>-3</sup> h. One should note, however, that this average represents an exposure estimate for a hypothetical individual. In reality, more than 19.5 individual students acquired the aggregate student exposure of  $6214 \times 10^3$  cm<sup>-3</sup> h. We know this because the maximum student occupancy was 26 recorded during observational monitoring. Because we were not able to track individual students, we cannot compute the true exposure acquired by individuals. Nevertheless, we believe that the cumulative exposure divided by average occupancy provides the best robust indicator of exposure conditions for students in the classroom. The average student exposure acquired while in the classroom per school day is obtained by dividing the average individual exposure for students  $(319 \times 10^3 \text{ cm}^{-3} \text{ h})$  by the number of school days monitored (4), yielding the result  $80 \times 10^3$  cm<sup>-3</sup> h/d. About 598/6214 = 10% of the students' exposure is attributable to the one quantified indoor source event, cooking pancakes, with the remainder caused by particles originating from outdoors. Analogous calculations have been performed for the teacher. The teacher's daily exposure rate in the classroom is higher than that of the students (141 versus  $80 \times 10^3$  cm<sup>-3</sup> h/d) primarily because of the teacher's longer duration of exposure: 8.6 versus 4.7 h/d. This evaluation of exposure does not consider time when students and teachers were on the school grounds but not in the classroom.

| Time (h)  | Activity or observation                                    | PN source? |
|-----------|--|------------|
| 19.3      | Overhead projector in use                                  | Ν          |
| 41.0      | Overhead projector in use                                  | Ν          |
| 42.0      | Occasional writing on white board                          | Ν          |
| 42.3      | Overhead projector in use                                  | Ν          |
| 42.8      | Printer used for 1 min, moderate use of white board        | N          |
| 43.1      | Overhead projector in use                                  | N          |
| 43.3      | Moderate use of white board                                | Ν          |
| 49.1      | Custodian sweeping   | N          |
| 64.9      | Intermittent use of white board                            | N          |
| 71.4      | Odor of pine-scented floor cleaner reaches classroom (from | ?          |
|           | hallway). Custodian vacuums.                               |            |
| 88.9-91.7 | Pancakes prepared in the morning (exact time unknown)      | Y          |

Table 4.6. Activity notes related to potential PN emission sources at S1.

| Table 4.7. Exposure analysis for students and teacher at classroom site S1, based on four school |
|--|
| days of observational monitoring.  |

| Parameter  | Students | Teacher |
|--|----------|---------|
| Hours with at least one member present                                     | 18.9     | 34.3    |
| Average occupancy when at least one present                                | 19.5     | 1       |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                     | 6214     | 563     |
| Total exposure from cooking peak $(10^3 \text{ cm}^{-3} \text{ h})$        | 598      | 27      |
| Average individual exposure $(10^3 \text{ cm}^{-3} \text{ h})$             | 319      | 563     |
| Average individual exposure rate $(10^3 \text{ cm}^{-3} \text{ h d}^{-1})$ | 80       | 141     |

#### 4.2.2 Site S2

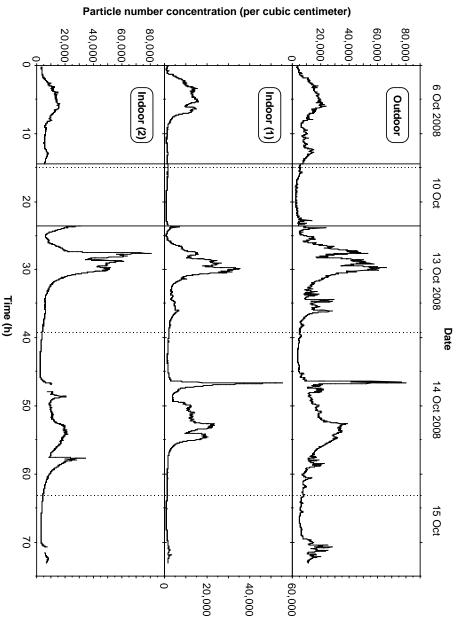
Observational monitoring was conducted at classroom site S2 for 73 hours during three periods: (i) 9:00-23:30 on 6 October 2008 (14.5 h); (ii) from 23:30 on 9 October through 8:30 on 10 October 2008 (9 h); and (iii) from 8:20 on 13 October until 9:50 on 15 October 2008 (49.5 h). For detailed background information about this school site, see §2.2.2. See also Figure C.9 (Appendix C) for a floor plan. The school site is the same one as for site S1. Classroom S2 is located on the second floor of a new two-story building. The classroom is situated such that three of its walls separate it from outdoors and it shares one wall with an adjacent classroom. The classroom has a mechanical air handling system that contains a particle filter. Often, the system fan is on, even when the thermostat does not call for heat or cooling. However, the teacher also reported that she turns off the fan occasionally when she is disturbed by the sound.

The primary indoor monitoring package was deployed inside the classroom, in the NW corner of the room. The outdoor package was set up to monitor in the same courtyard as used during monitoring in S1; this location is about 30-40 m away from the S2 classroom. The supplementary indoor monitor was placed in the cafeteria, which is located in the same older building on the school site as classroom S1. The cafeteria is about 15-20 m from the S2 entrance.

A summary of the time-averaged species concentration and cofactor values was presented in Table 4.1 (§4.1). The average particle number concentrations measured during the entire observational monitoring period were 12.9 (outdoors), 5.1 (indoors classroom), and 10.5 (indoors cafeteria) in units of 1000 particles per cm<sup>3</sup>. The average outdoor PN levels measured at this site were similar to the average observed at all school sites. The indoor levels were on the low side of the central tendency. Focusing on the main indoor monitor, the average for all six sites was  $6.9 \times 10^3$  particles cm<sup>-3</sup> about 35% higher than the average at this classroom. The supplementary indoor monitor, sampling from the cafeteria, produced a time-averaged PN concentration that was about twice as high as the classroom concentration. The supplementary monitor was unavailable for a portion of the observational monitoring period: the duration of sampling from the cafeteria was 62 h compared with 73 h from the other monitors. Restricting the comparison to the periods when the supplementary monitor was operating shifts the perspective only slightly: during these 62 h, the average outdoor PN level was  $14.1 \times 10^3$  cm<sup>-3</sup> and the average PN level recorded by the classroom monitor was  $5.6 \times 10^3$  cm<sup>-3</sup>.

Figure 4.8 shows the time-series of particle number concentrations measured at this site. The outdoor concentrations at this site display a diurnal pattern that is similar to that observed at several other sites, with markedly higher PN concentrations during midday periods than overnight. In addition, there is one prominent peak in the outdoor trace, which occurs on the morning of 14 October. The peak has a duration of ~ 25 min, beginning just after 7:00. (See Figure 4.9.) It is reflected not only in the outdoor monitor but also in the classroom PN signal. The supplementary monitor was not operating during a part of this episode. The cause of this peak is unknown.

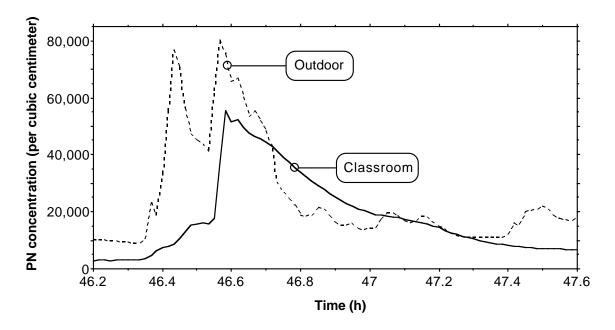
To quantify the diurnal pattern, we computed hourly average concentrations for each clock hour of the monitoring period and then averaged the results for all like hours over the fourday period. The resulting maximum was  $34.4 \times 10^3$  cm<sup>-3</sup> at 14:00-15:00, almost 9× the diurnal minimum,  $3.9 \times 10^3$  cm<sup>-3</sup>, at 3:00-4:00. The dominant temporal trend in the classroom PN level tracks the outdoor behavior, with higher concentrations during the daytime hours and lower levels overnight. There are no prominent spikes in the indoor signal other than the one that corresponds to the outdoor trace. As at S1, these traces suggest, and analysis supports, that the



main source of classroom PN levels is the penetration and persistence of particles that originate in outdoor air.

observational monitoring period. The time scale records the elapsed time of observational The vertical dashed lines denote midnight; the vertical solid lines mark breaks in the outdoor monitor sampled from the same courtyard as for S1, ~ 30-40 m from the S2 classroom. sampled from the school cafeteria about 20 m from the classroom, in a separate building. classroom site S2. Monitor "Indoor (1)" was located in the classroom; monitor "Indoor (2)" Figure 4.8. Time-series of particle number concentrations during observational monitoring at monitoring. The

classroom is vacant is ~ 1.8. Second, the ratio of indoor to outdoor concentrations tends to be primary indoor monitor, "in 1," we see that the average concentration when the classroom is occupied by students,  $8.8 \times 10^3$  cm<sup>-3</sup>, is more than twice as high as when the classroom is vacant, observational monitoring as sorted by occupancy status of the classroom. Focusing on the Figure 4.10 displays the time-averaged particle number concentrations measured during are higher. The ratio of outdoor average concentration with children present to when the higher during periods of occupancy because occupancy occurs during daytime hours when levels occupancy can be decomposed into two component factors. First, the outdoor concentrations are  $3.3 \times 10^3$  cm<sup>-3</sup>. As we observe at many school sites, the higher indoor concentrations during The occupancy time series for site S2 is presented in Appendix F (see Figure F.15). higher when the classroom is occupied. The in1:out ratio when children are present is 0.48, which is  $1.5 \times$  the value when the classroom is vacant. The higher indoor-outdoor ratio is probably a result of higher ventilation rates during occupancy, especially through open doors and windows.



**Figure 4.9.** Particle number concentrations measured outdoors and inside the classroom site S2 on the morning of 14 October 2008. The time 46.2 h corresponds to 7:00. The cause of the outdoor excursion is unknown, however the indoor concentration is most likely responding to the change in outdoor conditions.

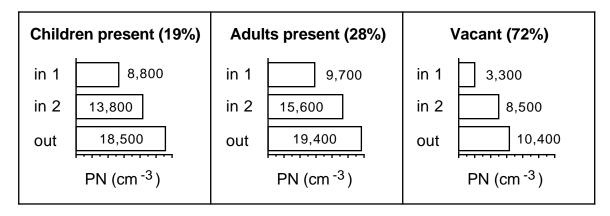
When indoor particles are only a result of the penetration and persistence of outdoor particles, we may think about the indoor-outdoor ratio  $(f_l)$  according to the simple expression:

$$f_1 \sim \frac{Pa}{a+k} \tag{4.1}$$

All else being equal, increasing the air-exchange rate, a, increases  $f_I$ . Furthermore, shifting more of the ventilation to occur through open doors and windows instead of through infiltration paths or through mechanical ventilation filters will increase the average penetration factor, P. We have also noted, though, that increasing occupancy might be expected to increase the indoor particle deposition rate, k. The empirical evidence from this site suggests that this effect, if it occurs, is less significant than the changes in air-exchange rate or penetration factor.

Table 4.8 summarizes the average copollutant levels at S2, sorted according to the occupancy status of the classroom. In Appendix F are presented time-series plots for the copollutants as follows: Figure F.16 for ozone; Figure F.17 for NO; Figure F.18 for carbon dioxide; and Figure F.19 for carbon monoxide (outdoor only). Ozone levels average about 20 ppb outdoors and the indoor level is about 25% of that outdoors. Nitric oxide are low, with only

one period of several h duration on 14 October where the concentrations are clearly above zero, rising to  $\sim$  10-20 ppb.



**Figure 4.10.** Time-averaged indoor and outdoor particle number concentrations measured during the observational monitoring period at classroom site S2 and sorted according to the classroom occupancy status. The period with children present is a subset of the period when adults are present. (Note that the average for the supplementary indoor monitor ("in 2") is based on sampling for a subset of the total monitoring period, about 62 h of 73 h total.) Noted in the upper portion of each frame is the percentage of the observational monitoring period for which the indicated state applies.

| Species (units)           | Students | Adults | Unoccupied       |
|---------------------------|----------|--------|------------------|
| NO_in (ppb)               | 5.4      | 4.1    | 0.0 <sup>b</sup> |
| NO_out (ppb)              | 11.8     | 10.4   | 3.7              |
| O <sub>3</sub> _in (ppb)  | 4.6      | 5.7    | 3.5              |
| O <sub>3</sub> _out (ppb) | 18.1     | 19.2   | 19.5             |
| CO <sub>2</sub> _in (ppm) | 750      | 682    | 437              |

**Table 4.8.** Copollutant levels at site S2 during observational monitoring.<sup>a</sup>

<sup>a</sup> Measurements sorted according to occupancy status of the building and then averaged. <sup>b</sup> Negative baseline; mean is less than zero

We made five determinations of the effective air-exchange rate (AER) based on an analysis of tracer gas decay using carbon dioxide. The results are summarized in Table 4.9. Two of these determinations were made during observational monitoring. The other three measurements (A1-A3) were made as part of the manipulation experiments. For two cases, the classroom was closed and the measured air-exchange rates were low, 0.3-0.4 h<sup>-1</sup>. For the other three assessments, the front door was open and, in two cases, a window was open. In these cases, much higher air-exchange rates were recorded, 2.8-3.9 h<sup>-1</sup>.

Outdoor air appears to be the dominant source of indoor particles at all times during observational monitoring. Furthermore, the air-exchange rate of the class is sensitive to the position of doors and windows. According to equation (4.1), then, we might expect the indoor proportion of outdoor particles to be influenced by door and window position. To investigate, we sorted the minute-by-minute PN concentration into groups, based on occupancy and door and window position, and computed average PN concentrations for the classroom and outdoor monitors for each of these groups. We then computed the classroom to outdoor ratio (=  $f_1$ ),

based on averages for each condition. As shown in Table 4.10, the effect of door and window position on  $f_I$  is strong. For example, when students are present (13.9 h total),  $f_I$  has a relatively low value of 0.16 when the room is closed (2.4 h), and increases to  $f_I = 0.27$  when only the door is open (1.5 h). On the other hand, when the door and one or two windows are open (5.9 h combined), the value of  $f_I$  is much higher, 0.60 or 0.63. Similarly, when the classroom is unoccupied,  $f_I = 0.19$  when the room is closed, but for the period of ~ 4 h when either one or two windows is open (with or without the door), the value of  $f_I$  takes on higher values of 0.44, 0.64, or 0.68.

| Table 4.9. Air-exchange rates (AER) determined by carbon dioxide concentration decay in |
|---|
| classroom site S2.  |

| Time (h)    | <b>AER</b> $(h^{-1})$ | <b>WS</b> (m $s^{-1}$ ) | $ \Delta \mathbf{T} $ (°C) | <b>Classroom configuration</b> <sup>a</sup> |
|-------------|-----------------------|-------------------------|----------------------------|---|
| 3.1-3.4     | 3.9                   | 2.2                     | 3.8                        | Door and one window open <sup>b</sup>       |
| 27.3-27.8   | 2.8                   | 4.0                     | 2.2                        | Door open <sup>b</sup>                      |
| A1 (9 Oct)  | 3.3                   | 3.6                     |                            | Door open; one window open (5 cm);          |
|             |                       |                         |                            | ventilation system fan on.                  |
| A2 (9 Oct)  | 0.3                   | 3.6                     |                            |   |
| A3 (10 Oct) | 0.4                   | 4.9                     |                            |   |

<sup>a</sup> Unless otherwise noted, the configuration of the classroom during these measurements was this: doors and windows closed, room vacant, mechanical air supply system off. The state (on or off) of the air supply during the observational monitoring intervals in this table was not determined.

<sup>b</sup> One researcher (adult female) was present continuously in the classroom during the observational monitoring intervals presented in the table. AER estimates incorporate an estimate of the effect of CO<sub>2</sub> emissions from this occupant.

In combination, the distinctly different  $f_1$  values combined with the differences by occupancy status in the fractional periods in the high or low  $f_1$  states can help explain the shapes of the cumulative distribution curves presented in Appendix G, Figures G.2a-G.2c. To be specific, when students are present, only for 3.9 h/13.9 h = 28% of the time is the average indoor concentration a small fraction (< 30%) of the outdoor value. For another 2.2 h/13.9 h = 16% of the time, the average I/O ratio is intermediate at ~ 40%. However, for 5.9 h/13.9 h = 42% of the time, the I/O ratio has a high value of ~ 60-70%. Figure G.2a shows a difference between the classroom and outdoor concentrations that is less than a factor of two for the upper 40% of the distribution and for the lower 25% of the distribution, the indoor concentration is lower than that outdoors by about a factor of four.

At this site, there was no clear indoor emission source of particles based on the evidence from observational monitoring. During the manipulation experiments, we tested whether the laser printer in the classroom might be a source. In this test, with the room closed and continuous monitoring of indoor and outdoor PN concentrations, we printed 350 pages over a 20-minute period. Figure 4.11 displays the time-dependent particle concentrations. Indoor particle levels rise during the printing period. However, the rise might be explained by the increasing outdoor concentrations with time, especially since indoor concentrations remain high during the post-printing period. Consequently, we interpret this evidence to indicate that the printer in this room was not a prominent source of indoor UFP.

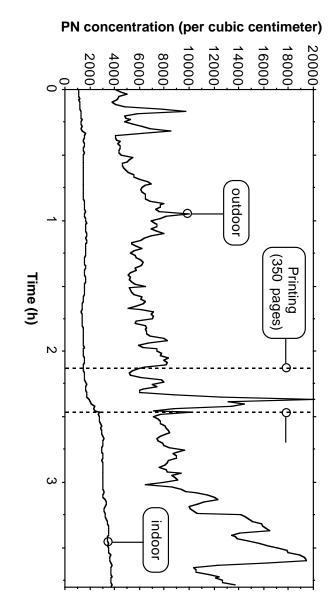
| State                        | <b>Duration</b> (h) | <b>WS</b> (m $s^{-1}$ ) | $ \Delta \mathbf{T}  (^{\circ} \mathbf{C})^{b}$ | $f_{I}(-)$ |  |  |
|------------------------------|---------------------|-------------------------|---|------------|--|--|
| At least one student present |                     |                         |   |            |  |  |
| All closed                   | 2.4                 | 1.7                     | 5.8   | 0.16       |  |  |
| Door open                    | 1.5                 | 3.0                     | 2.2   | 0.27       |  |  |
| Two windows open             | 2.2                 | 3.0                     | 6.4   | 0.45       |  |  |
| Door & 1 window open         | 2.0                 | 2.1                     | 3.8   | 0.63       |  |  |
| Door & 2 windows open        | 3.9                 | 2.3                     | 3.5   | 0.60       |  |  |
| Average                      | 13.9                | 2.0                     | 4.4   | 0.48       |  |  |
| At least one teacher prese   | nt                  |                         |   |            |  |  |
| All closed                   | 3.6                 | 1.8                     | 6.1   | 0.15       |  |  |
| Door open                    | 1.7                 | 3.0                     | 2.3   | 0.28       |  |  |
| Two windows open             | 3.7                 | 1.9                     | 6.6   | 0.53       |  |  |
| Door & 1 window open         | 3.7                 | 2.5                     | 3.2   | 0.58       |  |  |
| Door & 2 windows open        | 5.0                 | 2.4                     | 4.1   | 0.62       |  |  |
| Average                      | 20.2                | 2.2                     | 4.8   | 0.50       |  |  |
| Unoccupied                   |                     |                         |   |            |  |  |
| All closed                   | 32.7                | 2.2                     | 10.8  | 0.19       |  |  |
| Door open <sup>c</sup>       | 16.0                | 1.1                     | 10.0  | 0.37       |  |  |
| One window open              | 1.5                 | 2.6                     | 3.6   | 0.68       |  |  |
| Two windows open             | 1.2                 | 3.7                     | 2.7   | 0.44       |  |  |
| Door & 2 windows open        | 1.5                 | 2.2                     | 3.5   | 0.64       |  |  |
| Average                      | 52.9                | 1.9                     | 9.8   | 0.32       |  |  |

**Table 4.10.** Analysis of the indoor to outdoor and indoor to hallway particle concentration ratios at classroom site S2 for various door and window configurations.<sup>a</sup>

<sup>a</sup> Conditions with less than 1.0 h duration are omitted from the table, but not from the averages. <sup>b</sup> These averages are based on incomplete data. Overall, 14% of the *average student present* temperature data, 12% of the *average teacher present* temperature data, and 17% of the *average unoccupied* temperature data are missing.

<sup>c</sup> Almost all (15.5 h = 97%) of the unoccupied door open period occurs overnight. Because the door is an exterior one, it is unlikely that it was left open overnight. A more likely explanation for the data is a faulty sensor. Hence, the majority of data in this category may be better classified as "all closed."

Our field research staff maintained a log of activities that might possibly be associated with UFP emissions during observational monitoring. Table 4.11 summarizes the entries from that log. None of the recorded activities was associated with evident peaks in the classroom PN concentration.



rise in response to the printer operation. The rise that is displayed might be a consequence of the printer was used to print 350 pages. The indoor PN concentration trace does not show a marked experiment at site S2. During a 20-minute period beginning at about 2.1 h elapsed time, a laser rising outdoor concentrations. Figure 4.11. Time traces of particle concentrations during the source-investigation manipulation

| Time (h) | Activity or observation                                 | PN      |
|----------|---|---------|
|          |   | source? |
| 2.00     | Use of white board                                      |         |
| 2.50     | Pencil sharpened with electric pencil sharpener (twice) |         |
| 3.75     | Writing on chalk board                                  |         |
| 6.50     | Custodian sweeps room                                   |         |
| 48.12    | Use of white board                                      |         |
| 49.02    | Smell of perfume, aromatic personal care products       |         |
|          | (associated with guest teacher)                         |         |
| 53.98    | Washing up at sink (with fragrant dish soap)            |         |
| 54.33    | Custodian sweeps room                                   |         |
| 54.78    | Vacuuming   |         |
| 71.87    | Electric pencil sharpener used                          |         |
| 72.05    | Use of white board                                      |         |
| 72.57    | Electric pencil sharpener used                          |         |

Table 4.11. Activity notes related to potential PN emission sources at site S2

present in the classroom during the time when any student was present was 15.4. Aggregate one student was present in the classroom was 13.9 h or 4.6 h/d. The average number of students monitoring at site S2. For the three school days of monitoring, the cumulative time that at least Table 4.12 presents the results of the PN exposure analysis based on observational student PN exposure in the classroom was  $1882 \times 10^3$  cm<sup>-3</sup> h. Dividing this cumulative exposure by the average occupancy yields an average individual exposure for students of  $122 \times 10^3$  cm<sup>-3</sup> h. The average student exposure acquired while in the classroom per school day is obtained by dividing this average individual exposure for students ( $122 \times 10^3$  cm<sup>-3</sup> h) by the number of school days monitored (3), yielding the result  $41 \times 10^3$  cm<sup>-3</sup> h/d. We believe that essentially all of this exposure is caused by particles originating from outdoors. Analogous calculations have been performed for the teacher. The teacher's daily exposure rate is higher than the students (65 versus  $41 \times 10^3$  cm<sup>-3</sup> h/d) primarily because of the teacher's longer duration of exposure: 6.7 h/d versus 4.7 h/d. These exposures only consider time spent in the classroom.

| _school days of observational monitoring.                                  |          |         |
|--|----------|---------|
| Parameter  | Students | Teacher |
| Hours with at least one member present                                     | 13.9     | 20.2    |
| Average occupancy when at least one present                                | 15.4     | 1       |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                     | 1882     | 196     |
| Average individual exposure $(10^3 \text{ cm}^{-3} \text{ h})$             | 122      | 196     |
| Average individual exposure rate $(10^3 \text{ cm}^{-3} \text{ h d}^{-1})$ | 41       | 65      |

**Table 4.12.** Exposure analysis for students and teacher at classroom site S2, based on three school days of observational monitoring.

## 4.2.3 Site S3

Observational monitoring was conducted at classroom site S3 for 71 hours over three periods: (i) 9:00-15:21 on 20 October; 8:30 on 21 October through 7:34 on 23 October; and (iii) 15:17 on 23 October through 8:30 on 24 October 2008. For detailed background information about this school site, see §2.2.2. See also Figure C.10 (Appendix C) for a floor plan. Summarizing briefly, the school site is on the corner of a medium-trafficked urban street and a lightly used residential street. The nearest freeway is 1.7 km to the west. Although the campus dates from the 1950s, this classroom was built in the 1980s. The building contains three classrooms oriented around a center common room, known as a "pod." The classroom windows are inoperable. There are three doors, two to the outside and one to the pod. The classroom has a mechanical ventilation system that also serves the pod but is independent of the other two classrooms. The volume of the classroom is ~ 220 m<sup>3</sup>; the building volume is ~ 640 m<sup>3</sup>.

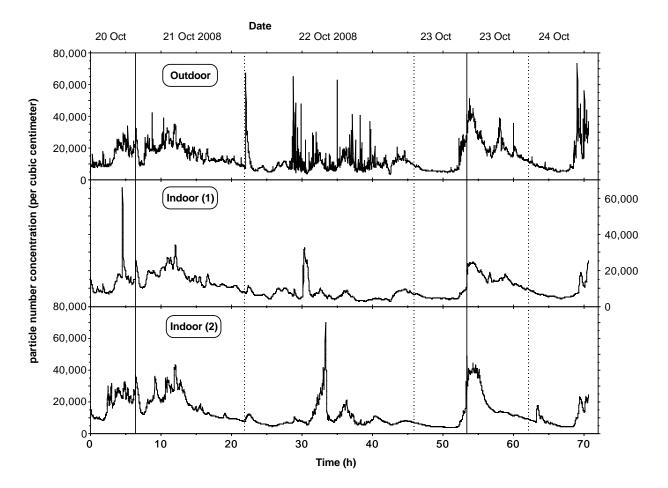
The primary indoor monitoring package was deployed inside the classroom, in the NW corner. The supplementary indoor monitor was placed in the cafeteria in a main building of the campus. The outdoor enclosure was placed in a secure area on the north edge of the campus about 100 m from the classroom. The outdoor monitor was only about 10 m from the more heavily trafficked street, whereas the classroom was about 100 m from that street.

A summary of the time-averaged species concentration and cofactor values was presented in Table 4.1 (§4.1). The average particle number concentrations measured during the observational monitoring period were 14.2 (outdoors), 10.5 (indoors classroom), and 12.6 (indoors cafeteria) in units of 1000 particles per cm<sup>3</sup>. The average outdoor PN levels measured at this site were similar to the average observed at all school sites. The indoor levels were the highest observed at any school site, about 50% higher than the average of  $6.9 \times 10^3$  particles cm<sup>-3</sup> for all six sites.

Figure 4.12 shows the time-series of particle number concentrations measured at this site. The outdoor concentrations at this site suggest higher average levels during the day than overnight, as we have seen at other sites. However, the breaks in monitoring erode what can be gleaned from direct visual observation. The outdoor profile also shows a prominent outdoor peak just after midnight on 22 October, something we have not observed at other sites.

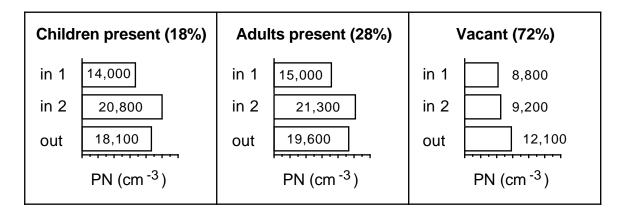
As for the other sites, we undertook a quantitative analysis of the diurnal concentration patterns outdoors. We computed the hourly average concentration for each clock hour for which we had complete data and then averaged the like hours across the full observational monitoring period. The resulting maximum was in the early afternoon, 13:00-14:00, at  $22.5 \times 10^3$  particles cm<sup>-3</sup>. The minimum occurred in the middle of the night, 3:00-4:00 and was several times smaller,  $5.5 \times 10^3$  particles cm<sup>-3</sup>.

The classroom PN level mainly tracks the outdoor behavior on a time-averaged basis, with the sharp spikes in the outdoor concentration profile absent from the indoor signal. There are two episodes where the classroom PN concentration spikes to a high value, one at time  $t \sim 4$  h and the second at time  $t \sim 30$  h. We believe that these episodes reflect the influence of indoor emission sources: a birthday candle in the first case and the use of the heater in the second. Investigations of these episodes as well as the influence of outdoor PN levels on indoor concentrations are explored subsequently.



**Figure 4.12.** Time-series of particle number concentrations during observational monitoring at classroom site S3. Monitor "Indoor (1)" was located in the classroom; monitor "Indoor (2)" sampled from the school cafeteria in a main campus building. The outdoor monitor sampled from a fenced area at the edge of the campus and about 100 m from the classroom site. The vertical dashed lines denote midnight. The time scale records the elapsed time of observational monitoring, which commenced at 9:00 on 20 October 2008. The two solid vertical lines indicate breaks in the monitoring period (15:21 on 20 October until 8:30 on 21 October; and 7:34-15:17 on 23 October).

The occupancy time series for site S3 is presented in Appendix F (Figure F.27). Figure 4.13 displays the time-averaged particle number concentrations measured during observational monitoring as sorted by occupancy status of the classroom. Focusing on the primary indoor monitor, "in 1," we see that the average concentration when the classroom is occupied by students,  $14.0 \times 10^3$  cm<sup>-3</sup>, is about 60% higher than when the classroom is vacant,  $8.8 \times 10^3$  cm<sup>-3</sup>. The classroom average concentration when students are present is 78% of the average outdoor concentrations drops to 72%. Overall, the ratio of average indoor to average does not appear to be the cause. Rather, the indoor air appears to be better coupled to outdoor air at this site than elsewhere owing to a high air exchange rate and, it seems, ineffective filtration.



**Figure 4.13.** Time-averaged indoor and outdoor particle number concentrations measured during the observational monitoring period at classroom site S3 and sorted according to the classroom occupancy status. The period with children present is a subset of the period when adults are present. Indicated in the upper portion of each frame is the percentage of the observational monitoring period for which the indicated state applies.

Table 4.13 summarizes the average copollutant levels at S3, sorted according to the occupancy status of the classroom. In Appendix F are presented time-series plots for the copollutants as follows: Figure F.28 for ozone; Figure F.29 for NO; Figure F.30 for carbon dioxide; and Figure F.31 for carbon monoxide. The time series data show that both ozone and NO levels indoors track the outdoor levels. As expected, owing to the fast homogeneous chemical reaction between NO and O<sub>3</sub>, these two species are anticorrelated. Ozone levels rise in the late morning and persist elevated into the mid afternoon. NO levels are elevated in the morning and again in the late afternoon to early evening period. The average indoor to outdoor ozone ratio is higher at this site than at any other school site, a bit below 50% when the classroom is occupied and close to 60% when it is unoccupied on a time-average basis.

| Species (units)           | Students | Adults | Unoccupied |
|---------------------------|----------|--------|------------|
| NO_in (ppb)               | 15.1     | 13.8   | 6.1        |
| NO_out (ppb)              | 16.6     | 19.0   | 15.2       |
| O <sub>3</sub> _in (ppb)  | 9.0      | 9.7    | 7.3        |
| O <sub>3</sub> _out (ppb) | 19.6     | 20.1   | 12.5       |
| CO <sub>2</sub> _in (ppm) | 577      | 549    | 443        |

**Table 4.13.** Copollutant levels at site S3 during observational monitoring.<sup>a</sup>

<sup>a</sup> Measurements sorted according to occupancy status of the building and then averaged.

Four air-exchange rate determinations were made at site S3 (see Table 4.14). Three of these determinations were made during observational monitoring at lunch, when the classroom was suddenly vacated with an elevated CO<sub>2</sub> level. The other measurement was made as a part of the manipulation experiments. The air-exchange rate of the classroom is consistently high, with individual determinations in the range 4.3-4.9 h<sup>-1</sup>, an overall average of 4.6 h<sup>-1</sup> and a standard deviation of  $0.3 h^{-1}$ . The high indoor to outdoor ratio for ozone and PN concentrations are consistent with this high air-exchange rate.

| Time (h)    | <b>AER</b> $(h^{-1})$ | <b>WS</b> (m $s^{-1}$ ) | $ \Delta \mathbf{T} $ (°C) |  |  |  |  |
|-------------|-----------------------|-------------------------|----------------------------|--|--|--|--|
| 3.0-3.5     | 4.4                   | 2.7                     | 3.4                        |  |  |  |  |
| 9.8-10.3    | 4.3                   | 1.8                     | 2.7 <sup>b</sup>           |  |  |  |  |
| 33.8-34.1   | 4.9                   | 3.2                     | 1.8 <sup>b</sup>           |  |  |  |  |
| A1 (27 Oct) | 4.8                   | 1.8                     | 4.6                        |  |  |  |  |

**Table 4.14.** Air-exchange rates (AER) determined by carbon dioxide concentration decay in classroom site S3. <sup>a</sup>

<sup>a</sup> The configuration of the classroom during these measurements was this: doors and windows closed, room vacant, mechanical air supply system on. The net indoor  $CO_2$  levels are referenced to a baseline value of 397 ppm, which is the minimum  $CO_2$  over the entire observational monitoring period.

<sup>b</sup> Onsite measurements of outdoor temperature are missing at these times.  $|\Delta T|$  is the mean of absolute values of the difference between the measured average classroom temperature and the reported outdoor temperature at the UC Berkeley central monitoring site.

The air-exchange rate measurements reported in Table 4.14 all occurred with the classroom doors closed. Our observational monitoring record indicates that the room was sometimes operated with one or two doors open. The indoor proportion of outdoor particles  $(f_1)$  might be influenced by door position and possibly by occupancy. When doors are closed, all ventilation air is provided either by mechanical ventilation, with the possibility of particle removal in a filter, or by infiltration, with the potential for particle loss along the infiltration air flow paths. When doors are open, air-exchange rates could increase, reducing the relative importance of deposition onto room surfaces. Also, there is no opportunity for particle loss from the air entering through an open doorway. These factors in combination lead us to expect a higher value of  $f_1$  when doors are open than when they are closed.

We investigated in the following manner. We started with the full time series of particle concentration measurements in the classroom and outdoors. We removed from the data set the two periods when we saw evidence that indoor sources caused elevated indoor concentrations: t = 4-5 h (candle); and t = 30-31.5 h (heater). Then, we sorted the data according to occupancy conditions and door position. For each state, we computed the average classroom and the average outdoor PN concentration. The value of  $f_1$  was computed as the ratio of these two averages, i.e. the average classroom concentration divided by the average outdoor concentration for the times when the particular condition applied. The results, presented in Table 4.15, show a pattern that conforms to expectations for a room with a high ventilation rate and ineffective filtration. When doors are open, the indoor/outdoor PN concentration ratio is in the range 0.74 (unoccupied) to 0.80 (students or teacher present). When the doors are closed, the ratio drops somewhat, to 0.66-0.71. When door position is taken into account, occupancy is seen to have little or no influence on the indoor proportion of outdoor particles at this site. The time-average value of  $f_1$  is higher for occupied periods (0.74 for students and 0.76 for teachers) compared with unoccupied periods (0.71), because the doors are open a much higher proportion of the time when the classroom is occupied.

Apart from two peak events, evidence at this site indicates that indoor sources contributed little to the PN concentrations in the classroom. Table 4.16 presents a list of activities recorded during observational monitoring that might be associated with UFP emissions. For each of the noted activities, we examined at a fine time scale the coincident indoor and outdoor PN concentration time series, looking for clear evidence of a sudden increase in indoor

concentrations that might indicate a source. Only two of the events were clear sources of PN in this classroom. The first occurred at  $t \sim 4.4$  h when a candle was lit briefly to commemorate the birthday of one of the students. The second occurred at  $t \sim 30.4$  h, when the heater was turned on. (Note that the heater was used on two other occasions during observational monitoring and these occurrences did not produce evident increases in the indoor PN concentration.) In addition to examining the PN concentration trace at the times of events recorded in the activity log, we also scanned the concentration profile looking for evidence of a sudden increase in indoor concentrations that could not be accounted for by penetration from outdoor air. No such episodes were detected at this site.

| State                        | <b>Duration</b> (h) | <b>WS</b> (m $s^{-1}$ ) | $ \Delta \mathbf{T}  (^{\circ} \mathbf{C})^{c}$ | $f_{I}(-)$ |  |  |
|------------------------------|---------------------|-------------------------|---|------------|--|--|
| At least one student present |                     |                         |   |            |  |  |
| One or both doors open       | 7.8                 | 2.4                     | 5.7   | 0.80       |  |  |
| Both doors closed            | 3.3                 | 3.5                     | 4.8   | 0.66       |  |  |
| Average                      | 12.0                | 2.7                     | 5.4   | 0.76       |  |  |
| At least one teacher present |                     |                         |   |            |  |  |
| One or both doors open       | 9.2                 | 2.3                     | 5.5   | 0.80       |  |  |
| Both doors closed            | 7.3                 | 2.7                     | 3.6   | 0.68       |  |  |
| Average                      | 18.0                | 2.5                     | 4.4   | 0.74       |  |  |
| Unoccupied                   |                     |                         |   |            |  |  |
| One or both doors open       | 1.6                 | 2.4                     | 3.4   | 0.74       |  |  |
| Both doors closed            | 47.9                | 1.6                     | 5.4   | 0.71       |  |  |
| Average                      | 50.1                | 1.6                     | 5.3   | 0.71       |  |  |

**Table 4.15.** Analysis of the indoor to outdoor particle concentration ratios at classroom site S3 for various door configurations.<sup>a,b</sup>

<sup>a</sup> Excluded from the analysis are two periods when there is evidence that indoor emissions influenced indoor PN concentrations: t = 4-5 h and t = 30-31.5 h.

<sup>b</sup> Windows were closed at all times.

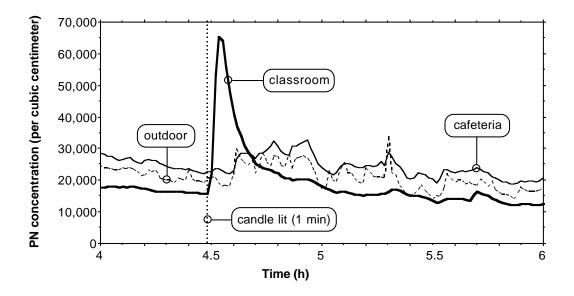
<sup>c</sup> Outdoor temperature data are not available for portions of the monitoring period. The averages reflect means for periods when data were available.

Figures 4.14 and 4.15 display detailed time series of the indoor and outdoor PN concentrations in relation to the use of the candle (Figure 4.14) and the heater (Figure 4.15). These figures show unambiguously that the indoor concentrations rise suddenly and significantly above the level in outdoor air with the onset of the event and then, afterward, decay fairly rapidly back to a baseline level that is less than the outdoor PN concentration. During manipulation experiments, we tested the use of a candle and the operation of the classroom heater and confirmed that both activities increased the indoor PN concentration in the classroom.

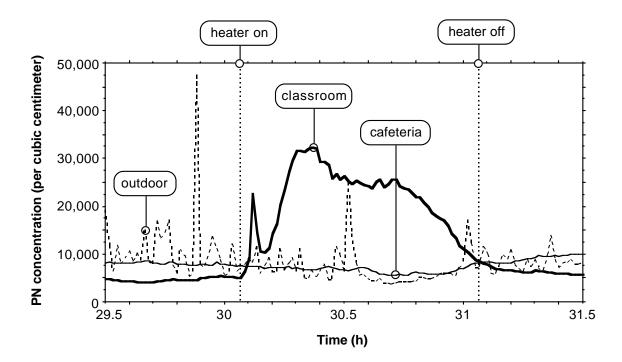
An analysis of the concentration peaks resulting from the source events was carried out, using the same approach as in houses. Table 4.17 presents a summary of the results. The decay coefficients were relatively high, 4-13 h<sup>-1</sup>. Other than the one very high value for the candle during the observational period, the other results appear consistent with the high air-exchange rates at this site (4.6 h<sup>-1</sup> average) plus a moderate rate (~1-2 h<sup>-1</sup>) associated with deposition to room surfaces. The overall particle emissions per event ( $\sigma$ ) are within the range of values observed in houses, ranging from 6 × 10<sup>12</sup> to 20 × 10<sup>12</sup> particles per event with a mean ± standard deviation of 14 ± 6 × 10<sup>12</sup> particles per event.

| Time (h) | Activity or observation                                       | PN source? |
|----------|---|------------|
| 0.0      | Heater is on  | Ν          |
| 4.4      | Candle lit for one minute, then blown out                     | Y          |
| 4.6      | Carpet is shaken by teacher                                   | Ν          |
| 7.8      | Heater is on  | Ν          |
| 7.9      | Teacher adjusts thermostat to reduce heat                     | Ν          |
| 7.9      | Earthquake drill. Students take shelter under desks.          | Ν          |
| 14.1     | High activity level; ten teachers work on Halloween costumes. | Ν          |
| 30.4     | Heater is on.   | Y          |
| 30.9     | Teacher turns off heater                                      | Ν          |
| 31.3     | Heater turned on.   | Ν          |
| 31.8     | Teacher turns off heater                                      | Ν          |
| 33.6     | Teacher sprays dish soap on tables                            | N          |

Table 4.16. Activity notes related to potential PN emission sources at site S3.



**Figure 4.14.** Detail of particle number concentration time-series measured at site S3 at the time that a candle was briefly lit in the room. The x-axis measures elapsed time during monitoring. The start time on the scale of 4.0 h corresponds to 13:00 on 20 October 2008.



**Figure 4.15.** Detail of particle number concentration time-series measured at site S3 at a time that a heater was used. The x-axis measures elapsed time during monitoring. The start time on the scale of 29.5 h corresponds to 7:39 on 22 October 2008.

**Table 4.17.** Analysis of UFP sources from observational monitoring and manipulation experiments.

| ID | Source activity | <b>Time/duration</b> (h) | $k + a (h^{-1})$ | $\sigma/V (10^3 \text{ cm}^{-3})$ | $\sigma$ (10 <sup>12</sup> ) |  |
|----|-----------------|--------------------------|------------------|-----------------------------------|------------------------------|--|
| a  | Candle          | 4.5-4.8                  | 13 <sup>a</sup>  | 70                                | 14                           |  |
| b  | Central heater  | 30.1-31.1                | 3.6              | 87                                | 18                           |  |
| M1 | Candle          | 0.6                      | 6.9              | 98                                | 20                           |  |
| M2 | Candle          | 0.6                      | 6.5              | 78                                | 16                           |  |
| M3 | Central heater  | 0.8                      | 5.2              | 42                                | 8.6                          |  |
| M4 | Central heater  | 0.7                      | 6.4              | 27                                | 5.6                          |  |

<sup>a</sup> Average k+a value. At the start of the decay period the door was open, and the decay rate was estimated to be 16 h<sup>-1</sup>. At the end of the decay period the door was closed and the decay rate was estimated as 10 h<sup>-1</sup>.

Table 4.18 presents the results of the PN exposure analysis based on observational monitoring at site S3. For the three school days of monitoring, the cumulative time that at least one student was present in the classroom was 13.0 h or 4.3 h/d. The average number of students present in the classroom during the time when any student was present was 16.6. For each minute when one or more students were present in the classroom, the increment of exposure was computed as the student occupancy multiplied by classroom PN level. The increments, summed over all periods of student occupancy during the observational monitoring period, totaled 2889 ×  $10^3$  cm<sup>-3</sup> h. Of this total, the candle indoor peak event contributed an estimated increment of 105 ×  $10^3$  cm<sup>-3</sup> h, or about 4% of the total. The heater event did not contribute to the exposure of

students because the complete peak occurred before they arrived for school in the morning. Dividing the students' cumulative exposure by the average occupancy yields an average individual exposure of  $174 \times 10^3$  cm<sup>-3</sup> h. The average student exposure acquired while in the classroom per school day is obtained by dividing the average individual exposure for students  $(174 \times 10^3 \text{ cm}^{-3} \text{ h})$  by the number of school days monitored (3), yielding the result  $58 \times 10^3 \text{ cm}^{-3}$  h/d. Other than the 4% attributable to the candle event, we believe that essentially all of the remainder of the indoor exposure is attributable to particles originating from outdoors. Analogous calculations have been performed for the teacher. The teacher's daily exposure rate is higher than the students (99 versus  $58 \times 10^3$  cm<sup>-3</sup> h/d) primarily because of the teacher's longer duration of exposure: 6.6 h/d versus 4.3 h/d. These exposures only consider time spent in the classroom.

| Table 4.18. Exposure analysis for students and teachers at classroom site S3, based on three |          |         |  |  |  |  |
|--|----------|---------|--|--|--|--|
| school days of observational monitoring.   |          |         |  |  |  |  |
| Parameter  | Students | Teacher |  |  |  |  |

| Parameter   | Students | Teacher |
|---|----------|---------|
| Hours with at least one member present                                  | 13.0     | 19.9    |
| Average occupancy when at least one present                             | 16.6     | 1       |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                  | 2889     | 297     |
| Cumulative exposure from candle peak $(10^3 \text{ cm}^{-3} \text{ h})$ | 105      | 5       |
| Cumulative exposure from heater peak $(10^3 \text{ cm}^{-3} \text{ h})$ | 0        | 6       |
| Average individual exposure $(10^3 \text{ cm}^{-3} \text{ h})$          | 174      | 297     |
| Avg. individual exposure rate $(10^3 \text{ cm}^{-3} \text{ h d}^{-1})$ | 58       | 99      |

## 4.2.4 Site S4

Observational monitoring was conducted at classroom site S4 for a continuous 72-hour period commencing at 6:45 on 4 November and ending at 7:05 on 7 November 2008. For detailed background information about this school site, see §2.2.2; see also Figure C.11 (Appendix C) for a floor plan. Summarizing briefly, the school site is on a residential street. Busy surface streets are located 100 m to the north and 200 m to the south, and the nearest freeway is ~ 1 km to the west. The classroom is located in an older building (> 100 y) and is equipped with a radiant heating system. The room ventilation and cooling is achieved through the use of windows. The classroom we monitored ( $V \sim 240 \text{ m}^3$ ) is on the second floor of the two-story building. One wall of the room has a door that opens to the hallway. The opposite wall has windows that look out onto a courtyard. The other two walls adjoin interior spaces, respectively, a classroom and a custodian closet.

The primary indoor monitoring package was deployed inside the classroom, along the S wall. The supplementary monitor was placed in the hallway near the classroom door. The outdoor package was set up to monitor in the nearby courtyard. We did not have access to secure outdoor electrical power at this school site. Consequently, we did not acquire data on outdoor PN or copollutant concentrations during the approximate period midnight to 7:00 each day during observational monitoring. The indoor packages operated continuously.

A summary of the time-averaged species concentration and cofactor values was presented in Table 4.1 (§4.1). The average particle number concentrations measured during the observational monitoring period were 21.3 (outdoors), 7.3 (indoors classroom), and 9.8 (indoors hallway) in units of 1000 particles per cm<sup>3</sup>. Note that the outdoor concentration represents an average for only a portion of the observational period. The average is probably biased high relative to the daily average because of the absence of data from the overnight periods. Table 4.19 substantiates this point. The table presents (a) the time-average PN level outside at all six school sites for only the hours 7:00-23:00, and (b) the daily average for all hours for the five sites where we have complete data. The all hours average is lower than the 7:00-23:00 average at each site, by values ranging from 10% to 23%. The outdoor levels for the 7:00-23:00 periods are higher at S4 than at any other site. The average across all sites during these hours is  $16.8 \times 10^3$ cm<sup>-3</sup>; the level at S4,  $22.0 \times 10^3$  cm<sup>-3</sup>, is about 30% higher than this average.

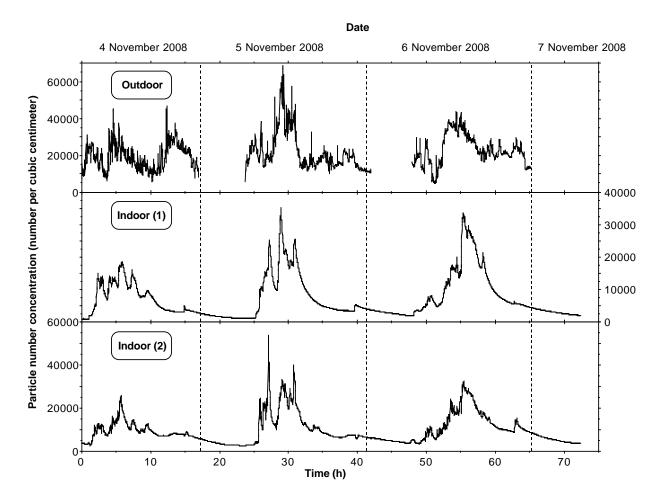
| 25.00      | 25.00 and for an monitored nours. |                |                           |  |  |  |
|------------|-----------------------------------|----------------|---------------------------|--|--|--|
| ID         | Avg. 7:00-23:00                   | Avg. all hours | <b>Ratio</b> <sup>a</sup> |  |  |  |
| <b>S</b> 1 | 20.0                              | 16.0           | 0.80                      |  |  |  |
| S2         | 16.7                              | 12.9           | 0.77                      |  |  |  |
| <b>S</b> 3 | 15.8                              | 14.2           | 0.90                      |  |  |  |
| <b>S</b> 4 | 22.0                              | na             |                           |  |  |  |
| S5         | 10.8                              | 9.7            | 0.90                      |  |  |  |
| <b>S</b> 6 | 15.4                              | 13.4           | 0.87                      |  |  |  |

| <b>Table 4.19.</b> Average outdoor PN concentration $(10^3 \text{ c})$ | cm <sup>-3</sup> ) at the six school sites for hours 7:00- |
|--|--|
| 23:00 and for all monitored hours.                                     |  |

<sup>a</sup> This column presents the ratio of the all-hours average to the average for the hours 7:00-23:00 alone.

The outdoor levels are higher during observational monitoring at this site compared with other school sites. Classroom concentrations are intermediate among the sites. The value at S4,  $7.3 \times 10^3$  cm<sup>-3</sup>, is close to the overall average of  $6.9 \times 10^3$  cm<sup>-3</sup> at all sites.

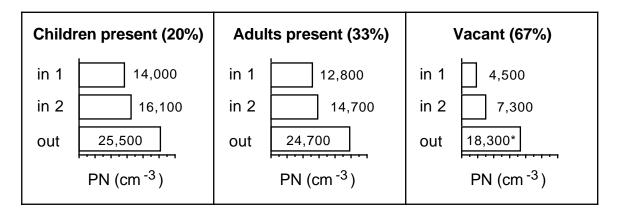
Figure 4.16 displays the time-series of particle number concentrations measured at this site. Even without a complete record, the outdoor concentrations measured at this site suggest a diurnal pattern with the highest concentrations during midday. During the hours monitored, the highest hourly average concentration on 5 November occurred 11:00-12:00 ( $46.1 \times 10^3$  cm<sup>-3</sup>). On 6 November, the highest hour was 12:00-13:00 ( $36.5 \times 10^3$  cm<sup>-3</sup>). On 4 November, there are two peaks, one 11:00-12:00 ( $26.5 \times 10^3$  cm<sup>-3</sup>) and the second occurring in the late afternoon, 19:00-20:00 ( $29.0 \times 10^3$  cm<sup>-3</sup>).



**Figure 4.16.** Time-series of particle number concentrations during observational monitoring at classroom site S4. Monitor "Indoor (1)" was located in the classroom; monitor "Indoor (2)" sampled from the hallway just outside the classroom door. The outdoor monitor sampled from a courtyard close to the school building. The vertical dashed lines denote midnight. The time scale records the elapsed time of observational monitoring referenced to the start at 6:45 on 4 November 2008.

The diurnal pattern of classroom PN levels shows much higher concentrations during the daytime hours than overnight. There are no prominent spikes in the indoor signal, suggesting that the main source of classroom PN levels is the penetration and persistence of particles that originate in outdoor air.

The occupancy time series for site S4 is presented in Appendix F (see Figure F.40). Figure 4.17 displays the time-averaged particle number concentrations measured during observational monitoring as sorted by occupancy status of the classroom. Focusing on the primary indoor monitor, "in 1," we see that the average concentration when the classroom is occupied by students,  $14.0 \times 10^3$  cm<sup>-3</sup>, is more than three times as high as when the classroom is vacant,  $4.5 \times 10^3$  cm<sup>-3</sup>. The classroom average concentration when students are present is 55% of the average outdoor concentration over the same time period.



**Figure 4.17.** Time-averaged indoor and outdoor particle number concentrations measured during the observational monitoring period at classroom site S4 and sorted according to classroom occupancy status. (Note that the average for the outdoor monitor during vacant periods is based on sampling for a subset of that time, 28 h of 48 h total.) Indicated in the upper portion of each frame is the percentage of time for which the indicated state applies.

Table 4.20 summarizes the average copollutant levels at S4, sorted according to the occupancy status of the classroom. In Appendix F are presented time-series plots for the copollutants as follows: Figure F.41 for ozone; Figure F.42 for NO; Figure F.43 for carbon dioxide; and Figure F.44 for carbon monoxide. At this site, the baseline CO<sub>2</sub> level when the classroom is unoccupied is elevated (Figure F.43) and the diurnal pattern of NO concentrations also suggests the possibility of a continuous low-level indoor emission source (Figure F.42). We note, as discussed in §2.2.2.4, that this classroom is sited above the school's boiler room. On the other hand, the indoor UFP level when the classroom is unoccupied does not appear to be elevated compared with levels at other classroom sites.

| Species (units)           | Students | Adults | Unoccupied |
|---------------------------|----------|--------|------------|
| NO_in (ppb)               | 6.4      | 7.3    | 16.0       |
| NO_out (ppb)              | 10.3     | 16.2   | 11.9       |
| O <sub>3</sub> _in (ppb)  | 4.7      | 5.4    | 6.7        |
| O <sub>3</sub> _out (ppb) | 21.7     | 18.1   | 9.4        |
| CO <sub>2</sub> _in (ppm) | 1162     | 961    | 722        |

Table 4.20. Copollutant levels at site S4 during observational monitoring.<sup>a</sup>

<sup>a</sup> Measurements sorted according to occupancy status of the building and then averaged.

Eight air-exchange rate determinations were made at site S4. The results are summarized in Table 4.21. Two of these determinations were made during observational monitoring on occasions when the classroom became suddenly unoccupied with an elevated  $CO_2$  level. The other six measurements (A1-A6) were made during the course of the manipulation experiments.

**Table 4.21.** Air-exchange rates (AER) determined by carbon dioxide concentration decay in classroom site S4.

| Time (h)    | <b>AER</b> $(h^{-1})^{a}$ | <b>WS</b> (m $s^{-1}$ ) | $ \Delta \mathbf{T} $ (°C) <sup>b</sup> | <b>Classroom configuration</b> <sup>c</sup> |
|-------------|---------------------------|-------------------------|---|---|
| 31.5-32.5   | 0.1                       | 2.9                     | 3.7                                     |   |
| 35.0-38.0   | 0.1                       | 1.4                     | 2.7                                     |   |
| A1 (7 Nov)  | 0.5                       | 1.6                     | 3.5                                     | One window open                             |
| A2 (7 Nov)  | 0.7                       | 1.0                     | 3.3                                     | Three windows open                          |
| A3 (7 Nov)  | 5.5                       | 1.3                     | 2.0                                     | Open door; three windows open               |
| A4 (10 Nov) | 0.2                       | 3.9                     | 1.6                                     |   |
| A5 (10 Nov) | 2.2                       | 3.6                     | 1.7                                     | Open door                                   |
| A6 (10 Nov) | 0.1                       | 2.8                     | 2.0                                     |   |

<sup>a</sup> The net indoor CO<sub>2</sub> levels are referenced to a baseline value of 530 ppm, based on observations from the CO<sub>2</sub> rebound experiment.

<sup>b</sup>  $\Delta$ T here represents the difference between the classroom and the hallway temperatures (classroom was always warmer during these experiments). The classroom-hallway temperature difference is key for governing buoyancy driven flow through the open doorway.

<sup>c</sup> Unless otherwise noted, the configuration of the classroom during these measurements was this: doors and windows closed, room vacant.

The air-exchange rate in this classroom varies across a large range, strongly influenced by the status (open or closed) of doors and windows. For four of the determinations, the classroom was closed. In this condition, the air-exchange rate was very low,  $0.1-0.2 \text{ h}^{-1}$ . At the other extreme, one measurement was made when the door and three windows were open; in this case, the air-exchange rate was  $5.5 \text{ h}^{-1}$ . In another case, with the door open but all windows closed, the air-exchange rate was  $2.2 \text{ h}^{-1}$ . For the two experiments with windows open and the door closed, the measured result was  $0.5 \text{ h}^{-1}$  when one window was open, and  $0.7 \text{ h}^{-1}$  with three open windows.

Proper interpretation of the air-exchange rate is confounded at this site, as at many other classroom sites, by the potentially significant yet likely incomplete coupling of the air space in the classroom with other interior spaces. When the school is occupied, an open doorway, leading to air-exchange between the hallway and the classroom, can serve as a net source of particles in the classroom, since the hallway commonly has higher PN concentrations than does the classroom. If the door and windows are open, providing for the possibility of a cross flow through the classroom, the direction of the flow could significantly influence the intrusion of  $CO_2$  (for determining AER) and particles from coupled indoor and from outdoor spaces. Despite these complications, it is evident that when its doors and windows are fully closed, the classroom is only weakly ventilated. Conversely, when doors and windows are open, especially in combination, the ventilation rate can be fairly high.

One expects then that the status of the doors and windows would not only influence the air-exchange rate but also the indoor proportion of outdoor particles. If the loss rate of UFP by means of deposition to indoor surfaces occurs at a rate of ~ 1 h<sup>-1</sup>, then the broad range of air-

exchange rates could result in a broad range of indoor-outdoor PN concentration ratios. With the room open, an air-exchange rate of AER ~ 5 h<sup>-1</sup>, and k ~ 1 h<sup>-1</sup>, the expected I/O ratio would be  $f_1$  ~ 5/6 = 0.83. With the room closed and an AER ~ 0.2 h<sup>-1</sup> and k ~ 1 h<sup>-1</sup>, the expected I/O ratio would be  $f_1 \sim 0.2/1.2 = 0.17$  (assuming that the penetration efficiency for UFP from outdoors is nearly unity in both cases). These estimates are based on equation 4.1.

We carried out an analysis of the indoor proportion of outdoor particles for conditions sorted by occupancy and by the status of the door and the windows. The results are summarized in Tables 4.22a (conditions with students present), 4.22b (teachers present), and 4.22c (classroom unoccupied and the outdoor WCPC operating). To generate these tables, the minuteby-minute data were sorted, first by occupancy and then by door and window conditions. Only configurations that cumulatively occurred for at least one hour during observational monitoring were included in the analysis. For each distinct configuration, we computed the average values of three environmental parameters:  $|\Delta T_1|$  represents the average of the absolute value of the temperature difference between the classroom and outdoors;  $|\Delta T_{12}|$  is the average of the absolute value of the temperature difference between the classroom and the hallway; and WS is the average wind speed (based on central station monitoring). Then, for each condition we determined the average PN concentrations in the classroom to the average outdoors;  $f_{12}$ represents the ratio of the average in the classroom to the average outdoors;  $f_{12}$ 

Table 4.22a shows some of the influence expected, with the highest value of  $f_1$  (0.84) occurring when the door and all four windows are open. However, with students present and the classroom completely closed, the average indoor/outdoor ratio is relatively high,  $f_1 = 0.57$ , similar to the ratio for all times of student occupancy. Worth noting is that the completely closed condition applied for only one hour while students were present.

| 15 Occup | is becupied by at least one student. |              |                     |                        |                 |         |          |
|----------|--------------------------------------|--------------|---------------------|------------------------|-----------------|---------|----------|
| Door     | Windows                              | <i>t</i> (h) | $ \Delta T_1 $ (°C) | $ \Delta T_{12} $ (°C) | <b>WS</b> (m/s) | $f_{I}$ | $f_{12}$ |
| Open     | 4 open                               | 1.7          | 3.9                 | 1.9                    | 1.1             | 0.84    | 1.06     |
| Open     | 1 open                               | 1.9          | 3.1                 | 2.7                    | 1.3             | 0.56    | 0.93     |
| Open     | All closed                           | 5.4          | 6.7                 | 4.0                    | 2.1             | 0.53    | 0.75     |
| Closed   | 3 open                               | 1.7          | 3.9                 | 4.2                    | 1.0             | 0.38    | 0.81     |
| Closed   | All closed                           | 1.0          | 5.4                 | 3.2                    | 1.4             | 0.57    | 0.94     |
| Occupi   | ed average                           | 14.5         | 4.8                 | 3.4                    | 1.6             | 0.55    | 0.87     |

**Table 4.22a.** PN concentration ratios for major room configurations at site S4, when classroom is occupied by at least one student.<sup>a</sup>

**Occupied average** | 14.5 | 4.8 | 3.4 | 1.6 | 0.55 | 0.87 ] <sup>a</sup> Parameter definitions: t = duration of the indicated condition;  $|\Delta T_1| =$  average of the absolute value of the classroom-outdoor temperature difference;  $|\Delta T_{12}| =$  average of the absolute value of the classroom-hallway temperature difference; WS = average windspeed measured at the nearest central monitoring station;  $f_1$  = ratio of average classroom to average outdoor PN concentrations;  $f_{12}$  = ratio of average classroom to average hallway PN concentrations. Only conditions with  $t \ge$ 1 h are included.

A clearer picture emerges from examining Table 4.22b, for conditions with the classroom occupied by a teacher (including those times when students are also present). Here, the overall average  $f_I$  value is 0.52. With the room completely closed, the lowest value of  $f_I = 0.28$  is obtained. Conversely, when the door and all four windows are open, the highest value of  $f_I = 0.87$  results. The results for unoccupied conditions are similar for like room configurations, as

displayed in Table 4.22c. For example, when the door is open and all windows are closed, the  $f_1$  value is 0.63 when the room is unoccupied and 0.43 when a teacher is present. Better agreement occurs when comparing closed-classroom conditions: with the door and all windows closed, the  $f_1$  value is 0.28 for conditions with a teacher in the room and 0.27 when the room is unoccupied. A point made clear in the final rows of these three tables is that the value of  $f_1$  is substantially lower when the room is unoccupied (0.33) than when the room is occupied (0.52 for teachers, 0.55 for students). The likely reason for this difference is the different distribution of classroom configuration states between unoccupied and occupied conditions. The door was closed 85% of the time when the room was unoccupied, a much higher percentage than when teachers (28%) or students (31%) were present.

| Door   | Windows    | <i>t</i> (h) | $ \Delta T_1 $ (°C) | $ \Delta T_{12} $ (°C) | <b>WS</b> (m/s) | $f_{I}$ | $f_{12}$ |
|--------|------------|--------------|---------------------|------------------------|-----------------|---------|----------|
| Open   | 4 open     | 4.5          | 3.1                 | 2.6                    | 1.4             | 0.87    | 1.05     |
| Open   | 2 open     | 1.3          | 6.3                 | 2.8                    | 1.8             | 0.62    | 1.05     |
| Open   | 1 open     | 2.3          | 3.0                 | 2.5                    | 1.2             | 0.56    | 0.92     |
| Open   | All closed | 8.6          | 7.2                 | 3.9                    | 1.9             | 0.43    | 0.74     |
| Closed | 3 open     | 1.7          | 3.9                 | 4.2                    | 1.0             | 0.38    | 0.81     |
| Closed | 1 open     | 1.3          | 2.0                 | 4.0                    | 1.5             | 0.45    | 0.81     |
| Closed | All closed | 2.6          | 8.7                 | 3.7                    | 2.3             | 0.28    | 0.79     |
| Occupi | ed average | 24.2         | 5.4                 | 3.4                    | 1.6             | 0.52    | 0.88     |

**Table 4.22b.** PN concentration ratios for major room configurations at site S4, when classroom is occupied by at least one teacher. <sup>a</sup>

<sup>a</sup> Parameter definitions: t = duration of the indicated condition;  $|\Delta T_1| =$  average of the absolute value of the classroom-outdoor temperature difference;  $|\Delta T_{12}| =$  average of the absolute value of the classroom-hallway temperature difference; WS = average windspeed measured at the nearest central monitoring station;  $f_1 =$  ratio of average classroom to average outdoor PN concentrations;  $f_{12} =$  ratio of average classroom to average hallway PN concentrations. Only conditions with  $t \ge 1$  h are included.

**Table 4.22c.** PN concentration ratios for major room configurations at site S4, when classroom is unoccupied and data on outdoor concentrations are available.<sup>a</sup>

| Door    | Windows    | <i>t</i> (h) | $ \Delta T_1 $ (°C) | $ \Delta T_{12} $ (°C) | <b>WS</b> (m/s) | $f_{I}$ | $f_{12}$ |
|---------|------------|--------------|---------------------|------------------------|-----------------|---------|----------|
| Open    | All closed | 3.4          | 7.0                 | 3.3                    | 2.3             | 0.63    | 0.84     |
| Closed  | 2 open     | 6.1          | 9.3                 | 3.3                    | 2.6             | 0.33    | 0.63     |
| Closed  | All closed | 17.6         | 8.6                 | 2.9                    | 2.8             | 0.27    | 0.59     |
| Overall | average    | 28.2         | 8.5                 | 3.0                    | 2.7             | 0.33    | 0.65     |

<sup>a</sup> Parameter definitions: t = duration of the indicated condition;  $|\Delta T_1| =$  average of the absolute value of the classroom-outdoor temperature difference;  $|\Delta T_{12}| =$  average of the absolute value of the classroom-hallway temperature difference; WS = average windspeed measured at the nearest central monitoring station;  $f_1$  = ratio of average classroom to average outdoor PN concentrations;  $f_{12}$  = ratio of average classroom to average hallway PN concentrations. Only conditions with  $t \ge 1$  h are included.

Evidence indicates that indoor sources contributed little to the PN concentrations in this classroom. During observational monitoring, we had a researcher in the classroom throughout each school day. Among the information observed and recorded was a log of activities that

might be associated with UFP emissions. Table 4.23 presents a summary. For each of the noted activities, we examined at a fine time scale the coincident indoor and outdoor PN concentration time series, looking for clear evidence of a sudden increase in indoor concentrations that might indicate a source. None of the events listed, with one possible exception, was a clear source of PN in this classroom. The exception was an occurrence on the first morning of monitoring (t = 2.1 h) in which a ballast overheated. This event produced a strong burning smell and, in response, the teacher opened windows. The PN concentration time series shows a sudden increase in concentrations. However, the indoor levels do not exceed the outdoor levels. We cannot discern whether the increase in indoor concentrations is a consequence of an increased supply rate of UFP from outdoor air, caused by the higher ventilation rate, or a consequence of emissions from the ballast.

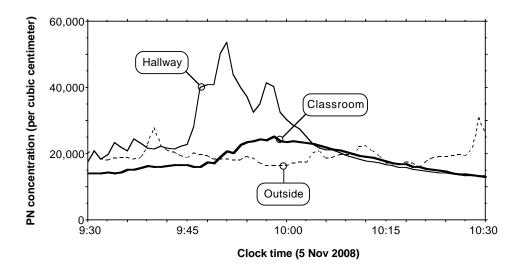
| <b>Time</b> (h) <sup>a</sup> | Observation  | PN source? |
|------------------------------|--|------------|
| 1.4                          | Printer used (1 page)  | N          |
| 1.8                          | Overhead projector in use  | Ν          |
| 2.0                          | Overhead projector off   | Ν          |
| 2.1                          | Lighting ballast overheated; strong burning smell, teacher opens windows | ?          |
| 3.2                          | Students writing with wet erase markers on desk mats                     | Ν          |
| 3.6                          | Electric pencil sharpener used   | Ν          |
| 24.3                         | Printing (a few sheets)  | Ν          |
| 24.3                         | Some white board writing/erasing   | N          |
| 25.8                         | Overhead projector on  | N          |
| 26.0                         | Overhead projector off   | N          |
| 26.5                         | Pencil sharpened   | Ν          |
| 28.2                         | Pencil sharpened   | Ν          |
| 28.4                         | Some writing on white board  | Ν          |
| 29.9                         | Printing   | N          |
| 30.1                         | Pencil sharpened a few times   | N          |
| 49.8                         | Overhead projector on; use wet erase marker                              | N          |
| 50.0                         | Projector off  | N          |
| 50.7                         | Pencil sharpened   | N          |
| 52.2                         | Pencil sharpened   | N          |
| 53.9                         | Printer used   | N          |

**Table 4.23.** Activity notes related to potential PN emission sources and ventilation conditions at site S4.

<sup>a</sup> Times referenced to the start of observational monitoring at 6:45 on 4 November 2008.

One incident during observational monitoring indicated the presence of an indoor PN source, but elsewhere in the school, rather than in the classroom. Figure 4.18 displays the data from this event, which occurred 9:45-10:15 on 5 November 2008. Note that at 9:30, before the event, the hallway and outdoor PN concentrations are similar and the classroom PN concentration is moderately lower. This was the common relationship among these traces. At 9:45, the hallway concentration rises sharply, increasing by more than a factor of two over a period of about five minutes. With a slight delay, the PN concentration in the classroom rises to a peak level that is less than observed in the hallway but above that in outdoor air. Hallway and

classroom concentrations both track closely on the decay side of the curve. The cause of this episode is unknown. It is interpreted as an indoor event that occurred in the school but outside of the classroom. Its impact on the classroom PN levels is estimated by integrating the area under the classroom PN time-series curve from ~ 9:45-10:30 and then subtracting from the result the expected contribution from outdoor particles (~  $f_1$  × outdoor PN concentration × event duration). The result, ~ 4 × 10<sup>3</sup> cm<sup>-3</sup> h, is incorporated in the exposure assessment. However, this contribution to exposure is relatively small compared with indoor events observed at house sites.

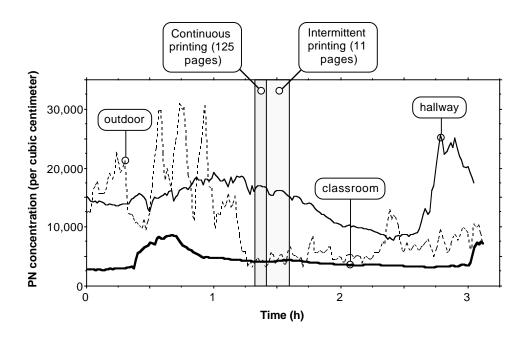


**Figure 4.18.** Detail of particle number concentration traces measured at site S4. For the time scale on the x-axis, 9:30 corresponds to an elapsed time of 26.75 h. The traces show evidence of an indoor emission event somewhere in the school that causes PN levels to rise in the hallway, which, in turn, causes the classroom PN level to briefly rise above that outdoors.

During the manipulation experiments at this site, we tested the classroom laser printer as a possible emission source. Within the closed classroom, we first continuously printed 125 pages over a period of  $\sim 5$  minutes and subsequently intermittently printed an additional 11 pages over  $\sim 10$  minutes. The monitored PN concentration traces, presented in Figure 4.19, do not indicate significant emissions from this activity. The PN concentration in the classroom remains essentially constant throughout the printing period.

Table 4.24 presents the results of the PN exposure analysis based on observational monitoring at site S4. For the three school days of monitoring, the cumulative time that at least one student was present in the classroom was 14.5 h or 4.8 h/d. The average number of students present in the classroom during the time when any student was present was 20.1. The cumulative exposure for students during the observational monitoring period totaled  $3955 \times 10^3$  cm<sup>-3</sup> h. Dividing this cumulative exposure by the average occupancy yields an average individual exposure of  $196 \times 10^3$  cm<sup>-3</sup> h. The average daily exposure acquired by students while in the classroom is obtained by dividing the average individual exposure ( $196 \times 10^3$  cm<sup>-3</sup> h) by the number of school days monitored (3), yielding the result  $66 \times 10^3$  cm<sup>-3</sup> h/d. The peak event at *t* ~ 27 h associated with an indoor emission source contributed about 2% to the average student exposure. We believe that essentially all of the remainder of the indoor exposure is

attributable to particles originating from outdoors. Analogous calculations have been performed for the teacher. The teacher's daily exposure rate is higher than the students (100 versus  $66 \times 10^3$  cm<sup>-3</sup> h/d) primarily because of the teacher's longer duration of exposure: 7.7 h/d versus 4.8 h/d. These exposures only consider time spent in the classroom.



**Figure 4.19.** Time traces of particle concentrations during the source-investigation manipulation experiment at site S4. During a 15-minute period beginning at 1.3 h elapsed time, a laser printer was used to print 136 pages. The indoor PN concentration trace does not show a marked rise in response to the printer operation.

**Table 4.24.** Exposure analysis for students and teachers at classroom site S4, based on three school days of observational monitoring.

| Parameter  | Students | Teacher |
|--|----------|---------|
| Hours with at least one member present                                     | 14.5     | 23.1    |
| Average occupancy when at least one present                                | 20.1     | 1       |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                     | 3955     | 299     |
| Exp. contribution from indoor event $(10^3 \text{ cm}^{-3} \text{ h})^a$   | 64       | 4       |
| Average individual exposure $(10^3 \text{ cm}^{-3} \text{ h})$             | 196      | 299     |
| Average individual exposure rate $(10^3 \text{ cm}^{-3} \text{ h d}^{-1})$ | 66       | 100     |

<sup>a</sup> This event occurred at  $t \sim 27$  h elapsed time (9:45 on 5 Nov 2008); the detailed PN concentration traces are presented in Figure 4.18. The table entries represent cumulative exposures for all present at the time.

## 4.2.5 Site S5

Observational monitoring was conducted at classroom site S5 for a continuous 71-hour period commencing at 8:58 on 18 November and ending at 8:02 on 21 November 2008. For detailed background information about this school site, see 2.2.2. See also Figure C.12 (Appendix C) for a floor plan. Summarizing briefly, the school is sited along a heavily traveled urban street. About 500 m to the west along this street is an on-ramp to an interstate highway. The school campus was established in the 1970s. The classroom we studied is in a single-story building that contains a cluster of classrooms that enclose an open courtyard. The classroom volume is estimated to be 258 m<sup>3</sup>.

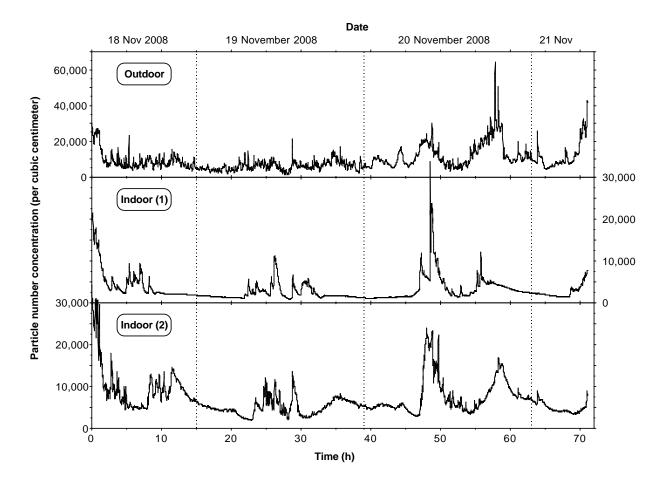
The classroom windows are inoperable. There are two doors, located on opposite sites of the room. The classroom has an independent mechanical ventilation system that is programmed to operate continuously between 6:45 and 18:15. A natural gas furnace controlled by a thermostat in the room provides heating when necessary. The ventilation system is equipped with a filter that the building maintenance person reported to have a "high efficiency" rating.

The primary indoor monitoring package was deployed inside the classroom, along the west wall. The supplementary indoor monitor was placed in a hallway in a different (nearby) building on the campus. The outdoor enclosure was placed in the inner courtyard.

A summary of the time-averaged species concentration and cofactor values was presented in Table 4.1 (§4.1). The average particle number concentrations measured during the observational monitoring period were 9.7 (outdoors), 3.2 (indoors classroom), and 7.1 (indoors hallway) in units of 1000 particles per cm<sup>3</sup>. The average outdoor PN and classroom PN levels measured at this site were lower than at any other school site. Furthermore, the ratio of classroom average to outdoor average PN levels, 0.33, was lower here than in any other S site.

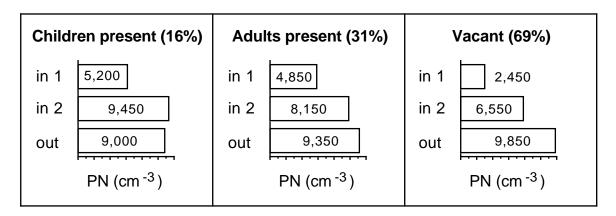
Figure 4.20 shows the time-series of particle number concentrations measured at this site. For a period extending shortly after the beginning of monitoring until almost two days later, the outdoor PN levels are consistently moderate, without any strong evidence of a diurnal pattern. The overall average outdoor PN level between 11:00 on 18 November and 7:00 on 20 November was  $7.1 \times 10^3$  particles cm<sup>-3</sup>. On 20 November, there are prominent morning and afternoon peaks with a dip in the middle of the day. The outdoor PN levels on this day peak at about 9:00 and again at 19:00, which would seem to be too late in each case to be directly a consequence of nearby traffic emissions. Using a 1-h running average, the maximum morning peak on 20 November was  $20.5 \times 10^3$  particles cm<sup>-3</sup> from 8:50-9:50. The maximum peak in the afternoon on this day was  $33.0 \times 10^3$  particles cm<sup>-3</sup> occurring 18:40-19:40.

The classroom PN levels ("indoor (1)") are very low and steady during the overnight periods when school is not in session. Higher concentrations and more fluctuations are seen during midday periods when school is in session. To a large extent, the shape of the concentration profiles during the school day track the outdoor levels, with dampened fluctuations. There is one prominent indoor peak, which occurs on the morning of 20 November. The cause of this peak, discussed in more detail below, is the use of the classroom heater for the first time in the season. Although the peak is relatively high, on an absolute scale it is fairly modest, reaching just above 30,000 particles cm<sup>-3</sup>.



**Figure 4.20.** Time-series of particle number concentrations during observational monitoring at classroom site S5. Monitor "Indoor (1)" was located in the classroom; monitor "Indoor (2)" sampled from the hallway of a separate, nearby campus building. The outdoor monitor sampled from a courtyard adjacent to the classroom. The vertical dashed lines denote midnight. The time scale records the elapsed time of observational monitoring, which commenced at 8:58 on 18 November 2008.

The occupancy time series for site S5 is presented in Appendix F (see Figure F.53). Figure 4.21 displays the time-averaged particle number concentrations measured during observational monitoring as sorted by occupancy status of the classroom. Focusing on the primary indoor monitor, "in 1," we see that the average concentration when the classroom is occupied by students,  $5.2 \times 10^3$  cm<sup>-3</sup>, is more than twice as high as when the classroom is vacant,  $2.5 \times 10^3$  cm<sup>-3</sup>. The classroom average concentration when students are present is 57% of the average outdoor concentration over the same time period. When students are absent, the ratio of average indoor to average outdoor concentrations drops to 25%.



**Figure 4.21.** Time-averaged indoor and outdoor particle number concentrations measured during the observational monitoring period at classroom site S5 and sorted according to the classroom occupancy status. The period with children present is a subset of the period when adults are present. Indicated in the upper portion of each frame is the percentage of the observational monitoring period for which the indicated state applies.

Table 4.25 summarizes the average copollutant levels at S5, sorted according to the occupancy status of the classroom. In Appendix F are presented time-series plots for the copollutants as follows: Figure F.54 for ozone; Figure F.55 for NO; Figure F.56 for carbon dioxide; and Figure F.57 for carbon monoxide. The indoor-outdoor ratio of ozone is low at this site, about 20% on average. The indoor NO level exceeded the outdoor level, especially during the unoccupied periods, suggesting the possibility of an indoor emission source. The time-series data (Figure F.55) supports this inference for the period starting with elapsed monitoring time  $t \sim 57$  h.

| Species (units)           | Students | Adults | Unoccupied |
|---------------------------|----------|--------|------------|
| NO_in (ppb)               | 17.2     | 12.2   | 15.1       |
| NO_out (ppb)              | 9.8      | 7.7    | 1.7        |
| O <sub>3</sub> _in (ppb)  | 2.3      | 3.1    | 3.9        |
| O <sub>3</sub> _out (ppb) | 13.0     | 15.7   | 19.6       |
| CO <sub>2</sub> _in (ppm) | 1080     | 856    | 481        |

Table 4.25. Copollutant levels at site S5 during observational monitoring.<sup>a</sup>

<sup>a</sup> Measurements sorted according to occupancy status of the building and then averaged.

Ten air-exchange rate determinations were made at site S5. The results are summarized in Table 4.26. One determination was made during observational monitoring, using occupant generated CO<sub>2</sub>. The other nine measurements (A1-A9) were made during manipulation experiments. The measured air-exchange rates of the classroom varied over a wide range, from 0.2 to 4.2 h<sup>-1</sup> (average  $\pm$  standard deviation = 1.6  $\pm$  1.1 h<sup>-1</sup>; median = 1.4 h<sup>-1</sup>; GM = 1.2 h<sup>-1</sup> GSD = 2.4).

The configurations under which the AER was measured can be sorted into four groups in a  $2 \times 2$  matrix, according to whether the mechanical ventilation system was on or off and whether the door was open or closed. Figure 4.22 presents the average and standard deviation of measurements for each of these groups. As expected, when a door is open, the air-exchange rate is much higher than when it is closed. With the ventilation system on and the doors closed, the

air-exchange rate averages  $1.2 \text{ h}^{-1}$ , which is fairly low for a classroom. Not surprisingly then, the CO<sub>2</sub> levels in the classroom rose to peak levels of ~ 1600 ppm when school was in session.

| Time (h)    | <b>AER</b> $(h^{-1})$ | <b>WS</b> (m $s^{-1}$ ) | $ \Delta \mathbf{T} $ (°C) | Classroom configuration notes          |
|-------------|-----------------------|-------------------------|----------------------------|--|
| 3.1-3.6     | 1.1                   | 2.7                     | 5.3                        | One adult occupant (researcher)        |
| A1 (21 Nov) | 1.5                   | 1.8                     | 5.9                        |  |
| A2 (21 Nov) | 1.2                   | 1.3                     | 5.6                        | Ventilation system fan off             |
| A3 (21 Nov) | 2.6                   | 1.3                     | 6.6                        | Door cracked open; ventilation fan off |
| A4 (21 Nov) | 4.2                   | 1.3                     | 6.9                        | Door wide open; ventilation fan off    |
| A5 (21 Nov) | 0.5                   | 1.3                     | 7.6                        | Heat on                                |
| A6 (24 Nov) | 1.2                   | 0.9                     | 7.6                        |  |
| A7 (25 Nov) | 1.5                   | 2.2                     | 4.5                        |  |
| A8 (25 Nov) | 0.2                   | 1.3                     | 4.3                        | Ventilation system fan off             |
| A9 (25 Nov) | 2.2                   | 0.9                     | 4.0                        | Front door cracked open                |

**Table 4.26.** Air-exchange rates (AER) determined by carbon dioxide concentration decay in classroom site S5. <sup>a, b</sup>

<sup>a</sup> Unless specified, the configuration of the classroom during these measurements was this: doors and windows closed, room vacant, ventilation system on.

<sup>b</sup> The net indoor CO<sub>2</sub> levels are referenced to a baseline value of 418 ppm, which is the minimum indoor CO<sub>2</sub> level measured over the entire observational monitoring period. When an adult occupant is present, we account for the metabolic emissions of CO<sub>2</sub> in the analysis, using a CO<sub>2</sub> emission rate of 0.013 mol/min (= 830 g CO<sub>2</sub>/d) for adults. The estimate is derived from ASHRAE Standard 62-1989, assuming a metabolic level of 1.2 met units, corresponding to "office (seated)" and a respiratory quotient of 0.83, corresponding to a "normal mix" diet.

Doors closedOne door open $V_{i}$ 1.2 ± 0.4 (5)2.2 (1) $V_{i}$ 0.7 ± 0.7 (2)3.4 ± 1.1 (2)

**Figure 4.22.** Average  $\pm$  standard deviation (number of measurements) for air-exchange rates (units = per hour) measured at S5 and sorted according to the position of the front door of the classroom and the status of the mechanical ventilation system fan. These results summarize the detailed presentation in Table 4.26.

We next explore the impact of outdoor PN concentrations on indoor levels. We first removed from the time-series data the period t = 48-51 h, when an indoor source (heater) significantly influenced the classroom PN levels. Then, we sorted the remaining data according to three criteria: (i) occupancy (students present, teacher present, or unoccupied); (ii) door position (at least one door open, or both doors closed); and (iii) operation of the ventilation

system fan (on or off). For the third criterion, we used the scheduled times of fan operation (6:45-18:15) as the basis for sorting.

For each grouping, we computed the time-averaged classroom and outdoor PN concentration. The ratio of these two averages is the parameter we refer to as  $f_1$ . It is a strong indicator of the impact of outdoor PN concentrations on classroom levels.

Table 4.27 presents a summary of the results. When the room is occupied by students,  $f_I$  has an overall average of ~ 0.51. With the doors closed,  $f_I$  drops to 0.41, whereas with at least one door open the value is 0.54. Results when a teacher is present are comparable. The higher value of  $f_I$  with open doors is expected because of the closer coupling of classroom air to the outdoors. But the quantitative extent of the effect is smaller than anticipated. The value of  $f_I$  is lower when the classroom is unoccupied, with the lowest value of 0.22 occurring when the ventilation system fan is off.

| <b>Table 4.27.</b> Analysis of the indoor proportion of outdoor particles $(f_1)$ at classroom site S5 in |
|---|
| relation to occupancy, door position, and ventilation system status. <sup>a,b</sup>                       |

| State                                      | <b>Duration</b> (h) | <b>WS</b> $(m s^{-1})$ | $ \Delta \mathbf{T} $ (°C) | $F_1(-)$ |  |
|--|---------------------|------------------------|----------------------------|----------|--|
| At least one student present and HVAC on   |                     |                        |                            |          |  |
| One or both doors open                     | 6.9                 | 2.1                    | 5.7                        | 0.54     |  |
| Both doors closed                          | 2.1                 | 2.6                    | 6.1                        | 0.41     |  |
| Average                                    | 9.1                 | 2.2                    | 5.8                        | 0.51     |  |
| At least one teacher prese                 | nt and HVAC on      |                        |                            |          |  |
| One or both doors open                     | 7.9                 | 2.2                    | 5.7                        | 0.55     |  |
| Both doors closed                          | 7.3                 | 2.4                    | 5.2                        | 0.45     |  |
| Average                                    | 19.1                | 2.3                    | 5.4                        | 0.49     |  |
| Unoccupied and HVAC on                     |                     |                        |                            |          |  |
| Both doors closed                          | 11.0                | 2.2                    | 6.2                        | 0.28     |  |
| Average                                    | 11.5                | 2.2                    | 6.2                        | 0.31     |  |
| Unoccupied and HVAC off; both doors closed |                     |                        |                            |          |  |
| Average                                    | 37.5                | 2.1                    | 6.2                        | 0.22     |  |

<sup>a</sup> Excluded from the analysis is one period when there is evidence that indoor emissions influenced indoor PN concentrations: t = 48-51 h.

<sup>b</sup> Windows were closed at all times.

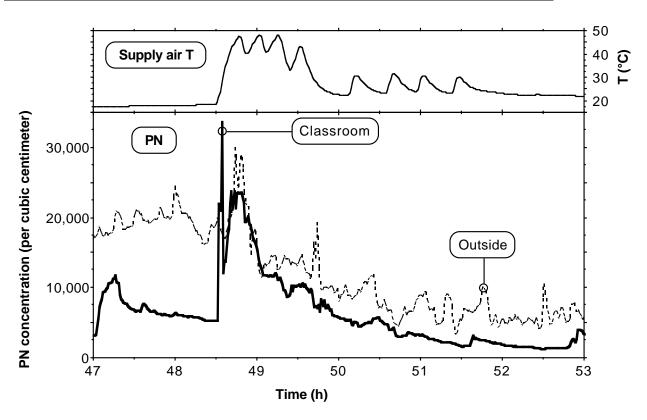
Apart from one brief peak event associated with heater use, evidence at this site indicates that indoor sources contributed little to the PN concentrations in the classroom. Table 4.28 presents a list of activities recorded during observational monitoring that might be associated with UFP emissions. Only the operation of the heater at  $t \sim 48.6$  h was a clear source of PN in this classroom. However, while the emissions from this event were sufficient to detect the occurrence, they were not sufficient to quantify emissions against the background contribution from outdoor air.

Figure 4.23 displays a detailed time series of the indoor and outdoor PN concentrations in relation to the use of the heater. The figure shows a sharp spike in indoor PN concentration, coincident with the beginning of heater use. It is clear that the narrow spike at t = 48.6 h is a consequence of heater use. Because the spike is so narrow, it contributes little to exposure (which is related to the area beneath the curve). A more significant contribution is associated with the subsequent few hours, when the indoor concentration rises to a second peak of ~ 25,000

cm<sup>-3</sup> and then slowly decays. In this portion of the time series, the indoor concentration tracks the shape of the outdoor PN concentration, with a gradually declining ratio of indoor to outdoor particle concentrations. At the time the heater was first turned on, which was the first use of the season, it produced a "burning hair" smell that led to the opening of classroom doors. We cannot discern whether the indoor particles beyond the first peak are solely a consequence of the penetration of particles from outdoor air or whether there is a persistent contribution from the heater. However, it is noteworthy that during the manipulation experiments we tested the operation of the classroom heater and did not detect indoor emissions of particles from it then.

| Time (h) | Activity or observation                            | PN source? |
|----------|--|------------|
| 0.0      | Thermostat at 72°F, air supply system fan on       | Ν          |
| 1.9      | Overhead projector turned on                       | Ν          |
| 48.6     | Heater on; perceived as creating a "burning" smell | Y          |

Table 4.28. Activity notes related to potential PN emission sources.



**Figure 4.23.** Detail of particle number concentration time-series measured at site S5 for elapsed (monitoring) times 47 h < t < 53 h. The upper frame shows the air temperature monitored at one of the discharge registers from the ventilation system. The classroom heater was turned on for the first time during the season at t = 48.6 h. The spike in indoor PN concentration at this time is almost certainly associated with emissions from the heater. The event produced a "burning hair" smell, which led to the opening of classroom doors to improve ventilation. We cannot distinguish in the subsequent trajectory of indoor PN concentrations whether there are continuing contributions from the heater or whether, instead, the indoor particles are solely due to intrusion from outdoor air.

Table 4.29 presents the results of the PN exposure analysis based on observational monitoring at site S5. For the three school days of monitoring, the cumulative time that at least one student was present in the classroom was 11.3 h or 3.8 h/d. The average number of students present in the classroom during the time when any student was present was 23.0. The cumulative student exposure during observational monitoring totaled  $1312 \times 10^3$  cm<sup>-3</sup> h. Dividing this result by the average occupancy yields an average individual exposure of  $57 \times 10^3$  cm<sup>-3</sup> h per student. The average student exposure per school day is obtained by dividing the average individual exposure for students ( $57 \times 10^3$  cm<sup>-3</sup> h) by the number of school days monitored (3), yielding the result  $19 \times 10^3$  cm<sup>-3</sup> h/d. We believe that almost all of this indoor exposure is attributable to particles originating from outdoors. Analogous calculations have been performed for the teacher. The teacher's daily exposure rate is higher than the students (36 versus  $19 \times 10^3$  cm<sup>-3</sup> h/d) primarily because of the teacher's longer duration of exposure: 7.3 h/d versus 3.8 h/d. These exposures only consider time spent in the classroom.

**Table 4.29.** Exposure analysis for students and teacher at classroom site S5, based on three school days of observational monitoring.

| Parameter  | Students | Teacher |
|--|----------|---------|
| Hours with at least one member present                                     | 11.3     | 21.9    |
| Average occupancy when at least one present                                | 23.0     | 1.0     |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                     | 1312     | 106     |
| Average individual exposure $(10^3 \text{ cm}^{-3} \text{ h})$             | 57       | 106     |
| Average individual exposure rate $(10^3 \text{ cm}^{-3} \text{ h d}^{-1})$ | 19       | 36      |

## 4.2.6 Site S6

Observational monitoring was conducted at classroom site S6 for a total of 70 hours. Monitoring occurred over four periods in late 2008: (i) from 8:15 on 2 December until 8:36 on 3 December; (ii) from 14:04 on 4 December until 8:30 on 5 December; (iii) from 12:25 on 5 December until 8:30 on 6 December; and (iv) from 8:42 until 15:48 on 8 December. For detailed background information about this school site, see §2.2.2; see also Figure C.13 (Appendix C) for a floor plan. The campus for S6 was the same as for S3. See §4.2.3 for a site description.

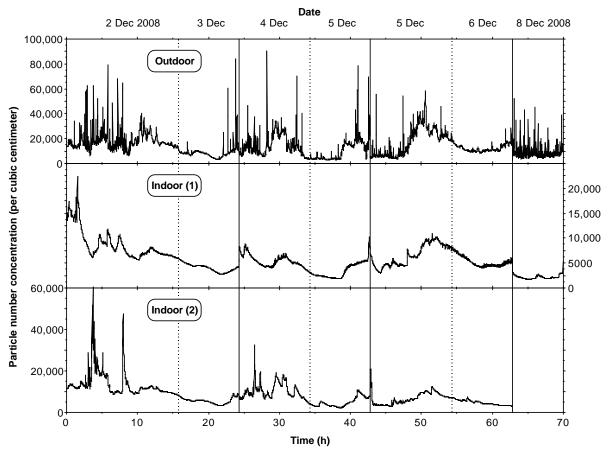
The classroom ( $V \sim 297 \text{ m}^3$ ) is located in a small building, constructed in the 1980s, which consists of only two classrooms. It is not equipped with mechanical ventilation; instead, air exchange occurs through infiltration and natural ventilation. One wall of the classroom has seven large, operable windows. They were not opened during the observational monitoring period, but the seals are leaky and so high infiltration rates would be expected. The same wall has the one doorway, which was sometimes open when school was in session.

The primary indoor monitoring package was deployed inside the classroom, near the middle of the room. The supplementary and outdoor monitoring packages were deployed in the same way as they were for S3. Specifically, the supplementary indoor monitor was placed in the cafeteria in a main building of the campus. The outdoor enclosure was placed in a secure area on the north edge of the campus about 150 m from the classroom. The outdoor monitor was only about 10 m from the street with heavier traffic at this site, whereas the classroom was about 150 m from that street.

A summary of the time-averaged species concentration and cofactor values was presented in Table 4.1 (§4.1). The average particle number concentrations measured during the observational monitoring period were 13.4 (outdoors), 5.6 (indoors classroom), and 8.0 (indoors cafeteria) in units of 1000 particles per cm<sup>3</sup>. The average outdoor PN levels measured at this site were similar to the average observed at all school sites. The indoor concentrations were about 20% below the mean  $(6.9 \times 10^3 \text{ cm}^{-3})$  for the six school sites.

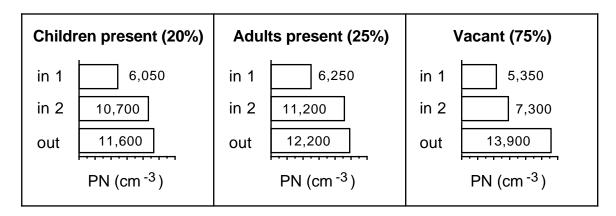
Figure 4.24 displays the time-series of particle number concentrations measured at this site. Because of the several interruptions in the monitoring period, it is difficult to discern visually a strong diurnal pattern to the outdoor PN concentrations. For the three midnight periods, the PN concentration is either steadily declining (2-3 Dec and 5-6 Dec) or much lower than average (4-5 Dec). We computed average concentrations for each clock hour that was completely monitored and averaged results for like hours over the multiple monitoring days. On average, the highest outdoor PN concentrations occurred in the evening, with the maximum of  $27.6 \times 10^3$  cm<sup>-3</sup> during 19:00-20:00. The minimum levels were about  $4 \times 10^{3} \text{ cm}^{-3}$  and occurred in the early morning, 3:00-4:00.

Like the case for the outdoor PN concentration, the intermittent monitoring obscures visual evidence of a diurnal pattern of PN concentrations in the classroom. However, one noteworthy point is evident from Figure 4.24. There is an absence of high concentration events: the highest concentration is about  $20 \times 10^3$  cm<sup>-3</sup>, much lower than the peak outdoor concentrations. As we have observed elsewhere, the indoor concentrations vary more smoothly over time. The high-frequency positive spikes that we observe in the outdoor PN signal are absent from the indoor concentration time series.



**Figure 4.24.** Time-series of particle number concentrations during observational monitoring at classroom site S6. Monitor "Indoor (1)" was located in the classroom; monitor "Indoor (2)" sampled from the school cafeteria in a main campus building. The outdoor monitor sampled from a fenced area at the edge of the campus and about 150 m from the classroom site. The vertical dashed lines denote midnight. The time scale records the elapsed time of observational monitoring, which commenced at 8:15 on 2 December 2008. The three solid vertical lines indicate breaks in the monitoring period (8:36 on 3 December until 14:04 on 4 December; 8:30-12:25 on 5 December; and 8:30 on 6 December until 8:42 on 8 December). No data were collected in the cafeteria for the final 7 h of the monitoring period owing to an instrument malfunction.

The occupancy time series for site S6 is presented in Appendix F (see Figure F.66). Figure 4.25 displays the time-averaged PN concentrations measured during observational monitoring, sorted by occupancy status of the classroom. Focusing on the primary indoor monitor, "in 1," we see that the average concentration when the classroom is occupied by students,  $6.0 \times 10^3$  cm<sup>-3</sup>, is about 10% higher than when the classroom is vacant,  $5.4 \times 10^3$  cm<sup>-3</sup>. Among the six classroom sites studied, this is by far the smallest ratio of occupied to vacant PN concentrations in the classroom. One factor is that this site, uniquely, had higher outdoor PN concentrations when the classroom was vacant  $(13.9 \times 10^3 \text{ cm}^{-3})$  than when it was occupied (11.6  $\times 10^3$  cm<sup>-3</sup>). As we have observed at all classroom sites, the ratio of classroom to outdoor concentrations is substantially higher for the occupied period (51%) than for the vacant period (37%).



**Figure 4.25.** Time-averaged indoor and outdoor particle number concentrations measured during the observational monitoring period at classroom site S6 and sorted according to classroom occupancy status. The period with children present is a subset of the period when adults are present. The "in 2" monitor, which was deployed in the cafeteria, did not operate during the final 7 h of the observational monitoring period. The loss of data for this monitor was 5.4 h for the period when children were in the classroom (8% of the total monitoring period), 5.8 h (8%) when adults were present, and 1.2 h (2%) when the classroom was vacant. The reported results for this monitor are based on the available 63 h of data. In the upper portion of each frame is the percentage of the observational monitoring period for which the indicated state applies.

Table 4.30 summarizes the average copollutant levels at S6, sorted according to the occupancy status of the classroom. In Appendix F are presented time-series plots for the copollutants as follows: Figure F.67 for ozone; Figure F.68 for NO; Figure F.69 for carbon dioxide; and Figure F.70 for carbon monoxide. This site had by far the highest outdoor NO levels of any of the classroom sites monitored. The indoor-outdoor ratios of ozone and NO are well below 1.0 during periods of occupancy.

| Species (units)           | Students | Adults | Unoccupied |
|---------------------------|----------|--------|------------|
| NO_in (ppb)               | 6.2      | 5.8    | 21.4       |
| NO_out (ppb)              | 26.3     | 27.8   | 52.6       |
| O <sub>3</sub> _in (ppb)  | 1.8      | 2.1    | 0.8        |
| O <sub>3</sub> _out (ppb) | 12.0     | 12.3   | 3.7        |
| CO <sub>2</sub> _in (ppm) | 1070     | 1000   | 512        |

**Table 4.30.** Copollutant levels at site S6 during observational monitoring.<sup>a</sup>

<sup>a</sup> Measurements sorted according to occupancy status of the building and then averaged.

Eleven air-exchange rate determinations were made at site S6; see Table 4.31. Five of these determinations were made during observational monitoring on occasions when the classroom became suddenly unoccupied with an elevated  $CO_2$  level. The other six measurements (A1-A6) were made during the course of the manipulation experiments.

Individual determinations of the air-exchange rate span more than an order of magnitude, from 0.4 to 5.9 h<sup>-1</sup>. The key factor influencing the outcome is the position of the classroom door. For the seven determinations made with the door closed (the base-case condition), the range of air-exchange rates was 0.4-0.8 h<sup>-1</sup> with a mean  $\pm$  standard deviation of 0.6  $\pm$  0.2 h<sup>-1</sup>. Each of the four measurements with the door open was considerably higher than any of the seven with the

door closed. The average  $\pm$  standard deviation for the door-open condition was  $4.0 \pm 1.7$  h<sup>-1</sup>. The operation of the HAC system was not a strong factor, presumably because it is a purely recirculating system. Temperature difference and wind speed would also be expected to influence the outcome. Regression analysis of the AER versus wind speed (WS) and temperature difference ( $\Delta$ T), separately for door open and door closed positions, shows that the AER correlates well with wind speed ( $r^2 = 0.63$ ) when the door is closed and with temperature difference ( $r^2 = 0.83$ ) when the door is open.

| classiooni sit | .0.50.                    |                                |                            |   |
|----------------|---------------------------|--------------------------------|----------------------------|---|
| Time (h)       | <b>AER</b> $(h^{-1})^{a}$ | <b>WS</b> (m s <sup>-1</sup> ) | $ \Delta \mathbf{T} $ (°C) | <b>Classroom configuration</b> <sup>b</sup> |
| 3.8-4.4        | 0.4                       | 2.2                            | 6.4                        | Researcher present                          |
| 6.9-7.3        | 3.9                       | 2.2                            | 7.5                        | Door open                                   |
| 25.6-25.9      | 1.7                       | 1.8                            | 6.7                        | Door open                                   |
| 45.9-46.9      | 0.5                       | 2.2                            | 6.6                        |   |
| 66.2-66.6      | 5.9                       | 3.1                            | 12.7                       | Door open; researcher present               |
| A1 (4 Dec)     | 0.7                       | 2.9                            | 7.5                        | Thermostat set at 18 °C (64 °F)             |
| A2 (4 Dec)     | 0.7                       | 2.7                            | 5.2                        | Thermostat set at 18 °C (64 °F)             |
| A3 (4 Dec)     | 0.5                       | 2.2                            | 4.7                        | Thermostat set at 18 °C (64 °F)             |
| A4 (6 Dec)     | 4.5                       | 1.3                            | na                         | Door open; thermostat set at 23-24 °C       |
| A5 (6 Dec)     | 0.8                       | 2.5                            | 4.9                        | HAC off; three portable fans on             |
| A6 (6 Dec)     | 0.8                       | 2.7                            | 5.9                        | Thermostat set at 24 °C (76 °F)             |

**Table 4.31.** Air-exchange rates (AER) determined by carbon dioxide concentration decay in classroom site S6.

<sup>a</sup> The net indoor  $CO_2$  levels are referenced to a baseline value of 431 ppm, which is the minimum classroom  $CO_2$  level over the entire observational monitoring period. When an occupant is present, we account for his or her metabolic emissions of  $CO_2$  in the analysis, using a  $CO_2$  emission rate of 0.013 mol/min (= 830 g  $CO_2/d$ ) for adults (researcher). This estimate is derived from ASHRAE Standard 62-1989, assuming a metabolic level of 1.2 met units, corresponding to "office (seated)" and a respiratory quotient of 0.83, corresponding to a "normal mix" diet.

<sup>b</sup> Unless specified, the configuration of the classroom during observational monitoring measurements was this: doors and windows closed, room vacant, wall mounted heating and air conditioning units (HAC) set to "fan on," but with no thermal conditioning. During manipulation experiments, unless otherwise specified, conditions were the same as for observational monitoring except that the HAC thermal conditioning mode was also on, under thermostatic control.

Because of the strong influence of door position on air-exchange rate, one expects that the indoor proportion of outdoor particles would also vary with door position. We investigated this by computing the classroom to outdoor time-averaged particle number concentration ratios for conditions sorted by occupancy and door position (see Table 4.32). To produce the table, the minute-by-minute data were sorted, first by occupancy and then by door position. For each distinct configuration, we computed the average values of two environmental parameters:  $|\Delta T|$ represents the average of the absolute value of the temperature difference between the classroom and outdoors; and WS is the average wind speed (based on central station monitoring). Then, for each condition we determined the average PN concentrations in the classroom and outdoors. The parameter  $f_1$  represents the ratio of the average in the classroom to the average outdoors. Table 4.32 shows significant influence of occupancy on  $f_I$  but, surprisingly, only modest influence of door position. When a teacher is present in the classroom, the overall value of  $f_I$  is 0.51, as compared with 0.38 when the room is unoccupied. When the room is occupied and the door is open, the value of  $f_I$  is 0.53 (teacher) or 0.60 (students); when the door is closed, the corresponding  $f_I$  values are only slightly smaller, 0.51 (teacher or student).

| State                        | <b>Duration</b> (h)          | $WS (m s^{-1})$ | $ \Delta \mathbf{T} $ (°C) | $f_{I}(-)$ |  |  |
|------------------------------|------------------------------|-----------------|----------------------------|------------|--|--|
| At least one student present |                              |                 |                            |            |  |  |
| Door open                    | 2.4                          | 2.7             | 6.7                        | 0.60       |  |  |
| Door closed                  | 10.6                         | 2.7             | 9.5                        | 0.51       |  |  |
| Average                      | 13.9                         | 2.7             | 8.9                        | 0.52       |  |  |
| At least one teach           | At least one teacher present |                 |                            |            |  |  |
| Door open                    | 2.9                          | 2.7             | 7.0                        | 0.53       |  |  |
| Door closed                  | 13.1                         | 2.5             | 9.0                        | 0.51       |  |  |
| Average                      | 17.5                         | 2.5             | 8.6                        | 0.51       |  |  |
| Unoccupied                   |                              |                 |                            |            |  |  |
| Door closed <sup>b</sup>     | 44.4                         | 1.3             | 10.3                       | 0.38       |  |  |
| Average                      | 52.5                         | 1.4             | 10.7                       | 0.38       |  |  |

**Table 4.32.** Analysis of the indoor to outdoor concentration ratios at classroom site S6 in relation to occupancy status and door position.<sup>a</sup>

<sup>a</sup> Windows were closed at all times.

<sup>b</sup> For 5.7 h of the unoccupied period, the door status is unknown (but probably closed).

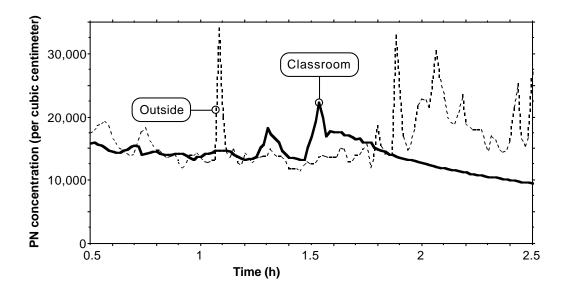
Available evidence at this site suggests that indoor sources did not contribute significantly to classroom PN concentrations. A list of activities recorded by our field researcher that might have contributed to indoor PN emissions is presented in Table 4.33. Close examination of the time-series of PN concentrations at the times noted did not reveal any noticeable coincident increase in classroom PN levels. The most prominent peak in the classroom PN concentration time series occurred early in the monitoring period, at an elapsed time, t, between 1.5 and 2 h. Figure 4.26 presents the detailed time-series of outdoor and classroom PN concentrations at this time. The classroom levels at the peak are not higher than the outdoor levels at nearby times. Taking into consideration the separation of ~ 150 m between the classroom and the outdoor monitoring site and the potential for spatial gradients in outdoor concentrations, we cannot discern whether the excursion in classroom PN concentrations was caused by emissions from an indoor source or from penetration of particles from outdoors.

Four source-characterization tests were conducted during the manipulation experiments at this site. Two involved the use of the industrial-grade vacuum cleaner that the custodian used in the classroom. A third involved use of the classroom laser printer. The fourth test entailed use of a "linen-scented" Lysol spray coincident with operating the classroom HAC units on an "air cleaning" setting. None of these tests produced increases in classroom PN concentrations that were clearly discernible against the background influence of outdoor particles.

Table 4.34 presents the results of the PN exposure analysis based on observational monitoring at site S6. At this site, we had two days (2 Dec and 8 Dec) on which monitoring results were available for the entire school day. For two additional days (4-5 Dec) monitoring was only available for portions of the day, in the afternoon. For Table 4.34, we analyzed the conditions on the two full days of monitoring and did not include data from the two partial days.

| Time (h) | Activity or observation                           | PN source? |
|----------|---|------------|
| 2.6      | Teacher sprays table with soap solution           | Ν          |
| 8.6      | Custodian enters                                  | Ν          |
| 9.5      | Custodian enters                                  | N          |
| 24.8     | Teacher uses rubbing alcohol to clean white board | N          |
| 28.3     | Custodian enters                                  | Ν          |
| 47.1     | Custodian enters                                  | N          |
| 51.5     | Custodian enters                                  | N          |

 Table 4.33. Activity notes related to potential PN emission sources at site S6.



**Figure 4.26.** Detail of particle number concentration traces measured at site S6. The time scale on the x-axis is referenced to the beginning of monitoring at 8:15 on 2 December 2008. This evidence is insufficient to conclude that the excursion in classroom concentrations at  $t \sim 1.5$  h was a consequence of indoor emissions.

For the two school days of monitoring, the cumulative time that at least one student was present in the classroom was 10.5 h or 5.2 h/d. The average number of students present in the classroom during the time when any student was present was 14.5. Cumulative classroom PN exposure summed over all students for the two days was  $963 \times 10^3$  cm<sup>-3</sup> h. Dividing this cumulative exposure by the average occupancy yields an average individual exposure of  $66 \times 10^3$  cm<sup>-3</sup> h. The average student exposure acquired while in the classroom per school day is half this value, i.e.  $33 \times 10^3$  cm<sup>-3</sup> h/d. We believe that essentially all of this exposure is attributable to particles originating from outdoors. Analogous calculations have been performed for the teacher, yielding a modestly higher daily exposure rate of  $41 \times 10^3$  cm<sup>-3</sup> h/d. The teacher's daily exposure rate is higher than the students (41 versus  $33 \times 10^3$  cm<sup>-3</sup> h/d) primarily because of the teacher's longer duration of exposure: 6.1 h/d versus 5.2 h/d. These exposures only consider time spent in the classroom.

**Table 4.34.** Exposure analysis for students and teachers at classroom site S6, based on two full school days of observational monitoring.<sup>a</sup>

| Parameter  | Students | Teacher |
|--|----------|---------|
| Hours with at least one member present                                     | 10.5     | 12.2    |
| Average occupancy when at least one present                                | 14.5     | 1       |
| Cumulative exposure $(10^3 \text{ cm}^{-3} \text{ h})$                     | 963      | 81      |
| Average individual exposure $(10^3 \text{ cm}^{-3} \text{ h})$             | 66       | 81      |
| Average individual exposure rate $(10^3 \text{ cm}^{-3} \text{ h d}^{-1})$ | 33       | 41      |

<sup>a</sup> Two ~ full school days were monitored (2 Dec, and 8 Dec; on these days monitoring was initiated at 8:15 and 8:43, respectively). In addition, on two school days monitoring commenced at 14:04 and 12:25 and continued until the end of the school day. These two partial days are not included in the exposure analysis presented here.

#### 4.3 Synthesis of the Research Findings from the Six Classroom Sites

In this subsection, we explore the aggregate study results from the six classroom sites. The presentation addresses the main themes that were discussed for each of the sites individually:

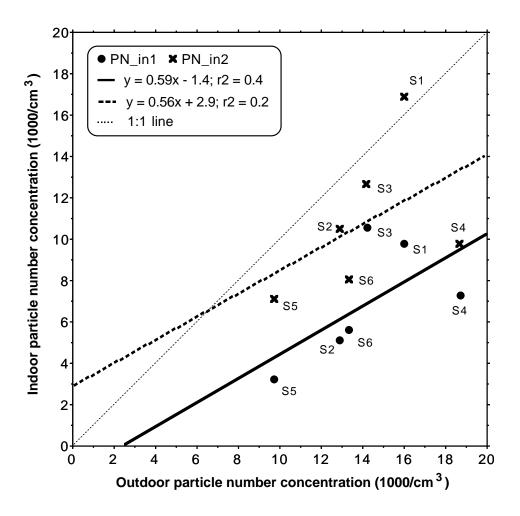
- Particle number PN concentrations
- Classroom air-exchange rates
- Influence of outdoor particles on indoor levels
- Indoor particle emission sources
- Exposure of classroom occupants to PN during observational monitoring

Table 4.1 (see §4.1) presented a summary of parameter values measured during observational monitoring at the six classroom sites. An overarching finding at the school sites is that, in terms of particle number (PN) concentrations, outdoor particles are the predominant source of indoor particles. This finding is in sharp contrast to the case in the houses we monitored, where indoor emission sources contributed strongly to indoor PN levels and especially to exposures.

We have seen that outdoor particles are attenuated in their influence on indoor PN levels. Three main effects can cause indoor PN levels to be less than outdoor levels in the absence of indoor sources: particles may be lost by deposition as air leaks into the building envelope; particles deposit on interior classroom surfaces; and particles may be removed from indoor air by means of active filtration, e.g. in a classroom's mechanical ventilation system or its recirculating thermal conditioning system. As in houses, an interesting and important question for classrooms is this: to what degree can indoor PN levels be predicted by measuring outdoor concentrations? Figure 4.27 displays a scattergram with the classroom (in1) PN concentrations as the dependent variable and the outdoor level as the independent variable. Each point represents a time-average value for the entire monitoring period. (Later in this section, we will consider the extent to which exposures are correlated with outdoor concentrations.) Separate data points are plotted for the primary indoor monitor (PN\_in1), which sampled from the classroom at each site, and for the supplemental indoor monitor (PN\_in2), which either sampled from a nearby hallway or from the school cafeteria.

The data do show the expected trend, that higher indoor PN levels are correlated with higher outdoor levels. However, the scatter about the trend lines is large, as reflected in the low correlation coefficients ( $r^2 = 0.4$  for the primary indoor site and  $r^2 = 0.2$  for the supplementary indoor site). All but one of the points lies below the 1:1 line, indicating that outdoor PN concentrations are generally higher than indoor PN levels. The one point with a slightly higher indoor concentration than outdoors was for monitoring the corridor at school site S1 and the difference between the indoor and outdoor levels was only about 6%, which lies within the range of measurement uncertainty.

Overall, the average classroom PN levels were about half of the average outdoor levels, based on the entire observational monitoring periods (see Table 4.35). The difference in the mean values  $(7.3 \times 10^3 \text{ cm}^{-3})$  is statistically significant at a very low *p* value, since it is about  $6 \times$  the standard error of either mean  $(1.2 \times 10^3 \text{ cm}^{-3})$ . Average levels measured by the supplementary indoor monitor were, on average, about halfway between the classroom and outdoor values.



**Figure 4.27.** Scattergram of time-averaged particle number concentrations measured indoors (classroom = PN\_in1; supplemental = PN\_in2) versus outdoors at six school sites. These results represent averages over the entire observational monitoring period at each site. The equations for the regression lines are labeled in the key and each point is labeled with the school site to which it corresponds. The supplemental indoor monitor was either sited in a corridor (S1, S4, S5) or in the cafeteria (S2, S3, S6) at each site. The outdoor value at S4 was adjusted from 21.3 to  $18.7 \times 10^3$  cm<sup>-3</sup> to account for the period not monitored overnight. The adjustment factor was based on the time-resolved data at all other sites.

There is considerable interest in the influence of UFP emissions from motor vehicles on levels near major roadways. The data from our study show an association between proximity to a freeway and the outdoor average PN concentration. Table 4.36 summarizes the data for school sites. Figure 4.28 presents a regression analysis incorporating data from the 13 field sites. Note that the  $r^2$  value is only 0.12 for this regression. If one removes from the analysis the data point at H5, based on the reasoning that this suburban site is qualitatively different than the other 12 urban sites, then the  $r^2$  value declines further to 0.04, indicating essentially no relationship. However, two important qualifications need to be made. First, studies of the influence of freeways on outdoor PN levels have generally found that the effect is significantly discernible only to a distance of about 0.3 km downwind. Second, as noted in §3.3 when reviewing overall findings in houses, each of these field sites was measured over a different 3+-day period.

Clearly, PN levels at any site can vary with time according to factors such as meteorological conditions. At the level of comparing one site to another, our study design does not allow us to separately consider the effects of time and space on outdoor PN levels.

**Table 4.35.** Overall summary statistics of particle number concentrations ( $\times 10^3$  cm<sup>-3</sup>) measured at the six classroom sites.

| Parameter                 | Average <sup>a</sup> | Std. Error | Range                 |
|---------------------------|----------------------|------------|-----------------------|
| Outdoor (PN_out)          | 14.2                 | 1.2        | 9.7-18.7 <sup>b</sup> |
| Classroom (PN_in1)        | 6.9                  | 1.2        | 3.2-10.5              |
| Secondary indoor (PN_in2) | 10.8                 | 1.4        | 7.1-16.9              |

<sup>a</sup> The time-weighted average is first computed for each site over the full observational monitoring period; the results for the six sites are linearly averaged to obtain the entries in this column.

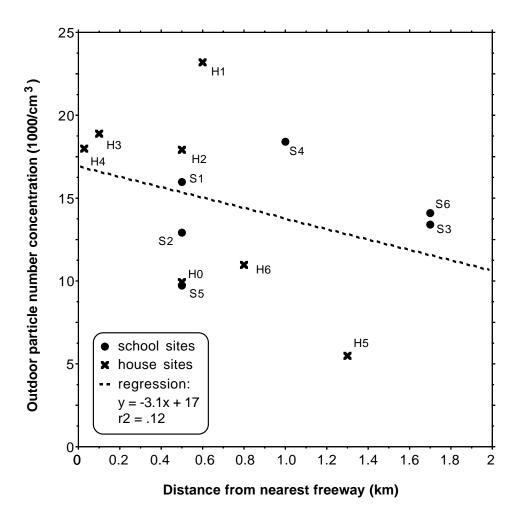
<sup>b</sup> The highest value corresponds to an estimated 24-h average concentration at S4. The adjustment factor was based on the time-resolved data at all other sites and was used to correct for the lack of observation data overnight at S4.

**Table 4.36.** Summary of average outdoor PN concentrations ( $\times 10^3$  cm<sup>-3</sup>) in relation to distance from closest freeway at the six classroom sites.

| Site       | Avg. PN_out | <b>Fwy location</b> | Note                                  |
|------------|-------------|---------------------|---------------------------------------|
| <b>S</b> 1 | 16.0        | 0.5 km E            | Second freeway is ~ 1 km NW of school |
| S2         | 12.9        | 0.5 km E            | Same campus as S1                     |
| <b>S</b> 3 | 14.2        | 1.7 km W            |                                       |
| <b>S</b> 4 | 18.7        | 1.0 km W            |                                       |
| <b>S</b> 5 | 9.7         | 0.5 km W            |                                       |
| <b>S</b> 6 | 13.4        | 1.7 km W            | Same campus as S3                     |

Air-exchange rate is expected to influence the indoor PN levels. In the classrooms, where outdoor particles are the dominant source, one expects that an increase in air-exchange rate would produce a higher proportion of indoor particles in relation to the outdoor level.

In aggregate, we made 43 determinations of the air-exchange rate at the six classroom sites. Table 4.37 summarizes these measurement results. We observed that two key factors had a strong influence on air-exchange rate: (a) whether an exterior classroom door was open or closed and (b) whether or not a mechanical ventilation system was operating. In presenting the data, we first sorted the determinations into two groups. For one group, the measurements were made with the (exterior) classroom doors closed and the mechanical ventilation system (if present) off. Except for S3, which was equipped with a ventilation system that we believe operated continuously, measurements were made at each site under both types of conditions. The arithmetic mean of the measurements under these conditions at the five sites ranged from 0.3 to 0.7 h<sup>-1</sup> (average = 0.5 h<sup>-1</sup> across all sites). The other measurement group, which applied at all six sites, was based on conditions in which either one or more exterior classroom doors was open or the mechanical ventilation system was operating. In this case, the arithmetic mean of the measurements at each site ranged from 1.9 to 4.6 h<sup>-1</sup> (average = 3.3 h<sup>-1</sup>). There is almost no overlap in the distributions of individual measurements.



**Figure 4.28.** Correlation between time-averaged outdoor PN level measured at thirteen field sites and the distance of each site from the nearest freeway. The outdoor level is based on the full period of observational monitoring at each site. The points are each labeled with a site ID.

| Table 4.57. All-exchange rates (if ) determined at six classroom's |                 |            |            |                 |             |             |
|--|-----------------|------------|------------|-----------------|-------------|-------------|
|  | Do              | ors closed | l, air off | Door(           | (s) open an | d/or air on |
| Site   | Ν               | AM         | Range      | Ν               | AM          | Range       |
| <b>S</b> 1   | 1               | 0.5        | 0.5        | 4               | 2.2         | 1.3-2.6     |
| S2   | 2               | 0.3        | 0.3-0.4    | 3               | 3.3         | 2.8-3.9     |
| <b>S</b> 3   | 0               |            |            | 4               | 4.6         | 4.3-4.9     |
| S4   | 6               | 0.3        | 0.1-0.7    | 2               | 3.9         | 2.2-5.5     |
| S5   | 2               | 0.7        | 0.2-1.2    | 8               | 1.9         | 1.1-4.2     |
| S6   | 7               | 0.6        | 0.4-0.8    | 4               | 4.0         | 1.7-5.9     |
| Avg.   | 18 <sup>b</sup> | 0.5        | 0.1-1.2    | 25 <sup>b</sup> | 3.3         | 1.1-5.9     |
| a <b>.</b> .   |                 | 0.1        |            |                 |             |             |

**Table 4.37.** Air-exchange rates (h<sup>-1</sup>) determined at six classroom sites. <sup>a</sup>

 $^{a}$  N = number of determinations; AM = arithmetic mean of individual measurements.

<sup>b</sup> Total number of AER determinations at the six sites for the conditions indicated.

For classrooms, occupancy is a key factor for evaluating and assessing indoor PN concentrations. Even though indoor sources do not appear to contribute substantially to indoor PN concentrations at the sites studied, occupancy still plays an important role influencing indoor PN levels. The key reason is because of ventilation conditions. Occupancy is associated with higher ventilation rates, often because of open doors and windows, which increases the indoor proportion of outdoor particles. A second and almost obvious key factor is that occupancy is an essential component of exposure. The indoor PN levels that matter are those that occur when students or teachers are present in the classrooms.

Table 4.38 presents summary statistics for classroom PN levels at the six study sites, sorted by occupancy. Presented are the lognormal parameters of the minute-by-minute concentration distributions, sorted by occupancy status. The condition with "students present" is a subset of the condition with "teacher present," since a teacher is always present when there are students in the classroom. The "unoccupied" condition is the complement of the case with "teacher present" such that the sum of the prevalence percentages for these two cases total 100%.

|                  | Stud | ents pre | esent | Tea  | cher pre | esent | U   | noccupi | ed  |
|------------------|------|----------|-------|------|----------|-------|-----|---------|-----|
| Site             | GM   | GSD      | %     | GM   | GSD      | %     | GM  | GSD     | %   |
| <b>S</b> 1       | 13.8 | 1.71     | 20%   | 14.0 | 1.78     | 36%   | 5.1 | 1.73    | 64% |
| S2               | 5.7  | 2.77     | 19%   | 6.4  | 2.75     | 28%   | 2.1 | 2.30    | 72% |
| <b>S</b> 3       | 12.1 | 1.73     | 18%   | 13.3 | 1.67     | 28%   | 7.7 | 1.63    | 72% |
| S4               | 12.5 | 1.63     | 20%   | 9.5  | 2.48     | 33%   | 3.7 | 1.86    | 67% |
| S5               | 3.8  | 2.19     | 16%   | 3.8  | 1.98     | 31%   | 2.1 | 1.64    | 69% |
| S6               | 4.6  | 2.12     | 20%   | 4.9  | 2.05     | 25%   | 5.0 | 1.49    | 75% |
| Avg <sup>b</sup> | 8.8  | 2.03     | 19%   | 8.7  | 2.12     | 30%   | 4.3 | 1.78    | 70% |

**Table 4.38.** Summary statistics of the lognormal parameters for classroom PN concentrations
 measured at the six school sites, with the data first sorted according to occupancy status.<sup>a</sup>

<sup>a</sup> GM = geometric mean, with units of  $10^3$  cm<sup>-3</sup>; GSD = geometric standard deviation; % = percentage of the observational monitoring period when the indicated state applied.

<sup>b</sup> This row presents the average of the indicated parameters across the six classroom sites.

Table 4.38 shows that the conditions with students present and with a teacher present are similar to one another at most sites and in terms of the overall average. A key finding is that the typical PN concentration when a classroom is occupied is 2× the value when it is unoccupied, as reflected in the ratio of the average geometric means  $(8.8 \times 10^3 \text{ cm}^{-3} \text{ when students are present})$ versus  $4.3 \times 10^3$  cm<sup>-3</sup> when unoccupied). Three factors contribute to this difference. The most important factor is that outdoor PN concentrations are higher when school is occupied than when it is not. The second important factor is that the indoor proportion of outdoor particles in the classroom is higher when the classroom is occupied (owing to higher ventilation rates) than when it is vacant. The third factor, of relatively minor significance, is the contribution of indoor emission sources, which correlate with occupancy, to the classroom PN concentration.

Table 4.39 explores the second factor, reporting the indoor proportion of outdoor particles at each of the six sites, sorted according to occupancy status of the classroom. Across the six sites, the average condition with the classroom occupied,  $f_1 = 0.57$ , is about 40% higher than the average when the classroom is vacant. At each of the six sites, the value of  $f_1$  is higher for both occupied categories than for the unoccupied case.

| Site       | Students | Teacher | Unoccupied | Details    |
|------------|----------|---------|------------|------------|
| <b>S</b> 1 | 0.58     | 0.64    | 0.54       | Table 4.5  |
| S2         | 0.48     | 0.50    | 0.32       | Table 4.10 |
| <b>S</b> 3 | 0.76     | 0.74    | 0.71       | Table 4.15 |
| S4         | 0.55     | 0.52    | 0.33       | Table 4.22 |
| <b>S</b> 5 | 0.51     | 0.49    | 0.24       | Table 4.27 |
| S6         | 0.52     | 0.51    | 0.38       | Table 4.32 |
| Avg        | 0.57     | 0.57    | 0.42       |            |

**Table 4.39.** The indoor proportion of outdoor particles, evaluated at the six classroom sites, according to occupancy status of the classroom.<sup>a</sup>

<sup>a</sup> Each entry represents the time-average classroom concentration divided by the time-average outdoor concentration, for periods sorted according to occupancy (one or more students present; the teacher present; or unoccupied). Data in which there is a clear influence of indoor sources on indoor PN levels are excluded from the analysis.

Table 4.40 investigates how the value of  $f_I$  varies with configuration condition of the classrooms during times when students occupy the room. All times when students are in the room are sorted into two groups, taking into consideration the position of exterior door(s) and the operation of a mechanical ventilation system, if present. For the "doors closed; air off" case, no ventilation system is operating and the exterior doors are all closed. Across the six sites, this configuration occurs an average of 23% of the time (range: 0-76%). For the four sites where the proportion of time exceeds zero, the average indoor proportion of outdoor particles under this configuration is  $f_I = 0.38$  (range: 0.16-0.51). When one or more exterior doors is open and/or the mechanical ventilation system is on, the air-exchange rate is much higher, and so too is the indoor proportion of outdoor particles. For this more ventilated condition, the average is  $f_I = 0.60$  (range: 0.51-0.76). The importance of classroom configuration in determining the outcome is seen in that there is almost no overlap in the range of  $f_I$  values between the two categories.

The broad range of ventilation conditions reflected in different classroom configurations presents an opportunity to evaluate the performance of a simple model for predicting time averaged indoor PN concentrations that result from particles in outdoor air. The model representation is a simplified version of equation (4.1):

$$f_1 \sim \frac{a}{a+k} \tag{4.2}$$

In this representation, we are making two key assumptions: (i) that particles penetrate effectively from outdoors along with ventilation air ( $P \sim 1$ ); (ii) that there are two dominant removal mechanisms for particles indoors, either ventilation (proportional to the air-exchange rate, a), or deposition (proportional to the rate constant, k). In applying the model here, we will further assume that the values of k are approximately constant from one site to another.

We applied the data in Table 4.40 to simultaneously obtain a best estimate of k and also to test the performance of this simple model. For each of the two configurations and each of the six sites, we computed a "model predicted" value of  $f_1$  and compared it with the measured value. We adjusted the value of k, assumed to be the same at each site, to obtain the best overall agreement between model and measurement, using a least-squares metric. Figure 4.29 presents the result of this analysis, plotting the measured against the modeled values of  $f_1$  for each

configuration and classroom site. The best-estimate value of k is 1.4 h<sup>-1</sup>, which is within the range of expectations. With this value of k, the agreement between model and measurement is good, but not great. The average absolute value of the deviation between model and measurement is 0.12, which is 23% of the average measured value of  $f_1$ .

**Table 4.40.** Room configuration during student occupancy, average air-exchange rate by configuration, and corresponding indoor proportion of outdoor particles ( $f_1$ ) for two groups of configuration conditions at the classroom sites. <sup>a</sup>

| Doors closed; air off |                       |               | Door(s) open             | and/or air o          | n             |           |
|-----------------------|-----------------------|---------------|--------------------------|-----------------------|---------------|-----------|
| Site                  | Time (%) <sup>b</sup> | $AER(h^{-1})$ | $f_{I}()$                | Time (%) <sup>b</sup> | $AER(h^{-1})$ | $f_{I}()$ |
| <b>S</b> 1            | 3%                    | 0.5           | 0.39                     | 96%                   | 2.2           | 0.59      |
| S2                    | 35%                   | 0.4           | 0.16                     | 53%                   | 3.3           | 0.54      |
| <b>S</b> 3            | 0%                    | —             | —                        | 100%                  | 4.6           | 0.76      |
| S4                    | 25%                   | 0.3           | 0.46                     | 68%                   | 3.9           | 0.59      |
| S5                    | 0%                    |               | —                        | 100%                  | 1.9           | 0.51      |
| S6                    | 76%                   | 0.6           | 0.51                     | 17%                   | 4.0           | 0.60      |
| Avg.                  | 23%                   | <b>0.45</b> ° | <b>0.38</b> <sup>c</sup> | 72%                   | 3.1           | 0.60      |

<sup>a</sup> For all cases considered in this table, students were present in the classroom. The "doors closed; air off" condition implies that all exterior doors to the classroom were closed and that the mechanical ventilation system, if present, was off. The "door(s) open and/or air on" is the complementary case.

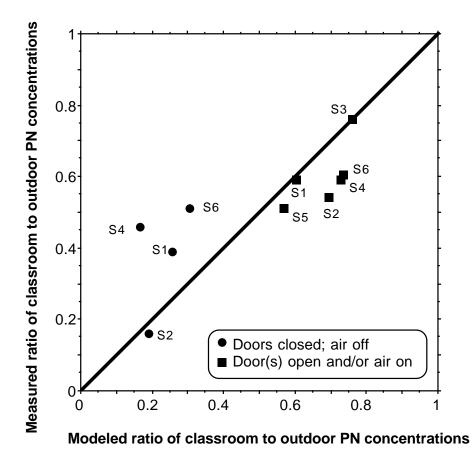
<sup>b</sup> These columns report the proportion of all minutes during student occupancy that the indicated ventilation condition prevailed. Minutes during which a change of state occurred are excluded, which is why the proportions don't sum to 100% at all sites. In addition, for 5.4% of the time at S2, the door position sensor was not recording; this unknown state is also excluded from the table entries.

<sup>c</sup> For the configuration of "doors closed; air off", S3 and S5 are excluded from the average AER and  $f_I$  calculations.

Based on all of the evidence gathered, we infer that indoor emission sources contributed little to the particle levels observed at each of the classroom sites. At the seven house sites, we identified 59 peak events resulting from episodic indoor emissions. At the six classroom sites, only six events were identified, as summarized in Table 4.41. Four of these six events were quantitatively characterized. Their overall contributions to the exposure of classroom occupants to PN concentrations were relatively small. The total cumulative exposure increment across all four characterized events was ~  $770 \times 10^3$  cm<sup>-3</sup> h for students and ~  $40 \times 10^3$  cm<sup>-3</sup> h for teachers. The result is higher for students than for teachers because of the degree of occupancy, with ~ 20 students typically in a classroom compared with one primary teacher. To put these numbers in perspective, note that the aggregate total exposure in classrooms during the 18 observational monitoring days at all six sites was  $17,200 \times 10^3$  cm<sup>-3</sup> h for students and  $1,500 \times 10^3$  cm<sup>-3</sup> h for teachers. Thus, the indoor episodic emission events contributed only 4% and 3%, respectively, to the cumulative exposure of students and teachers during observational monitoring.

In total, we conducted nine source-characterization manipulation experiments at the six classroom sites. As summarized in Table 4.42, only three of these experiments generated a clear indoor PN concentration signal: mopping with a pine-oil based cleaner, using a candle, and using a classroom heater. We refer the reader to the other subsections of the report (§4.2.1 and §4.2.3)

for additional information about these sources. We tested the classroom laser printer at three sites and did not detect any significant increase in classroom PN levels from these uses.



**Figure 4.29.** Regression analysis of measured indoor proportion of outdoor particles  $(f_1)$  against the modeled value  $(f_1 \sim a/(k+a))$  considering each classroom site and two room configurations. The best fit value of k = 1.4 h<sup>-1</sup> is used in the comparison, with that value determined by minimizing the sum of the squared differences between modeled and measured  $f_1$  values for each of the ten points.

The final subject considered in this subsection is exposure to particle number concentrations in the classrooms, based on observational monitoring at the six sites. Table 4.43 presents a summary of the daily average per-person exposure rate at each of the six sites, separately assessed for students and for teachers. Figure 4.30 displays this information graphically.

There are a few key points to make from the results of the analysis aggregated across all classroom sites. First, the range of exposures is moderate. For students, the average per person exposure rate is  $50 \times 10^3$  cm<sup>-3</sup> h/d with a relative standard deviation of 45%. The range spans a factor of four, from  $19 \times 10^3$  cm<sup>-3</sup> h/d to  $80 \times 10^3$  cm<sup>-3</sup> h/d. Relatively speaking, the results are similar for teachers, with a mean exposure rate of  $80 \times 10^3$  cm<sup>-3</sup> h/d, a relative standard deviation of 50%, and a range of ~ 4× (from  $36 \times 10^3$  cm<sup>-3</sup> h/d to  $141 \times 10^3$  cm<sup>-3</sup> h/d). Second, the daily exposure rates for teachers are substantially higher than for students (80 versus  $50 \times 10^3$  cm<sup>-3</sup>

h/d). However, the difference is almost entirely a consequence of the greater number of hours per day spent by teachers in the classroom than by students (7.1 versus 4.6 h/d on average). We stress that the analysis here only considers exposure that occurs in the monitored classrooms. Exposures that occur elsewhere on the school grounds are not included in the assessment.

|            |   | <b>Exposure contribution</b> <sup>4</sup> |         |  |
|------------|---|---|---------|--|
| Site       | Event   | Students                                  | Teacher |  |
| <b>S</b> 1 | Cooking pancakes on an electric griddle             | 598                                       | 27      |  |
| <b>S</b> 1 | Use of pine-scented cleaner elsewhere in building   | na  | na      |  |
| <b>S</b> 3 | Candle to celebrate student birthday                | 105                                       | 5       |  |
| <b>S</b> 3 | Heater  | 0   | 6       |  |
| S4         | Unidentified source; particles entered from hallway | 64  | 4       |  |
| S5         | Heater  | small                                     | small   |  |

**Table 4.41.** Identified peaks in classroom PN concentrations resulting from indoor episodic

 emissions during observational monitoring at the six sites.

<sup>a</sup> The contribution to exposure is in units of  $10^3$  cm<sup>-3</sup> h, summed over all exposed individuals present in the classroom during the event.

**Table 4.42.** Summary of results from the source-characterization manipulation experiments in classroom sites.

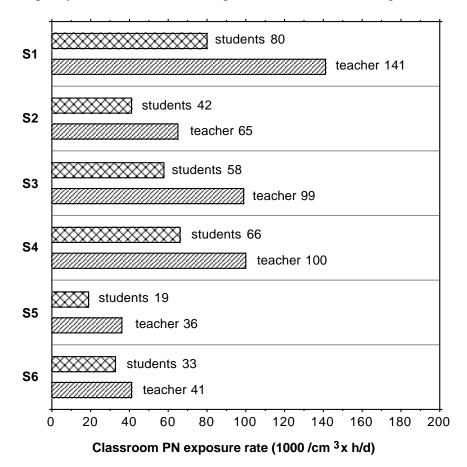
| Site       | Activity or product tested              | PN source? |
|------------|---|------------|
| <b>S</b> 1 | Mopping with pine-oil based cleaner     | Y          |
| S2         | Laser printer                           | Ν          |
| <b>S</b> 3 | Candle                                  | Y          |
| <b>S</b> 3 | Heater                                  | Y          |
| S4         | Laser printer                           | Ν          |
| S5         | Heater                                  | Ν          |
| <b>S</b> 6 | Vacuum cleaner                          | Ν          |
| <b>S</b> 6 | Laser printer                           | Ν          |
| S6         | Scented spray plus ionizing air cleaner | N          |

A third important point emerges from comparing the results in classrooms with the results in houses, as summarized in §3.1 and §3.3. In their classroom, an average student acquires  $50 \times 10^3$  cm<sup>-3</sup> h/d of PN exposure. For the seven children included as subjects in the seven houses we studied, the average in-home PN exposure rate was about  $6 \times$  higher at  $316 \times 10^3$  cm<sup>-3</sup> h/d (see Table 3.4). A large part of the difference occurs because of the different extents of occupancy. The average time spent indoors at home for our seven children subjects was 16.9 h/d, almost 4 × as long as the average time spent in the classroom. In addition, even allowing for the much lower overnight concentrations, the average PN concentrations in the houses we monitored were higher during occupancy than the corresponding values in classrooms. We note further that on an annual basis, the classroom exposure would only apply for roughly 180 school days per year, whereas the residential exposure can be expected to apply for a large majority of days in a typical case. Consequently, assuming the evidence from our study is approximately true in general, the accumulated PN exposure in a child's home might well be about an order of magnitude higher than the accumulated exposure in the classroom at school on an annual average basis.

|            |                       | Stud     | lents    | Tea      | cher     |
|------------|-----------------------|----------|----------|----------|----------|
| Site       | <i>N</i> ( <b>d</b> ) | Exposure | Duration | Exposure | Duration |
| S1         | 4                     | 80       | 4.7      | 141      | 8.6      |
| S2         | 3                     | 42       | 4.6      | 65       | 6.7      |
| <b>S</b> 3 | 3                     | 58       | 4.3      | 99       | 6.0      |
| S4         | 3                     | 66       | 4.8      | 100      | 8.1      |
| S5         | 3                     | 19       | 3.8      | 36       | 7.3      |
| <b>S</b> 6 | 2                     | 33       | 5.3      | 41       | 6.1      |
| Avg.       | 3                     | 50       | 4.6      | 80       | 7.1      |

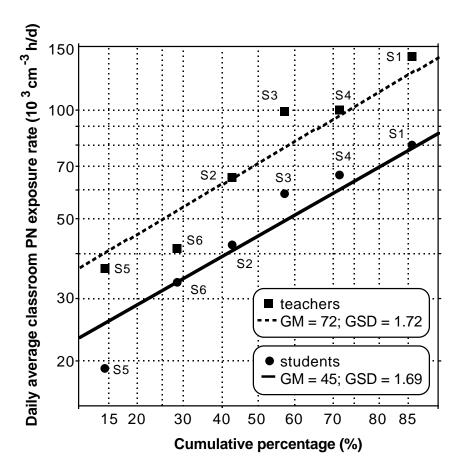
 Table 4.43. Classroom PN exposure rates for students and teachers.

<sup>a</sup> N = Number of days of observational exposure determination conducted at each site; exposures are reported in units of  $10^3$  cm<sup>-3</sup> h/d; and duration is the average number of hours per day of occupancy of the classroom during observational monitoring.



**Figure 4.30.** Average PN exposure occurring in classrooms per student and for the classroom teacher for each of the six classroom sites studied. The length of the bars in each case only considers exposure that occurs within the classroom on a per-school day basis and can be understood as the product of the average indoor PN concentration while in the classroom (1000/cm<sup>3</sup>) multiplied by the daily average time spent in the classroom (h/d).

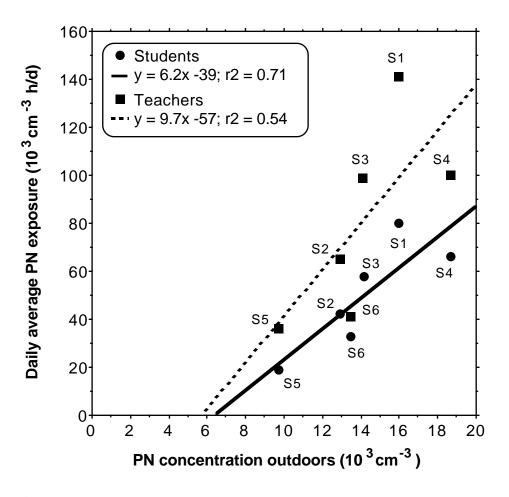
Figure 4.31 presents the distribution of daily classroom PN exposures for teachers and for students. The distributions for each group conform reasonably well to lognormals (as reflected by the degree to which the data points follow the straight lines).



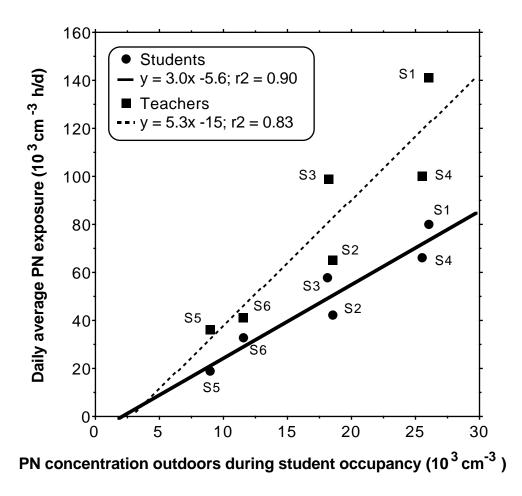
**Figure 4.31.** Cumulative distribution functions of classroom PN exposure rates for teachers (squares) and for students (circles). The points are labeled with the classroom site. The straight lines correspond to lognormal distributions. The parameter values (GM, GSD) are computed directly from the empirical data.

To conclude this section, we consider an important issue in exposure assessment. To what extent can classroom exposures to ultrafine particles (as measured by particle number concentration) be predicted based on outdoor concentration measurements? Figures 4.32 and 4.33 explore the answer to this question. In Figure 4.32, we have plotted the daily average exposure rate in each classroom site, separately for the teacher and for students, against the average outdoor PN concentration measured over the entire observational monitoring period at each site. The results exhibit a trend in the expected direction, although the regression lines do not pass near the origin, as one might expect. The correlation coefficients are fair to good. Since the classroom is only occupied for a portion of each day, it makes sense to consider the extent to which the outdoor concentrations averaged during occupancy are predictive of exposures. That analysis, presented in Figure 4.33, shows a much more favorable result, with good  $r^2$  values of

0.90 (for students) and 0.83 (teachers) and with intercepts that represent small fractions of the average exposure rates.



**Figure 4.32.** Scatterplot and regressions of measured classroom exposure rates against the average on-site outdoor PN concentration measurements. The outdoor concentrations represent the average over the entire period of observational monitoring.



**Figure 4.33.** Scatterplot and regressions of measured classroom exposure rates against the average on-site outdoor PN concentration measurements. The outdoor concentrations represent the averages only when students are present in the classroom.

#### 5. SUMMARY AND CONCLUSIONS

In this project, we measured particle number concentrations and associated factors in seven houses and six classrooms in the East Bay of the San Francisco Bay Area. The data were interpreted to characterize the important factors that affect particle number concentrations indoors. We also assessed the exposures of building occupants to particles at these sites and apportioned those exposures to major source classes: particles originating outdoors, particles emitted from episodic indoor sources, and particles emitted from continuous indoor sources.

Particle number concentration was measured using a recently developed water-based condensation particle counter (WCPC). The instrument continuously measures the total number concentration of airborne particles whose diameter is larger than about 6 nm. We recorded data with a one-minute time resolution. Although the condensation particle counter is a mature technology, traditional instruments used alcohol, such as butanol, as a working fluid. Toxicity and odor concerns with alcohols make these instruments difficult to use for monitoring ordinary occupied spaces. Consequently, relatively few data have been reported on particle number concentrations indoors. The advent of the WCPC, which uses ordinary water as the working fluid in place of alcohol, facilitates sampling in indoor environments under conditions of normal occupancy, an important aspect of the present research project.

Airborne particles vary widely in size, and size is an important particle attribute. Among other factors, chemical composition, regional lung deposition, and environmental persistence are all strongly influenced by particle size. Ultrafine particles are commonly considered to be those particles with a diameter smaller than 100 nm. In general, ultrafine particles contribute little to total particle mass concentrations and yet they dominate the airborne number concentration. Consider, for example, a typical condition in urban air with a fine-particle mass concentration  $(PM_{2.5})$  of 10 µg m<sup>-3</sup> and a particle number concentration of 10<sup>4</sup> cm<sup>-3</sup>. If the number concentration were all represented by ultrafine particles with a 30-nm diameter and a density of 1 g cm<sup>-3</sup>, their contribution to the total particle mass would be only 0.1  $\mu$ g/m<sup>3</sup>, i.e., just 1% of the total. Increases or decreases in the particle number concentration by a factor of two could not be detected by a mass-based measurement against the ordinary variability in particle mass concentration. Conversely, if all of the PM2.5 mass were in particles of 300 nm diameter, then the corresponding number concentration of these particles would be 700 cm<sup>-3</sup>, i.e., just 7% of the total number concentration. The point of these illustrative calculations is to demonstrate that ultrafine particle number concentration and fine particle mass concentration represent two almost independent attributes of air quality. Concentrations, exposures, and health consequences of ultrafine particles need to be studied separately from PM<sub>2.5</sub>. Particle number concentrations, as measured with a condensation particle counter, are a good approximation for ultrafine particle number concentrations because the number concentration of atmospheric particles is generally dominated by the ultrafine mode.

In this project, we measured particle number (PN) concentration inside and outside of seven houses and six classrooms. Because people spend most of their time indoors, concentrations in indoor air have a strong influence on human exposure. If there are no indoor emission sources, then indoor concentrations can still differ from outdoor levels owing to removal processes that occur indoors, such as particle deposition onto room surfaces and particle removal in active filtration systems. The ratio of indoor particles of outdoor origin to outdoor particles, a parameter that we have labeled  $f_I$  in this report, varies from one building to another and can also vary with time within any given building. Characterizing the  $f_I$  parameter for any

pollutant is an important aspect of understanding the relationship between monitored or modeled ambient air pollution and human exposure.

In addition, ultrafine particles can be emitted directly into indoor air. To the extent that indoor PN levels are influenced by indoor sources, then outdoor PN concentration measurements will be poorly predictive of the exposures that occur indoors. Whether one is interested in total human exposure to ultrafine particles or only that portion of exposure attributable to outdoor sources, it is important to differentiate the influence of indoor and outdoor sources on indoor PN levels.

We conducted field monitoring and experimentation at the thirteen sites over a span of 16 months, beginning in November 2007 and ending in February 2009. The duration of the main investigation at each site was about one week and was divided into two phases: observational monitoring and manipulation experiments. During observational monitoring, typically three days in duration, we recorded highly time-resolved data on indoor and outdoor PN concentrations and many cofactors with the house or classroom in normal use. In the second phase, we conducted manipulation experiments to acquire additional information on key factors that influence indoor PN levels: air-exchange rates, penetration and persistence of outdoor particles ( $f_1$ ), and emission of particles from indoor sources.

Most of the monitoring equipment was assembled into three packages, two primary units and one supplementary unit. The primary units contained a WCPC and monitors for ozone, nitric oxide, carbon dioxide, carbon monoxide, temperature, and relative humidity. One primary unit was deployed in a main location indoors and the second was deployed outdoors on the building grounds. The supplementary unit contained a third WCPC and was deployed to collect complementary indoor PN concentration data at the field site. Additional information was collected using portable temperature and proximity sensors with integral data loggers. The temperature sensors monitored the use of devices that might be PN emission sources, such as cooking appliances, and also monitored the use of a heater or air conditioner. The proximity sensors monitored door and window openings. Information was also acquired through the use of questionnaires (in houses) and direct observation (in schools). This information focused on timeresolved occupancy status of the buildings, on the use of possible emission sources, and on the building configuration (door and window position) or operation (e.g., thermal conditioning system) that could affect PN concentrations and fates.

The thirteen field sites were not selected to be statistically representative of any specific population of buildings. Rather, their selection combines the goal of meeting certain specific criteria along with ease of access and the cooperation of occupants. Overall, the aim was to study environments where the indoor PN levels might be higher than average, but within a normal range. Selection factors included proximity to major roadways and also the presence and indoor use of suspected UFP emission sources.

A key strength of this study is the simultaneous acquisition of particle number concentration data and of related information in multiple buildings over multiple days of normal occupancy. This information permitted us to go beyond simply reporting the PN levels inside and outside of the houses and classrooms monitored. We were able to interpret the data so that we could apportion the indoor particles among broad source categories: those that originated outdoors, those that originated from persistent indoor sources (unvented natural-gas pilot lights), and those that originated from indoor release events such as cooking. We were able to quantify the emissions source strength of the indoor release events and the rate of decay of PN concentration owing to processes such as ventilation and deposition after the events. The data also permitted us to not only characterize the indoor PN concentrations and their causes, but also to characterize the exposure of building occupants. This last contribution is particularly significant as exposures are an important link in the causal chain from sources to adverse health effects.

The most important limitations of the study are associated with the relatively small number of sites sampled over a relatively short period per site. Even though limited in scope, the evidence in this study indicates that indoor PN concentrations and exposures can vary markedly with time and from one building to another. This study was conducted in only one geographic area in California and each building was monitored in only one season. Different regions of California have different outdoor air pollution levels, different climatic conditions, different building stocks, and different demographics. Each of these features could certainly influence the indoor PN levels and the resulting exposures. One should be very cautious in extrapolating from the particular results obtained in this research to more general truths about ultrafine particle concentrations and resulting exposures in schoolrooms and homes throughout California.

Nevertheless, the field program produced a large quantity of data. The analysis of these data has provided insights into PN concentrations in houses and schools, the exposures that result from those concentrations, and the factors that influence both concentrations and exposures. In the following paragraphs, we emphasize some key findings. We encourage the reader who is interested in more information about the overall results to read §3.3 of the report on houses and §4.3 on schools. We stress that while we are certain that these findings are true for the buildings monitored in this study, their validity more generally remains to be verified.

# Key finding 1: Particle levels in classrooms and in houses are much higher when occupied than when vacant.

Figure 5.1 displays the arithmetic average PN concentrations inside and outside houses, sorted according to occupancy when one of two conditions is met: (a) all occupants are at home awake (left frame); or (b) all occupants are away from home. Consider the indoor concentrations. At each of the seven sites, the concentration for "awake at home" is higher than the concentration for "away from home" by a substantial margin. For "awake at home," the arithmetic average value across the seven households is  $33 \times 10^3$  cm<sup>-3</sup> (range: 6-81 × 10<sup>3</sup> cm<sup>-3</sup>). For "away from home," the arithmetic average is  $9 \times 10^3$  cm<sup>-3</sup> (range: 2-14 × 10<sup>3</sup> cm<sup>-3</sup>). The ratio of the "awake at home" average concentration to that for "away from home" is consistently large compared to 1.0, ranging from 1.7 at H2 to 8.2 at H1.

Figure 5.2 presents similar information for classrooms. Here, again focusing on the indoor data, we can compare the average indoor concentration with the classroom occupied by one or more students with the case when the classroom is vacant. When students are in the classroom, the arithmetic average across all six sites is  $11 \times 10^3$  cm<sup>-3</sup> (range:  $5 \cdot 17 \times 10^3$  cm<sup>-3</sup>); however, when the classrooms are unoccupied, the arithmetic average is  $5 \times 10^3$  cm<sup>-3</sup> (range:  $2 \cdot 9 \times 10^3$  cm<sup>-3</sup>). The effect of occupancy is not as pronounced as in houses, but it is consistent across all school sites. At only one site, S6, are the indoor concentrations comparable between occupied and unoccupied conditions.

The underlying reasons for this finding are different in the two categories of environments. In houses, indoor emission sources are important contributors to indoor PN concentrations. The more important emission sources are episodic and the releases correlate with occupancy. When people are awake at home, emissions from indoor sources cause indoor PN levels to rise. When the house is unoccupied, indoor emissions are much less important. Consequently, indoor PN levels correlate with occupancy.

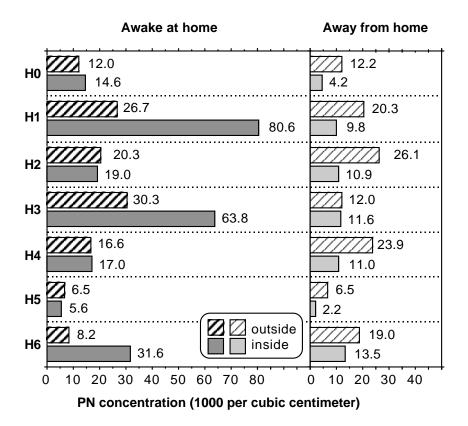
Two different key factors are at play in classrooms. First, classrooms are occupied during the daytime and are vacant at night. In our study, outdoor PN concentrations exhibited a diurnal pattern with higher concentrations occurring during the day and lower levels overnight. Thus, outdoor PN levels correlate with classroom occupancy and because outdoor particles are the main source of indoor particles, indoor levels also correlate with occupancy. The second factor amplifies this effect. Classroom ventilation rates tend to be higher when classrooms are occupied than when they are vacant. Higher ventilation rates produce higher concentrations of indoor particles when outdoor air is the dominant source. Together, these two factors account for this key finding in classrooms.

The importance of this finding is tied to the fundamental concern about potential adverse health effects of exposure to ultrafine particles. To the extent that there is any health risk, it is related to human exposure to the ultrafine particles. Exposure results from the simultaneous presence of people and particles in a given microenvironment. In characterizing houses and classrooms as sites of potential exposure, it is important to take full account of the higher concentrations occurring when people are present than when people are not. As a specific example, daily average PN concentrations measured in schools or in houses would not be a good indicator of the average exposure concentration encountered by occupants of these environments.

# *Key finding 2. Indoor emission sources are important contributors to PN levels in houses, but not in classrooms.*

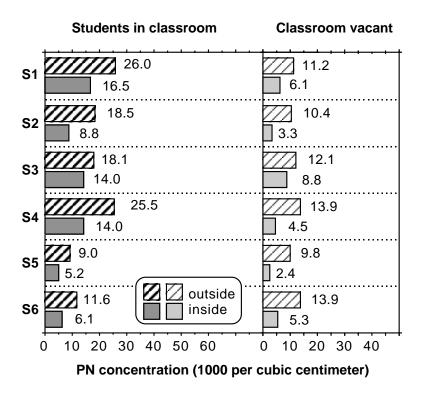
In each of the seven houses studied, we found that indoor sources contributed substantially to indoor PN levels. Specific sources varied among houses, as did the extent of their influence. Cooking was a consistently important source and emissions were detected from both stoves (ranges) and ovens, whether gas-fired or electric. Continuous pilot lights on gas cooking appliances were significant sources at two of the seven sites. Other sources that played a significant role at some houses included the toaster, the furnace, and candles.

The importance of indoor emission sources can be seen from comparing indoor and outdoor levels during occupancy at the house sites (Figure 5.1, left frame; see also Table 3.2). At three sites — H1, H3, and H6 — indoor PN levels are much higher than the corresponding outdoor levels. At the other four sites, the respective indoor and outdoor levels are similar. Overall, when the houses are occupied, the average of the indoor levels across the seven houses  $(33.2 \times 10^3 \text{ cm}^{-3})$  is almost twice as high as the corresponding outdoor average  $(17.2 \times 10^3 \text{ cm}^{-3})$ . When houses are unoccupied, the average of the indoor levels  $(9.0 \times 10^3 \text{ cm}^{-3})$  declines to a value much lower than the outdoor level, which remains about the same  $(17.1 \times 10^3 \text{ cm}^{-3})$ . For each of the seven house sites, when the house is occupied, the average indoor PN concentration is higher than the average outdoor level, and the reverse is true when the house is unoccupied.



**Figure 5.1.** Average particle number concentrations measured inside and outside of the seven house sites during observational monitoring, sorted according to occupancy status of the house. The data reported here apply when all occupants of the house are in the status noted: awake at home (left frame) or away from home (right frame). The indoor data are based on the primary indoor monitor, In1.

For schools, we see a different pattern. Whether the classroom is occupied or not, we see that the average indoor concentrations are consistently lower than the corresponding outdoor concentrations. The difference between the indoor and outdoor conditions is proportionately smaller when the classroom is occupied than when it is not. Averaging across all six classrooms, when the classroom is occupied, the average indoor concentration,  $10.8 \times 10^3$  cm<sup>-3</sup>, is 60% of the average outdoor level ( $18.1 \times 10^3$  cm<sup>-3</sup>). (See also Table 4.2.) When the classrooms are vacant, the average indoor concentration,  $5.1 \times 10^3$  cm<sup>-3</sup>, is a smaller proportion, roughly 40%, of the corresponding average outdoor level. The main cause of the higher indoor/outdoor ratio during occupancy is the higher average ventilation rate when the classroom is occupied. Episodic indoor emissions were detected at several of the school sites, but their contributions to the time-average indoor PN concentration were consistently small.



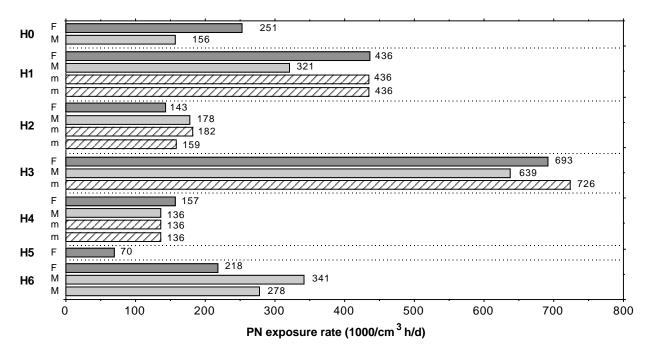
**Figure 5.2.** Average particle number concentrations measured inside and outside of the six classrooms during observational monitoring, sorted according to occupancy status of the classroom. The data reported here apply when one or more students are occupying the classroom (left frame) or when the classroom is vacant (right frame). Note that the outside average PN concentration for site S4 includes an estimate for the overnight conditions (approximately midnight to 7:00) when monitoring was suspended.

# Key finding 3. Daily average PN exposures per person are much higher in houses than in schools.

Exposure to an air pollutant can be quantified as the product of two terms: the average concentration encountered multiplied by the duration of the encounter. We have estimated PN exposures that occurred in their home for each of the 21 occupants of the seven house sites we studied. We have also estimated average PN exposure rates in the classroom for students and for teachers at each of the six classroom sites. Figures 5.3 and 5.4 present summaries of the results expressed in terms of daily exposure rate in the respective microenvironments. A brief restatement explaining the unusual units is merited. Exposure rates are expressed in units of 1000 cm<sup>-3</sup> h/d. A value of 100 on this scale means that the product of the average PN concentration (in 1000s of particles per cm<sup>3</sup>) during exposure multiplied by the duration of exposure in hours per day equals 100. So, for example, 4 h/d of exposure in the given microenvironment multiplied by an average PN concentration of 25,000 cm<sup>-3</sup> would produce an exposure rate of  $100 \times 1000$  cm<sup>-3</sup> h/d.

As shown in Figure 5.3, the average PN exposure rate for the 21 residents of our seven house sites spans about an order of magnitude, from 70 to  $726 \times 10^3$  cm<sup>-3</sup> h/d, with an average of  $297 \times 10^3$  cm<sup>-3</sup> h/d. For the seven children, the range is  $137-726 \times 10^3$  cm<sup>-3</sup> h/d with an average of  $316 \times 10^3$  cm<sup>-3</sup> h/d. Most of this exposure occurs while the occupants are awake at home. For

the 21 occupants, the average "awake at home" exposure rate was  $233 \times 10^3$  cm<sup>-3</sup> h/d, 79% of the total. For the seven children, the average exposure rate while awake was the same,  $234 \times 10^3$  cm<sup>-3</sup> h/d, which was 74% of the average total at-home exposure rate.

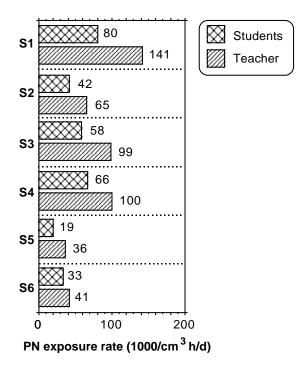


**Figure 5.3.** Average daily exposure rate to particle number concentration inside one's home for the 21 residents of the seven house sites investigated in this project. The exposure rate can be understood as the product of the average concentration indoors when that person is home (in 1000 particles per cm<sup>3</sup>) multiplied by the average daily time spent indoors at home. The left axis indicates the house site and the gender and age class of the individual (F = female adult, M = male adult, m = male child). Bars are labeled with the numerical value of the exposure rate plotted.

The exposure rates in classrooms are typically smaller (Figure 5.4). For students, the average across the six sites is in the range  $19-80 \times 10^3$  cm<sup>-3</sup> h/d, with an overall average of  $50 \times 10^3$  cm<sup>-3</sup> h/d. For teachers, the corresponding results are somewhat higher than for students, with range  $36-141 \times 10^3$  cm<sup>-3</sup> h/d and average  $80 \times 10^3$  cm<sup>-3</sup> h/d. The higher values for teachers than for students are a consequence of the longer amount of time per day that teachers spend in the classroom.

Comparing the students in the classroom with the children in houses, we see that the classroom exposure rates are much lower, averaging  $50 \times 10^3$  cm<sup>-3</sup> h/d, as compared with the average of  $316 \times 10^3$  cm<sup>-3</sup> h/d in houses ( $234 \times 10^3$  cm<sup>-3</sup> h/d while awake at home). Two components contribute to the factor of six differences between houses and classrooms. First, children spend more time at home than in school. In our study, the average time spent at home by the children subjects was 16.9 h/d (see data in Table 3.3) compared with an average of 4.6 h/d of classroom occupancy by students. The second factor is the higher average PN concentrations in houses than in classrooms.

The significance of the difference between classroom and at-home PN exposure rates is amplified when one considers that only about 50% of the days in a year are school days whereas many people reside in their homes for 90-100% of the days in a year.



**Figure 5.4.** Average daily exposure rate to particle number concentration inside the classroom for the six classroom sites investigated in this project. The exposure rate can be understood as the product of the average concentration indoors when an individual is in the classroom (in 1000 particles per cm<sup>3</sup>) multiplied by the average daily time spent in that classroom. Bars are labeled with the numerical value of the exposure rate plotted.

# Key finding 4. Indoor proportion of outdoor particles $(f_1)$ tends to be higher in classrooms than in homes.

There may be reasons to be concerned specifically with exposures to particles of outdoor origin even if they only constitute a portion of total UFP exposures. The factor  $f_I$  that we have evaluated in this study is a useful measure for that purpose. This factor is a measure of the time-averaged indoor PN concentration attributable to particles of outdoor origin normalized by the time-averaged outdoor PN concentration. In the absence of indoor sources, the indoor PN concentration is estimated as  $f_I$  times the outdoor PN concentration, both determined over an averaging time of hours or longer. Even with indoor sources present, the indoor PN concentration that is attributable to outdoor sources would be estimated in the same way.

The evaluation of  $f_I$  at each of the houses and school sites was described in detail in §3.2 and §4.2, respectively. Summary tables for houses (Table 3.58) and for schools (Tables 4.39 and 4.40) were presented in §3.3 and §4.3 respectively. Here, the key results are recapitulated. In houses, the  $f_I$  values ranged from 0.11 to 0.51. The average  $\pm$  standard deviation for the seven sites was 0.38  $\pm$  0.14. In classrooms, considering times when students are present, the  $f_I$  values ranged from 0.48 to 0.76. The average  $\pm$  standard deviation for the six sites was 0.57  $\pm$  0.10. In summary, this project was undertaken with the goal of advancing information about ultrafine particles in houses and in schoolrooms in California. The study design entailed detailed investigation, incorporating both observational monitoring and manipulation experiments, at 13 sites: seven houses and six classrooms. With this limited number, we did not intend to collect data that are statistically representative of broader classes of California buildings. On the other hand, the intensive monitoring generated rich sets of data that could be mined for insights about not only what was observed but also the underlying causes. The study has provided significant information and important insight about the interplay among human occupancy, building characteristics, and pollutant dynamic behavior related to the occurrence of ultrafine particles in schoolrooms and houses and the associated exposures.

#### 6. RECOMMENDATIONS

In this section, we present and discuss four recommendations for themes that could be fruitfully pursued in research that builds on the project reported here. These themes relate to important unresolved issues for human inhalation exposure to environmental ultrafine particles in the built environment. The recommendations take advantage of lessons learned from the research undertaken in this project.

#### Theme 1: Additional monitoring of ultrafine particle levels in classrooms and houses.

The building stock is richly diverse, even when one's attention is restricted to schools and houses. It is not possible that any study of thirteen sites could thoroughly characterize UFP conditions in any significant subsection of the building stock. To the extent that UFP exposures indoors remain a concern, then additional studies can be fruitfully undertaken using a streamlined version of the research approach employed here. It would be useful to expand the geographic scope of such studies to other populous areas of California, such as the Los Angeles/Orange County area and the San Joaquin Valley. Studies could be usefully focused on how indoor UFP concentrations and exposure conditions might vary in houses and in schools with the degree of urbanization. Two important factors that could influence indoor UFP levels are expected to correlate with degree of urbanization. First, the levels and diurnal patterns of ambient UFP levels could be influenced by the relative contributions of outdoor sources such as vehicle emissions and atmospheric nucleation events. Second, features of the building stock that influence indoor UFP levels, such as air tightness and prevalence of air conditioning, could also be expected to correlate with degree of urbanization. Studies could also be pursued across a broader range of housing types, for example specifically including apartments. Additional research on air pollutant exposure conditions in schools would be very valuable, as this is an area that is substantially understudied. Ultrafine particles would be worth including as one of several pollutants to monitor.

The present effort featured rich elements for monitoring pollutants and characterizing conditions at field sites. These elements were designed to provide information about the levels of ultrafine particles in classrooms and in houses and the factors that influence those levels. Some of the approaches proved very useful in pursuit of the main goals and others less so. The basic design of the observational monitoring campaign was effective, with highly time-resolved monitoring. It was essential to measure both indoors and outdoors and it was wise to do so with matched instrument packages. The information we gathered about occupancy was extremely important not only for characterizing exposures but also for interpreting the time-dependent PN concentrations. Among the real-time monitoring instruments, the WCPCs effectively produced valuable data. We had to address overheating concerns and the ultimate effort to package the instruments in actively cooled enclosures provided a good solution to the combined challenge of controlling noise and ensuring security while maintaining a thermally suitable environment. Indoor CO<sub>2</sub> monitoring was also valuable as it gave us the opportunity to use a "tracer of opportunity" approach to quantify air-exchange rates during observational monitoring as well as detecting the presence of unvented combustion (e.g. from pilot lights) in houses. Monitoring other copollutants provided potentially useful information about the copollutants monitored, but, relative to the effort and cost, did not prove highly informative for the main goals of characterizing factors that influence ultrafine particle levels. In a future study focusing on ultrafine particle characterization, for a given cost, it would probably be more advantageous to

characterize a larger number of sites with a simpler instrument package, and perhaps omit NO, ozone, and CO monitoring.

The sensors with integral data loggers that we used to monitor device temperatures and door and window position proved quite useful for augmenting activity logs to help identify source activities and characterize building configuration. In a future study, we would recommend expanding the use of these sensors to achieve more complete monitoring of building and appliance operational conditions.

In the research reported here, we devoted approximately equal field effort to observational monitoring and to manipulation experiments. With the benefit of experience, we can conclude that the observational monitoring was essential and that the manipulation experiments were relatively less informative than expected. In a future project, one could consider increasing the fraction of field effort devoted to observational monitoring from 50% to perhaps 75%. In particular, we had three goals for the manipulation experiments: to characterize air-exchange rates; to characterize the penetration and persistence of outdoor particles; and to characterize emissions from some representative sources. The air-exchange rate measurements and the source-characterization manipulation experiments were generally useful, whereas the outdoor penetration and persistence experiments were not sufficiently informative to justify the effort. An unanticipated bonus from the study was the high quality characterization information that could be gleaned from observational monitoring. At every site, information about all three of the important dynamic factors — air-exchange rate, indoor proportion of outdoor particles, and source characterization — were revealed by analysis of the observational monitoring data. Key factors in the success of these efforts were these: observational monitoring over multiday periods; good supplementary information from occupant activity logs or, in the case of schools, from direct researcher observation in the classroom; and sensor-based confirmation of important source-oriented and configuration-oriented parameters.

#### Theme 2: Effects of spatial and temporal variability on pollutant exposure

In human exposure studies for airborne particles, it has been frequently noted that measurements of concentrations using personal samplers produce higher exposure estimates than would be derived using microenvironmental measurements combined with time-activity data. This phenomenon is referred to as the "personal cloud" effect, implicitly suggesting that the underlying cause is spatial variability in airborne levels (Wallace, 2000). The thinking goes that people are spatially associated with certain particle emission activities. Because mixing takes time, spatial gradients are expected such that people are exposed to systematically higher concentrations, on average, than would be recorded, for example, by a monitor that measures the air in the room.

Our data show that the temporal correlation between occupancy and indoor UFP levels can cause exposure concentrations to be higher than the microenvironmental time average even in the absence of spatial gradients. When people are present in their house, the PN levels are higher than when they are absent. A monitor that determined an average concentration over, say, a 24-hour period would produce a lower result from microenvironmental monitoring in the house than the average that would be obtained during occupancy.

It would be a valuable contribution to the overall field of exposure science, not only for particles, but also for other air pollutants, to design and conduct a study whose purpose was to sort out the relative importance of near-field spatial gradients versus temporal variability as contributors to the phenomenon known as the personal cloud effect. Elements of such a study

could include personal monitoring along with intensive monitoring of a species such as UFP in, for example, all of the rooms of a house. The current generation of WCPCs is not capable of being used as a personal sampler, so some instrumentation development would be required. Such a study would be most valuable if the instruments were capable of making highly time-resolved measurements rather than integrated measurements over many hours. It would also be inherently more interesting (and challenging) to work with a dynamic pollutant such as ultrafine particles, rather than a conserved species such as carbon monoxide.

#### Theme 3: Near-field effects of motor vehicle emissions on indoor UFP levels

In preparing the proposal for this project, we were strongly encouraged by ARB staff to include some sites that were very close to freeways or other heavily trafficked roadways. We did include such sites in the ensemble studied. What we found was little if any clear effect of proximity of the site to major roadways even for outdoor PN levels. A key challenge, which our research was not designed to overcome, is that the temporal variability in overall UFP levels at any one site is much larger than the spatial variability expected when comparing an urban site 100 m from a freeway with one 0.5-1 km away.

Scientific and public interest is very high concerning the potential role that near-field exposures to vehicle emissions might have on health outcomes. Better characterization of exposure conditions would certainly make an important contribution to inform this interest. An interesting and important question is whether the indoor proportion of outdoor particles is different for ultrafine particles close to roadways than it is for the background urban aerosol. Such an effect might occur if there are some systematic differences in the building stock that correlate with freeway proximity. It might also occur if the freshly emitted UFP on freeways exhibits different penetration and persistence than the likely more aged background urban aerosol.

The approach employed in the present research could be adapted for a focused study on the effect of roadway proximity on indoor UFP levels. For example, one could consider sampling from two houses (or two schools) simultaneously, both indoors and outdoors, with a key difference being the distance downwind of a freeway. Alternatively, one could focus the research on buildings close to heavily traveled roadways, but not within an urban core region, so that larger spatial gradients in UFP concentrations would be anticipated. Conducting such targeted sampling over long enough duration to capture meteorological variability would be important. A key consideration is that such a project would need to be focused on understanding the effects of proximity as one of very few primary objectives, rather than as one of many diverse goals of the research.

#### Theme 4: Toward understanding emissions and exposures to UFP from cooking activities

When we initiated this research project, it was already known that cooking could be an important source of ultrafine particles indoors. However, this was one of many potentially significant indoor emission sources, with others including candles, laser printers, clothes dryers, and ozone interactions with terpene-based cleaning products or air fresheners. The results of our research are at least suggestive that cooking may be the most important of the indoor emission sources in terms of its contribution to exposures. For each of the 21 occupants of the seven house sites we studied, episodic emissions from cooking made a strong contribution to the cumulative PN exposure while at home. At the end of each subsection §3.2.1-§3.2.7, a table was presented detailing the exposure of household occupants and the apportionment to sources.

Considering only the episodic emissions that were unambiguously associated with cooking, the percentage contribution from cooking to total residential exposures ranged from 16% to 71% (average = 39%) for the 21 household study subjects. By comparison, the estimated contribution to residential exposure from particles originating outdoors was similar, ranging from 11% to 69% (average = 37%) for these 21 subjects. Hence, for this study at least, cooking was as important as outdoor air as a source of human exposure to ultrafine particles in residences.

The specific sources of particles related to cooking were diverse. Natural gas appliances, when present, were consistent sources whenever they were used. However, cooking on an electric range, or in an electric oven, or in some (but not all) cases with an electric toaster also constituted significant particle emission sources.

Because of the prevalence and the quantitative significance of UFP exposures from cooking, it would be valuable to conduct further studies. A combination of further field investigations along with controlled laboratory-based characterization studies could be beneficial. Additional studies that quantify the emissions rate and the deposition rate constants of UFP emitted from well-characterized cooking events would be useful for modeling and analysis. It also would be useful to characterize the chemical composition of the UFP emitted from cooking events.

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## 8. GLOSSARY OF TERMS, ABBREVIATIONS, AND SYMBOLS

| a                | air-exchange rate   |
|------------------|---|
| A1-A9            | label for manipulation experiments to determine air-exchange rate                 |
| ad               | adult   |
| AER              | air-exchange rate   |
| AM               | arithmetic mean   |
| avg              | average   |
| BCPC             | butanol-based condensation particle counter                                       |
| BTU              | British thermal unit  |
| С                | particle number concentration in indoor air                                       |
| $C_o$            | particle number concentration in outdoor air                                      |
| cm               | centimeter  |
| CO               | carbon monoxide   |
| $CO_2$           | carbon dioxide  |
| conc             | concentration   |
| CPC              | condensation particle counter   |
| $C_{pilots}$     | indoor particle number concentration attributable to emissions from pilot lights  |
| d                | day   |
| dev              | deviation   |
| DNA              | deoxyribonucleic acid   |
| $D_{50}$         | lower cutoff diameter for a condensation particle counter                         |
| $E_{30}$<br>E(t) | particle number emission rate (particles per time) from an indoor source          |
| encl             | enclosure   |
| En1-En3          | designation code for instrument enclosures  |
| exp              | exposure  |
| F                | female adult  |
| $f_1$            | time-averaged indoor concentration attributable to outdoor particles, as measured |
| <i>J</i> 1       | by the primary indoor monitor, normalized by the time-averaged outdoor            |
|                  | concentration   |
| $f_{12}$         | time-averaged indoor concentration, as measured by the primary indoor monitor,    |
| J12              | normalized by the time-averaged indoor concentration, as measured by the          |
|                  | supplementary monitor   |
| $f_2$            | time-averaged indoor concentration attributable to outdoor particles, as measured |
| J2               | by the supplementary indoor monitor, normalized by the time-averaged outdoor      |
|                  | concentration   |
| fwy              | freeway   |
| g                | grams   |
| GM               | geometric mean  |
| GSD              | geometric standard deviation  |
| h                | hour  |
| HAC              | heating and air-conditioning (system)   |
| H0-H6            | identification number for house sites   |
| HEPA             | high-efficiency particle arrestance (very efficient filter)                       |
| HM               | harmonic mean   |
| Hrs              | duration in hours   |
| HVAC             | heating, ventilating and air-conditioning (system)                                |
|                  | nound, continuing and an conditioning (system)                                    |

| I/O                | indoor – outdoor ratio  |
|--------------------|---|
| ID                 | identification  |
| In1                | indoor measurement result at the primary indoor monitor   |
| In2                | indoor measurement result at the supplementary indoor monitor                                   |
| Indoor(1)          | synonymous with "In1"   |
| Indoor(2)          | synonymous with "In2"   |
| IPOP               | indoor proportion of outdoor particles  |
| k                  | loss rate coefficient for particles from indoor air by means other than ventilation             |
| $k_d$              | loss rate coefficient for particles from indoor air by means of deposition onto indoor surfaces |
| km                 | kilometer   |
| Li-COR             | model designation of a carbon dioxide monitor   |
| LR                 | living room   |
| M1-M6              | designation identifier for manipulation experiment devoted to characterizing                    |
|                    | emissions from an indoor particle source  |
| m                  | meter   |
| m                  | male child  |
| M                  | male adult  |
| ME                 | microenvironmental  |
| min                | minute  |
| MVF                | microvascular function  |
| Ν                  | number concentration of particles in indoor air   |
| N                  | negative or "no"  |
| na                 | not available   |
| NAAQS              | national ambient air quality standard   |
| nm                 | nanometer (= $10^{-9}$ m)   |
| $N_{net}$          | number concentration of particles indoors attributable to indoor emission sources               |
| $N_o$              | number concentration of particles in outdoor air  |
| NO                 | nitric oxide  |
| NO_in              | nitric oxide concentration in indoor air  |
| NO_out             | nitric oxide concentration in outdoor air   |
| $N_{pilot}$        | number concentration of particles in indoor air attributable to emissions from pilot            |
| 1 ( puoi           | lights  |
| OSHA               | Occupational Safety and Health Administration   |
| O <sub>3</sub>     | ozone   |
| O <sub>3</sub> _in | ozone concentration in indoor air   |
| $O_3_{3}$ out      | ozone concentration in outdoor air  |
| P                  | efficiency of penetration of outdoor particles into indoor air                                  |
| PEL                | personal exposure limit   |
| Pkd                | shorthand notation for particle concentration-rebound experiment designed to                    |
|                    | characterize the parameters P and $k_d$   |
| PM <sub>2.5</sub>  | mass concentration of particulate matter smaller than 2.5 $\mu$ m in diameter                   |
| $PM_{10}$          | mass concentration of particulate matter smaller than 10 $\mu$ m in diameter                    |
| PN                 | particle number   |
| PN_in1             | particle number concentration measured indoors by primary monitor                               |
| PN_in2             | particle number concentration measured indoors by supplementary monitor                         |
| —                  |   |

| PN_out       | particle number concentration measured outdoors                                  |
|--------------|--|
| ppb          | parts per billion  |
| ppm          | parts per million  |
|              | d identifier of specific WCPC units  |
| Q-Trak       | designation of an instrument for measuring $CO_2$ , CO, temperature and relative |
| C C          | humidity   |
| RH           | relative humidity  |
| R1-R4        | identification codes for household occupants                                     |
| $r^2$        | correlation coefficient from linear regression analysis                          |
| S            | second   |
| S1-S6        | identification code for school sites   |
| std          | standard   |
| t            | time variable  |
| $t^*$        | time marking the beginning of an analysis period                                 |
| T_in         | indoor air temperature   |
| T_out        | outdoor air temperature  |
| T85          | synonymous with Q-Trak   |
| $t_e$        | time at the end of an analysis period  |
| $t_s$        | time at the start of an analysis period  |
| TWA          | time-weighted average  |
| UC           | University of California   |
| UFP          | ultrafine particles (particles with diameter smaller than 100 nm)                |
| USEPA        | United States Environmental Protection Agency                                    |
| V            | volume   |
| WCPC         | water-based condensation particle counter  |
| WS           | wind speed   |
| Х            | horizontal coordinate  |
| У            | year   |
| У            | vertical coordinate  |
| Y            | yes  |
| μg           | microgram  |
| μm           | micrometer   |
| %            | percent  |
| λ            | air-exchange rate  |
| σ            | total PN emissions from an episodic indoor source                                |
| $ \Delta T $ | absolute value of the indoor-outdoor temperature difference                      |
| $\Delta T$   | indoor-outdoor temperature difference  |
| °C           | degrees centigrade   |
| °F           | degrees farenheit  |
| 2B2a-2B2b    | designation of instrument for measuring ozone                                    |
| 2B4a-2B4b    | designation of instrument for measuring nitric oxide                             |
|              |  |

#### **APPENDIX A. CALIBRATIONS AND SIDE-BY-SIDE INSTRUMENT COMPARISONS**

This appendix provides a summary of steps taken to ensure the quality of our field experimental data. The information is presented in five sections:

- Side-by-side results from the pilot-phase monitoring at H0
- Summary of instrument calibration data
- Summary of side-by-side data acquired for particle number concentrations at field sites
- Description of methods used to adjust the raw field data
- Validation of ozone and nitric oxide monitor results using central station monitoring.

#### A.1. Side-by-side results from pilot-phase monitoring at H0.

House site H0 was designated as a pilot site. In the proposal, three objectives were stated for the pilot site: (a) side-by-side monitoring of the water-based CPC against a butanol-based instrument; (b) side-by-side monitoring of the battery of instruments in the field monitoring packages we assembled; and (c) testing of observational and manipulation experiment protocols. Because of contract delays, the project was started one year later than originally planned. In the meantime, effective performance of water-based CPCs against butanol-based CPCs was well demonstrated in other studies (Hering, 2006). Also, we found that the field protocols were suitable for the purposes of this project with only very minor adjustment. Because of this, we decided to treat the monitoring at this field site as a seventh home and analyze and report the results in a manner that parallels the other six house sites. See §3.2.1 for the reporting of results.

In this section, we report on the side-by-side instrument monitoring results at H0. Because this project is focused on characterization of ultrafine particle levels, we are particularly interested in the performance of the WCPCs.

In advance of an observational monitoring period at H0, we carried out side-by-side monitoring with the two instrument packages plus the supplementary WCPC indoors for approximately 4 days (21-25 November 2007) and outdoors for approximately 3 days (25-28 November). Here, we present an analysis of the side-by-side data for three WCPCs indoors (QMEa, QMEb, and QMEc) and two WCPCs outdoors (QMEa and QMEb; QMEc was unavailable). We present an analysis of the side-by-side data indoors for three CO<sub>2</sub> analyzers (LI-COR and two Q-Traks). We also present an analysis of the side-by-side data outdoors for two ozone monitors (2B2a and 2B2b).

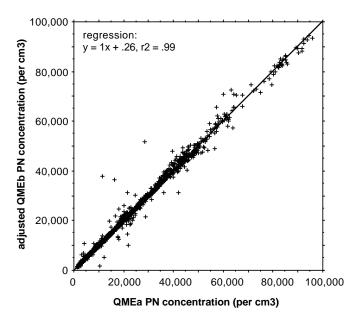
At the time of this effort, one of the NO analyzers was being repaired, so no side-by-side data are available for NO. The indoor ozone levels were consistently less than 10 ppb. Instrument 2B2a recorded an average of 5.8 ppb and instrument 2B2b recorded an average of 4.8 ppb. These data are not analyzed further. Similarly, the CO results averaged less than 1.5 ppm as recorded by both instruments both indoors and outdoors. This level is below the specified accuracy of the Q-Trak CO monitors and so we did not analyze the results further. Also, we did not analyze the comparison data for the outdoor  $CO_2$  monitor being of limited interest for this project.

To compare the results from the WCPCs, we followed this procedure, using data collected with one-minute resolution. Treating QMEa as the reference instrument, we carried out a regression analysis of the results of each of the other monitors against this instrument. The results:

Indoors: QMEb = 1.05 QMEa + 40 cm<sup>-3</sup> 
$$r^2 = 0.99$$
  
Indoors: QMEc = 1.16 QMEa - 82 cm<sup>-3</sup>  $r^2 = 0.98$ 

Outdoors: QMEb = 0.99 QMEa + 750 cm<sup>-3</sup>  $r^2 = 0.88$ 

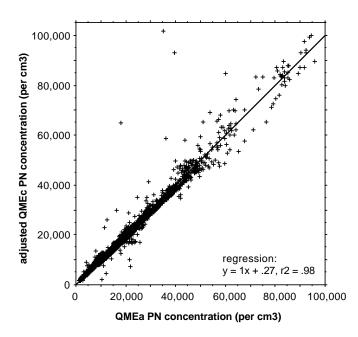
We then used these regression relationships to compute adjusted PN concentrations for QMEb (indoors and outdoors) and for QMEc (indoors). The scatterplots of adjusted QMEb and QMEc versus QMEa PN concentration measurements are presented in Figures A.1-A.3. Because of the way they were constructed, the slope of each line is 1.0. The scatter about the line provides an indication of the degree of comparability between instruments in minute-by-minute WCPC data.



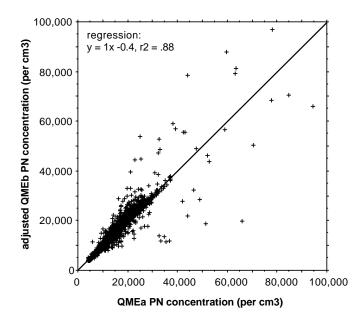
**Figure A.1.** Regression of the adjusted response of the QMEb instrument against the QMEa instrument when deployed side-by-side to measure indoor PN concentrations at site H0. The individual data are resolved and plotted at one-minute resolution.

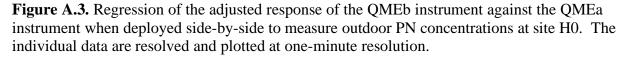
To determine the precision with which we could expect to make measurements of PN concentrations with one-minute time resolution, we computed the absolute value of the difference between adjusted QMEb (and adjusted QMEc) and QMEa for each minute of the side-by-side comparison periods. For the indoor side-by-side period, the average deviation was 310 cm<sup>-3</sup> for QMEb (adjusted) compared with QMEa; the average was 640 cm<sup>-3</sup> for QMEc (adjusted) compared with QMEa. These averages correspond to 3% and 5%, respectively of the mean indoor PN concentration of 11,800 cm<sup>-3</sup> during the side-by-side monitoring period. The corresponding comparison outdoors for QMEb was an average deviation of 1070 cm<sup>-3</sup>, which corresponded to 8% of the mean PN concentration of 14,200 cm<sup>-3</sup> measured during this time.

In summary, the slope of the uncorrected regressions from side by side monitoring indicated deviations in average instrument response in the range 1-16% (average deviation in slope = 7%). By adjusting the response factors of the instruments during side-by-side monitoring periods at each site, we expect to reduce the effect of average response errors to well below 10% and probably below 5%. At the level of time-resolved data with one-minute resolution, the adjusted instrument responses agreed to better than 10%.



**Figure A.2.** Regression of the adjusted response of the QMEc instrument against the QMEa instrument when deployed side-by-side to measure indoor PN concentrations at site H0. The individual data are resolved and plotted at one-minute resolution.

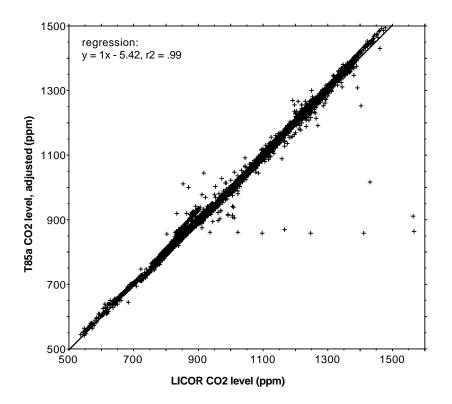




An analogous investigation was carried out of the performance of the three  $CO_2$  monitors using side-by-side monitoring indoors at H0. The LI-COR  $CO_2$  monitor was treated as the reference instrument. Results from the other two instruments produced these correlations when regressed against the readings from the LI-COR:

| Indoors: T85a = 0.95 LI-COR – 56 ppm                    | $r^2 = 0.98$ |
|---|--------------|
| Indoors: $T85b = 1.04 \text{ LI-COR} - 1.2 \text{ ppm}$ | $r^2 = 0.98$ |

Figures A.4 and A.5 present scattergrams of the adjusted  $CO_2$  data measured with one of the Q-Traks against the LI-COR. For the adjusted data, with one-minute time resolution, the average absolute deviation between each Q-Trak and the LI-COR averaged 11 ppm and 8 ppm, respectively, which was about 1% of the average reading of 1010 ppm indoors at H0 during the side-by-side comparison period. This evidence indicates that these analyzers could be expected to perform well in measuring indoor  $CO_2$  levels.

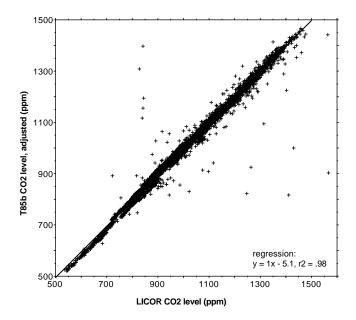


**Figure A.4.** Regression of the adjusted response of the Q-Trak T85a instrument against the LI-COR instrument when deployed side-by-side to measure indoor  $CO_2$  levels at site H0. The individual data are resolved and plotted at one-minute resolution.

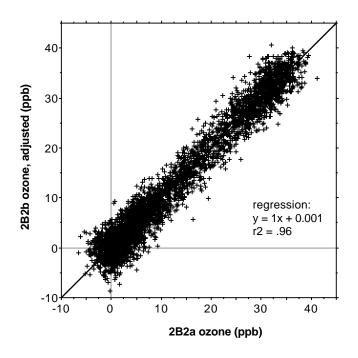
The two ozone analyzers were compared in their side-by-side performance measuring ozone outdoors. Treating 2B2a as the reference instrument and regressing the one-minute resolution data from 2B2b against this instrument produced the following expression:

Outdoors: [2B2b] = 0.98 [2B2a] - 1.5 ppb  $r^2 = 0.96$ 

The adjusted 2B2b data are plotted against the 2B2a data in Figure A.6. Compared with the WCPC and  $CO_2$  data presented in Figures A.1-A.5, the ozone data are much noisier. The average absolute value of the deviation between adjusted 2B2b and 2B2a was 2.1 ppb, which was 16% of the average reading of 12.8 ppb.



**Figure A.5.** Regression of the adjusted response of the Q-Trak T85b instrument against the LI-COR instrument when deployed side-by-side to measure indoor  $CO_2$  levels at site H0. The individual data are resolved and plotted at one-minute resolution.



**Figure A.6.** Regression of the adjusted response of the 2B2b instrument against the 2B2a instrument when deployed side-by-side to measure outdoor ozone levels at site H0. The individual data are resolved and plotted at one-minute resolution.

## A.2. Summary of instrument calibration data: gas analyzers

To the extent practical, instrument calibrations were conducted throughout the period of the field monitoring campaign. The approach is described in the main report §2.1.2. The calibration results are presented in Tables A.1 (ozone), A.2 (nitric oxide), A.3 (carbon dioxide), and A.4 (carbon monoxide).

|                   | Instrument 2B2a |              | Instrume | nt 2B2b      |
|-------------------|-----------------|--------------|----------|--------------|
| Date              | Slope           | Offset (ppb) | Slope    | Offset (ppb) |
| October 2007      | 0.94            | -0.6         | 0.95     | -1.9         |
| March 2008        | 1.11            | 1.1          | 1.13     | -2.3         |
| 3 April 2008      |                 | -0.6         |          | -0.9         |
| 22 May 2008       |                 | -1.8         |          | -0.8         |
| 17 June 2008      | —               | -0.1         | —        | -0.5         |
| 3 July 2008       | —               | -1.7         |          | -1.7         |
| 15 July 2008      | 0.98            | -1.7         | 1.00     | -1.7         |
| 25 September 2008 |                 | -1.5         |          | —            |
| 15 October 2008   |                 | -1.5         | _        | -0.9         |
| 11 November 2008  | —               | -1.8         |          | 0.1          |
| 26 November 2008  |                 | -0.7         |          | 0.4          |
| Average           | 1.01            | -1.0         | 1.03     | -1.0         |

**Table A.1**. Calibration parameters for ozone monitors

|                | Instrument 2B4a |              | Instrume | nt 2B4b      |
|----------------|-----------------|--------------|----------|--------------|
| Date           | Slope           | Offset (ppb) | Slope    | Offset (ppb) |
| October 2007   | 1.08            | 1.5          | 0.85     | -0.8         |
| February 2008  | 1.02            | -5.2         | 1.03     | 7.0          |
| March 2008     | 1.01            | -4.3         | 1.02     | 8.0          |
| April 2008     | —               | -4.3         |          | 17           |
| May 2008       | 1.07            | -6.0         | 1.05     | 7.4          |
| June 2008      |                 | -6.8         |          | 8.2          |
| July 2008      | 1.06            | -7.7         | 1.05     | 4.7          |
| September 2008 | 0.91            | -4.4         | 0.94     | 0.7          |
| October 2008   |                 | -6.4         |          | -5.0         |
| November 2008  | 0.89            | -14.7        | 0.90     | -1.6         |
| November 2008  |                 |              | 0.90     | -4.0         |
| Average        | 1.01            | -5.8         | 0.97     | 3.8          |

|              | LI-COR | LI-COR 820   |       | ( <b>T85</b> a) | Q-Trak | x (T85b)     |
|--------------|--------|--------------|-------|-----------------|--------|--------------|
| Date         | Slope  | Offset (ppm) | Slope | Offset (ppm)    | Slope  | Offset (ppm) |
| October 2007 | 1.00   | -2.9         | 1.06  | 20              | 1.11   | 6            |
| January 2008 |        | —            | 0.92  | -38             | 1.03   | 17           |
| March 2008   |        |              | 0.89  | -9              | 1.03   | 22           |
| April 2008   | 1.00   | 1.2          | 0.92  | -112            | 0.98   | 22           |
| May 2008     | 1.03   | -5.1         | 0.94  | -123            | 1.03   | 7            |
| June 2008    | 1.06   | 3.5          | 0.92  | -132            | 1.02   | 32           |
| 3 July 2008  |        |              | 0.90  | -145            | 1.03   | 13           |
| Sept 2008    | 1.00   | 0.2          |       |                 | —      |              |
| 15 Oct 2008  | 0.99   | 2.2          |       |                 |        |              |
| 11 Nov 2008  | 0.99   | 5.2          | 0.92  | 0               | 0.97   | 6            |
| 26 Nov 2008  | 1.00   | 6.5          | 1.00  | 43              | 1.07   | 22           |
| Average      | 1.01   | 1.4          | 0.94  | -55             | 1.03   | 16           |

**Table A.3**. Calibration parameters for CO<sub>2</sub> monitors.

Table A.4. Calibration parameters for CO monitors.

|                  | Q-Trak (T | Q-Trak (T85a) |       | T85b)        |
|------------------|-----------|---------------|-------|--------------|
| Date             | Slope     | Offset (ppm)  | Slope | Offset (ppm) |
| October 2007     | 1.14      | -1.7          | 1.10  | -2.9         |
| February 2008    | 1.09      | 1.0           | 1.07  | -0.4         |
| March 2008       | 1.13      | 0.4           | 1.02  | 1.1          |
| April 2008       | 1.11      | -0.8          | 0.93  | 1.5          |
| May 2008         | 1.11      | -0.4          | 1.09  | 0.1          |
| June 2008        | 1.09      | 0.4           | 1.06  | 0.0          |
| July 2008        | 1.13      | -0.7          | 1.10  | 0.1          |
| 15 October 2008  | 1.06      | —             | 1.06  | _            |
| 11 November 2008 | 1.13      | 4.4           | 1.13  |              |
| 26 November 2008 | 1.16      | 3.9           | 1.09  | -0.2         |
| Average          | 1.12      | 0.7           | 1.07  | -0.1         |

# A.3. Summary of side-by-side data acquired for PN concentrations

At each field site, we conducted several hours of side-by-side monitoring with the instrumentation packages colocated. The results were used to adjust the PN concentration data from the different WCPC instruments. At each site, one of the instruments was designated as the reference instrument. After correcting for flow calibrations done in the field, the response of the other WCPCs was adjusted based on the side-by-side monitoring period so that the average response of the adjusted instrument matched the average response of the reference instrument. Table A.5 presents the adjustment factors. The last few lines of this table indicate that instruments' average response based on side-by-side monitoring over all sites agreed to within 5%, but there was variability that averaged 10-15% in the side-by-side comparison at each site.

| Site            | QMEb              | QMEc              | QMEd |
|-----------------|-------------------|-------------------|------|
| H0 (inside)     | 1.05              | 1.16              |      |
| H0 (outside)    | 0.99              | —                 |      |
| H1              | 1.00              | 0.75              |      |
| H2              | 0.81 <sup>a</sup> | 0.82 <sup>a</sup> | —    |
| H3              | 0.88 <sup>a</sup> | 0.87 <sup>a</sup> |      |
| H4              |                   | 0.98              | 1.27 |
| H5              |                   | 1.12              | 1.02 |
| H6 <sup>b</sup> |                   | 1.14              | 1.07 |
| S1              |                   | —                 | 1.03 |
| S2              |                   | 1.16              | 1.19 |
| S3              | —                 | 1.08              | 0.99 |
| S4              | —                 | 0.92              | 0.89 |
| S5 <sup>c</sup> |                   | 1.12              | 0.82 |
| S6              |                   | 1.08              | 1.06 |
| Average         | 0.95              | 1.02              | 1.04 |
| St. Dev.        | 0.10              | 0.14              | 0.14 |

**Table A.5.** Slope of PN data from QMEb, QMEc or QMEd regressed against PN data from the transfer standard, QMEa.

<sup>a</sup> At H2 and H3, QMEa was not unavailable, therefore QMEd was used as the transfer standard. <sup>b</sup> A manufacturer upgrade was applied to instruments prior to work at H6. (This site was

sampled after S1 and before S2-S6.)

<sup>c</sup> Instrument nozzles were replaced prior to work at S5

## A.4. Methods used to adjust raw field data

Raw monitoring data from the field were adjusted in analysis with two aims: (a) for instrument responses to be consistent in so far as possible with calibration data; and (b) for the relative response of the instruments to agree as much as possible with one another when sampling the same conditions. Achieving (a) would guarantee (b); but (b) was treated as an important subsidiary goal, of value even when we could not ensure (a) with a high level of accuracy and reliability.

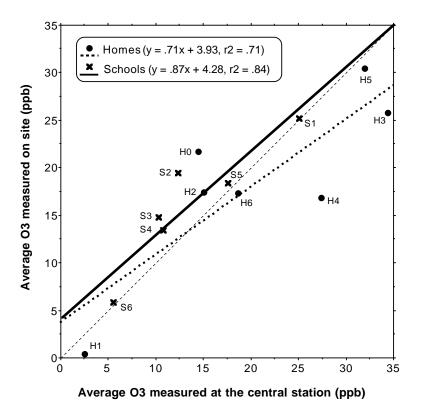
Overall, monitors for PN, NO and  $CO_2$  were adjusted using the same general method; specifically, calibration data was used to adjust one of the instruments, which in turn was used as a reference instrument for adjusting the other monitors(s) based on site-specific side-by-side (SBS) monitoring data. This approach could not be used for CO because the levels measured during field SBS monitoring were often too close to zero to be meaningfully resolved. Hence, we relied solely on calibration data for adjusting the CO monitors. For ozone, we could not obtain meaningful SBS data at each site, so we treated each instrument as having a uniform response factor, i.e. a slope of 1.0. We believe this treatment to be reasonable as it agrees within a few percent with calibration data (see Table A.1). The ozone calibration intercepts were obtained by per-instrument and per-site zero checks.

For SBS comparisons, the averaging time for matching instrument responses was chosen based on consideration of instrument response stability. We used 1-min data for  $CO_2$ , 5-min averages for PN and 1-h averaging for NO.

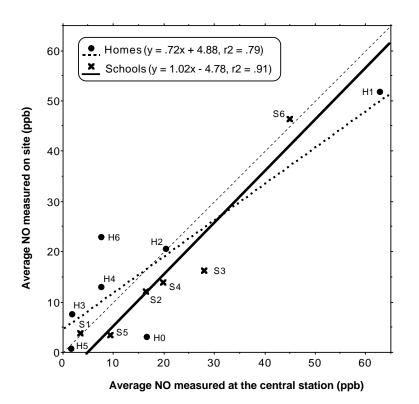
# A.5. Validation of ozone and nitric oxide monitor results using central station monitoring

As a further check on the performance of the nitric oxide and ozone monitors, the outdoor monitoring data we obtained during observational monitoring with central-station monitoring results for the same time period. The central station (CS) data were obtained from the Bay Area Air Quality Management District (<u>www.baaqmd.gov</u>). For sites H0, H1, H6 and all six classroom sites, we used CS data from the Berkeley 6<sup>th</sup> Street monitoring station. For sites H2, H3, and H4, we used the Oakland monitoring station on International Boulevard. For H5, we used data from the Livermore station (Rincon). In each case, we compared the time-average monitoring result on site against the central station average for the entire observational monitoring period.

Figures A.7 and A.8 present the results for ozone and NO, respectively. The data are well correlated. The average absolute deviation of the on-site measurement results as compared with the CS data was 3.8 ppb for ozone, representing 22% of the CS average for all 13 sites of 17.4 ppb. The analogous result for NO was 6.3 ppb, 34% of the average of 18.5 ppb across all 13 sites. Note that perfect agreement would not be expected for no other reason than that the on-site monitoring is not collocated with the CS monitors.



**Figure A.7.** Regression of time-averaged ozone level measured on-site during observational monitoring against the central-station monitoring results averaged for the same time period.



**Figure A.8.** Regression of time-averaged nitric oxide level measured on-site during observational monitoring against the central-station monitoring results averaged for the same time period.

## APPENDIX B. FORMS USED IN FIELD RESEARCH

This appendix presents the forms that were used to acquire data in the field. Five forms are presented here:

- Form A: House site characterization
- Form B: House site questionnaire Initial information
- Form C: Home site observational monitoring Daily survey
- Form D: Home site observational monitoring Daily activity log
- Form SA: School site characterization

The information in the forms as used is the same as presented in this appendix. The format has been revised to save space.

# FORM A: HOUSE SITE CHARACTERIZATION

| House ID:        |           |       |  |
|------------------|-----------|-------|--|
| Researcher name: |           | Date: |  |
| Start time:      | End time: |       |  |

#### 1) Type and layout of house

- a. House characteristics
  - i. Exterior wall finish (e.g. masonry, stucco, wood siding):
  - ii. Interior wall construction/finish (e.g. sheetrock, plaster):
  - iii. Number of levels/floors:
  - iv. Has fireplace or woodstove (specify which)?
- b. Sketch a floor plan of the house, indicating:
  - i. Type of room and dimensions
  - ii. Type of flooring in each room
  - iii. Doors and windows (D1, D2, W1, W2 etc.) specify dimensions
  - iv. Location of stove(s) (S1, S2 etc.)
  - v. Location of other combustion and heating devices (e.g. toaster, fireplace, candles or incense, portable/wall-mounted heaters)
  - vi. Location of portable air cleaners (*if any*)
  - vii. Location of return and supply vents (for central air system).
  - viii. Add location of instrument packages (and distance from stove and fireplace) once this is determined

## 2) Door and window openings

a. For each door and window in the sketch, indicate whether it was closed or open at the time of this survey. If the window is open, measure the opening area. Also indicate if the window is horizontal sliding, vertical sliding, or swinging, and whether a door is hinged or sliding.

| Code (from sketch) | Closed or<br>open<br>(area)*? | Type of<br>Window/<br>Door | Code (from sketch) | Closed or open (area)*? | Type of<br>Window/Door |
|--------------------|-------------------------------|----------------------------|--------------------|-------------------------|------------------------|
|                    |                               |                            |                    |                         |                        |

\* If the window is of the "swinging" type, measure the area of the plane defined by the outer edge of the swinging glass pane and the outer edge of the window opening.

## 3) Central air-handling system associated with heating, cooling, or ventilation

- a. Describe the type of heating system in the house (e.g., forced air, local combustion based, electrical resistance, etc.). Indicate type of heat source (natural gas, electricity, etc.)
- b. Does the house have air conditioning? If so, describe system.
- c. Is there a visible particle filter in the return duct or elsewhere? If so, describe it in terms of type (HEPA, etc.) and condition. *Note: Filter is likely to be near the furnace. Use labeling on the side of filter to indicate the type.*
- d. In what locations are the return air vents located (indicate on sketch)?
- e. In what locations are the supply vents located?
- f. How is the system controlled?
- g. Is the system on at present? If so, at what setting?
- h. Other observations and comments:

#### 4) Characteristics of stove and oven

- a. Is there a range hood vent?
- b. [If a = yes] Does it exhaust to the outdoors?
- c. Is the oven attached to the stove, such that they are both exhausted by the range hood? Is the oven vented separately?
- d. Is the stove electric or gas? Is the oven electric or gas?
- e. Number of burners on stove?
- f. Is there a pilot light for the stove?
- g. Degree of cleanliness of stove burners?
- h. Approximate age of stove?
- i. Does the oven have a broiler?
- j. Is there a pilot light for the oven?
- k. Degree of cleanliness of oven?
- 1. Approximate age of oven?

#### 5) Other kitchen appliances that might be particle sources

- a. Is there a microwave? If so, where is it located?
- b. Is there a toaster or toaster oven? If so, where is it or are they located?

c. Are there any other visible appliances that generate heat? (e.g. coffee pot) If so, where are they located?

#### 6) Fireplace or wood stove

- a. Is there a fireplace? If so, does it have a glass door?
- b. What type of fuel is used (e.g. wood logs, wood pellets, gas)?
- c. Is there a woodstove? If so, what is its approximate age?
- d. Does the woodstove appear to be airtight?

#### 7) Candles

a. Are there any visible candles that appear to have been used? [If yes] where are they located (*indicate on sketch*)?

#### 8) Incense

a. Is there any visible incense that appears to have been used? [If yes] where is it located *(indicate on sketch)*?

#### 9) Water heater

- a. Where is it located (*indicate on sketch*)?
- b. What type of heat source is used (gas or electric)?

#### 10) Clothes dryer

- a. Is there a dryer?
- b. [If yes] Where is it located (*indicate on sketch*)?
- c. What type of heat source is used (gas or electric)?

#### 11) Portable/wall-mounted heaters

- a. If there are local (room) heaters, indicate where they are located (*indicate on sketch*),
- b. What type of heater is it, and what is the relative size?

#### 12) Air cleaners

- a. If there are any portable air cleaners, where are they located (indicate on sketch)?
- b. List the model and make of the air cleaner(s):
- c. What are control settings options on the air cleaner(s)?
- d. Indicate if air cleaner(s) are on at time of visit and, if so, at what setting.

#### 13) Exhaust fans

a. Where are they located (e.g. bathroom, attic, kitchen)? (*Indicate on sketch*) Note if they are always on.

#### 14) Ambient sources

- a. Distance to nearest freeway (If not obvious, obtain distance from maps.google.com)
- b. Amount of traffic on street in front of the home (cars/min based on 5-min count)
- c. Other nearby ambient sources? (e.g. port, industry, smoke from neighbors chimney)

## FORM B: HOUSE SITE QUESTIONNAIRE—INITIAL INFORMATION (DAY 1)

| House ID:        |           |       |  |
|------------------|-----------|-------|--|
| Researcher name: |           | Date: |  |
| Start time:      | End time: |       |  |

- 1) How many people live in this house at present?
- 2) Does anyone smoke, among those who live here or regularly visits this residence?
- 3) How old is your house?
- 4) Would you consider your house *drafty*, *somewhat drafty*, or *tight*? (NOTE: drafty is defined as uncontrolled airflow through the house when the windows and doors are closed.)
- 5) When was your heating or air conditioning system installed? Has it been recently serviced?
- 6) Do you know what type of particle filter you have in your air-system? [If yes] What type?
- 7) During the previous month (30 days), have you used your fireplace: *never*, *occasionally*, *often*, *every day*?
- 8) Where is the clothes dryer located?
- 9) Where is the water heater located?
- 10) Are your appliances natural gas or electric?

a.If natural gas, is there a pilot light?

- 11) How old is your oven? How old is your stove?
- 12) Additional comments

## FORM C: HOUSE SITE OBSERVATIONAL MONITORING —DAILY SURVEY

For each of the question areas, consider a 30-h period, beginning at 6 AM yesterday and ending at noon today. (If the questionnaire is filled out before noon, then the period of interest should begin at 6 AM yesterday and end at the current time.)

| House ID:                 | Study Day (circle): | 2 | 3 | 4 |
|---------------------------|---------------------|---|---|---|
| Researcher name:          | Date:               |   |   |   |
| Questionnaire start time: | End time:           |   |   |   |

1) Occupant Activity. For each occupant of the home, at what times during that period was the occupant at home awake and at home asleep.

| Occupant ID | Hours at home | Hours asleep |
|-------------|---------------|--------------|
|             |               |              |

2) Guests/others (e.g. house employees) For each occupant of the home, at what times during that period was any other person in the residence.

| Person ID | Hours in the home |  |  |
|-----------|-------------------|--|--|
|           |                   |  |  |

## 3) Central air handling system associated with heating, cooling, or ventilation

- a. Did you use your air handling system for heating or cooling (specify which)?
- b. [If yes] Did you close the windows/doors while air-system was on
- c. [*If yes*] How did you interact with your air handling system:
  - i. Thermostat:
    - adjusted throughout day
    - at set point (specify) for entire day
    - at set point, on timer
    - at set point, manually turned on and off
  - ii. No thermostat, manual adjustment

#### 4) Door and window openings

a. Were any doors and/or windows that were ever partially open (1), or wide open (2)? For what hours were they in the open state? (Use codes from Form A sketch)

| Door or | Partially (1) or | Open during | Door or | Partially (1) | Open during  |  |  |
|---------|------------------|-------------|---------|---------------|--------------|--|--|
| Window  | wide (2) open?   | which       | Window  | or wide (2)   | which hours? |  |  |
| Code    | _                | hours?      | Code    | open?         |              |  |  |
|         |                  |             |         |               |              |  |  |

#### 5) Cooking activities

#### a. Stove use

| What was prepared? | How many burners? | Approx. time of day? | [ <i>if vent present</i> ] Was vent used? |
|--------------------|-------------------|----------------------|---|
|                    |                   |                      |   |

#### b. Oven use

| What was prepared? | How long was it on? | Approx. time of day? | [ <i>if vent present</i> ]<br>Was vent used? |
|--------------------|---------------------|----------------------|--|
|                    |                     |                      |  |

c. Other kitchen appliances (toaster, coffee maker, waffle iron etc.)

| What appliance? | When was it used? | en was it used? |  |  |
|-----------------|-------------------|-----------------|--|--|
|                 |                   |                 |  |  |

#### 6) Fireplace or woodstove

- a. Did you use the fireplace or woodstove?
- b. [If yes] What kind of fuel did you use (e.g. wood logs, wood pellets, natural gas)?
- c. [If yes] Did you have a door closed over the fire during use?
- d. [If yes] What were the times and duration of use?

#### 7) Candles and incense

- a. Did you burn candles?
- b. [*If yes*] Where (i.e. in which rooms) and when did you burn them?
- c. [*If yes*] Were they scented?
- d. Did you burn incense?
- e. [*If yes*] Where (i.e. in which rooms) and when did you burn it?

#### 8) Cleaning

a. What type of cleaning did you engage in? (List the type of cleaning activity and the products used):

| Type of cleaning | Products used | Time of activity |  |
|------------------|---------------|------------------|--|
|                  |               |                  |  |

- b. Did you ventilate the space during cleaning activities?
- c. [*If yes*] what is the ventilation method (fan, window etc.)?

## 9) Air fresheners

- a. Did you use any air fresheners?
- b. *[If yes]* List types, times, and locations used

| Type of air freshener | Times used: | Location of use: |
|-----------------------|-------------|------------------|
|                       |             |                  |

#### 10) Air cleaners

- a. Did you use any portable air cleaners?
- b. *[If yes]* List types, locations, and time of day and duration of use.

|                     | ··· , ·· · · · · · · · · · · · · · · · |                 |
|---------------------|--|-----------------|
| Type of air cleaner | Time of day and duration of use:       | Location of use |
|                     |  |                 |

#### 11) Other sources

- a. Did you use your clothes washer and/or dryer? What time of day?
- b. Did you use a portable or wall-mount heater? Where? What time of day?
- c. Were any cigarettes smoked on the premise? If so, approximately when and where?

# **12)** Additional information (If information pertains to a specific question, specify which question.)

## FORM D: HOUSE OBSERVATIONAL MONITORING — DAILY ACTIVITY LOG

**Cook**= Cooking activity (includes toaster and microwave). Please specify what was cooked, and appliances used.

**Comb**= Combustion activity, not cooking: candle, incense, etc. Please specify the combustion device, and the room it was used.

**Elec** = Electric heated appliance (unrelated to cooking): electric space heater, hair blow dryer. Please specify the type of appliance, and the room it was used.

**Clean**= Cleaning activity. Please specify location of cleaning activity, and product used. **Vent**=Use of kitchen or hood fan, door, or window for ventilation purposes

| Time interval       | Cook | Comb | Elec | Clean | Vent | Details                 |
|---------------------|------|------|------|-------|------|-------------------------|
| E.g.<br>7:00-7:30am |      |      | Х    |       |      | Used hair blow-dryer in |
| 7:00-7:30am         |      |      |      |       |      | master bathroom         |
|                     |      |      |      |       |      |                         |
|                     |      |      |      |       |      |                         |
|                     |      |      |      |       |      |                         |
|                     |      |      |      |       |      |                         |

## **OCCUPANCY LOG**

List the times of day that individuals were at home, and any hours for which they were asleep. Specify if the hours awake were of low or high activity

| Occupant name | Time intervals at home.      | Time intervals asleep |
|---------------|------------------------------|-----------------------|
|               | (Low or high activity level) |                       |
| E.g. Tommy    | 5-7pm: high                  | 10pm to 7am           |
|               | 7pm-8am: low                 |                       |
|               |                              |                       |
|               |                              |                       |
|               |                              |                       |
|               |                              |                       |

# FORM SA: SCHOOL SITE CHARACTERIZATION

| School ID:       |           |       |
|------------------|-----------|-------|
| Researcher name: |           | Date: |
| Start time:      | End time: |       |

## 1) Building construction and layout

- a. Construction characteristics:
- i. Interior wall construction/finish (e.g. concrete or wood/metal stud and sheetrock or plaster):
  - ii. Number of levels/floors:
- b. Sketch a rough floor plan of the school, indicating: (no dimensions required)
  - iii. General shape of the school building. Indicate whether the building falls into one of these three categories:
    - 1. Portable
    - 2. Row building
    - 3. Multistory/urban
  - iv. Basic orientation of the school, relative to near-by roads or hills
  - v. Location of parking lots, bus drop-off/pick-up zones, parent drop-off/pickup zones
  - vi. Approximate location of the classroom within the school building
  - vii. Rooms immediately adjacent to the classroom (indicate shape and relative size of adjacent rooms, and routes by which air may flow between the rooms).
  - viii. Doors and windows on exterior of school building in the vicinity of the classroom
  - ix. Doors and windows on exterior walls of classroom
  - x. Add location of outdoor instrument once this is determined

#### 2) Classroom layout

a. Draw a floor plan of the classroom, indicating: (dimensions required)

- i. Shape and dimensions of the classroom
- ii. Size and break-up of different "zones" within the room. Zones may include: teacher's desk/lecture area, student desks area, science lab space, students reading area etc.
- iii. Chalk board/ white board (indicate which)
- iv. Doors and windows
- v. For *central* thermal conditioning system:
  - 1. Supply and return vent locations (note return grill may be in the door)
  - 2. Location of thermostat

- vi. For *package* thermal condition unit:
  - 1. Location of the package unit
- vii. Potential particle sources (e.g. candles, incense, air fresheners, Bunsen burners).
- viii. Indicate type of flooring (carpet, vinyl tile etc.)
- ix. Locations of any electronic equipment: computers, projectors etc.
- x. Add location of indoor instrument packages once this is determined

## 3) Door and window openings within classroom

a. For each door and window in the classroom sketch, indicate whether it was closed, or open at the time of this survey. If the window is open, measure the area of the opening. Also indicate if the window is horizontal sliding, vertical sliding, or swinging.

| Code (from sketch) | Closed or<br>open<br>(area)*? | Type of<br>Window/<br>Door | Code (from sketch) | Closed or open (area)*? | Type of<br>Window/<br>Door |
|--------------------|-------------------------------|----------------------------|--------------------|-------------------------|----------------------------|
|                    | ((12 0 11) 1                  | 2001                       |                    |                         | 2001                       |

\* If the window is of the "swinging" type, measure the area of the plane defined by the outer edge of the swinging glass pane and the outer edge of the window opening.

## 4) Air handling system associated with heating, cooling, or ventilation

- a. Does the school have a central system for:
  - i. Heating
  - ii. Cooling
  - iii. Ventilation
- b. [*If yes to* (*a*)] In what locations within the classroom are the return air vents located (*specify whether they are on the ceiling or wall*)?
- c. [*If yes to* (*a*)] In what locations within the classroom are the supply vents located (*specify whether they are on the ceiling or wall*)?
- d. [*If yes to (a)*] Is there thermostat control within the classroom? [If yes] What are the setting options on the thermostat?
- e. [*If yes to* (*a*)] Is it on at present? At what setting?
- f. [*If no to* (*a*)] Is there a packaged/wall unit in the classroom for:
  - iv. Heating
  - v. Cooling
- g. [*If yes to (f)*] Is it on at present? At what setting?
- h. Other comments or observations:

## 5) Candles

- a. Are there any visibly located candles? [If yes] where are they located (*locate on sketch*)?
- b. Condition (esp. wick)

## 6) Incense

a. Is there any visibly located incense? [If yes] where is it located (*locate on sketch*)?

## b. Recently used?

## 7) Portable/wall-mounted heaters

- a. Where is the heater(s) located (*locate on sketch*)?
- b. What type of heater is it, and what is the relative size?

#### 8) Air cleaners

- a. Where are they located (*locate on sketch*)?
- b. What is the model and make of the air cleaner?
- c. How many settings does each air cleaner have?

## 9) Air fresheners

- a. Where are they located (*locate on sketch*)?
- b. What type of air freshener (e.g. spray or plug in)?
- c. What is the brand and scent of the air freshener?

## **10)** Portable mixing/cooling fans?

- a. Are there any portable or ceiling fans in the room? [*If yes*] Where are they located?
- b. What is the approximate diameter of the fan blades?
- c. How many settings are there?

## 11) Electronic equipment?

- a. What type of electronic equipment is visible in the room (e.g. computers, projectors, printers)?
- b. Where is this equipment located?
- c. Does it appear to have been recently used?

## 12) Other sources

- a. What is/are the sources?
- b. Where are they located (*locate on sketch*)?
- c. Other comments:

## 13) Ambient sources

- a. Proximity to freeway (*If not obvious, obtain distance from maps.google.com*):
- b. Density of traffic on street in front of the school (cars/min):
- c. Density of cars going through the school drop-off zone in morning/afternoon (cars/min):
- d. Density of buses going through the drop-off zone in morning/afternoon (buses/min):
- e. Other nearby ambient sources? (e.g. port, industry)

# 14) Additional comments

# APPENDIX C. FLOOR PLANS FOR HOUSES AND CLASSROOM SITES

This appendix presents floor plans of the thirteen study sites. Plans for the seven house sites are displayed as Figures C.1-C.7 and plans for the six classrooms are shown as Figures C.8-C.13.

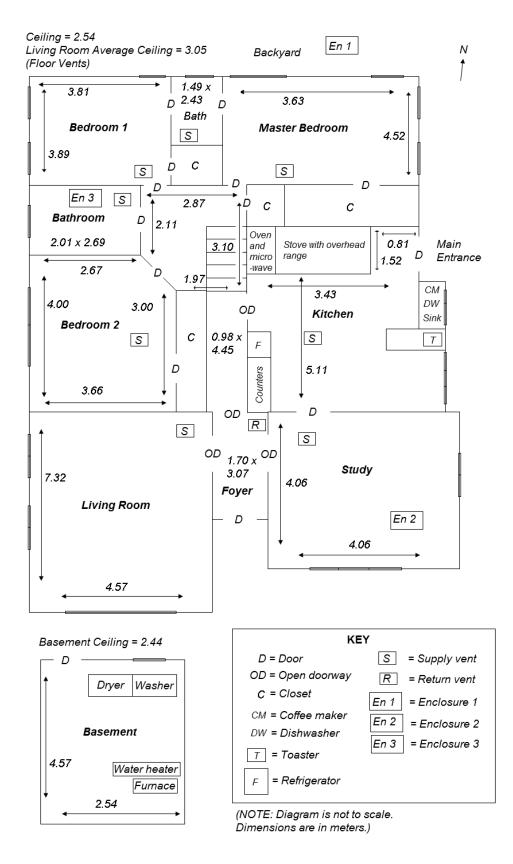


Figure C.1. Floor plan for house site H0.

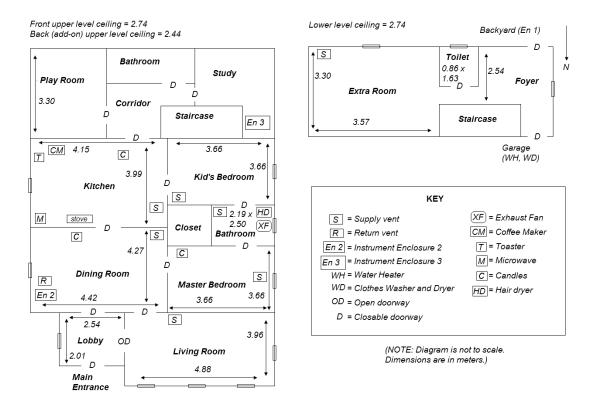


Figure C.2. Floor plan for house site H1.

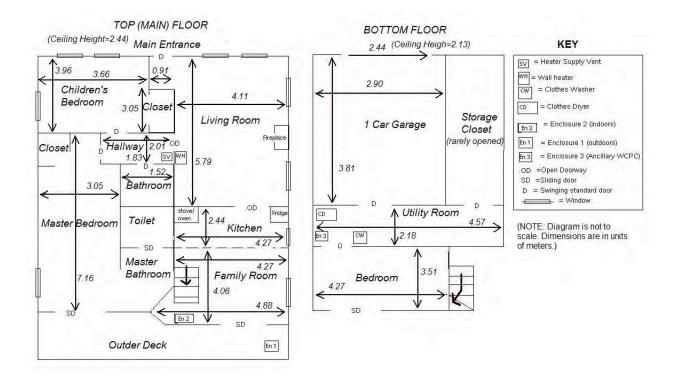


Figure C.3. Floor plan for house site H2.

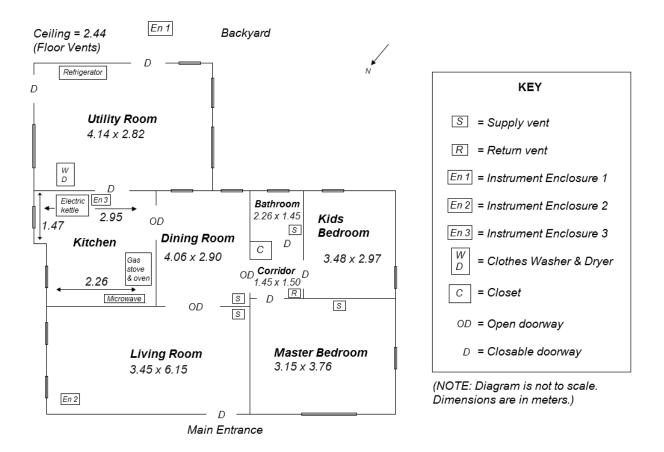


Figure C.4. Floor plan for house site H3.

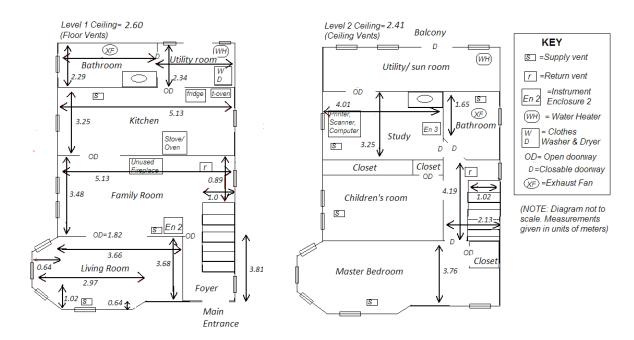


Figure C.5. Floor plan for house site H4.

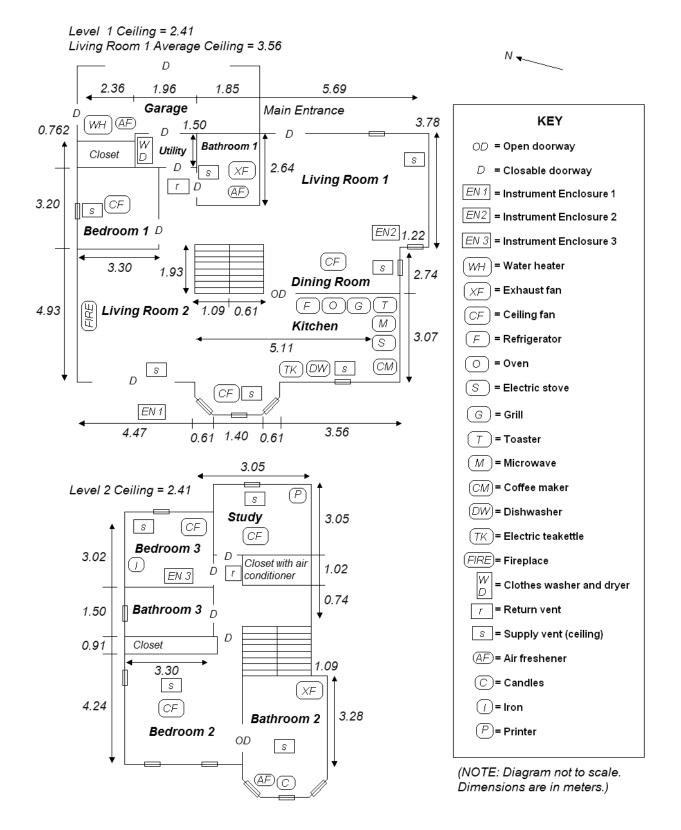


Figure C.6. Floor plan for house site H5.

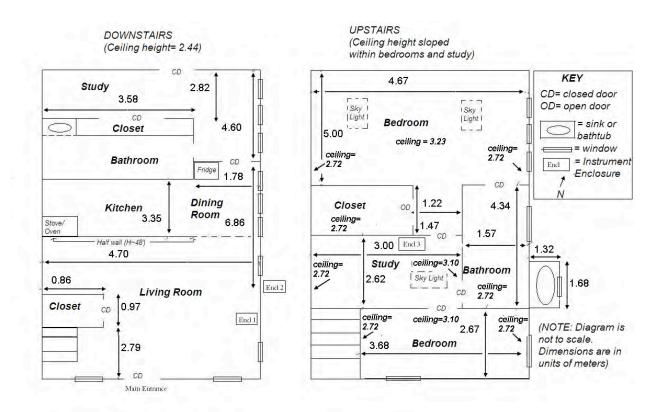


Figure C.7. Floor plan for house site H6.

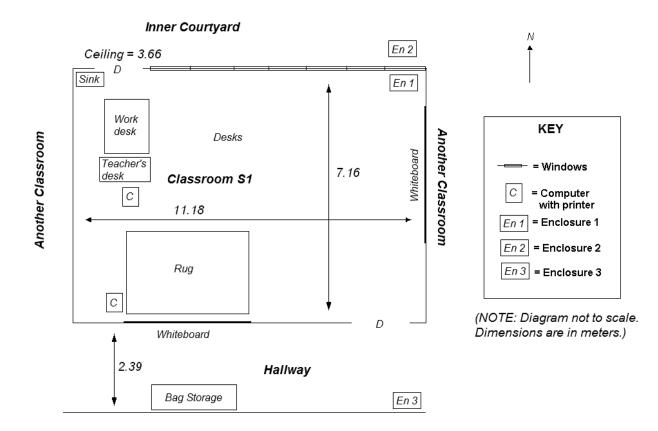


Figure C.8. Classroom floor plan for site S1.

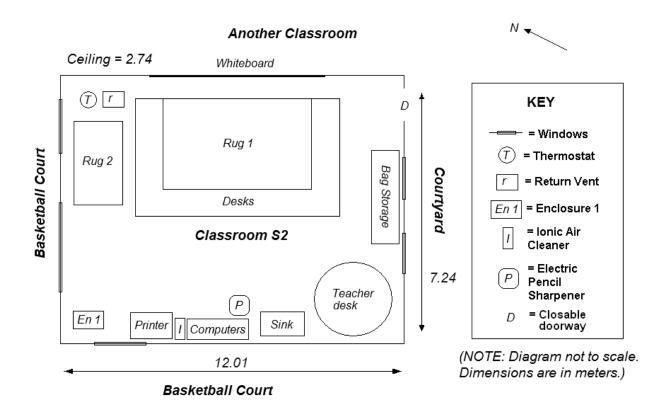


Figure C.9. Classroom floor plan for site S2.

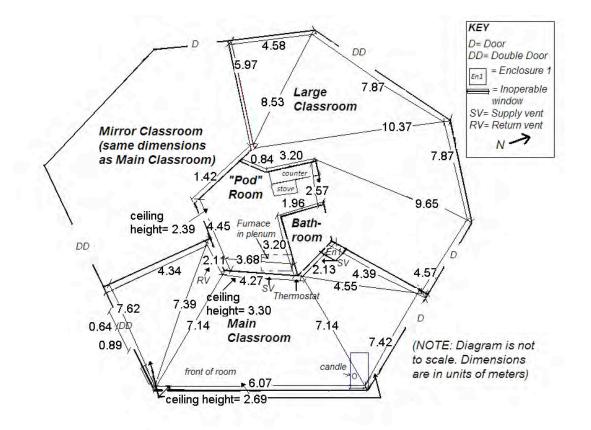


Figure C.10. Classroom floor plan for site S3.

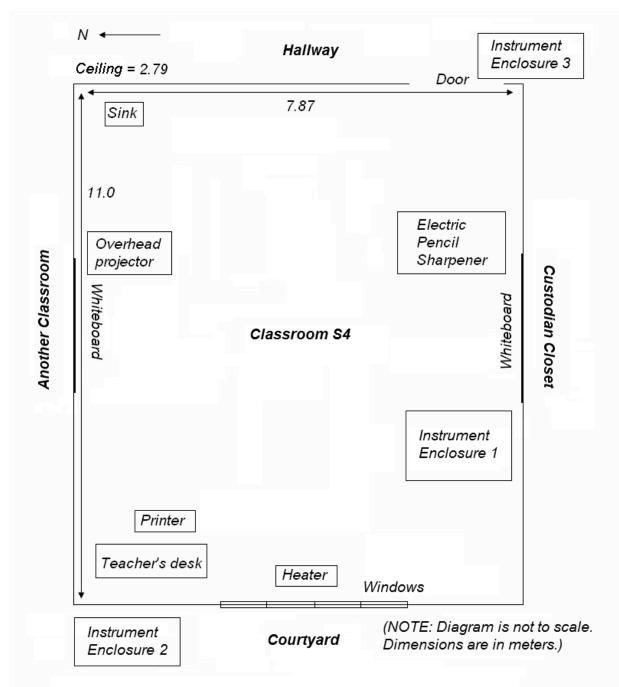


Figure C.11. Classroom floor plan for site S4.

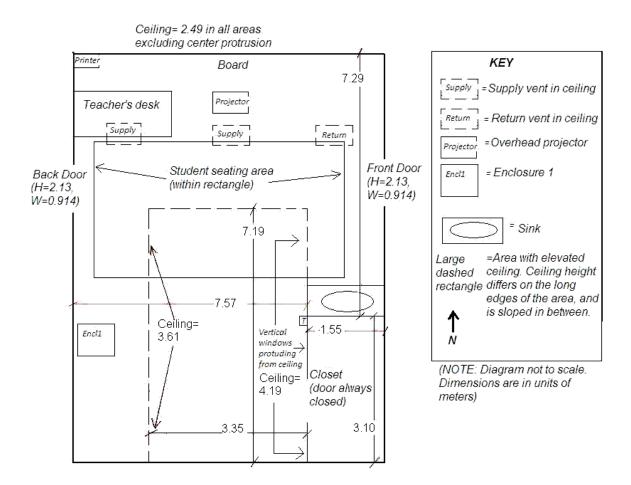


Figure C.12. Classroom floor plan for site S5.

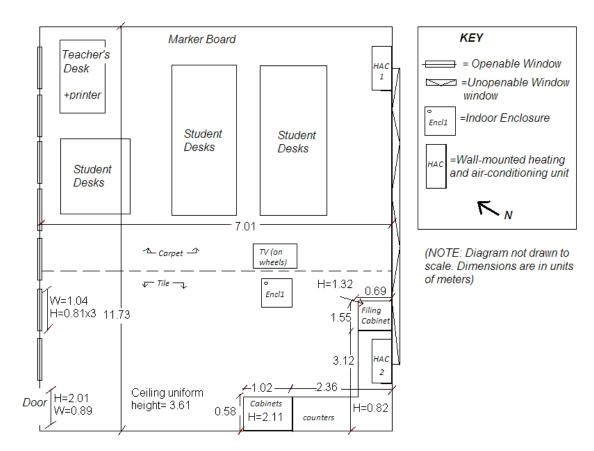


Figure C.13. Classroom floor plan for site S6.

## **APPENDIX D. TIME-SERIES PLOTS AT HOUSE SITES**

In this appendix, we present time-series plots from observational monitoring at the seven house sites. In §3.2, time-series plots were presented for each house showing the particle number concentrations on a linear scale. Those figures are not repeated here. Table B.1 lists the thirteen sets of parameters that are plotted. Generally, these are organized into thirteen figures per house with one house per section. At site H0, because the monitoring extended over almost six days, the time-series data are organized into pairs of plots, covering, respectively, the first half and second half of the monitoring period. In addition, at a few sites, we acquired sensor temperature or door and window position sensor data for more than three parameters and have organized the presentation of the respective data with figures in two parts.

In the narrative portion of each section, basic information is presented about the dates and times of monitoring. In each figure, the time scale is represented as elapsed time (hours), referenced to the start of monitoring. At some of the sites, monitoring proceeded continuously for the entire observational-monitoring period. However, at other sites, there were interruptions in the monitoring period. In these cases, a solid vertical line marks a break and, in effect, the elapsed time clock stops until monitoring resumes. The dashed vertical lines in each figure mark midnight and the calendar date is noted along the top horizontal axis. Note that sites H0 and H1 were monitored in 2007; the other sites were monitored in 2008.

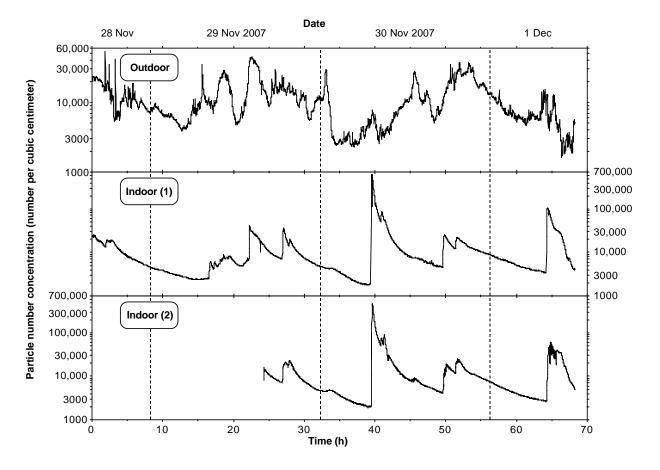
| each house site.   |
|--|
| Particle number concentration, logarithmic (primary indoor, secondary indoor, outdoor)       |
| Particle number ratios (primary indoor/outdoor, secondary indoor/outdoor, secondary/primary) |
| Occupancy  |
| Ozone concentration (outdoor, indoor)  |
| Nitric oxide concentration (outdoor, indoor)   |
| Carbon dioxide (outdoor, indoor) <sup>a</sup>  |
| Carbon monoxide (outdoor, indoor)  |
| Temperature (outdoor, indoor)  |
| Relative humidity (outdoor, indoor)  |
| Sensor temperature   |
| Door and window position sensors   |
| Central station air pollutants (ozone, NO, CO, PM <sub>2.5</sub> )                           |
| Central station meteorology (wind speed, wind direction)                                     |

**Table D.1.** Time-series plots presented in this appendix, based on observational monitoring at each house site.

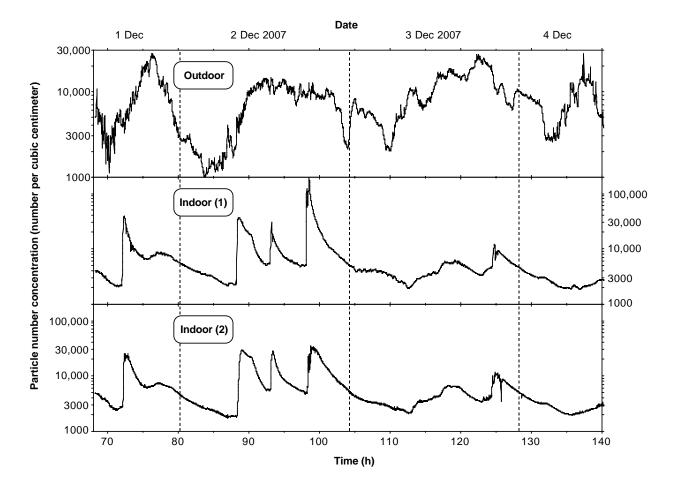
<sup>a</sup> Although outdoor carbon dioxide levels were measured, the Q-Trak appears to exhibit temperature sensitivity that makes it unreliable for quantifying the fairly narrow range of outdoor concentrations expected. Outdoor  $CO_2$  levels are presented here based on Q-Trak measurements, but these measurements are not deemed reliable. In analysis, whenever an outdoor value was needed for a calculation result, it was assumed to be the global average background level of 380 ppm.

#### D.1 Site H0

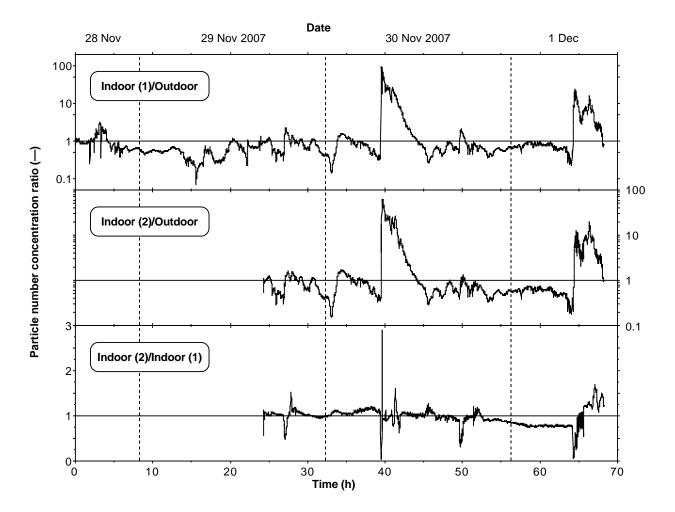
Observational monitoring at site H0 commenced at 15:45 on 28 November 2007 and terminated at 12:00 on 4 December 2007. The time-series figures are organized into parts a and b, which respectively cover the first and second halves of the observational monitoring periods.



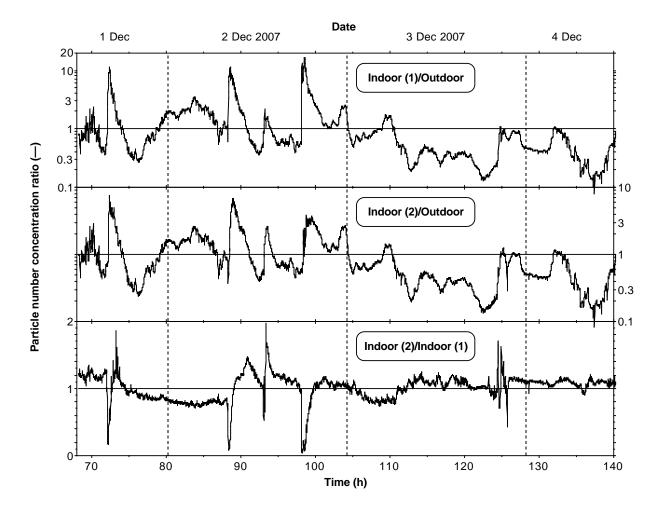
**Figure D.1a.** Time series of particle number concentrations plotted on a logarithmic scale for the first half of observational monitoring at site H0.



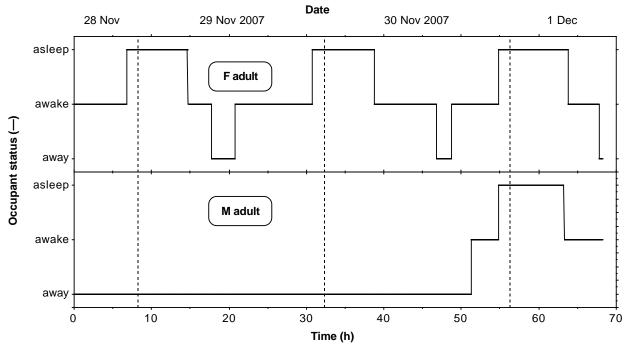
**Figure D.1b.** Time series of particle number concentrations plotted on a logarithmic scale for the second half of observational monitoring at site H0.



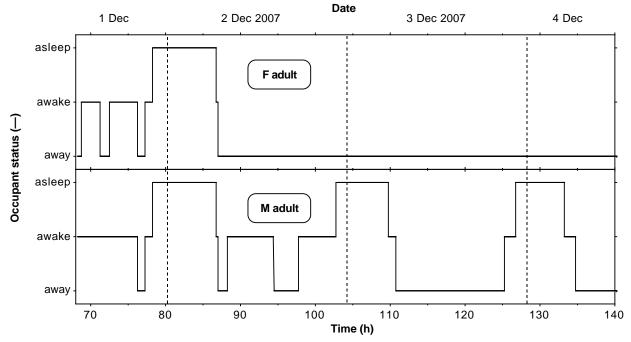
**Figure D.2a.** Time series of ratios of particle number concentrations for the first half of observational monitoring at site H0.



**Figure D.2b.** Time series of ratios of particle number concentrations for the second half of observational monitoring at site H0.



**Figure D.3a.** Time series data for occupancy status during the first half of observational monitoring at site H0.



**Figure D.3b.** Time series data for occupancy status during the second half of observational monitoring at site H0.

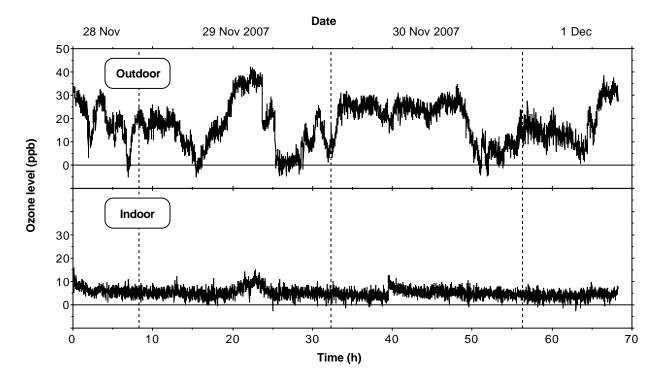
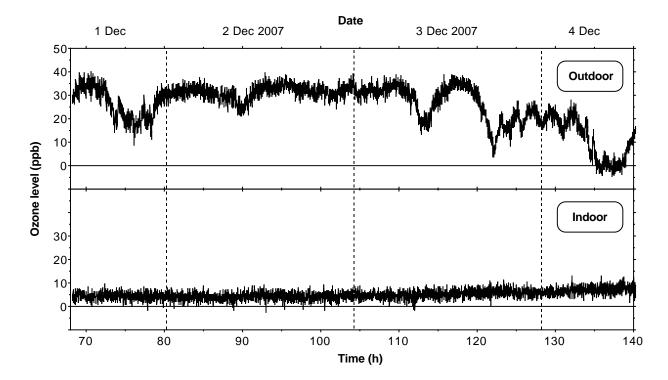
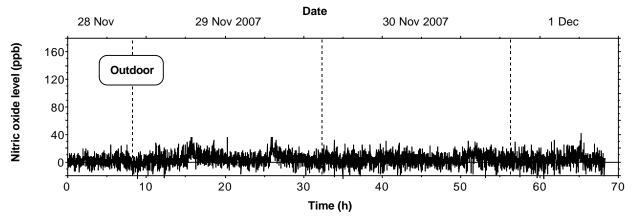


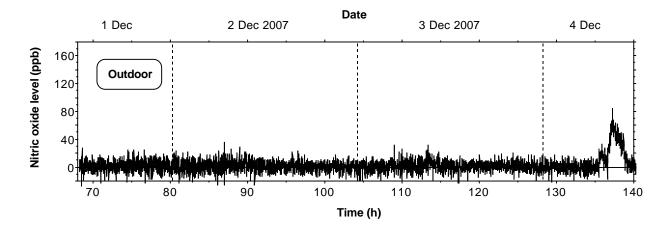
Figure D.4a. Time series of ozone levels for the first half of observational monitoring at site H0.



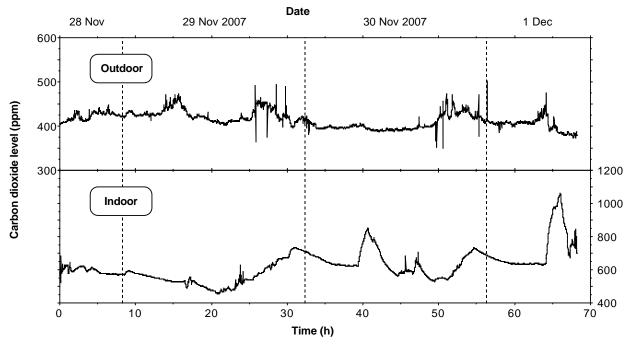
**Figure D.4b.** Time series of ozone levels for the second half of observational monitoring at site H0.



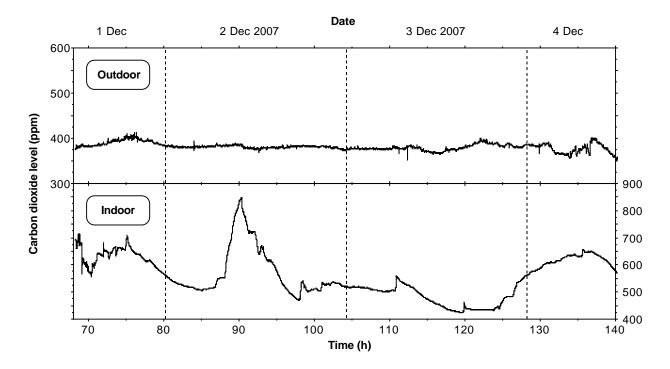
**Figure D.5a.** Time series of nitric oxide levels for the first half of observational monitoring at site H0.



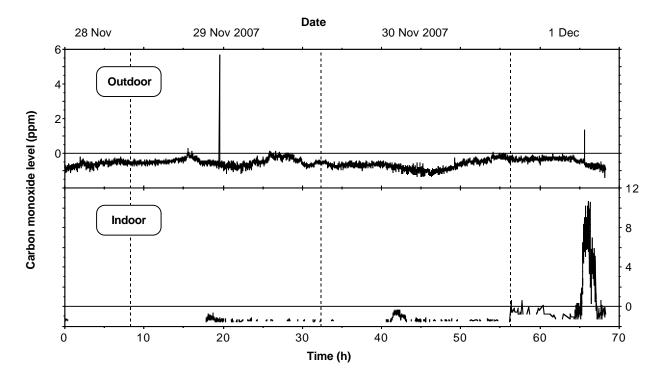
**Figure D.5b.** Time series of nitric oxide levels for the second half of observational monitoring at site H0.



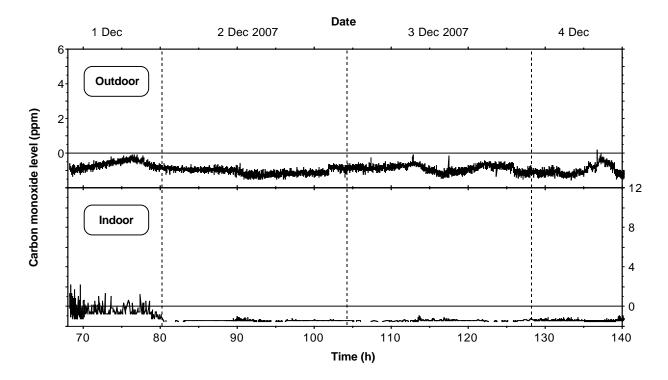
**Figure D.6a.** Time series of carbon dioxide levels for the first half of observational monitoring at site H0.



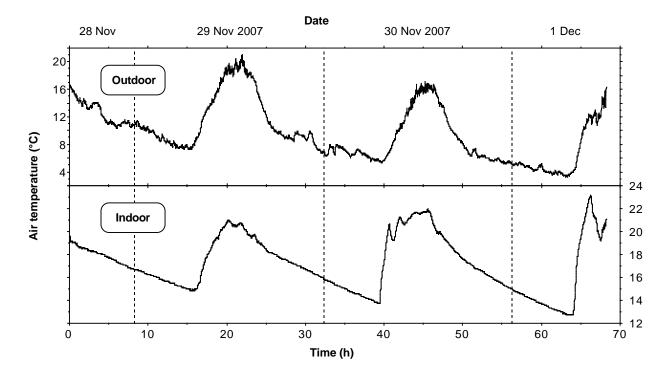
**Figure D.6b.** Time series of carbon dioxide levels for the second half of observational monitoring at site H0.



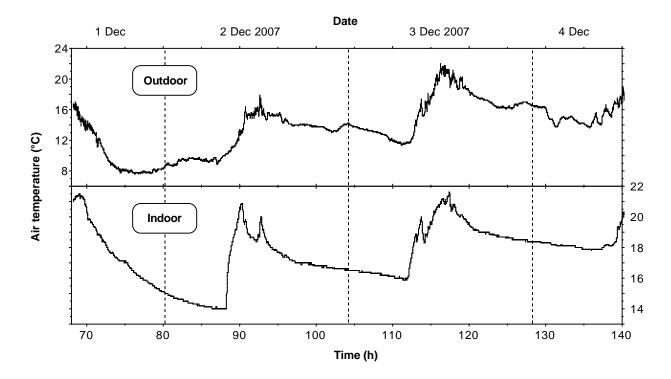
**Figure D.7a.** Time series of carbon monoxide levels for the first half of observational monitoring at site H0.



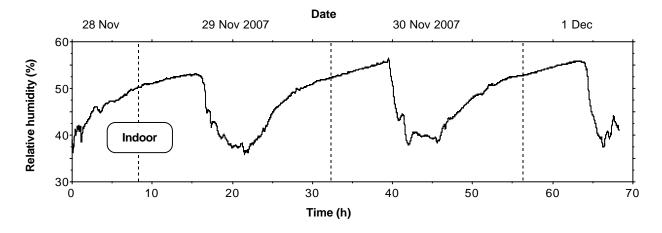
**Figure D.7b.** Time series of carbon monoxide levels for the second half of observational monitoring at site H0.



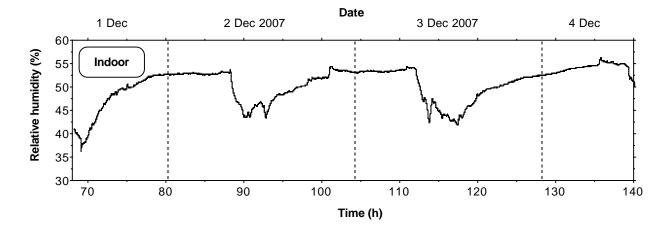
**Figure D.8a.** Time series of air temperatures for the first half of observational monitoring at site H0.



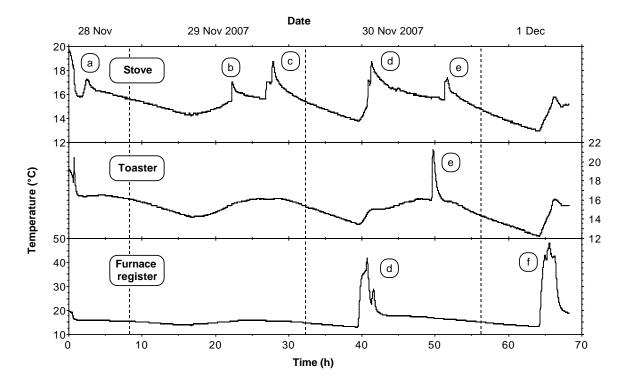
**Figure D.8b.** Time series of air temperatures for the second half of observational monitoring at site H0.



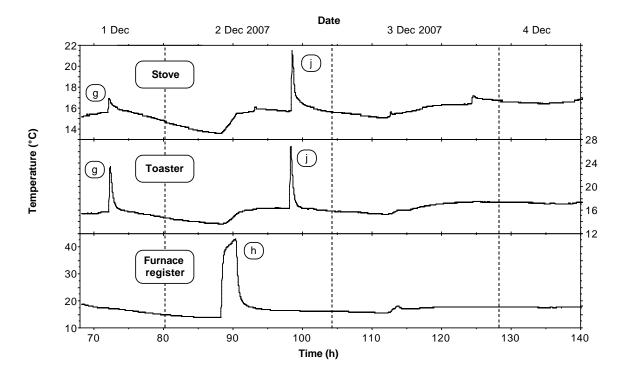
**Figure D.9a.** Time series of indoor relative humidity for the first half of observational monitoring at site H0.



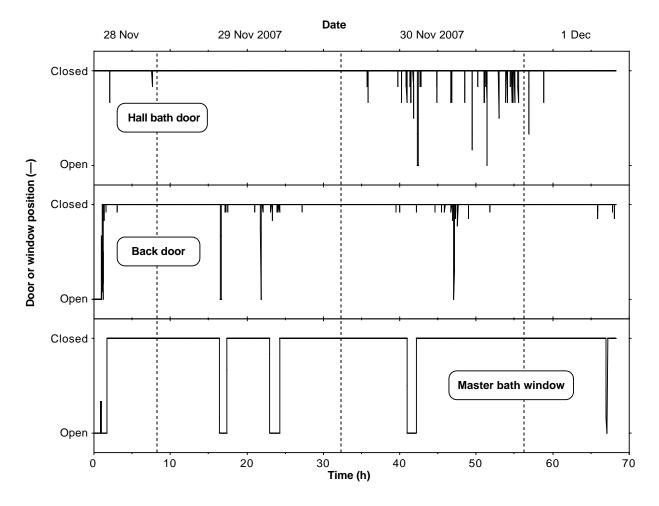
**Figure D.9b.** Time series of indoor relative humidity for the second half of observational monitoring at site H0.



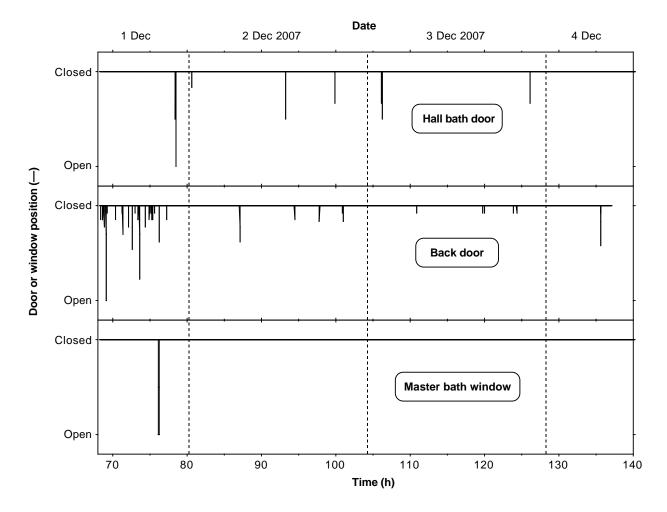
**Figure D.10a.** Time series of sensor temperatures for the first half of observational monitoring at site H0. The peak labels correspond to PN concentration peaks discussed in §3.2.1.



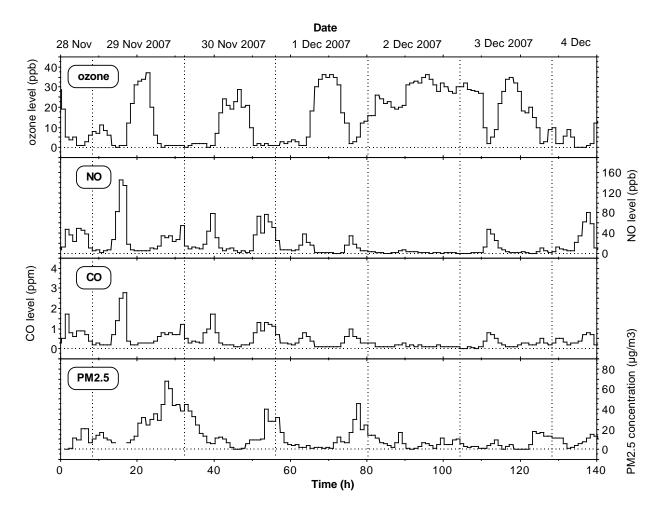
**Figure D.10b.** Time series of sensor temperatures for the second half of observational monitoring at site H0. The peak labels correspond to PN concentration peaks discussed in §3.2.1.



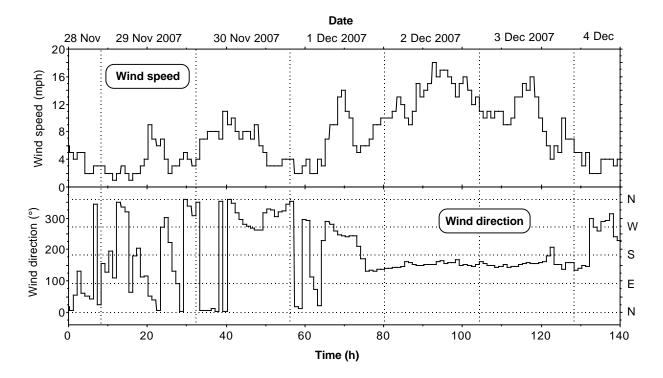
**Figure D.11a.** Time series of door and window positions for the first half of observational monitoring at house site H0. Vertical spikes in the plot denote changes in the open or closed state that persisted for less than a clock minute.



**Figure D.11b.** Time series of door and window positions for the second half of observational monitoring at house site H0. Vertical spikes in the plot denote changes in the open or closed state that persisted for less than a clock minute.



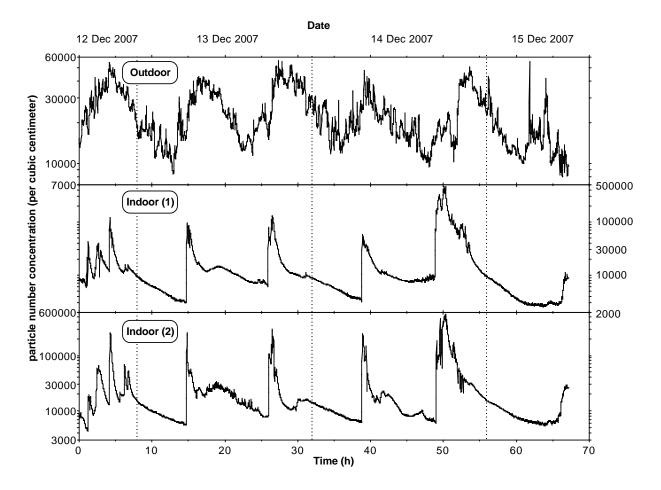
**Figure D.12.** Time series of pollutant concentrations based on central-station monitoring for the period of observational monitoring at house site H0.



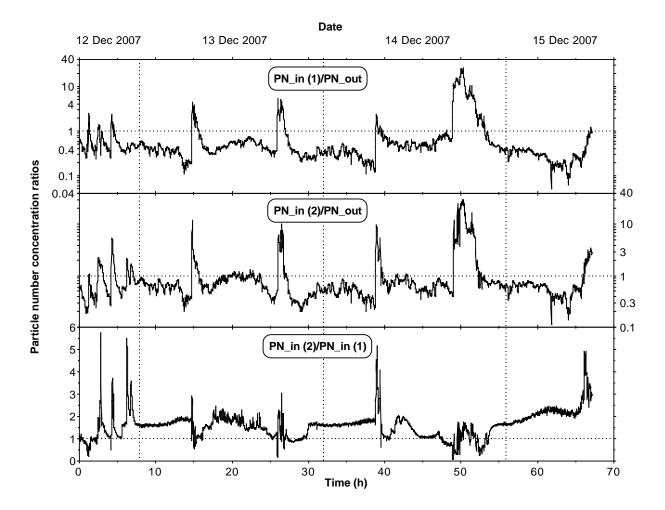
**Figure D.13.** Time series of wind speed and wind direction based on central-station monitoring for the period of observational monitoring at house site H0

## D.2 Site H1

Observational monitoring at site H1 commenced at 16:04 on 12 December 2007 and terminated at 11:15 on 15 December 2007.



**Figure D.14.** Time series of particle number concentrations plotted on a logarithmic scale for observational monitoring at site H1.



**Figure D.15.** Time series of ratios of particle number concentrations for observational monitoring at site H1.

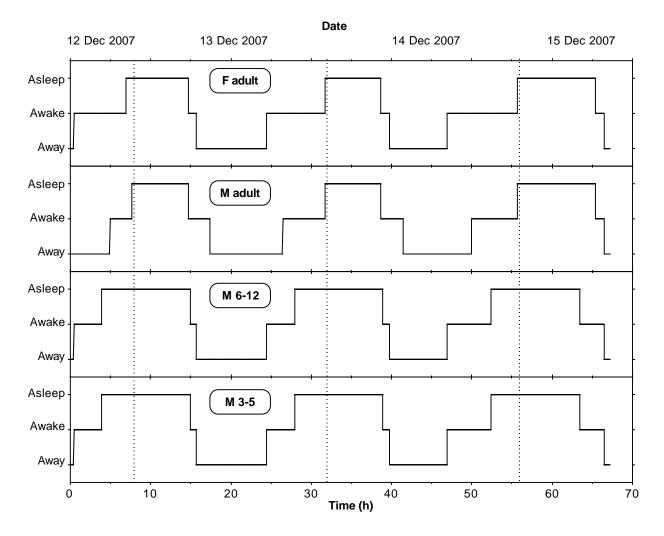


Figure D.16. Time series data for occupancy status during observational monitoring at site H1.

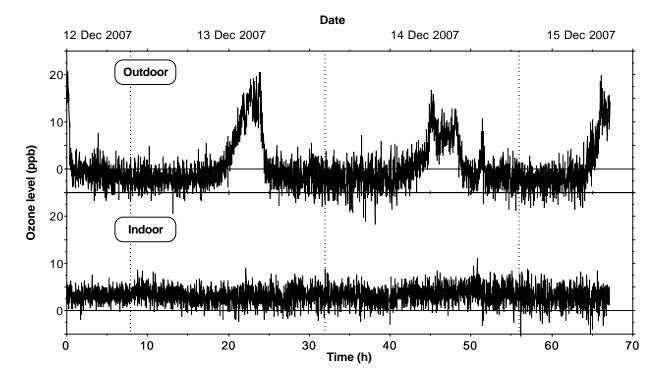
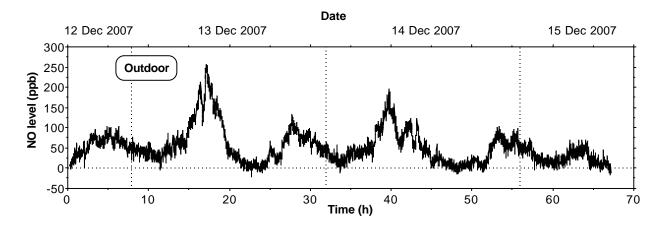


Figure D.17. Time series of ozone levels for observational monitoring at site H1.



**Figure D.18.** Time series of nitric oxide levels for observational monitoring at site H1. No instrument was available for monitoring indoor NO levels at this site.

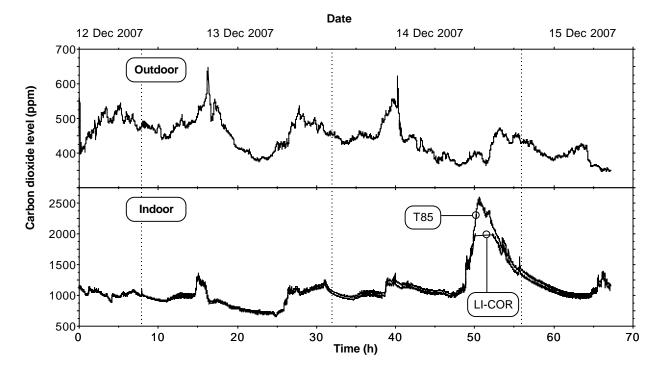


Figure D.19. Time series of carbon dioxide levels for observational monitoring at site H1.

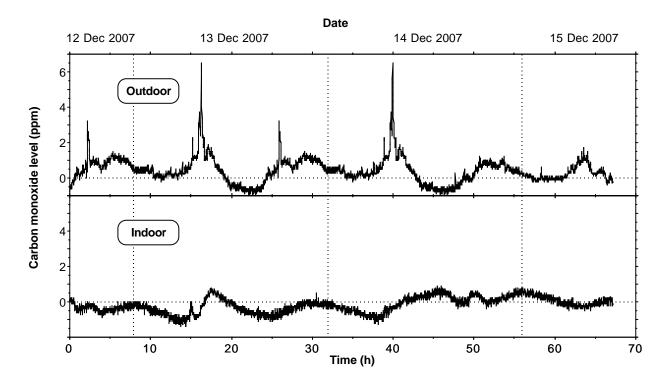


Figure D.20. Time series of carbon monoxide levels for observational monitoring at site H1.

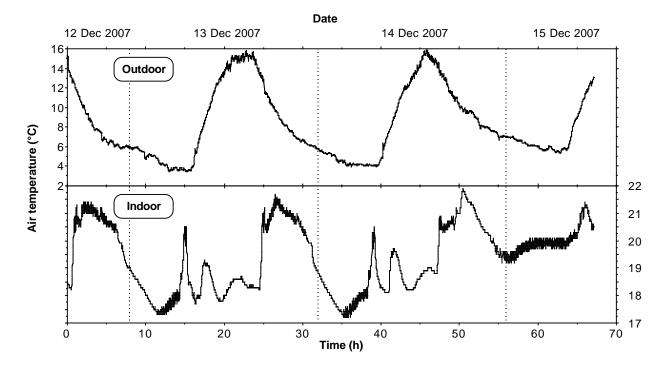


Figure D.21. Time series of air temperatures for observational monitoring at site H1.

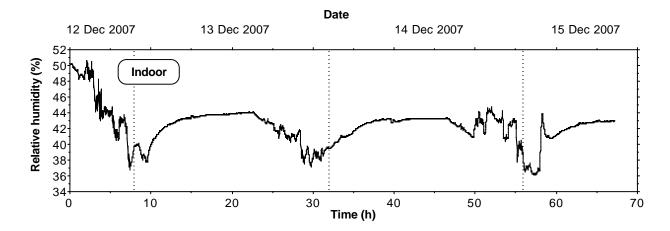


Figure D.22. Time series of indoor relative humidity for observational monitoring at site H1.

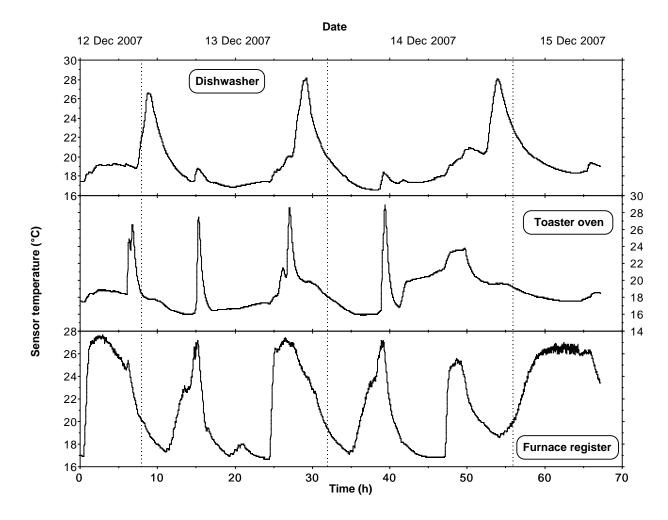
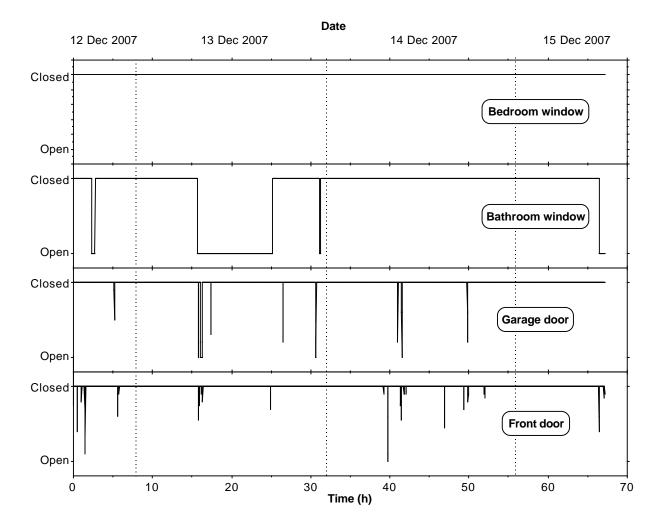
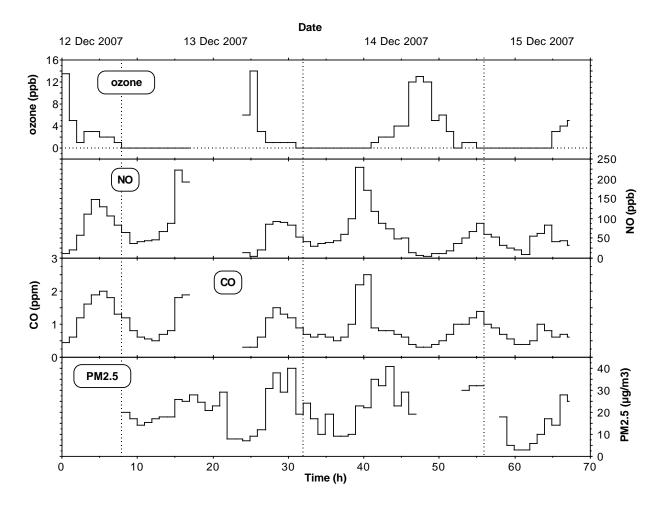


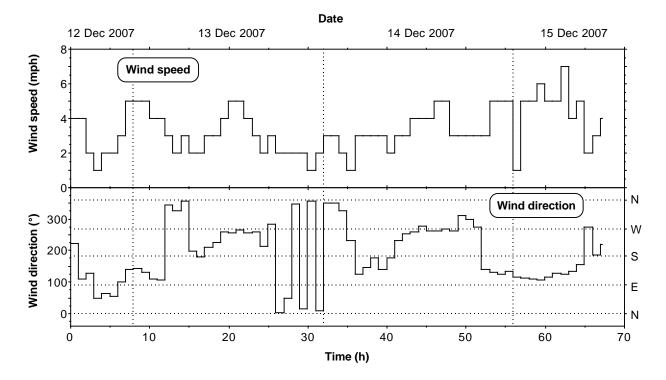
Figure D.23. Time series of sensor temperatures for observational monitoring at site H1.



**Figure D.24.** Time series of door and window positions for observational monitoring at site H1. Vertical spikes in the plot denote changes in the open or closed state that persisted for less than a clock minute.



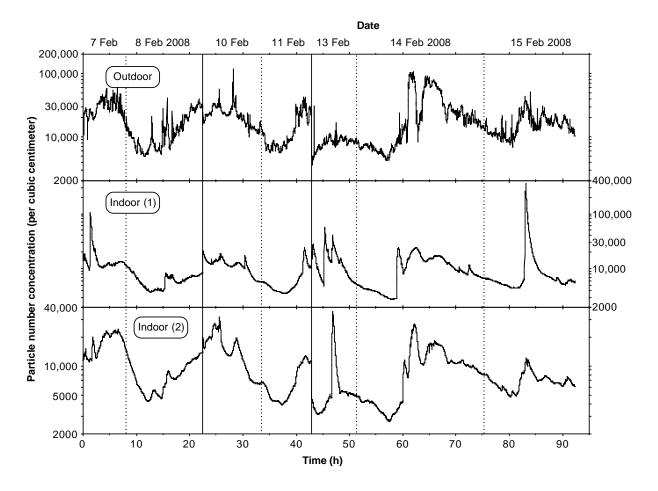
**Figure D.25.** Time series of pollutant concentrations based on central-station monitoring for the period of observational monitoring at house site H1.



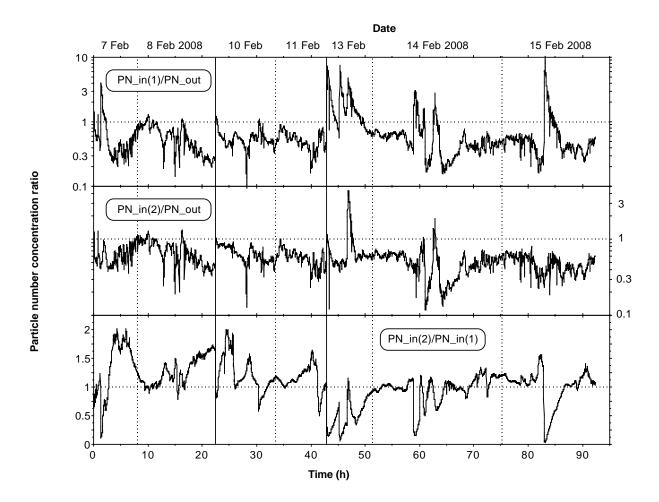
**Figure D.26.** Time series of wind speed and wind direction based on central-station monitoring for the period of observational monitoring at house site H1.

## D.3 Site H2

Observational monitoring at site H2 was conducted over three periods for a total of 92 hours. The first period commenced at 16:01 on 7 February 2008 and ended at 14:30 on 8 February 2008. The second period commenced at 13:04 on 10 February 2008 and terminated at 9:24 on 11 February 2008. The third period commenced at 15:27 on 13 February 2008 and ended at 16:54 on 15 February 2008.



**Figure D.27.** Time series of particle number concentrations plotted on a logarithmic scale for observational monitoring at site H2.



**Figure D.28.** Time series of ratios of particle number concentrations for observational monitoring at site H2.

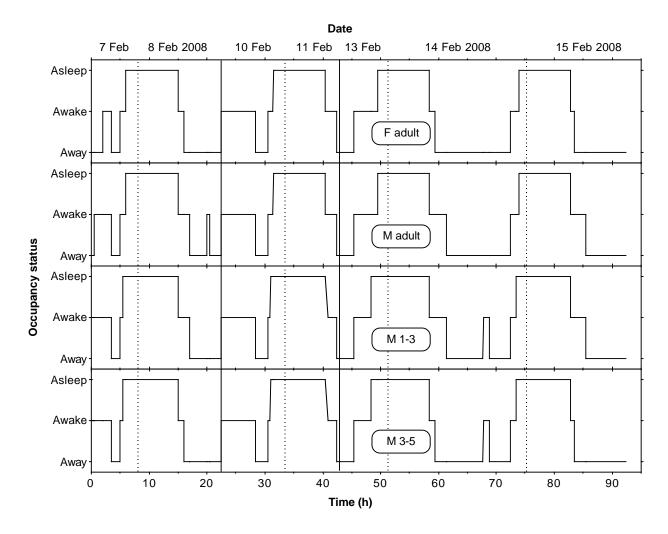


Figure D.29. Time series data for occupancy status during observational monitoring at site H2.

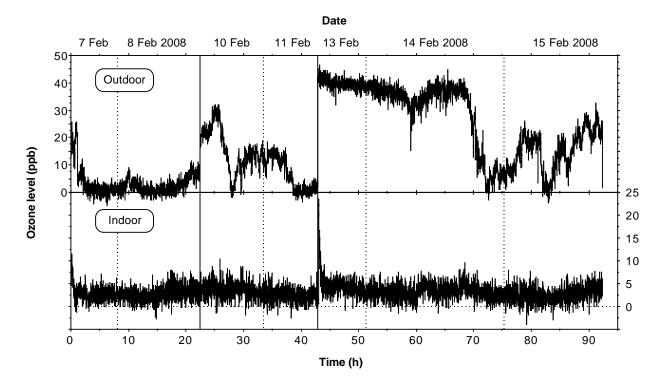


Figure D.30. Time series of ozone levels for observational monitoring at site H2.

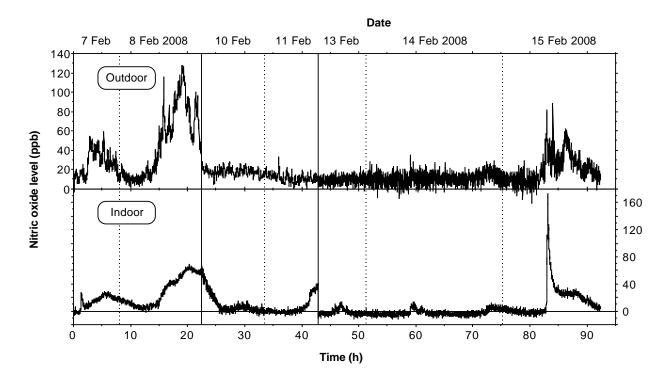


Figure D.31. Time series of nitric oxide levels for observational monitoring at site H2.

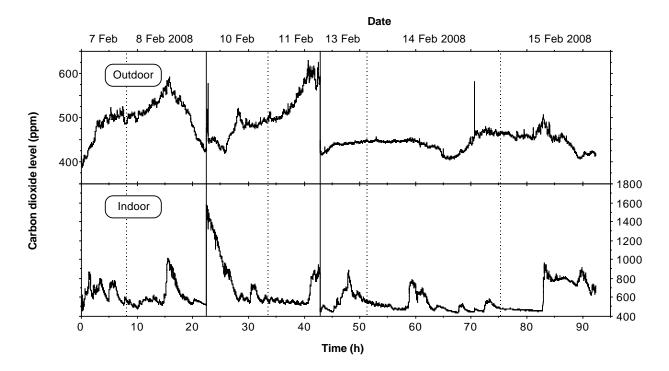


Figure D.32. Time series of carbon dioxide levels for observational monitoring at site H2.

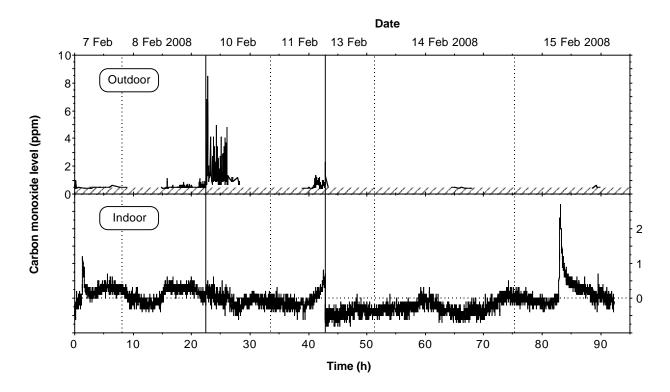


Figure D.33. Time series of carbon monoxide levels for observational monitoring at site H2.

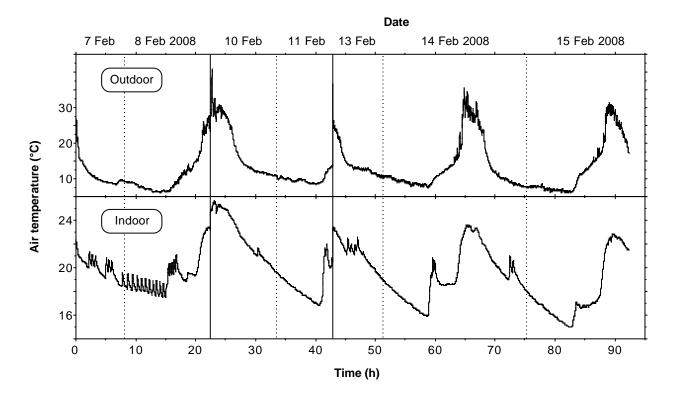


Figure D.34. Time series of air temperatures for observational monitoring at site H2.

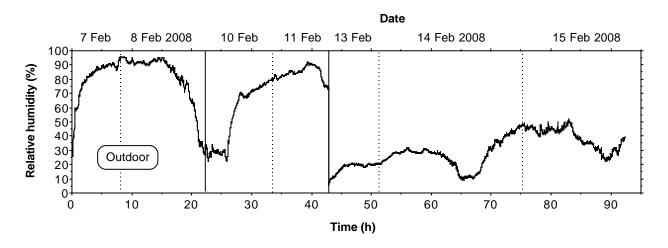


Figure D.35. Time series of outdoor relative humidity for observational monitoring at site H2.

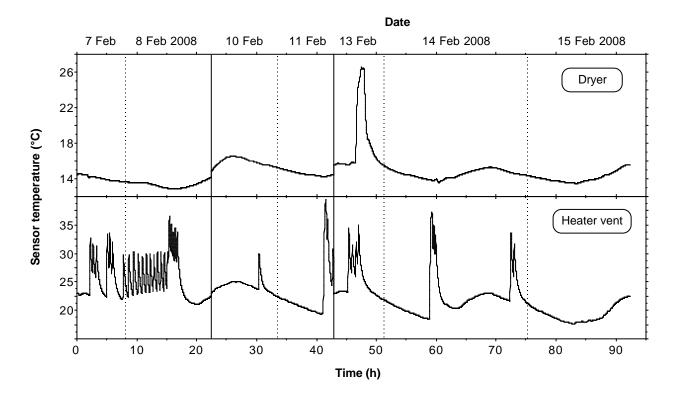
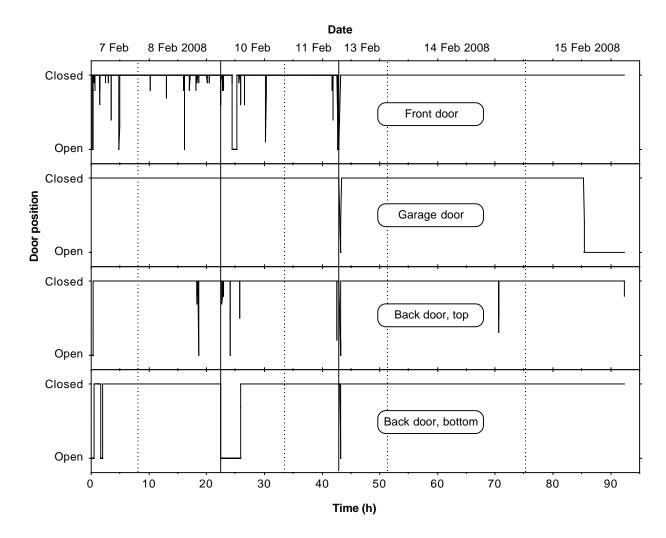
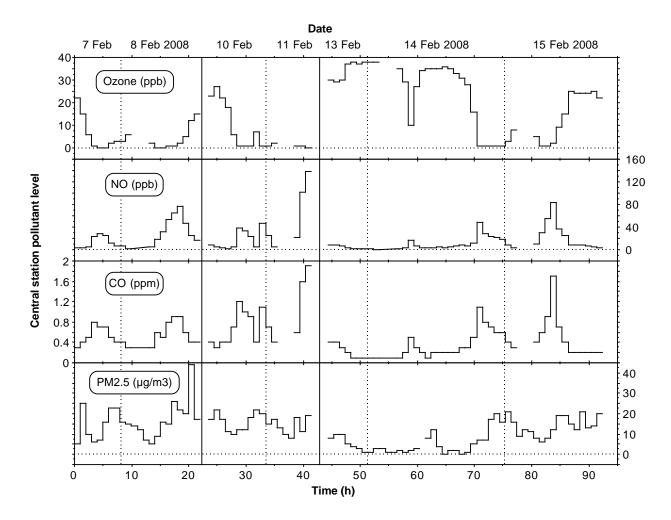


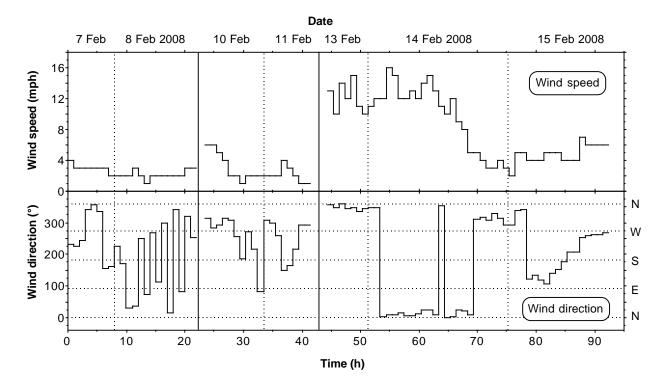
Figure D.36. Time series of sensor temperatures for observational monitoring at site H2.



**Figure D.37.** Time series of door positions for observational monitoring at site H2. Vertical spikes in the plot denote changes in the open or closed state that persisted for less than a clock minute.



**Figure D.38.** Time series of pollutant concentrations based on central-station monitoring for the period of observational monitoring at house site H2.



**Figure D.39.** Time series of wind speed and wind direction based on central-station monitoring for the period of observational monitoring at house site H2.

## D.4 Site H3

Observational monitoring at site H3 commenced at 13:19 on 13 March 2008 and ended at 18:00 on 16 March 2008.

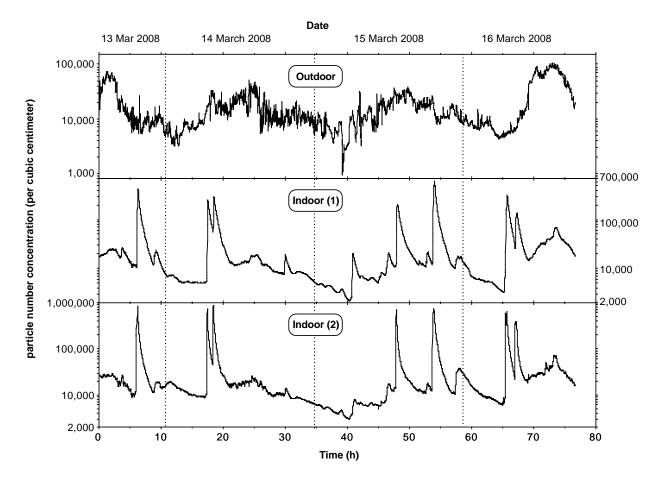
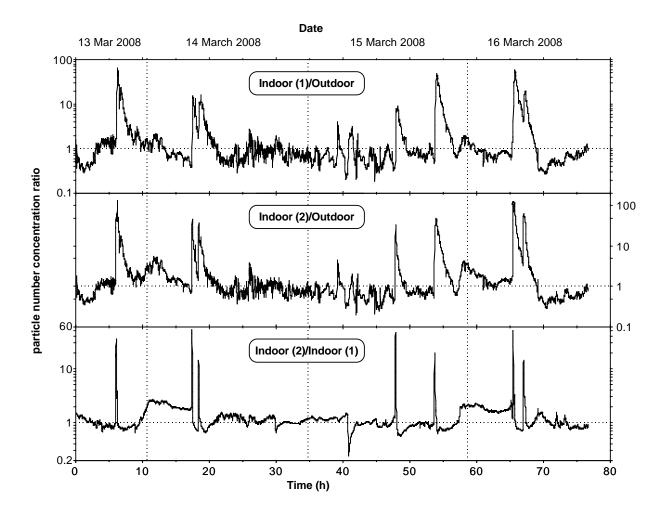


Figure D.40. Time series of particle number concentrations plotted on a logarithmic scale for observational monitoring at site H3.



**Figure D.41.** Time series of ratios of particle number concentrations for observational monitoring at site H3.

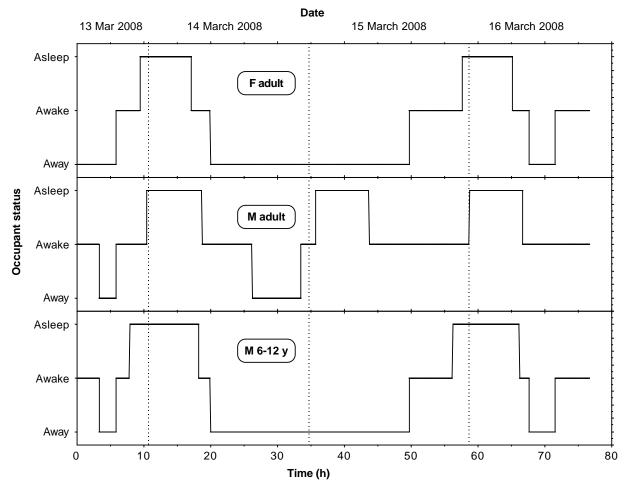


Figure D.42. Time series data for occupancy status during observational monitoring at site H3.

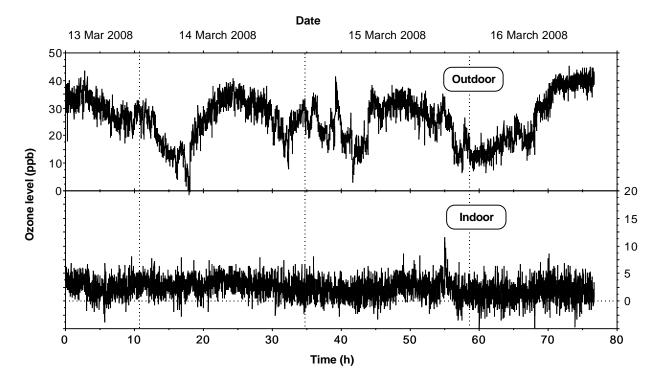


Figure D.43. Time series of ozone levels for observational monitoring at site H3.

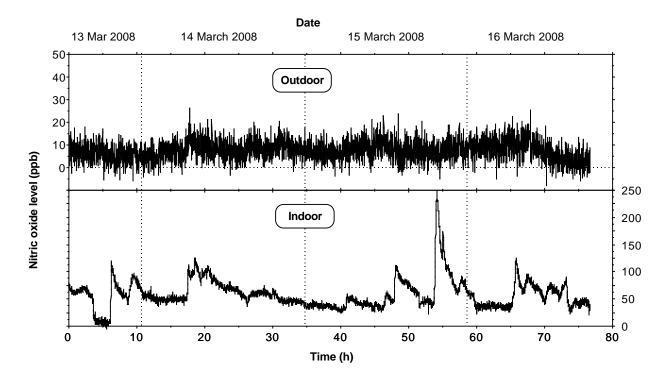


Figure D.44. Time series of nitric oxide levels for observational monitoring at site H3.

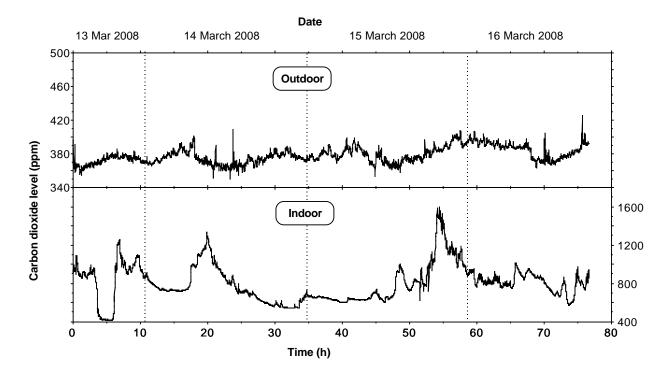


Figure D.45. Time series of carbon dioxide levels for observational monitoring at site H3.

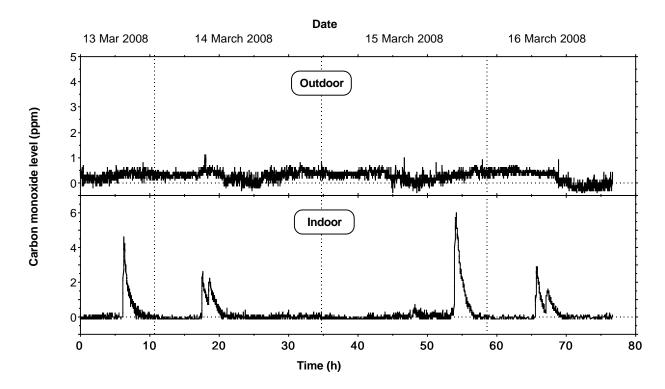


Figure D.46. Time series of carbon monoxide levels for observational monitoring at site H3.

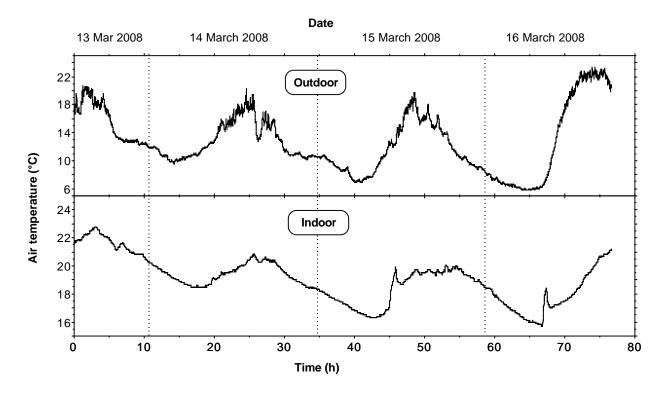


Figure D.47. Time series of air temperatures for observational monitoring at site H3.

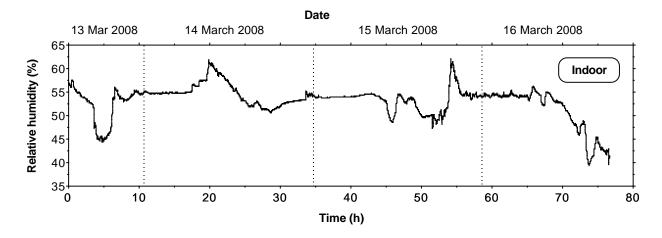


Figure D.48. Time series of indoor relative humidity for observational monitoring at site H3.

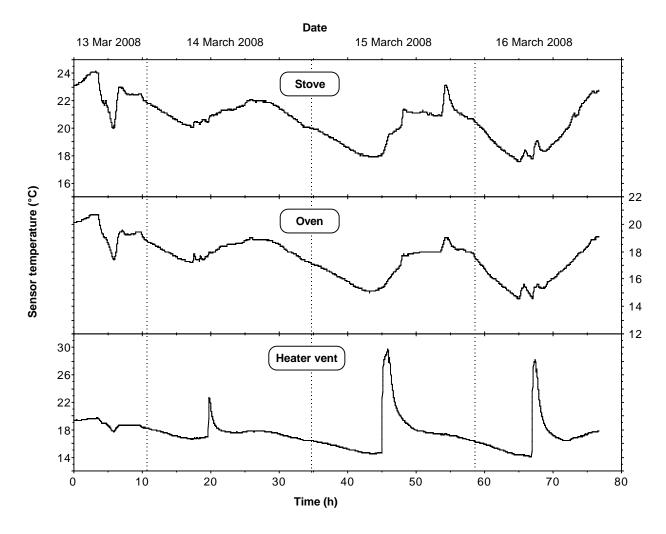
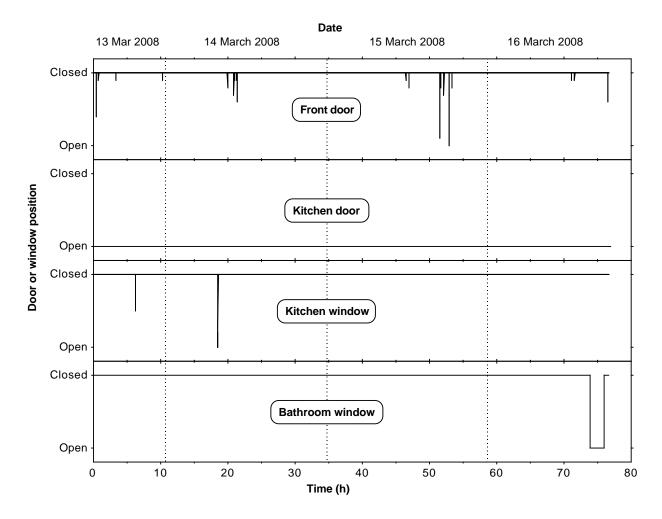
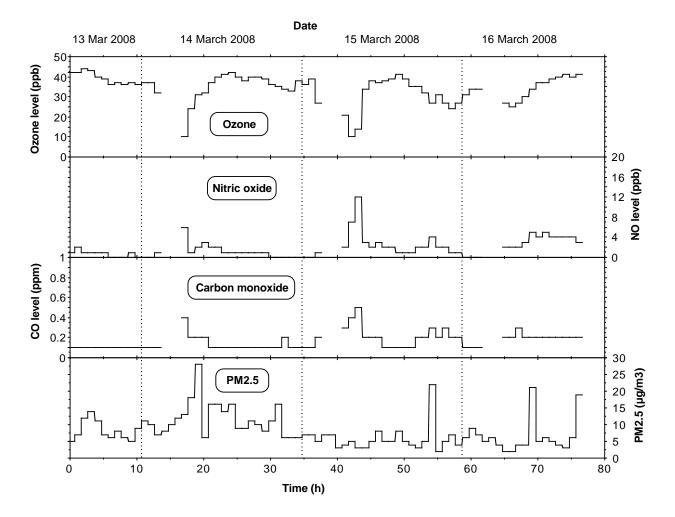


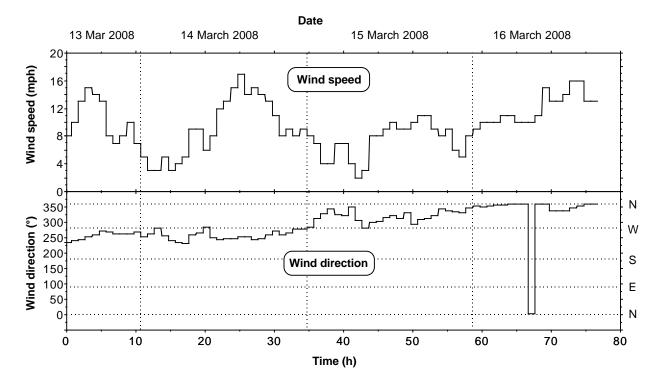
Figure D.49. Time series of sensor temperatures for observational monitoring at site H3.



**Figure D.50.** Time series of door and window positions for observational monitoring at site H3. Vertical spikes in the plot denote changes in the open or closed state that persisted for less than a clock minute.



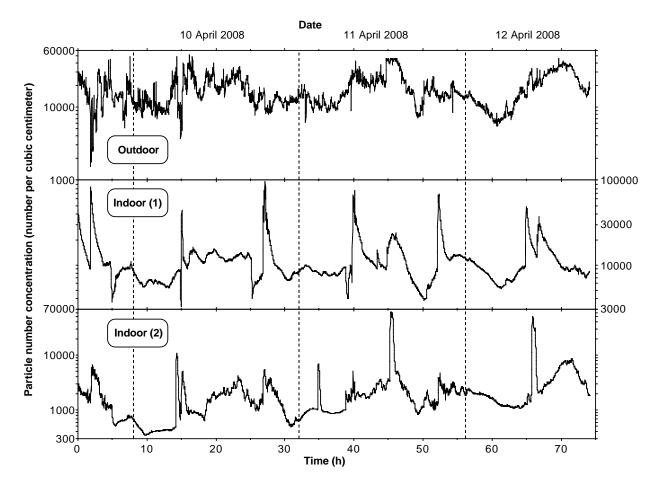
**Figure D.51.** Time series of pollutant concentrations based on central-station monitoring for the period of observational monitoring at house site H3.



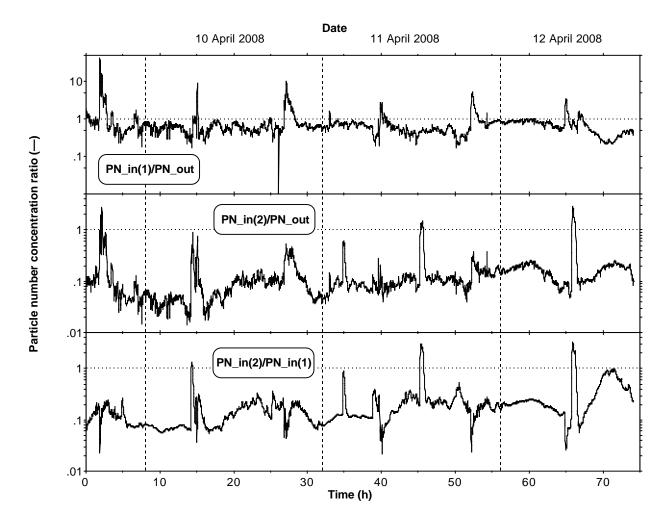
**Figure D.52.** Time series of wind speed and wind direction based on central-station monitoring for the period of observational monitoring at house site H3.

## D.5 Site H4

Observational monitoring was conducted at house site H4 for a continuous 74-h period, commencing at 15:55 on 9 April 2008 and ending at 18:05 on 12 April 2008.



**Figure D.53.** Time series of particle number concentrations plotted on a logarithmic scale for observational monitoring at site H4.



**Figure D.54.** Time series of ratios of particle number concentrations for observational monitoring at site H4.

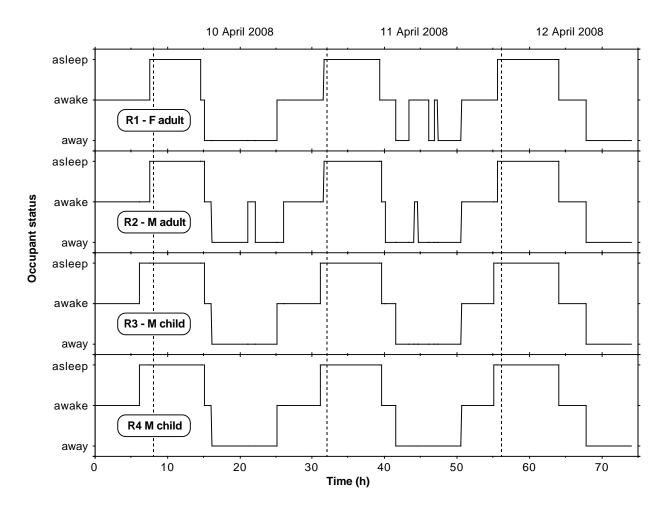


Figure D.55. Time series data for occupancy status during observational monitoring at site H4.

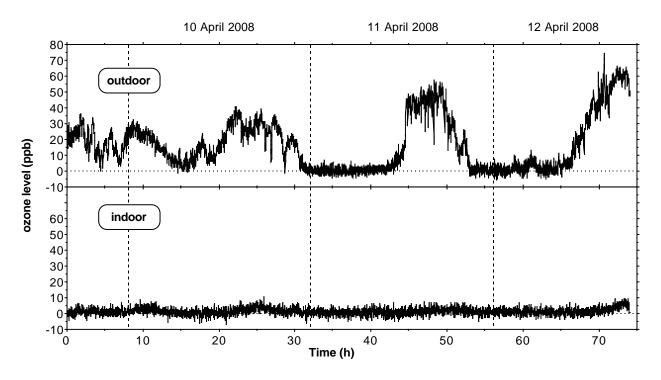


Figure D.56. Time series of ozone levels for observational monitoring at site H4.

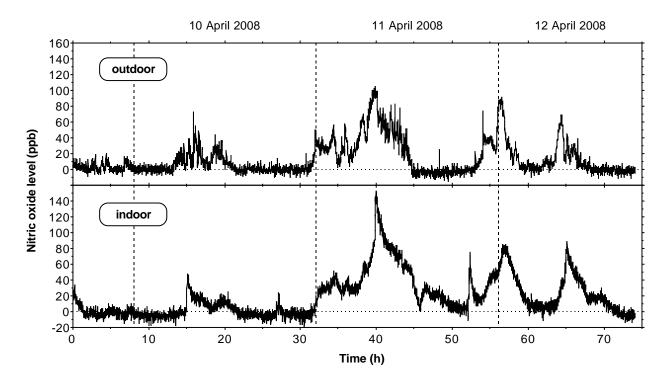


Figure D.57. Time series of nitric oxide levels for observational monitoring at site H4.

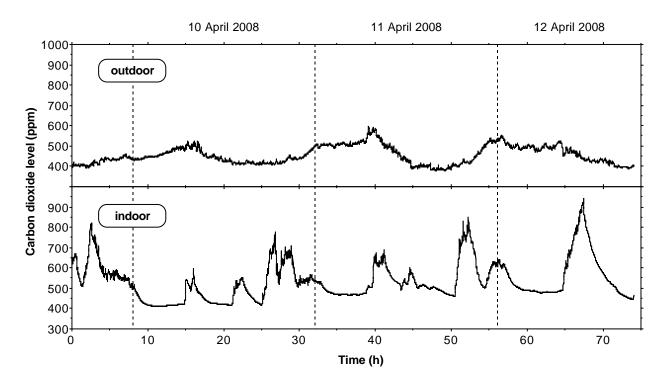


Figure D.58. Time series of carbon dioxide levels for observational monitoring at site H4.

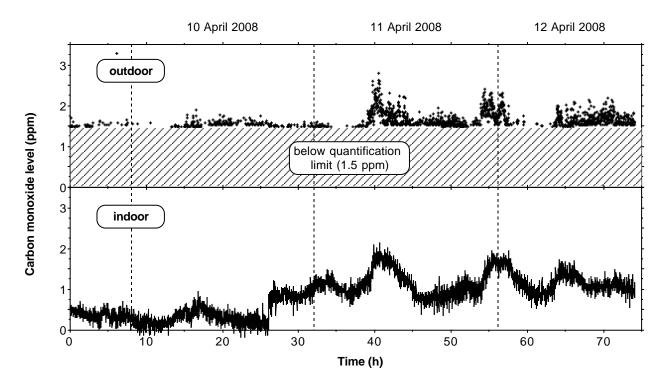


Figure D.59. Time series of carbon monoxide levels for observational monitoring at site H4.

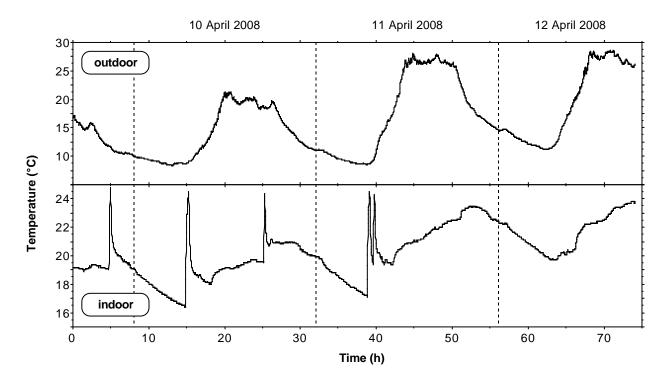


Figure D.60. Time series of air temperatures for observational monitoring at site H4.

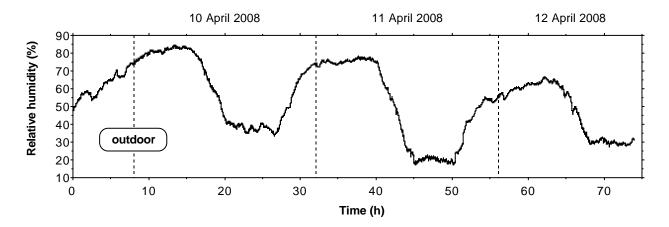


Figure D.61. Time series of outdoor relative humidity for observational monitoring at site H4.

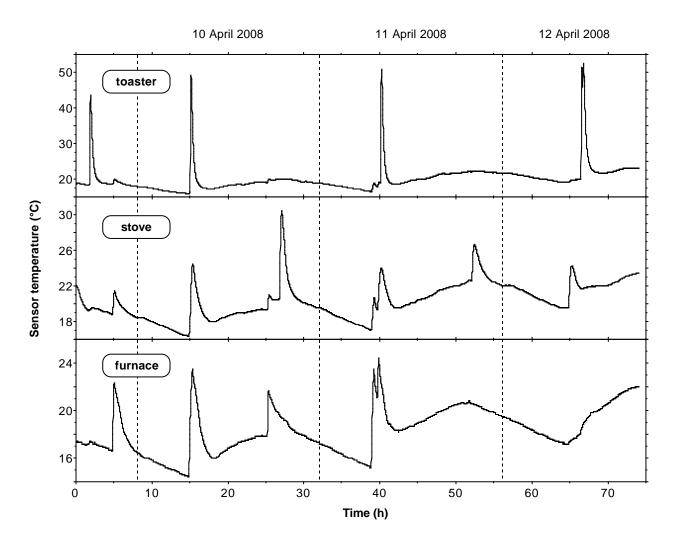
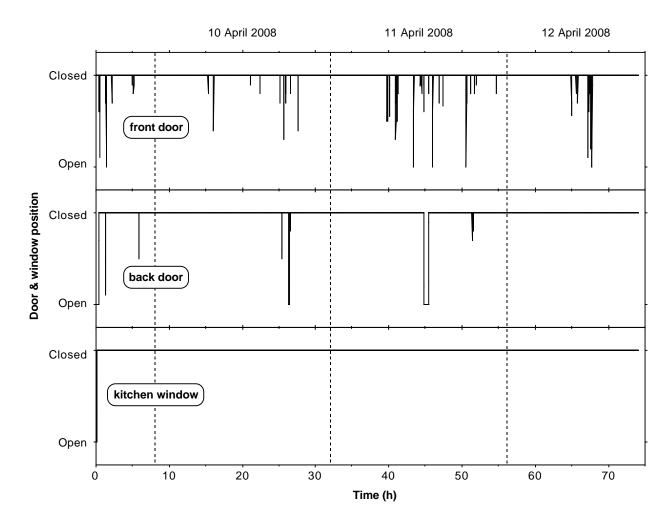
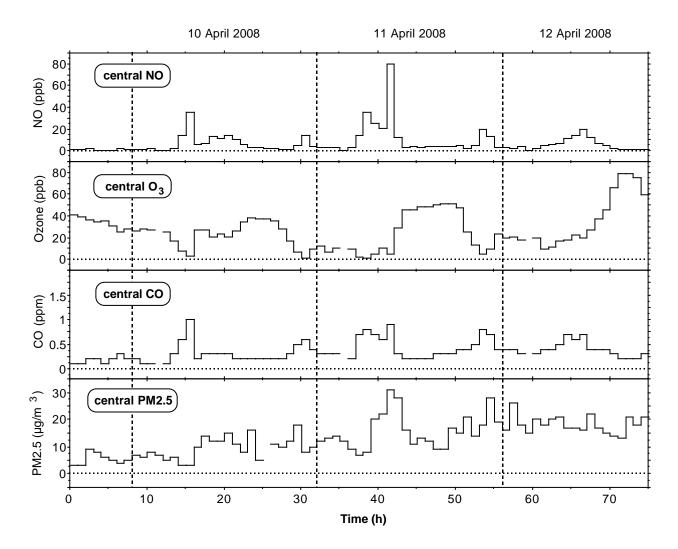


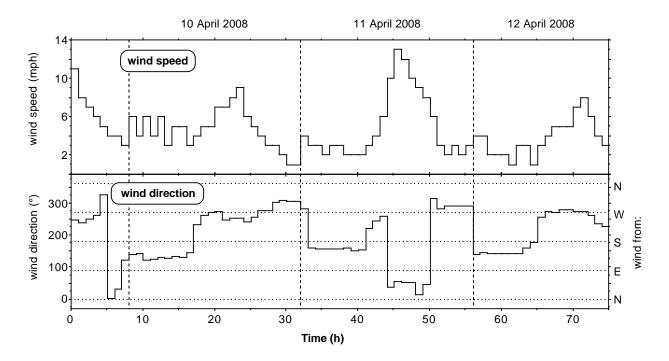
Figure D.62. Time series of sensor temperatures for observational monitoring at site H4.



**Figure D.63.** Time series of door and window positions for observational monitoring at site H4. Vertical spikes in the plot denote changes in the open or closed state that persisted for less than a clock minute.



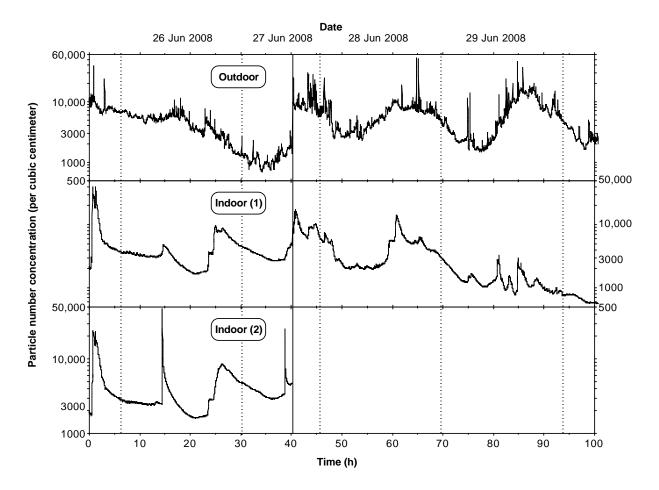
**Figure D.64.** Time series of pollutant concentrations based on central-station monitoring for the period of observational monitoring at house site H4.



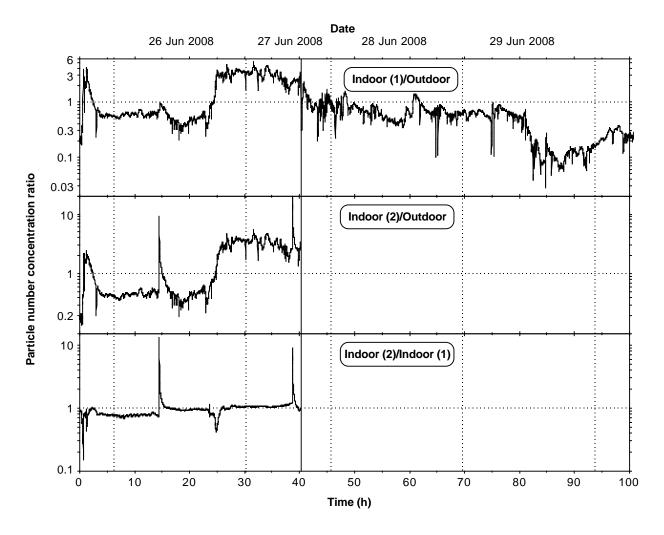
**Figure D.65.** Time series of wind speed and wind direction based on central-station monitoring for the period of observational monitoring at house site H4.

## D.6 Site H5

Observational monitoring commenced at house site H5 at 17:48 on 25 June 2008. There was an interruption of 8.5 h duration beginning at 10:00 on 27 June. Monitoring was terminated at 7:00 on 30 June 2008, so that the cumulative duration of observational monitoring was 100 h.



**Figure D.66.** Time series of particle number concentrations plotted on a logarithmic scale for observational monitoring at site H5. The solid vertical line marks a break in observational monitoring beginning at 10:00 on 27 June caused by a malfunction of the outdoor WCPC. The indoor (2) unit was moved outdoors and monitoring was resumed at 18:30.



**Figure D.67.** Time series of ratios of particle number concentrations for observational monitoring at site H5.

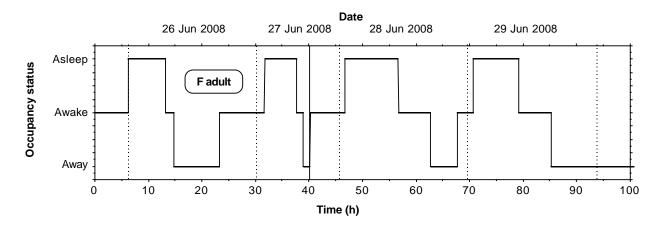


Figure D.68. Time series data for occupancy status during observational monitoring at site H5.

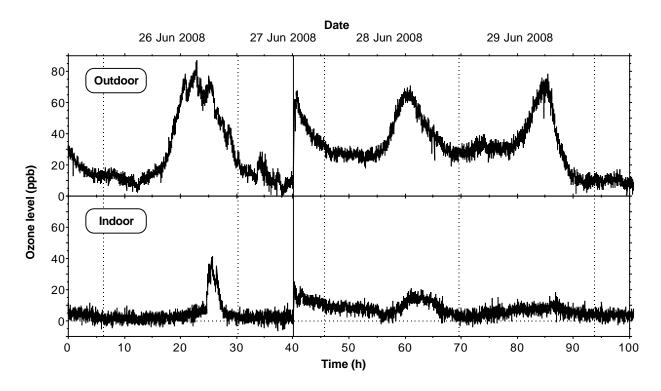


Figure D.69. Time series of ozone levels for observational monitoring at site H5.

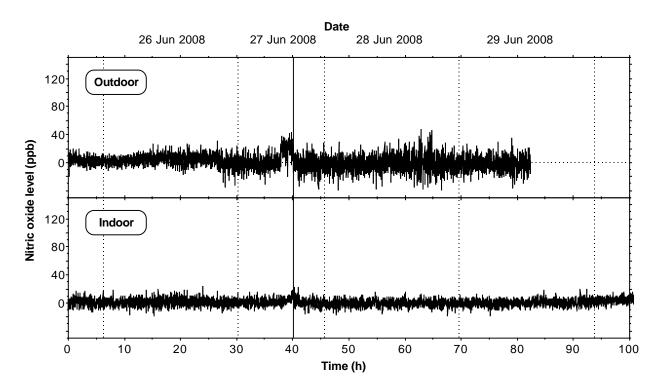


Figure D.70. Time series of nitric oxide levels for observational monitoring at site H5.

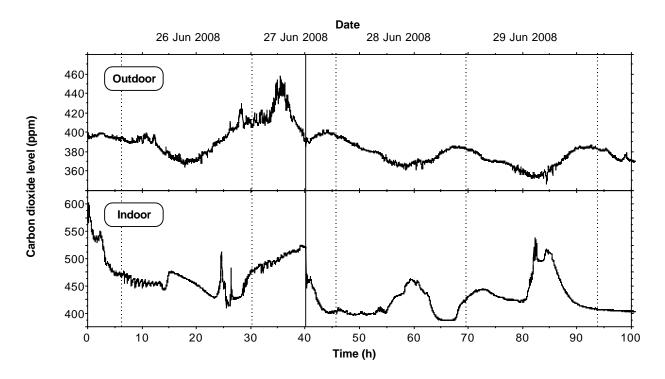


Figure D.71. Time series of carbon dioxide levels for observational monitoring at site H5.

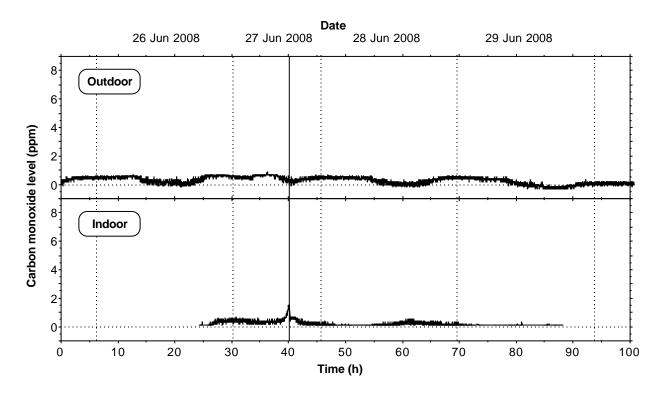


Figure D.72. Time series of carbon monoxide levels for observational monitoring at site H5.

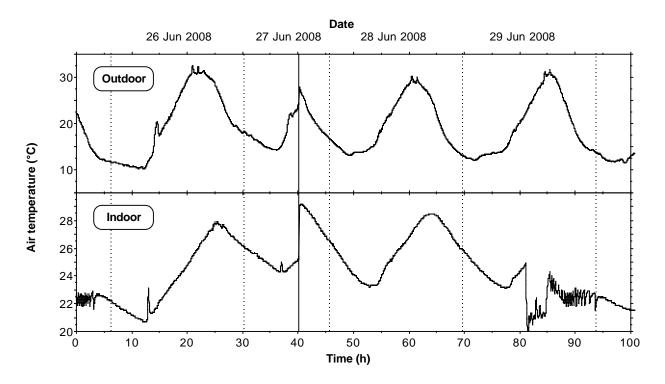


Figure D.73. Time series of air temperatures for observational monitoring at site H5.

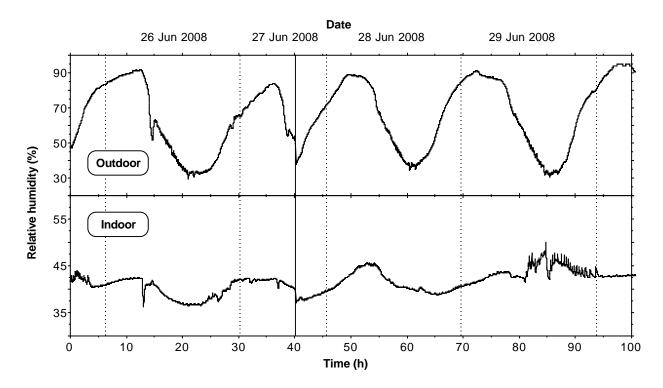


Figure D.74. Time series of relative humidities for observational monitoring at site H5.

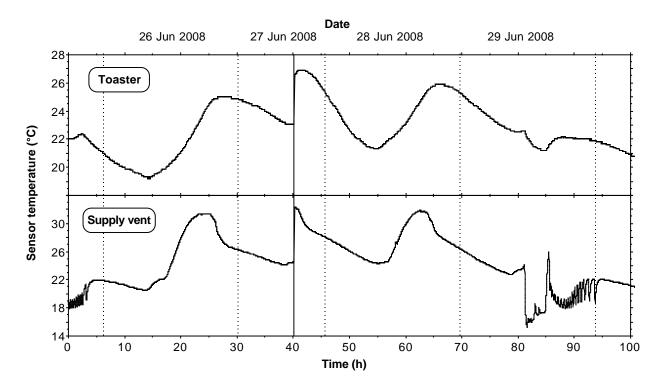


Figure D.75a. Time series of sensor temperatures on toaster and central air supply vent for observational monitoring at site H5.

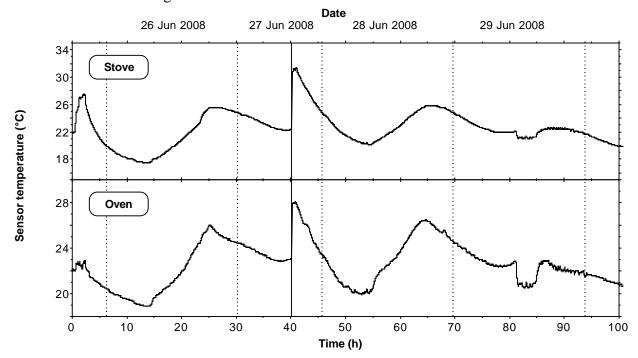
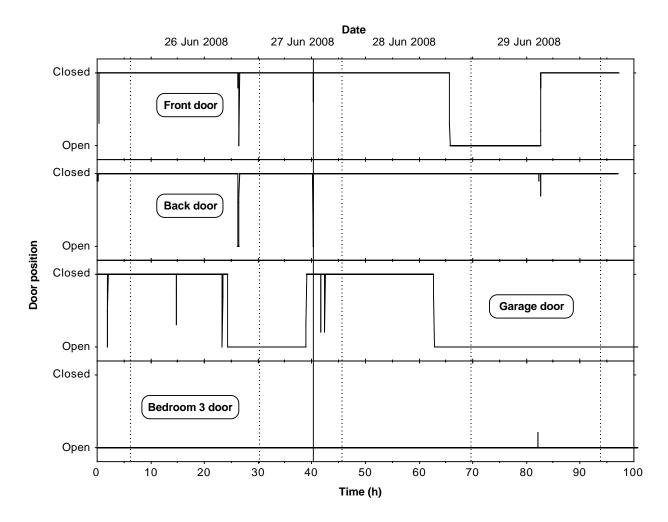
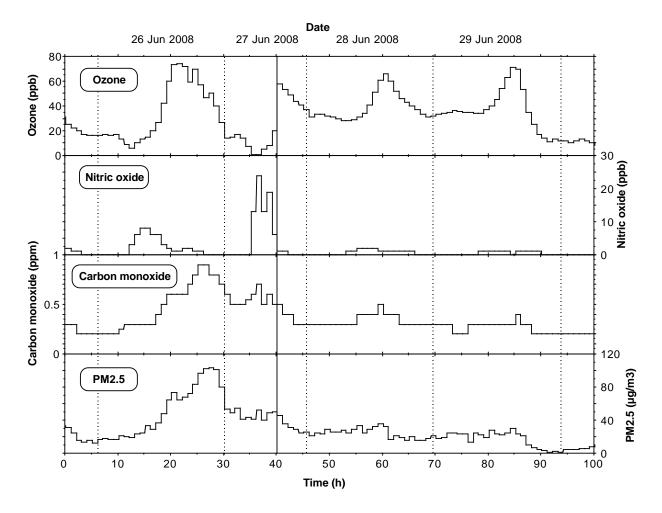


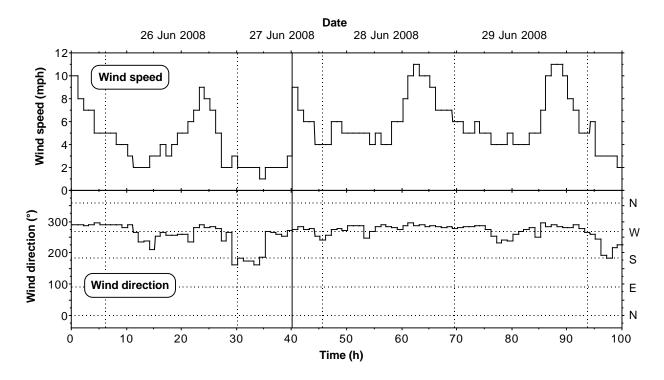
Figure D.75b. Time series of sensor temperatures on stove and oven for observational monitoring at site H5.



**Figure D.76.** Time series of door and window positions for observational monitoring at site H5. Vertical spikes in the plot denote changes in the open or closed state that persisted for less than a clock minute.



**Figure D.77.** Time series of pollutant concentrations based on central-station monitoring for the period of observational monitoring at house site H5.



**Figure D.78.** Time series of wind speed and wind direction based on central-station monitoring for the period of observational monitoring at house site H5.

## D.7 Site H6

Observational monitoring was conducted at house site H6 for a continuous 73-h period commencing at 12:33 on 28 September 2008 and ending at 14:11 on 1 October 2008.

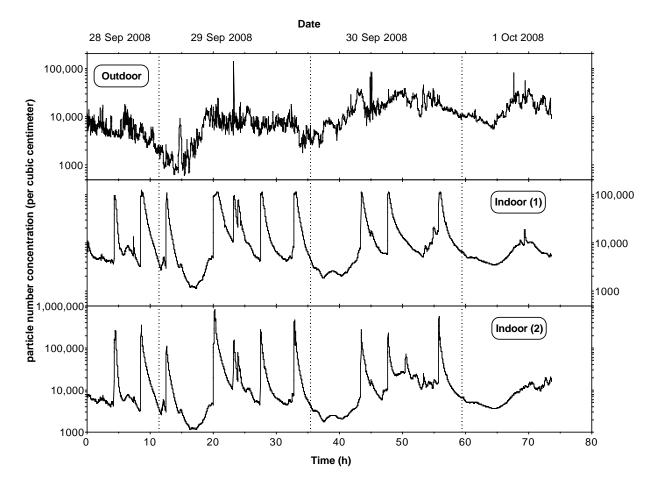
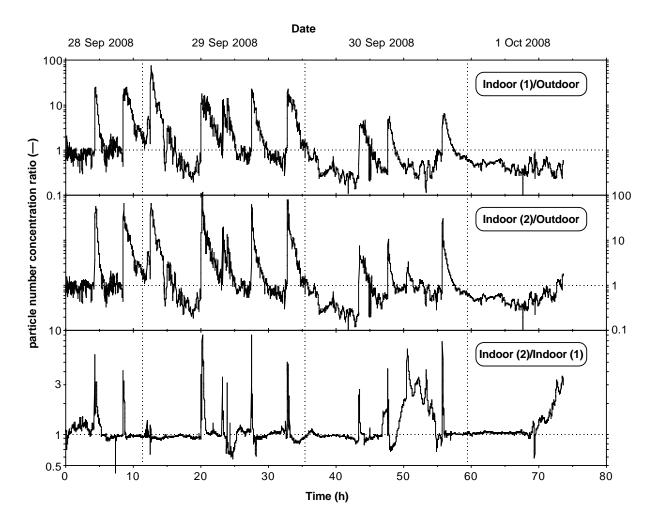


Figure D.79. Time series of particle number concentrations plotted on a logarithmic scale for observational monitoring at site H6.



**Figure D.80.** Time series of ratios of particle number concentrations for observational monitoring at site H6.

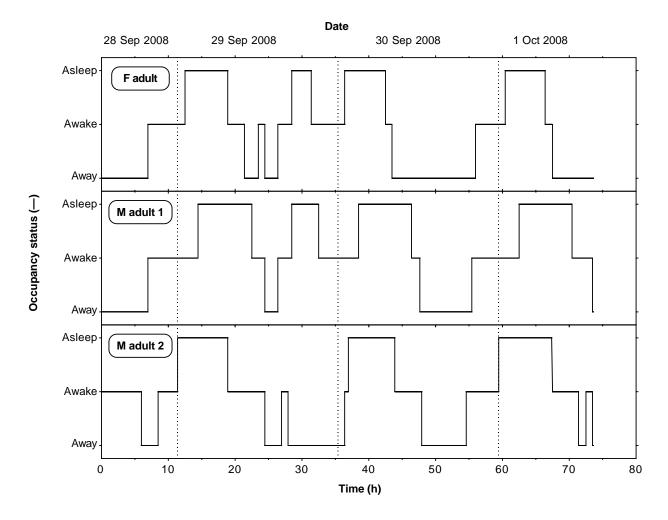


Figure D.81. Time series data for occupancy status during observational monitoring at site H6.

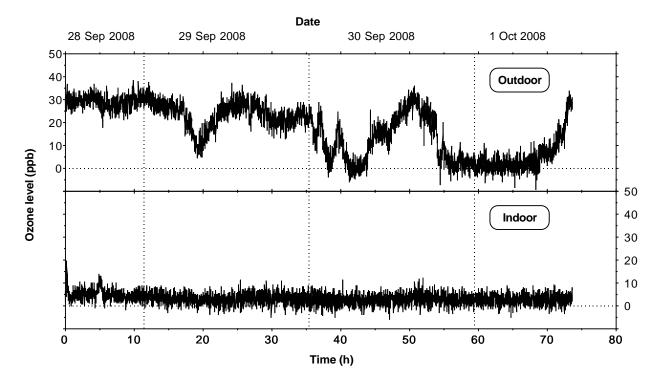


Figure D.82. Time series of ozone levels for observational monitoring at site H6.

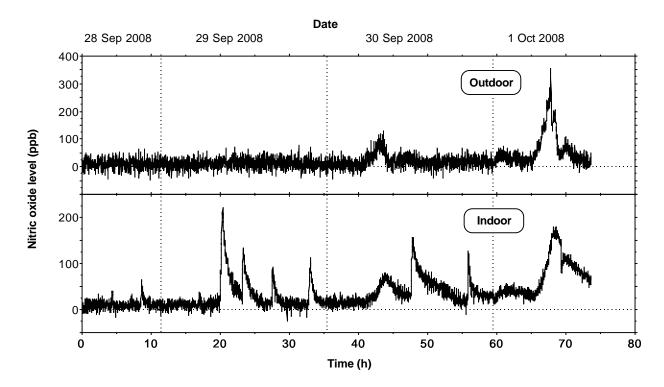


Figure D.83. Time series of nitric oxide levels for observational monitoring at site H6.

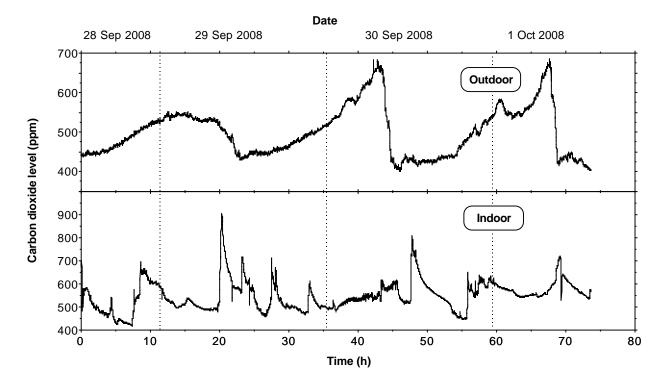


Figure D.84. Time series of carbon dioxide levels for observational monitoring at site H6.

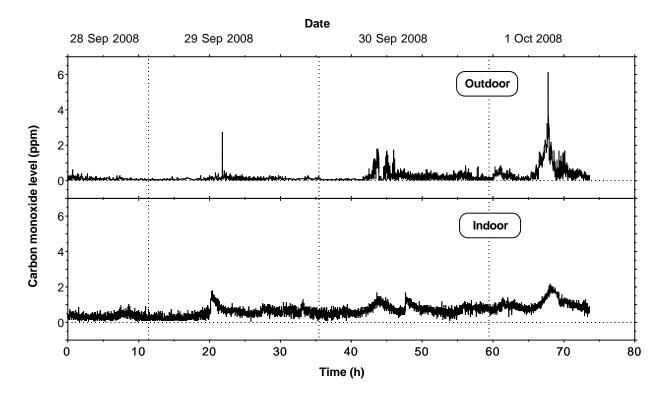


Figure D.85. Time series of carbon monoxide levels for observational monitoring at site H6.

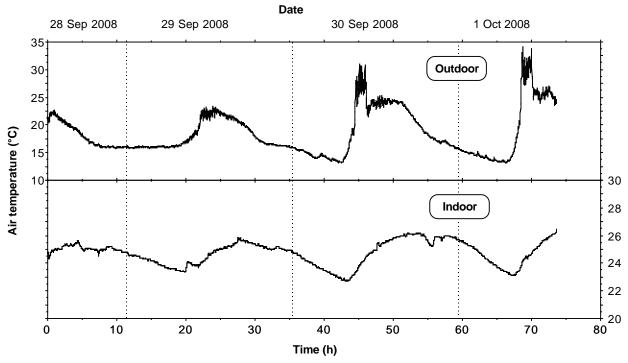


Figure D.86. Time series of air temperatures for observational monitoring at site H6.

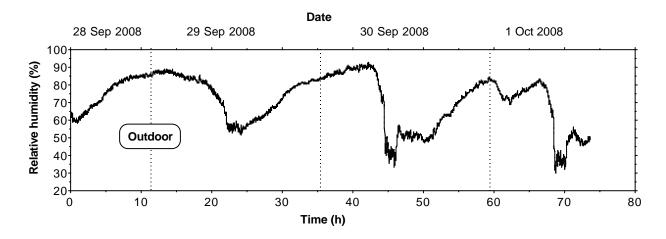


Figure D.87. Time series of outdoor relative humidity for observational monitoring at site H6.

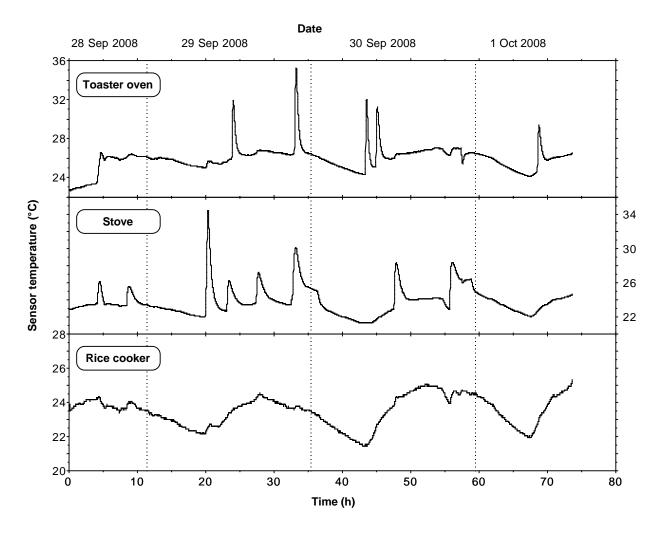
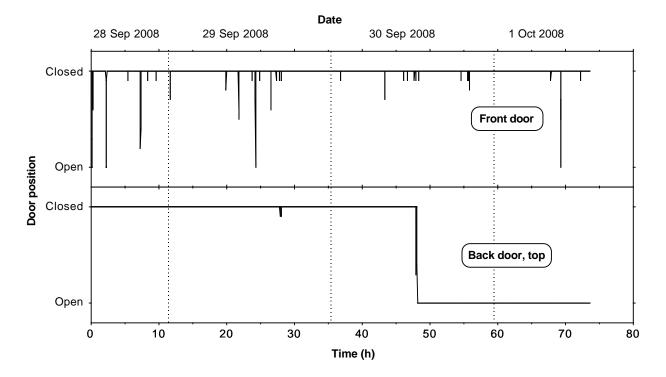


Figure D.88. Time series of sensor temperatures for observational monitoring at site H6.



**Figure D.89a.** Time series of door positions for observational monitoring at site H6. Vertical spikes in the plot denote changes in the open or closed state that persisted for less than a clock minute.

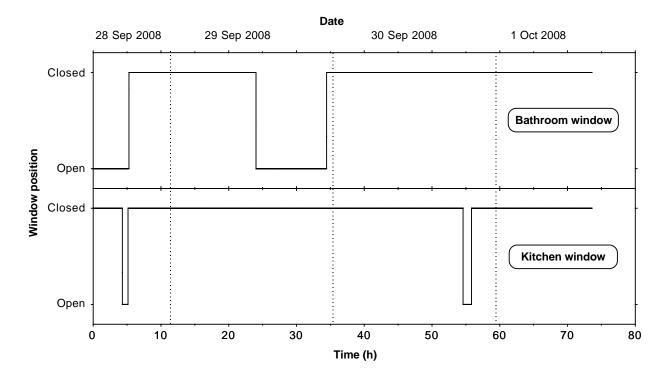
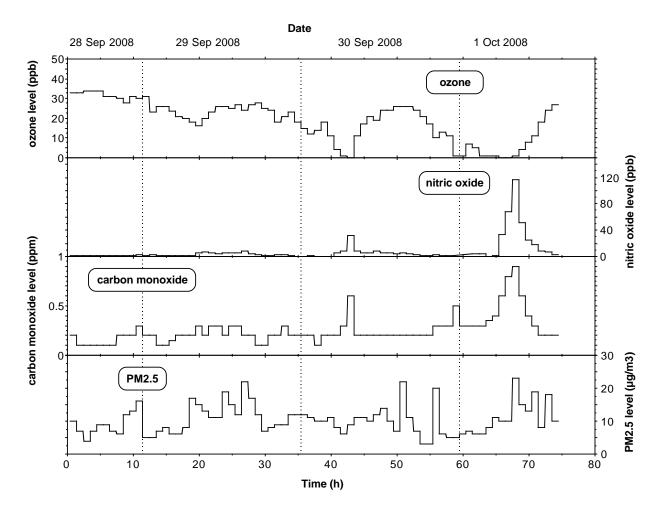
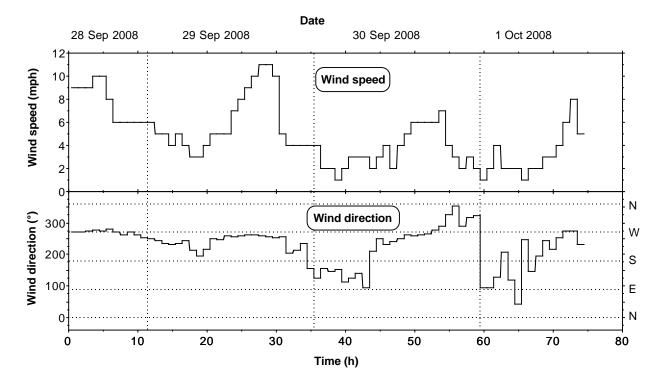


Figure D.89b. Time series of window positions for observational monitoring at site H6.



**Figure D.90.** Time series of pollutant concentrations based on central-station monitoring for the period of observational monitoring at house site H6.



**Figure D.91.** Time series of wind speed and wind direction based on central-station monitoring for the period of observational monitoring at house site H6.

## APPENDIX E. PROBABILITY DISTRIBUTIONS OF PARTICLE NUMBER CONCENTRATIONS IN HOUSES

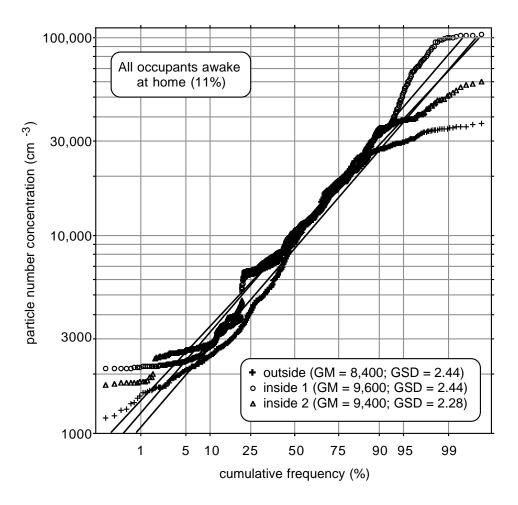
We observed that occupancy and occupant activities were important factors influencing indoor particle levels. In pursuing a goal of trying to understand sources and dynamic behavior of UFP indoors, we separately consider periods here in which the house is fully occupied with awake residents, fully occupied with asleep residents, and vacant. This appendix presents cumulative distribution functions of the minute-by-minute record of indoor and outdoor particle number concentrations for each house site, sorted according to occupancy status. For each house site, three figures are presented, one with the occupants at home awake, a second with the occupants at home asleep, and the third with the house vacant. To be clear, for an entry to be included in the "awake" grouping, all residents must report having been awake at home for that minute. Times when occupancy status varies among residents are not included in the analysis results presented here.

To produce these plots, the occupancy status was used to sort the minute-by-minute readings of PN concentrations from the full observational monitoring period. Once the data were grouped, results from each monitor were sorted in rank order for plotting the distribution. The geometric mean (GM) and geometric standard deviation (GSD) were computed for each monitor and each group, and lognormal distributions were plotted from these parameters on the cumulative distribution plots. In this coordinate system, a parameter that conforms to a lognormal distribution will follow a straight line. To the extent that the data do conform to a lognormal distribution considerable economy in reporting the information is gained, since two parameters, the GM and GSD, completely describe the distribution.

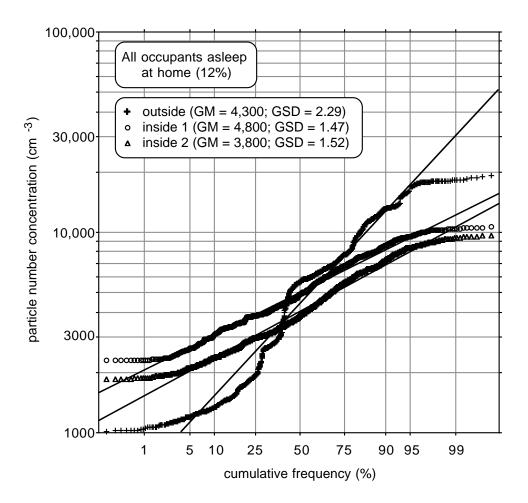
Figures E. 1a-E. 1c present distribution data for house H0. Figure E. 1a shows distributions for times when both occupants of H0 are awake at home. The influence of indoor sources can be seen most clearly in the upper end of the distribution, where the indoor distributions diverge above the outdoor level. (Note: the highest indoor levels do not necessarily correspond in time to the occurrence of the highest outdoor levels.) The figure also reinforces clearly an important point: that the indoor and outdoor particle concentrations exhibit considerable variability in time. For example, the central 90% of the distributions span more than an order of magnitude in each case, from the 5<sup>th</sup> percentile at ~  $2-3 \times 1000$  particles cm<sup>-3</sup> to the 95<sup>th</sup> percentile at ~  $30-50 \times 1000$  particles cm<sup>-3</sup>. The lognormal distributions provide a fairly good, but not excellent fit to the data.

Figure E.1b shows the cumulative distributions when both occupants are at home asleep. Overall, the levels are lower than when occupants are awake, as reflected in the GM values being about a factor of 2 lower. The dynamic range of concentrations during the "asleep" period is also smaller, as indicated by the smaller GSDs. An interesting feature of these data is that the lowest 35-40% of the outdoor distribution is below the lowest 35-40% of the indoor level. It is conceivable that this difference indicates the presence of a low-level continuous indoor emission source which contributes ~ 1000 cm<sup>-3</sup> to the indoor concentration. However, an equally probable hypothesis is that, because of low ventilation rates in this house, the indoor level persists at a somewhat elevated value overnight even as the outdoor level continues to decline into the dawn hours.

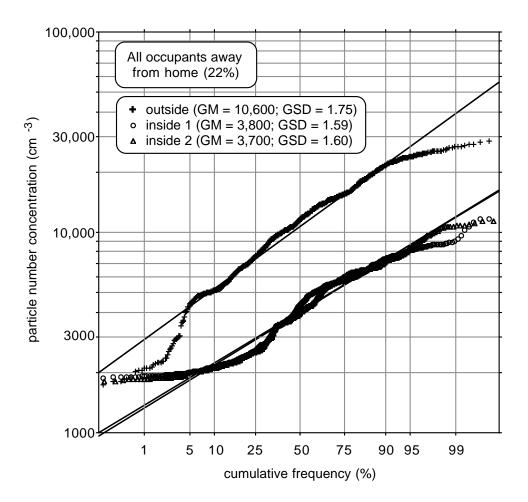
Figure E.1c shows that when the house is vacant the indoor concentration diminishes to a level of 35-40% of the outdoor value and that this ratio prevails about 95% of the time, on average.



**Figure E.1a.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H0 when both occupants were awake and at home (11% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the legend.



**Figure E.1b.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H0 when both occupants were asleep at home (12% of the observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the legend.



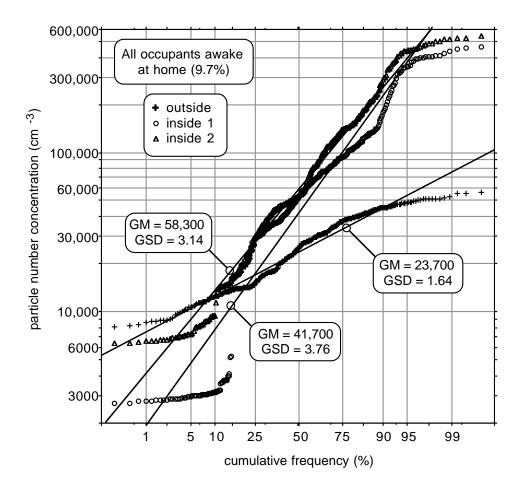
**Figure E.1c.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H0 when both occupants were away from home (22% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the legend.

Figures E.2a-E.2c present data on indoor and outdoor PN concentrations sorted according to the aggregate occupancy status of the house inhabitants for site H1. Figure E.2a shows distributions for times when all four occupants are awake at home. The influence of indoor sources can be seen in the fact that indoor concentrations exceed outdoor concentrations for the upper 80-90 percent of the distributions. When the house is fully occupied, the indoor concentrations span a large range. The central 90% of the distributions vary from 2900 cm<sup>-3</sup> to 345,000 cm<sup>-3</sup> (for "inside 1") and from 7300 cm<sup>-3</sup> to 438,000 cm<sup>-3</sup> (for "inside 2"). The corresponding range for the outdoor monitor is much narrower, with higher concentrations at the 5<sup>th</sup> percentile, but much lower concentrations at the 95<sup>th</sup> percentile: 10,700 cm<sup>-3</sup> to 48,000 cm<sup>-3</sup>.

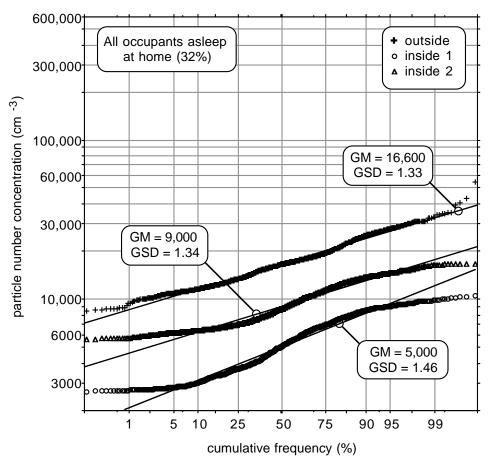
Figure E.2b shows the cumulative distributions when all occupants are at home asleep. Overall, the levels are much lower than when occupants are awake, as reflected in the GM values for the indoor monitors being smaller by factors of 6 ("indoor 1") or 8 ("indoor 2"). The concentrations recorded by the outdoor monitor are also smaller when occupants are asleep than awake, but the GM decreases only by about a third, from 24,000 cm<sup>-3</sup> to 17,000 cm<sup>-3</sup>. The

dynamic range of concentrations during the "asleep" period is also much smaller, as indicated by the small GSDs.

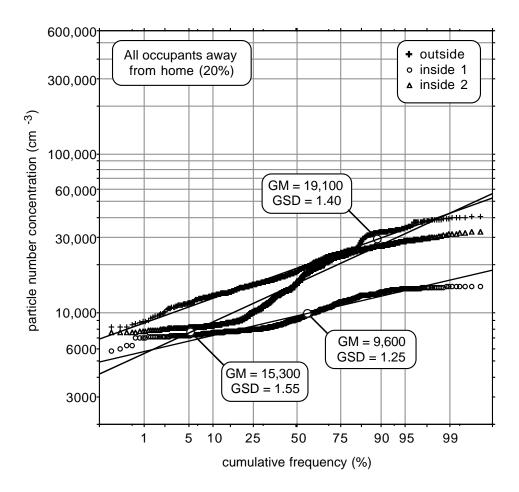
Figure E.2c shows the cumulative PN concentration distributions when the house is vacant. The overall trends are more like the conditions when occupants are asleep, as compared to conditions when the occupants are at home awake. As compared to conditions when occupants are asleep, however, the PN concentrations are higher when the house is vacant. The respective GMs for outdoor air are  $19.1 \times 10^3$  cm<sup>-3</sup> (away) compared with  $16.6 \times 10^3$  cm<sup>-3</sup> (asleep) and  $23.7 \times 10^3$  cm<sup>-3</sup> (awake). For the primary indoor monitor, the corresponding GMs are  $9.6 \times 10^3$  cm<sup>-3</sup> (away),  $5.0 \times 10^3$  cm<sup>-3</sup> (asleep), and  $41.7 \times 10^3$  cm<sup>-3</sup> (awake). And, for the supplemental indoor monitor, the GMs are  $15.3 \times 10^3$  cm<sup>-3</sup> (away),  $9.0 \times 10^3$  cm<sup>-3</sup> (asleep) and  $58.3 \times 10^3$  cm<sup>-3</sup> (awake).



**Figure E.2a.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H1 when all occupants were awake and at home (9.7% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes.

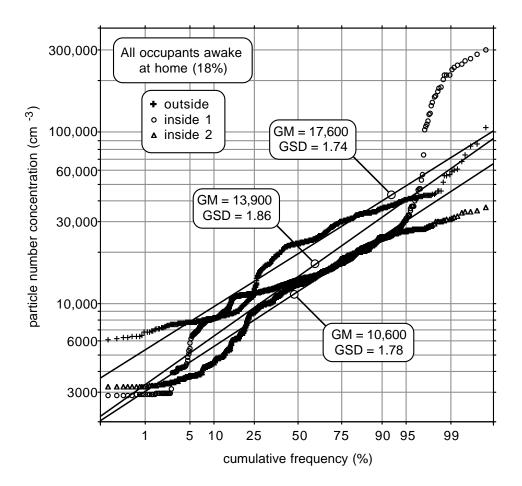


**Figure E.2b.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H1 when all occupants were asleep at home (32% of the observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes.



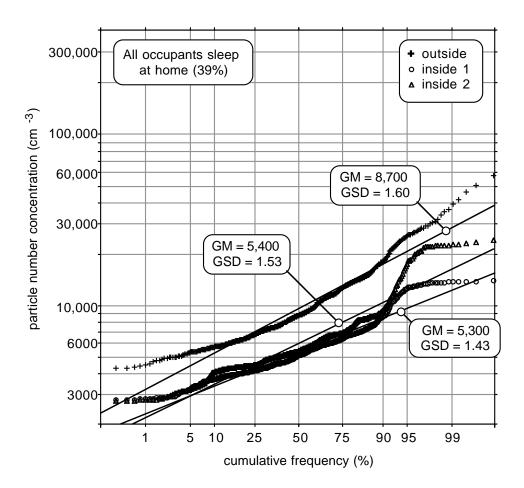
**Figure E.2c.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H1 when all occupants were away from home (20% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes.

Figures E.3a-E.3c present the distribution functions for house site H2. Figure E.3a shows distributions for times when all four occupants are awake at home. The influence of indoor sources can be seen in the upper part of the distribution, about the 95<sup>th</sup> percentile and above, where the indoor concentrations ("inside 1") depart upward from the main trend and rise to levels above the high end of the outdoor concentration distribution. When the house is fully occupied with awake residents, the central 90% of concentration distribution on the main floor indoors varies only over a moderate range, from 5400 cm<sup>-3</sup> to 30,000 cm<sup>-3</sup>. The corresponding range for the outdoor monitor is comparable: 7100 cm<sup>-3</sup> to 37,000 cm<sup>-3</sup>.

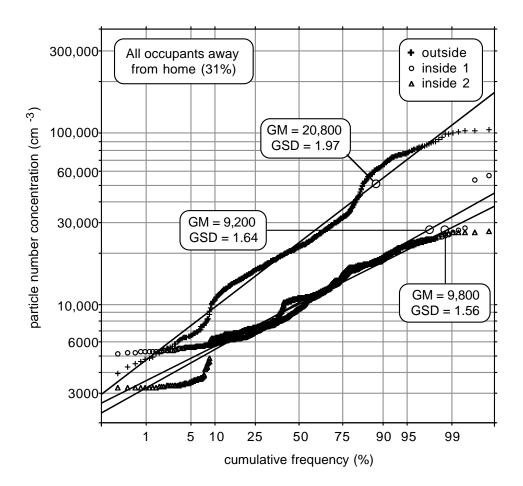


**Figure E.3a.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H2 when all occupants were awake and at home (18% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes.

Figure E.3b shows the cumulative distributions when all occupants are at home asleep. Overall, the levels are somewhat lower than when occupants are awake. Figure E.3c shows the cumulative PN concentration distributions when the house is vacant. The overall trends are more like the conditions when occupants are asleep, as compared to conditions when the occupants are at home awake. As compared to conditions when occupants are asleep, however, the PN concentrations are higher when the house is vacant. The respective GMs for outdoor air are 20.8  $\times 10^3$  cm<sup>-3</sup> (away) compared with 8.7  $\times 10^3$  cm<sup>-3</sup> (asleep) and 17.6  $\times 10^3$  cm<sup>-3</sup> (awake). For the primary indoor monitor, the corresponding GMs are 9.8  $\times 10^3$  cm<sup>-3</sup> (away), 5.3  $\times 10^3$  cm<sup>-3</sup> (asleep), and 13.9  $\times 10^3$  cm<sup>-3</sup> (awake).

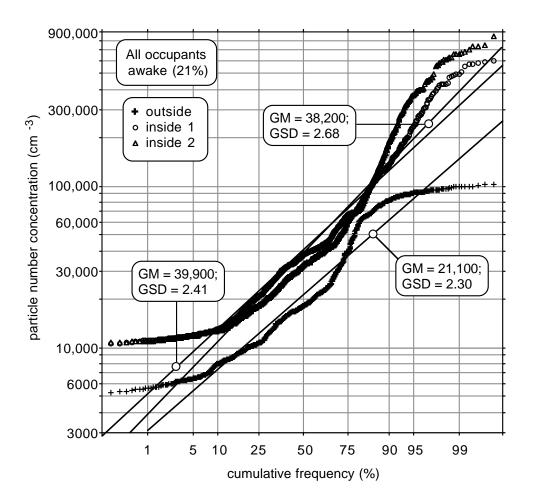


**Figure E.3b.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H2 when all occupants were asleep at home (39% of the observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes.



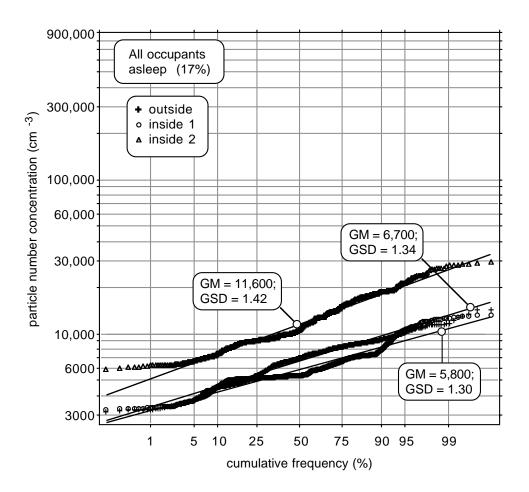
**Figure E.3c.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H2 when all occupants were away from home (31% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes.

Figures E.4a-E.4c present cumulative distribution functions for particle number concentrations at house site H3. Figure E.4a shows distributions for times when all three occupants are awake at home. The influence of indoor sources can be seen in the fact that indoor concentrations exceed outdoor concentrations across the entire distribution. When the house is fully occupied and the residents are awake, the indoor concentrations span a large range. The central 90% of the distributions vary from ~ 12,000 cm<sup>-3</sup> to 224,000 (inside 1) or 360,000 cm<sup>-3</sup> (inside 2). The corresponding range is also large for the outdoor monitor: 6500 cm<sup>-3</sup> to 90,000 cm<sup>-3</sup>.



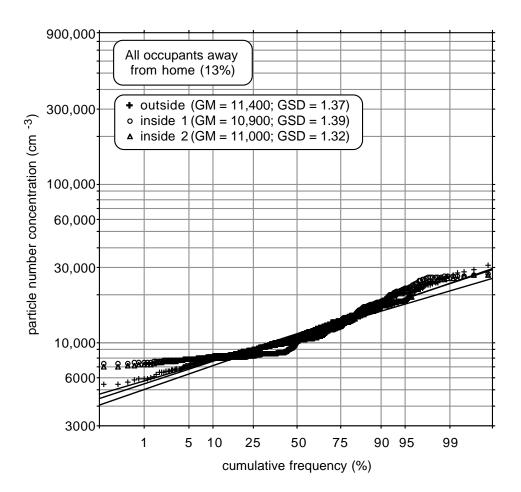
**Figure E.4a.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H3 when all occupants were awake and at home (21% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes.

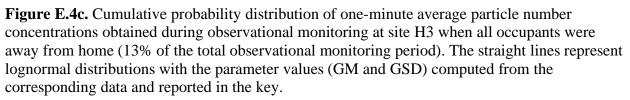
Figure E.4b shows the cumulative distributions when all occupants are at home asleep. Overall, the levels are much lower than when occupants are awake, as reflected in the GM values for the indoor monitors being smaller by factors of 7 ("inside 1") or 3 ("inside 2"). The concentrations recorded by the outdoor monitor are also smaller by about a factor of three when occupants are asleep than awake. The dynamic range of concentrations during the "asleep" period is much smaller than when occupants are "awake," as indicated by the smaller GSDs, ~ 1.3-1.4 instead of 2.3-2.7. An interesting feature of the "asleep" period distributions is that the supplemental indoor monitor, sited in the kitchen, records substantially higher PN concentrations than the primary indoor monitor, sited in the living room. This characteristic might reflect the influence of PN emissions from the continuous pilot lights on the gas stove and oven combined with poor mixing conditions at night.



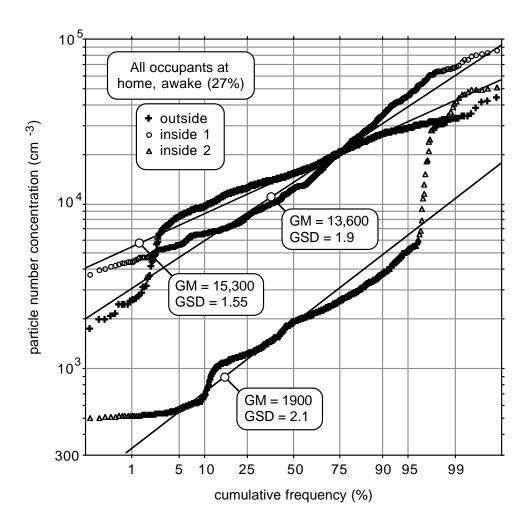
**Figure E.4b.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H3 when all occupants were asleep at home (17% of the observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes.

The cumulative probability distributions of PN concentrations are very similar to one another during times when the house is unoccupied, as shown in Figure E.4c. It would be tempting to infer from this evidence that all indoor particles are of outdoor origin when the house is unoccupied. Such an inference would be wrong. Evidence from other sites and even from this house consistently reveal that the indoor proportion of outdoor ultrafine particles is considerably less than one, owing to penetration and deposition losses indoors (and sometimes augmented by active filtration). We believe that the indoor concentrations at H3 during times when the house is unoccupied mainly reflect the superposition of contributions from outdoor particles, attenuated by indoor loss mechanisms, combined with emissions from the continuous pilot lights in the kitchen. It is not clear, however, why the distributions from the two indoor monitors agree closely when occupants are away from home (daytime periods) and yet consistently diverge when the occupants are alseep at home (nighttime periods). It is interesting that the distributions in the kitchen ("inside 2") are similar for these two periods, with "asleep" GM =  $11.6 \times 10^3$  cm<sup>-3</sup> and GSD = 1.42 versus "away" GM =  $11.0 \times 10^3$  cm<sup>-3</sup> and GSD = 1.32.





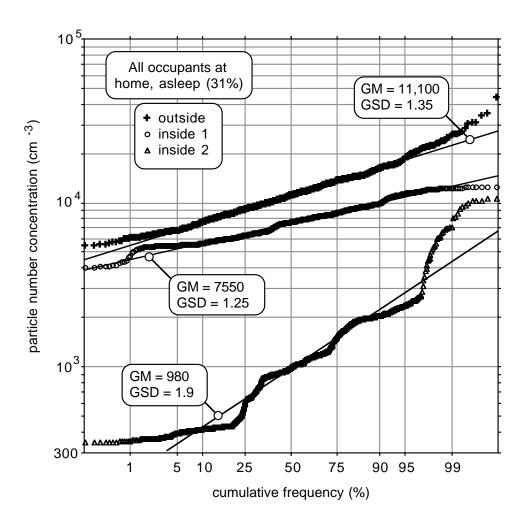
Figures E.5a-E.5c present cumulative distributions for minute-by-minute indoor and outdoor PN concentrations sorted according to the aggregate occupancy status of the house inhabitants at site H4. Figure E.5a shows distributions for times when all four occupants are awake at home. The influence of indoor sources can be seen in the upper part of the distribution, about the 75<sup>th</sup> percentile and above, where the indoor concentrations ("inside 1") rise to levels above the high end of the outdoor concentration distribution. When the house is fully occupied with awake residents, the central 90% of concentration distribution on the main floor indoors varies over a moderate range, from 5700 cm<sup>-3</sup> to 45,000 cm<sup>-3</sup>. The corresponding range for the outdoor monitor is narrower: 8400 cm<sup>-3</sup> to 28,000 cm<sup>-3</sup>. The distribution of measurements from the upstairs monitor ("inside 2") is strongly displaced downward except for the upper few percent of the distribution, when the upstairs levels rise to comparable values observed at the high end outdoors and downstairs. Otherwise, for the central 90% of the distribution, the upstairs levels at H4 are the lowest seen anywhere in this project, ranging from 600 to 5200 cm<sup>-3</sup>.



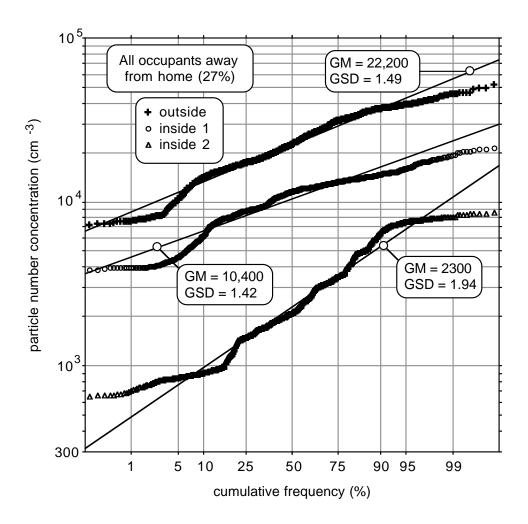
**Figure E.5a.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H4 when all occupants were awake and at home (27% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes.

Figure E.5b shows the cumulative distributions when all occupants are at home asleep. The distributions outdoors and downstairs span relatively narrow ranges, as indicated by the small GSDs. The indoor levels upstairs, where the occupants are sleeping, are again much lower than those downstairs or outdoors, with a central 90% of the distribution ranging from 400 to  $2400 \text{ cm}^{-3}$ .

When occupants are away from home (Figure E.5c), the downstairs and outside distributions track in an almost completely parallel fashion, supporting a view that the particles outdoors are the dominant source of the particles indoors at these times. The geometric mean of the downstairs indoor level is 47% of outdoor value,  $10.4 \times 10^3$  cm<sup>-3</sup> versus  $22.2 \times 10^3$  cm<sup>-3</sup>. Again, the upstairs level is substantially lower than that downstairs, indicative of the effectiveness of the recirculating air filtration system in controlling PN concentrations.



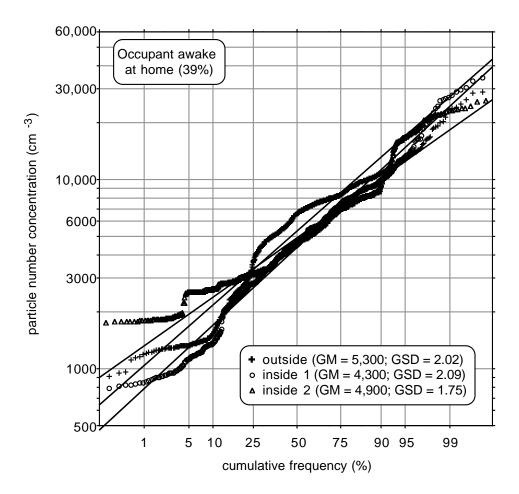
**Figure E.5b.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H4 when all occupants were asleep at home (31% of the observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes.



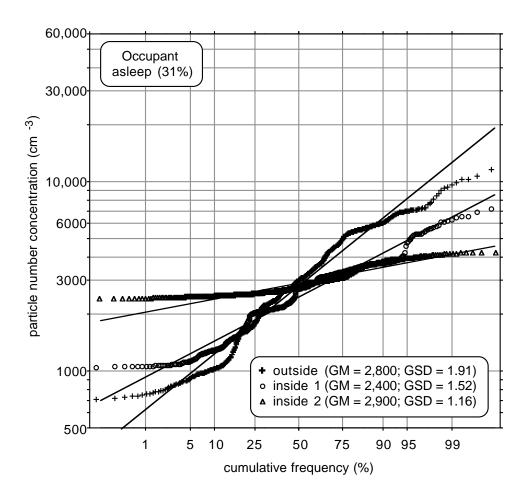
**Figure E.5c.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H4 when all occupants were away from home (27% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes.

Figures E.6a-E.6c present cumulative distribution data on indoor and outdoor PN concentrations sorted according to the aggregate occupancy status of the one inhabitant of house H5. Figure E.6a shows distributions for times when the occupant is awake at home. The three distributions are similar with the 5<sup>th</sup> percentile occurring at ~ 1,000-2,000 cm<sup>-3</sup> and the 95<sup>th</sup> percentile roughly an order of magnitude larger, at ~ 15,000 cm<sup>-3</sup>.

Figure E.6b shows the cumulative distributions when R1 is at home asleep. The central tendencies of all three distributions are similar, but the degree of variability is quite different, with the indoor distributions spanning a narrower range of concentrations than is observed outdoors.

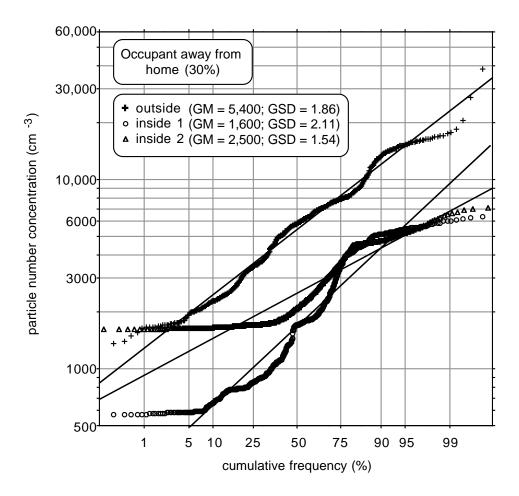


**Figure E.6a.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H5 when occupant R1 was awake and at home (39% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 5,300; inside 1 GM = 4,300; inside 2 GM = 4,900 cm<sup>-3</sup>). For "outside" and "inside 1", the analysis period is the full 100 h of observational monitoring; for "inside 2" the analysis period is the first 40 h.



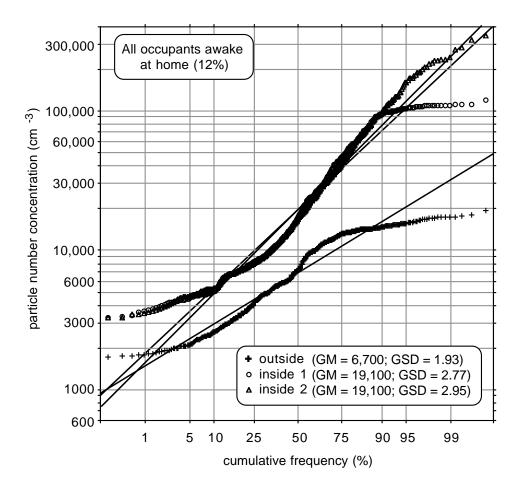
**Figure E.6b.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H5 when occupant R1 was asleep at home (31% of the observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes: outside GM = 2,800; inside 1 GM = 2,400 and inside 2 GM = 2,900 cm<sup>-3</sup>. For "outside" and "inside 1", the analysis period is the full 100 h of observational monitoring; for "inside 2" the analysis period is the first 40 h.

When occupant R1 is away from home, the indoor PN concentrations are considerably less than those outside, as illustrated in Figure E.6c. Focusing on the comparison between the outside and "inside 1" distributions, we observe strong similarity in the shapes, with an offset of similar downstairs and outside distributions track in an almost completely parallel fashion, supporting a view that the particles outdoors are the dominant source of the particles indoors at these times. The geometric mean of the downstairs indoor level is ~ 30% of outdoor value, similar to the ratio of arithmetic means (2200/6500 = 0.34).



**Figure E.6c.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H5 when occupant R1 was away from home (30% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes: outside GM = 5,400; inside 1 GM = 1,600; inside 2 GM = 2,500 cm<sup>-3</sup>. For "outside" and "inside 1", the analysis period is the full 100 h of observational monitoring; for "inside 2" the analysis period is the first 40 h.

Figures E.7a-E.7c present data on indoor and outdoor PN concentrations sorted according to the aggregate occupancy status of the inhabitants of H6. Figure E.7a shows distributions for times when all three occupants are awake at home (12% of the monitoring period). The influence of indoor sources is dramatically evident in these distributions: the indoor traces are substantially higher than the outdoor concentrations at all percentiles. The distributions for the two indoor monitors overlap substantially except for the upper 10% of the distribution, for which the upstairs monitor ("inside 2") exhibits much higher concentrations than the downstairs unit ("inside 1"). For the outdoor monitor, the central 90% of the distribution spans the range 2000-15,300 cm<sup>-3</sup>. For the downstairs indoor monitor, the corresponding range is 4600-103,000 cm<sup>-3</sup>.

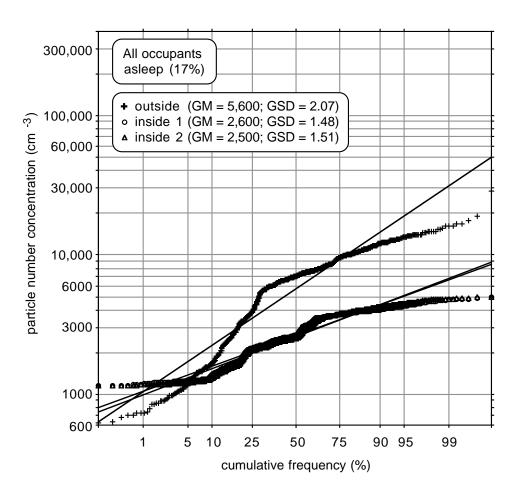


**Figure E.7a.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H6 when all three occupants were awake and at home (12% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 6,700; inside 1 GM = 19,100; inside 2 GM = 19,100 cm<sup>-3</sup>).

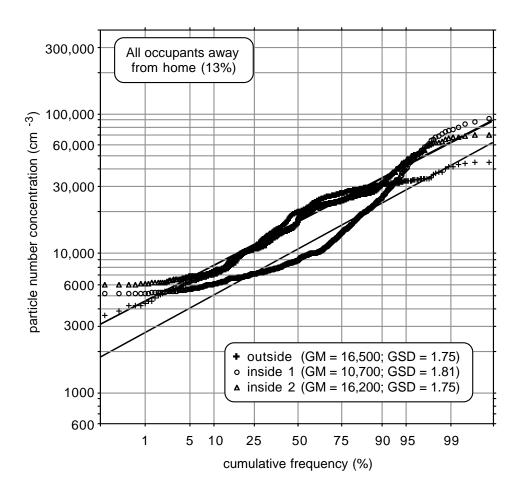
Figure E.7b shows the cumulative distributions when all three occupants are at home asleep. In comparison with conditions when occupants are awake (Figure E.7a), here the situation is reversed over most of the distribution: the outdoor concentrations exceed those indoors. For the outdoor distribution, the central 90% of values spans a range from ~ 1100-13,000 cm<sup>-3</sup>. The two indoor distributions are almost identical, and the central 90% of this distribution varies over a narrower range than outdoors, from 1200 to ~ 4400 cm<sup>-3</sup>.

When all occupants are away from home (13% of the monitoring period), the three distributions are the most similar to one another, as shown in Figure E.7c. Roughly, the central 90% of the distributions in these cases span the range 6,000-40,000 cm<sup>-3</sup>. Probably, though, the similar indoor and outdoor distributions is largely coincidental. The three occupants are simultaneously away only for two significant periods, for a few hours on the afternoon of 29 September and then for several hours on the following afternoon. The vacant periods follow

shortly after the occurrences of peaks e and i and before the indoor particle concentrations decay back to their background levels. Meanwhile, the second, longer vacant period also coincides with a period of high outdoor particle concentrations. One should not expect that the resulting distributions in PN concentrations are causally connected.



**Figure E.7b.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H6 when all three occupants are asleep at home (17% of the observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes: outside GM = 5,600; inside 1 GM = 2,600; and inside 2 GM = 2,500.



**Figure E.7c.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site H6 when the three occupants are all away from home (13% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes: outside GM = 16,500; inside 1 GM = 10,700; inside 2 GM = 16,200 cm<sup>-3</sup>.

## **APPENDIX F. TIME-SERIES PLOTS AT CLASSROOM SITES**

In this appendix, we present time-series plots from observational monitoring at the six classroom sites. In §4.2, time-series plots were presented for each classroom showing the particle number concentrations on a linear scale and the occupancy patterns. Those figures are not repeated here. Table F.1 lists the thirteen sets of parameters that are presented in this appendix. Generally, these are organized into twelve to thirteen figures per classroom site with one classroom per section.

In the narrative portion of each section, basic information is presented about the dates and times of monitoring. In each figure, the time scale is represented as elapsed time (hours), referenced to the start of monitoring. At some of the sites, monitoring proceeded continuously for the entire observational-monitoring period. However, at other sites, there were interruptions in the monitoring period. In these cases, a vertical black line marks a break and, in effect, the elapsed time clock stops until monitoring resumes. The vertical dashed lines in each figure mark midnight and the calendar date is noted along the top horizontal axis. All classroom sites were monitored in 2008.

| <b>Table F.1.</b> Time-series plots presented in this appendix, based on observational monitoring at |
|--|
| each classroom site.   |

Particle number concentration, logarithmic (primary indoor, secondary indoor, outdoor)

Particle number ratios (primary indoor/outdoor, secondary indoor/outdoor, secondary/primary) Occupancy status

Ozone concentration (outdoor, indoor)

Nitric oxide concentration (outdoor, indoor)

Carbon dioxide (outdoor, indoor)<sup>a</sup>

Carbon monoxide (outdoor, indoor)

Temperature (outdoor, indoor)

Relative humidity (outdoor, indoor)

Sensor temperature <sup>b</sup>

Door and window position sensors

Central station air pollutants (ozone, NO, CO, PM<sub>2.5</sub>)

Central station meteorology (wind speed, wind direction)

<sup>a</sup> Although outdoor carbon dioxide levels were measured, the Q-Trak appears to exhibit temperature sensitivity that makes it unreliable for quantifying the fairly narrow range of outdoor concentrations expected. Outdoor  $CO_2$  levels are presented here based on Q-Trak measurements, but these measurements are not deemed reliable. In analysis, whenever an outdoor value was needed for a calculation result, it was assumed to be the global average background level of 380 ppm.

<sup>b</sup> Sensor temperature data only collected at a subset of classroom sites.

## F.1 Site S1

Observational monitoring at site S1 commenced at 15:40 on 2 June 2008 and terminated at 15:00 on 6 December 2008.

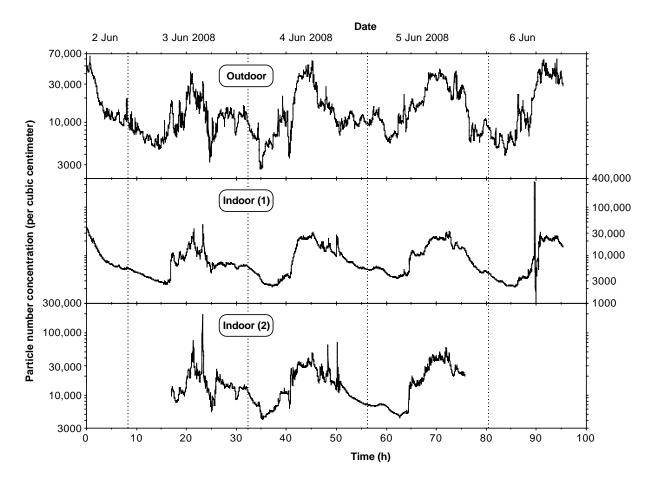


Figure F.1. Time series of particle number concentrations plotted on a logarithmic scale for observational monitoring at site S1.

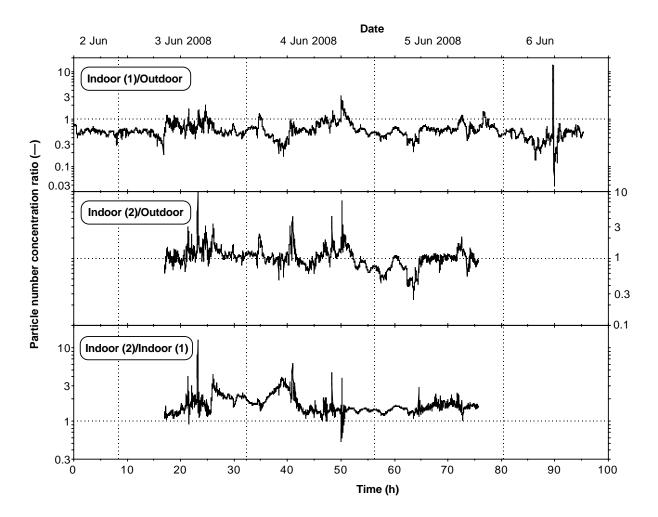


Figure F.2. Time series of ratios of particle number concentrations for observational monitoring at site S1.

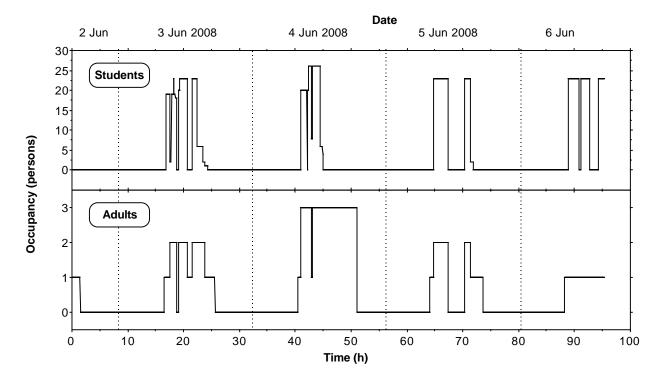


Figure F.3. Time series of occupancy status for observational monitoring at site S1.

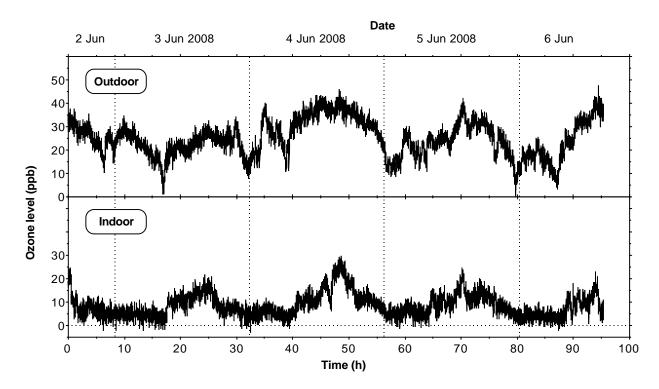


Figure F.4. Time series of ozone levels for observational monitoring at site S1.

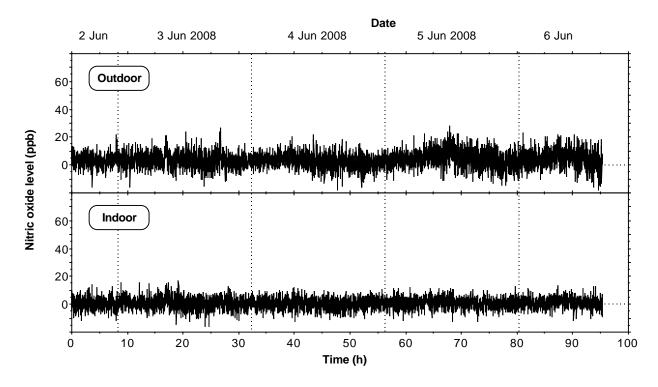


Figure F.5. Time series of nitric oxide levels for observational monitoring at site S1.

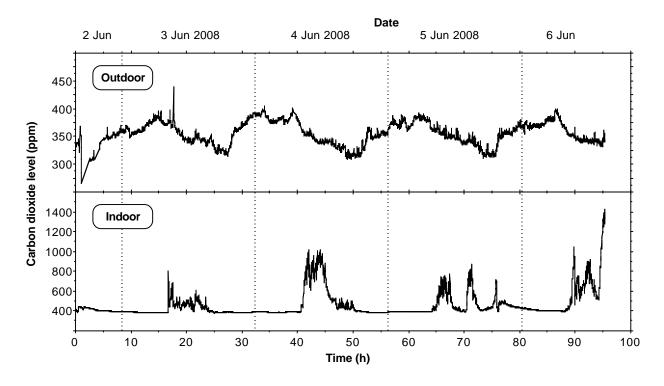


Figure F.6. Time series of carbon dioxide levels for observational monitoring at site S1.

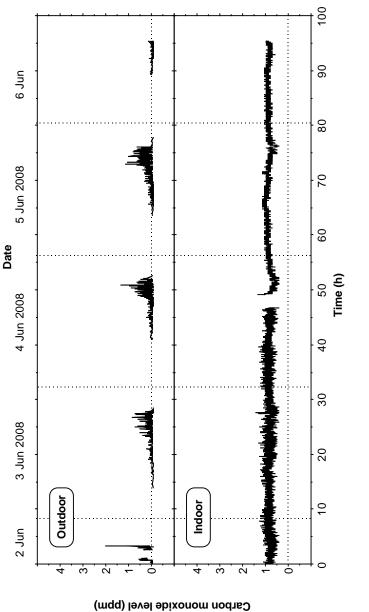


Figure F.7. Time series of carbon monoxide levels for observational monitoring at site S1.

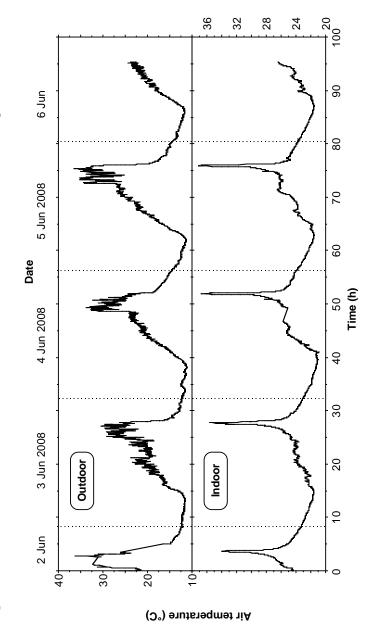


Figure F.8. Time series of air temperatures for observational monitoring at site S1.

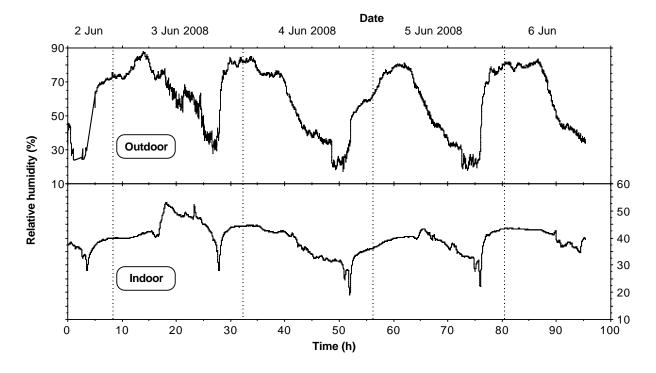
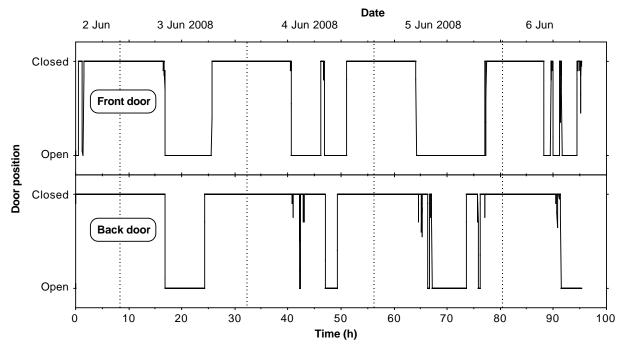
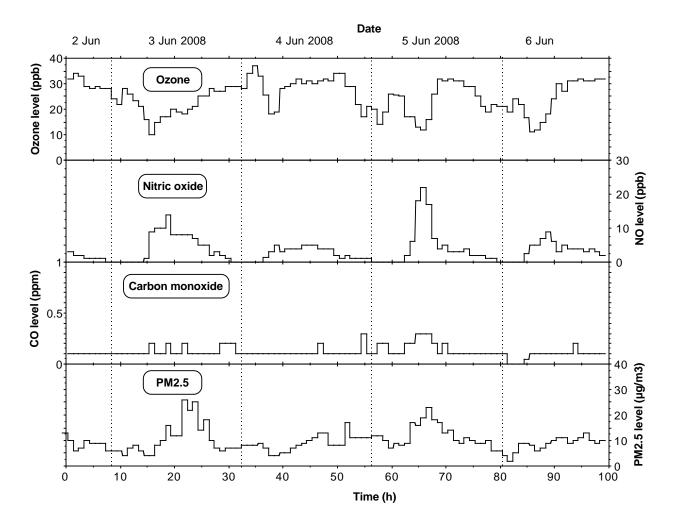


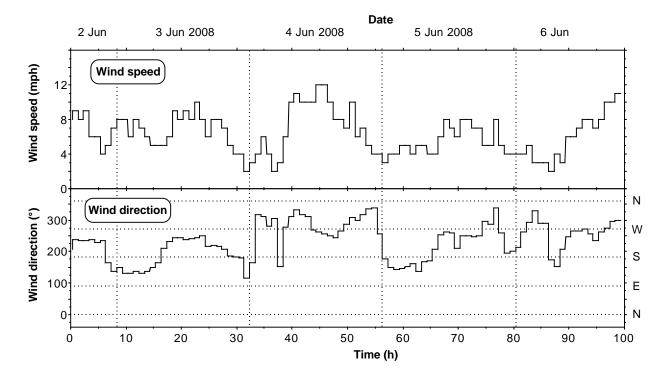
Figure F.9. Time series of indoor relative humidity for observational monitoring at site S1.



**Figure F.10.** Time series of door positions for observational monitoring at site S1. Vertical spikes in the plot denote changes in state that persisted for less than a clock minute.



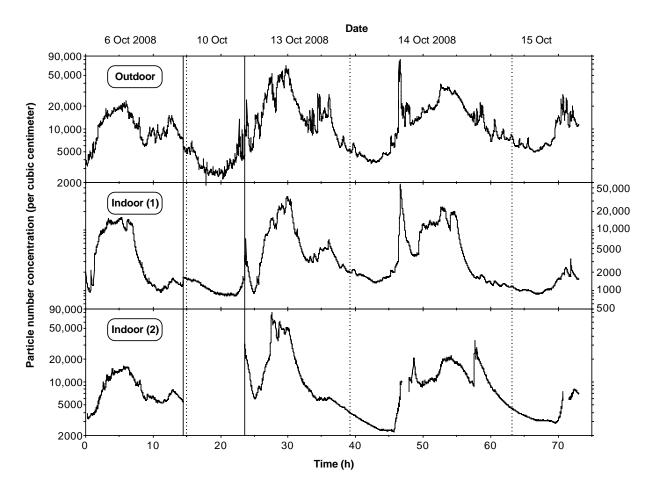
**Figure F.11.** Time series of pollutant concentrations based on central-station monitoring for the period of observational monitoring at school site S1.



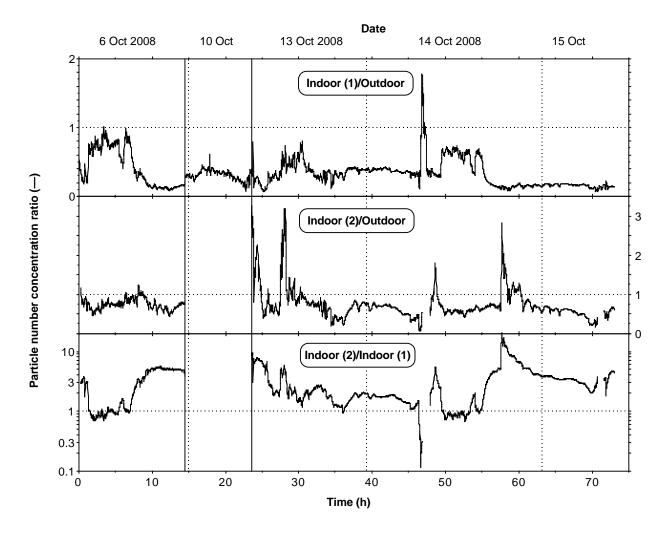
**Figure F.12.** Time series of wind speed and wind direction based on central-station monitoring for the period of observational monitoring at school site S1.

## F.2 Site S2

Observational monitoring was conducted at classroom site S2 for 73 hours in total during three periods: (i) 9:00-23:30 6 October 2008 (14.5 h); (ii) from 23:30 on 9 October through 8:30 on 10 October 2008 (9 h); and (iii) from 8:20 on 13 October until 9:50 on 15 October 2008 (49.5 h).



**Figure F.13.** Time series of particle number concentrations plotted on a logarithmic scale for observational monitoring at site S2.



**Figure F.14.** Time series of ratios of particle number concentrations for observational monitoring at site S2.

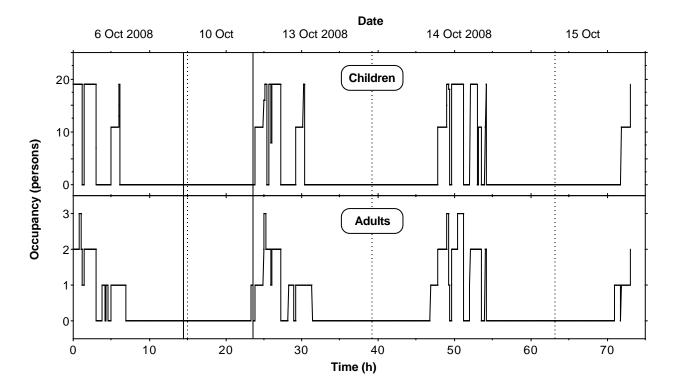


Figure F.15. Time series of occupancy status for observational monitoring at site S2.

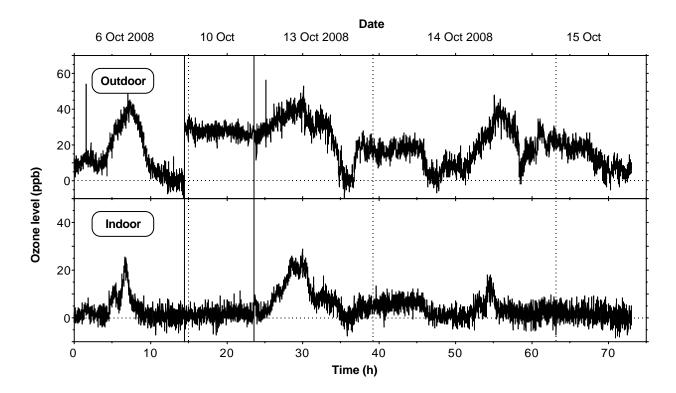


Figure F.16. Time series of ozone levels for observational monitoring at site S2.

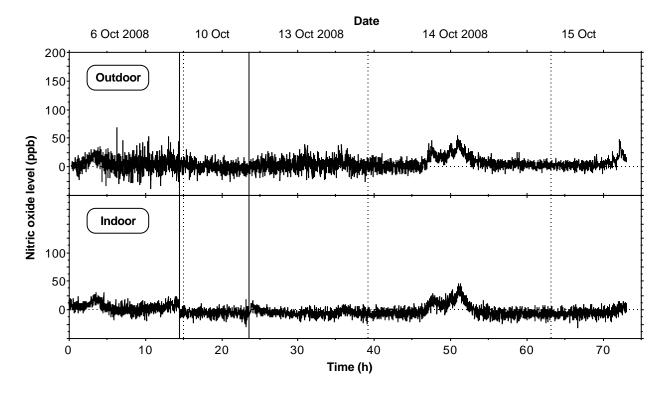


Figure F.17. Time series of nitric oxide levels for observational monitoring at site S2.

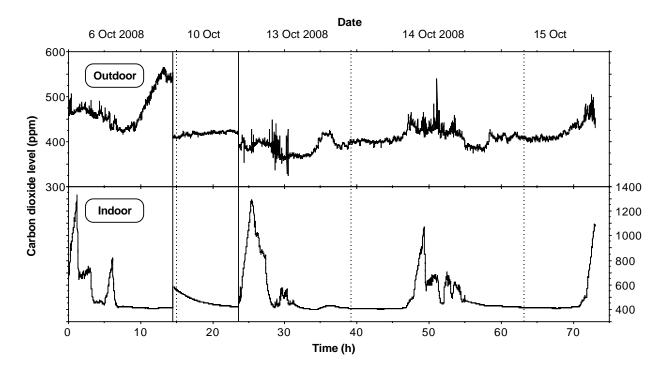
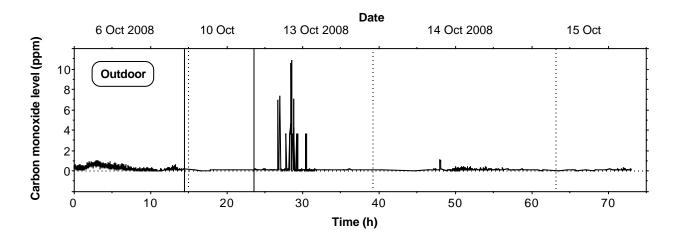


Figure F.18. Time series of carbon dioxide levels for observational monitoring at site S2.



**Figure F.19.** Time series of outdoor carbon monoxide levels for observational monitoring at site S2. No indoor monitoring data are available as the instrument had malfunctioned and was at the manufacturer for repair.

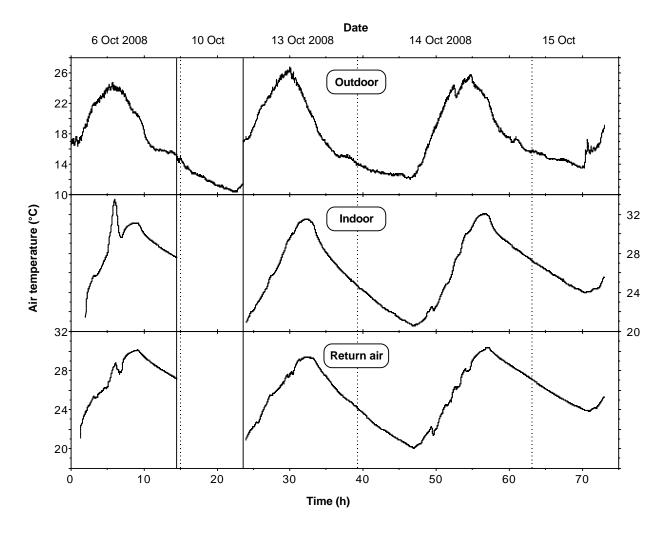


Figure F.20. Time series of air temperatures for observational monitoring at site S2.

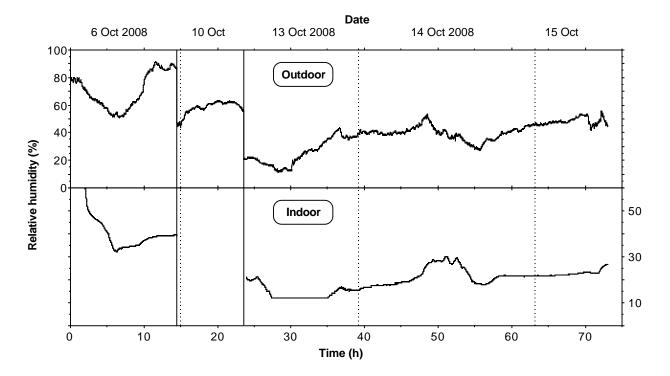
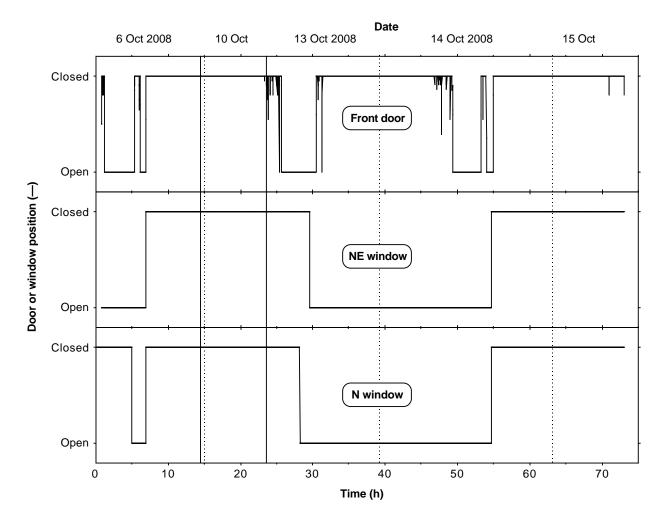
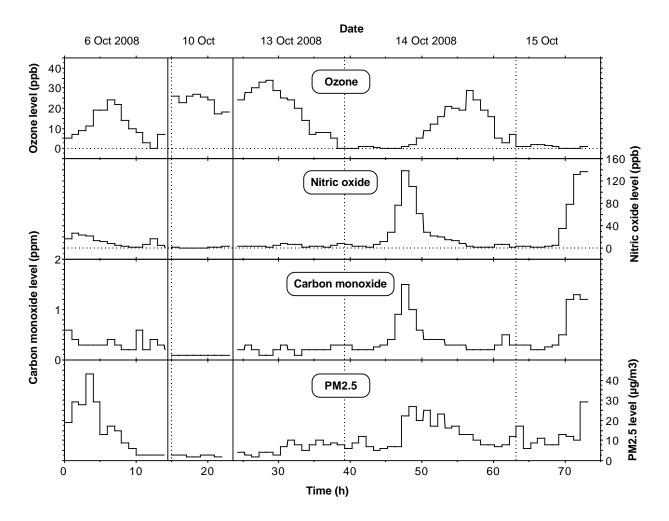


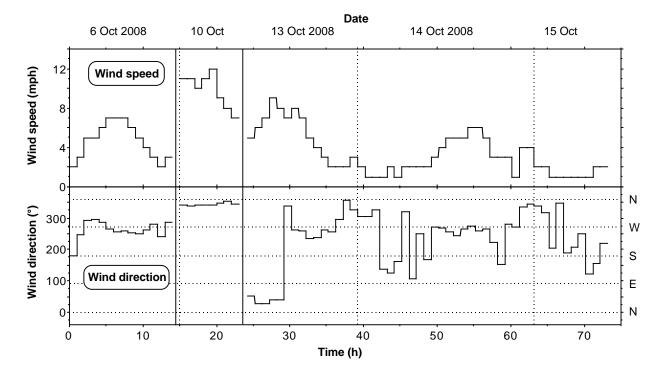
Figure F.21. Time series of relative humidities for observational monitoring at site S2.



**Figure F.22.** Time series of door and window positions for observational monitoring at site S2. Vertical spikes in the plot denote changes in the open or closed state that persisted for less than a clock minute.



**Figure F.23.** Time series of pollutant concentrations based on central-station monitoring for the period of observational monitoring at classroom site S2.



**Figure F.24.** Time series of wind speed and wind direction based on central-station monitoring for the period of observational monitoring at classroom site S2.

## F.3 Site S3

Observational monitoring was conducted at classroom site S3 for 71 hours over three periods: (i) 9:00-15:21 on 20 October; (ii) 8:30 on 21 October through 7:34 on 23 October; and (iii) 15:17 on 23 October through 8:30 on 24 October 2008.

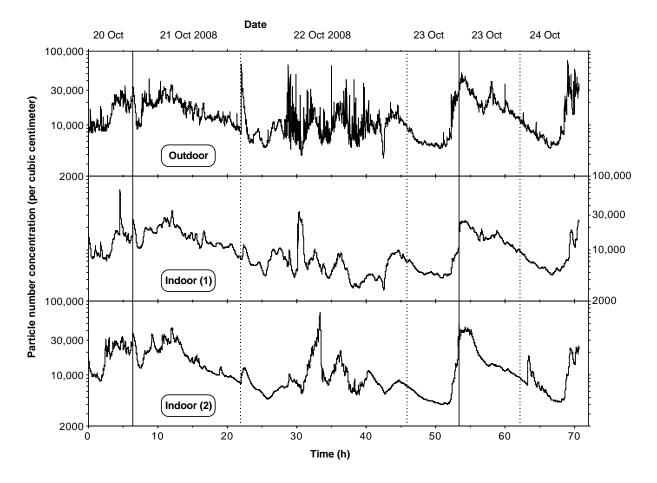
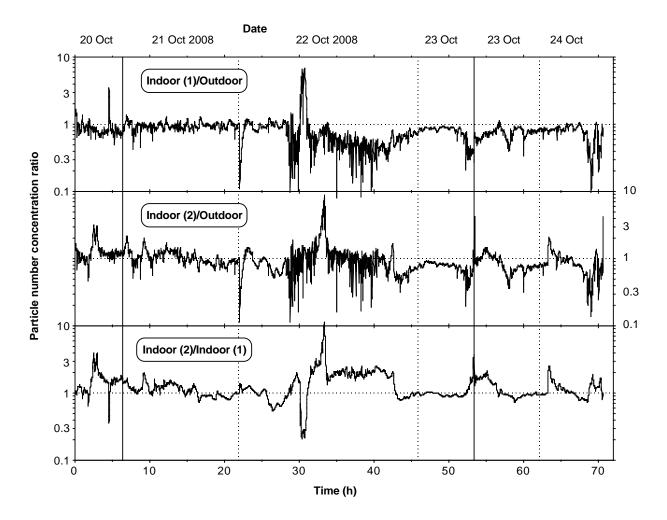


Figure F.25. Time series of particle number concentrations plotted on a logarithmic scale for observational monitoring at site S3.



**Figure F.26.** Time series of ratios of particle number concentrations for observational monitoring at site S3.

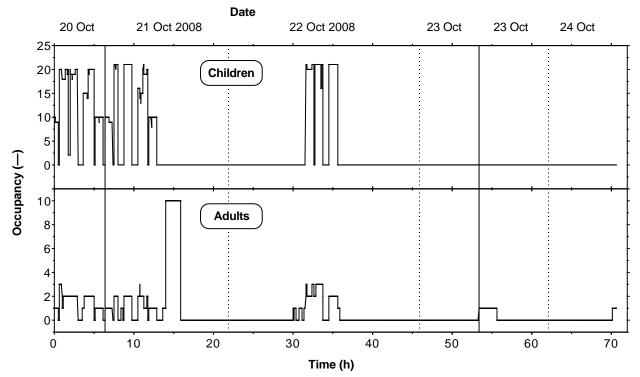


Figure F.27. Time series of occupancy status for observational monitoring at site S3.

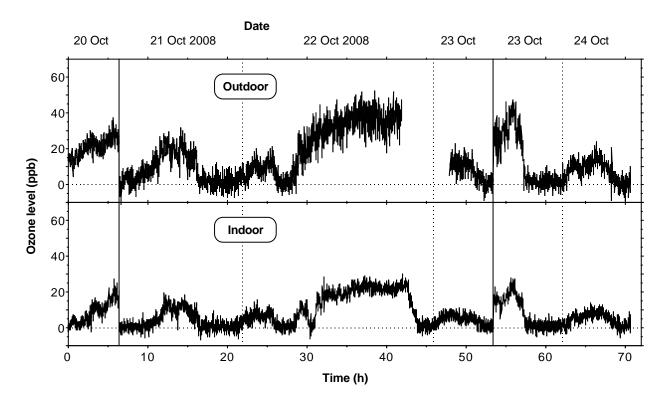


Figure F.28. Time series of ozone levels for observational monitoring at site S3.

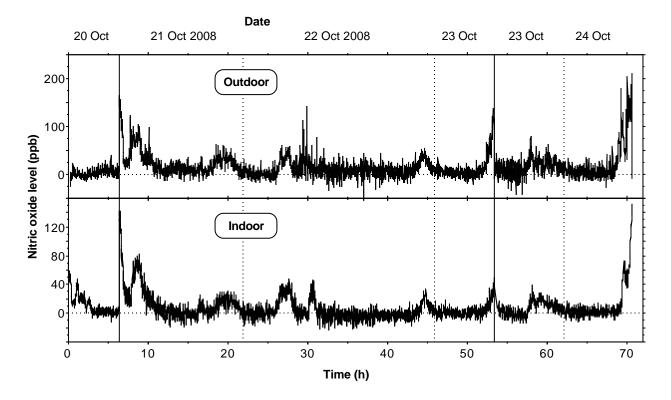


Figure F.29. Time series of nitric oxide levels for observational monitoring at site S3.

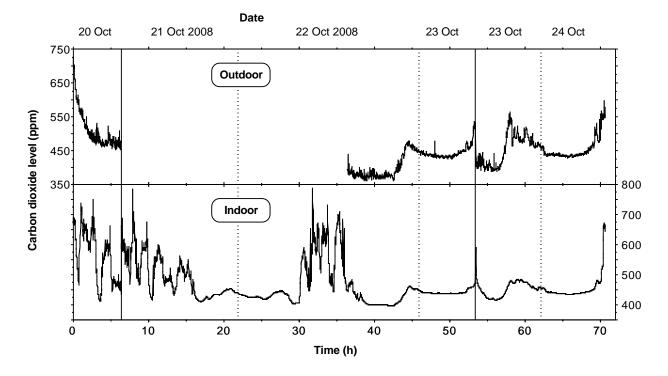


Figure F.30. Time series of carbon dioxide levels for observational monitoring at site S3.

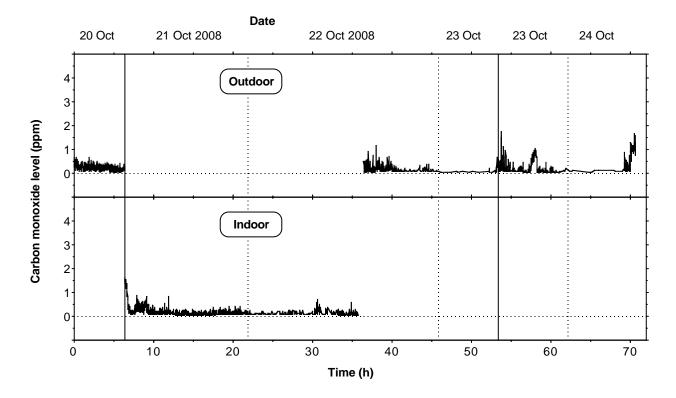


Figure F.31. Time series of carbon monoxide levels for observational monitoring at site S3.

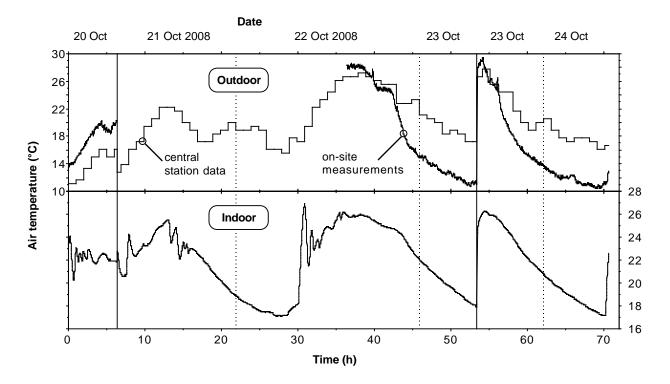


Figure F.32. Time series of air temperatures for observational monitoring at site S3.

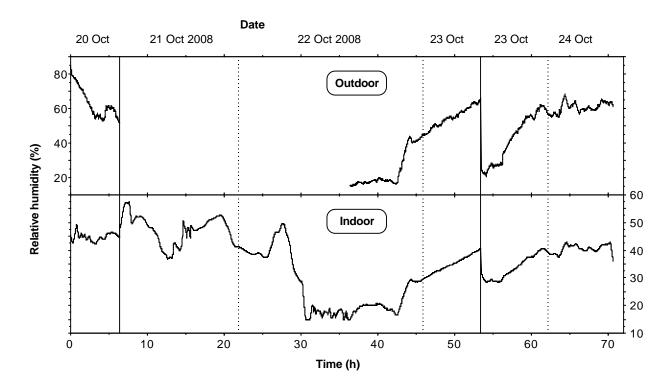


Figure F.33. Time series of relative humidities for observational monitoring at site S3.

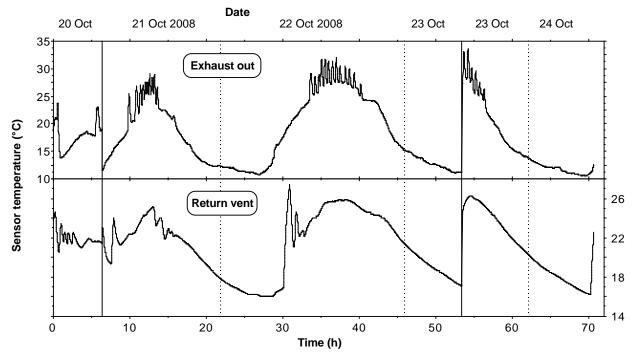
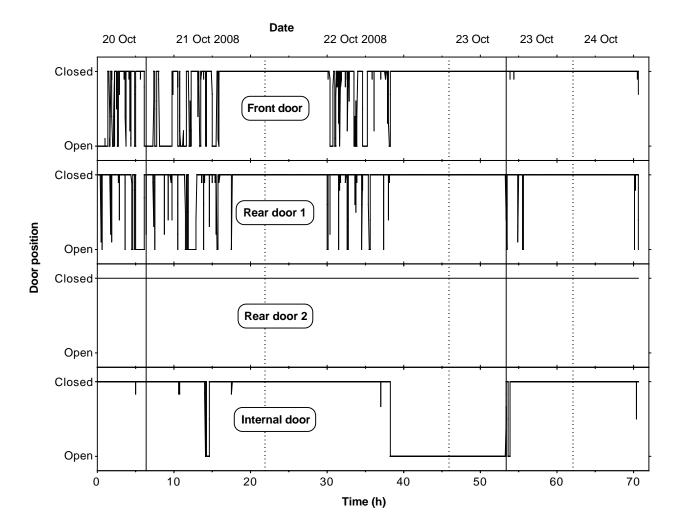
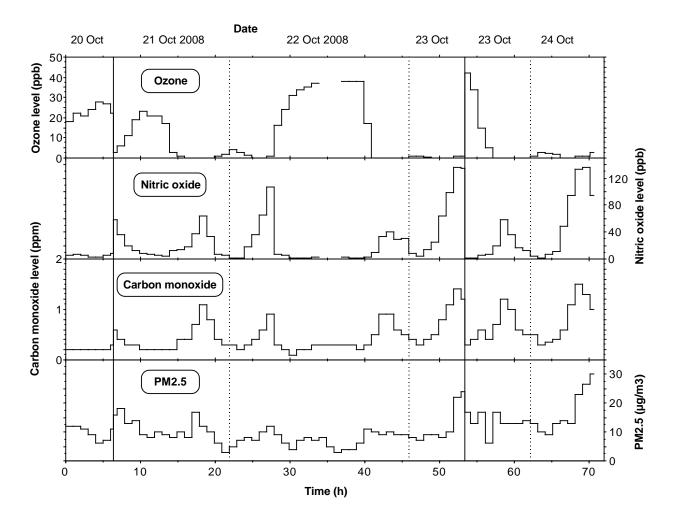


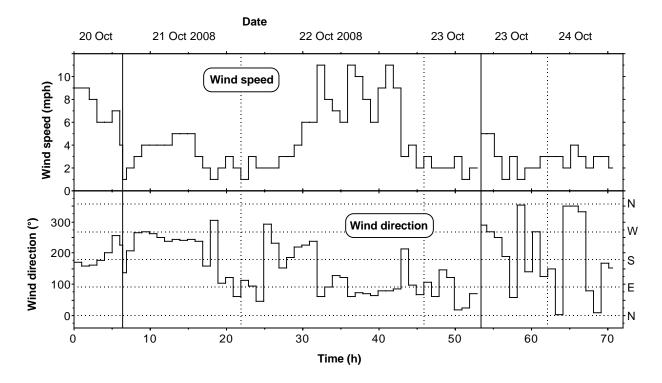
Figure F.34. Time series of sensor temperatures for observational monitoring at site S3.



**Figure F.35.** Time series of door positions for observational monitoring at site S3. Vertical spikes in the plot denote changes in the open or closed state that persisted for less than a clock minute.



**Figure F.36.** Time series of pollutant concentrations based on central-station monitoring for the period of observational monitoring at classroom site S3.



**Figure F.37.** Time series of wind speed and wind direction based on central-station monitoring for the period of observational monitoring at classroom site S3.

## F.4 Site S4

Observational monitoring was conducted at classroom site S4 for a continuous 72-hour period commencing at 6:45 on 4 November and ending at 7:05 on 7 November 2008.

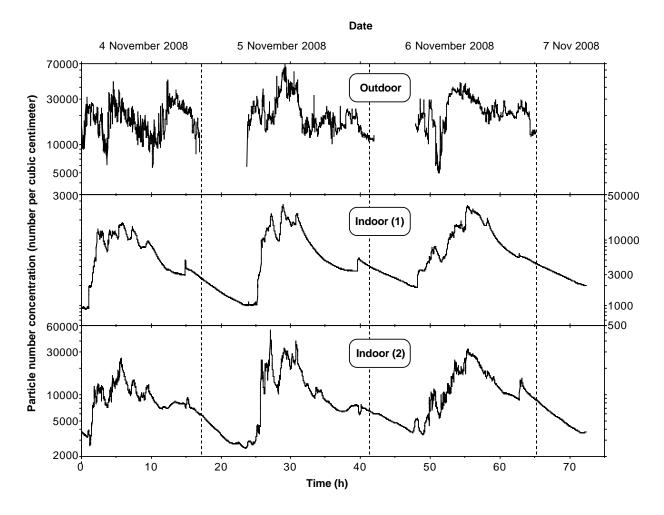
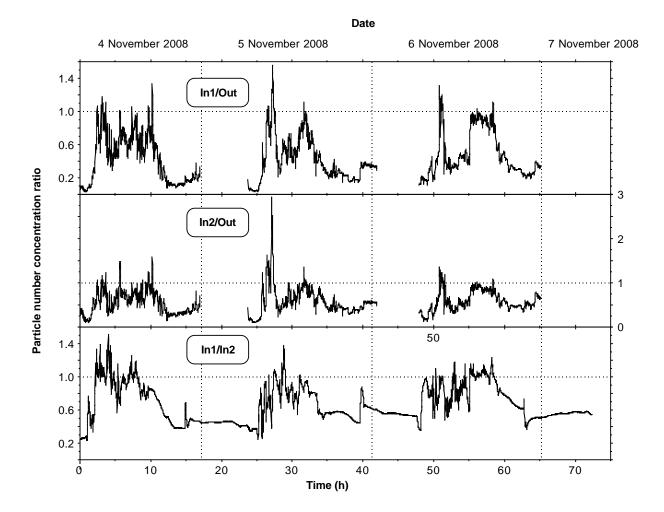


Figure F.38. Time series of particle number concentrations plotted on a logarithmic scale for observational monitoring at site S4.



**Figure F.39.** Time series of ratios of particle number concentrations for observational monitoring at site S4.

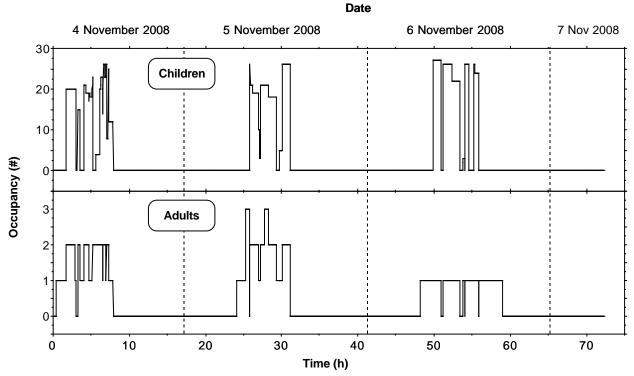


Figure F.40. Time series of occupancy status for observational monitoring at site S4.

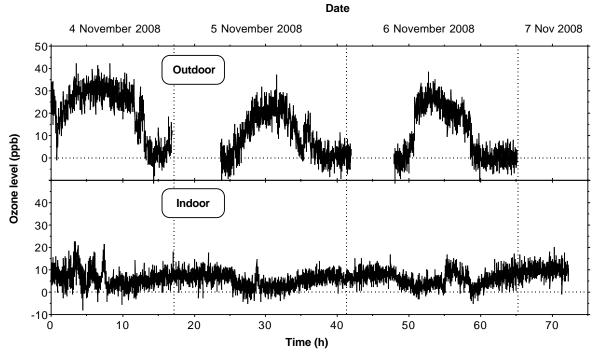


Figure F.41. Time series of ozone levels for observational monitoring at site S4.

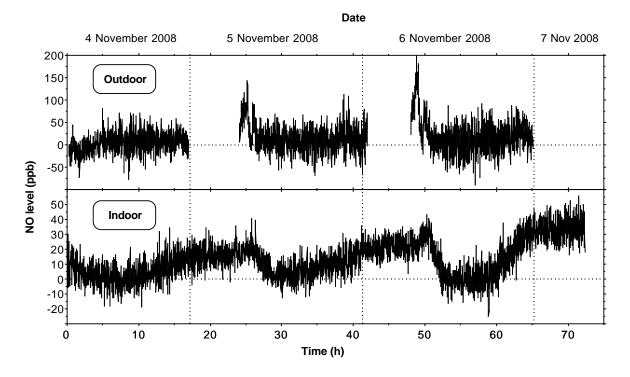


Figure F.42. Time series of nitric oxide levels for observational monitoring at site S4.

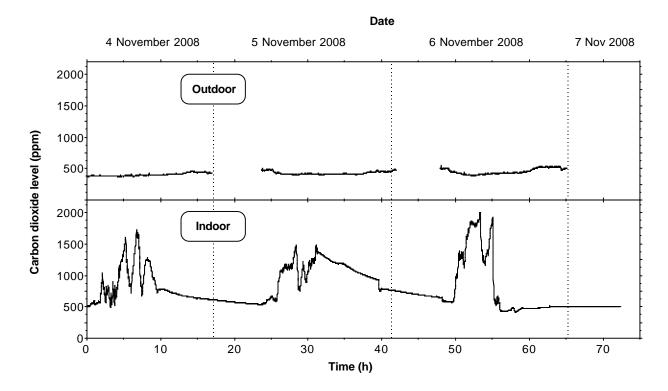


Figure F.43. Time series of carbon dioxide levels for observational monitoring at site S4.

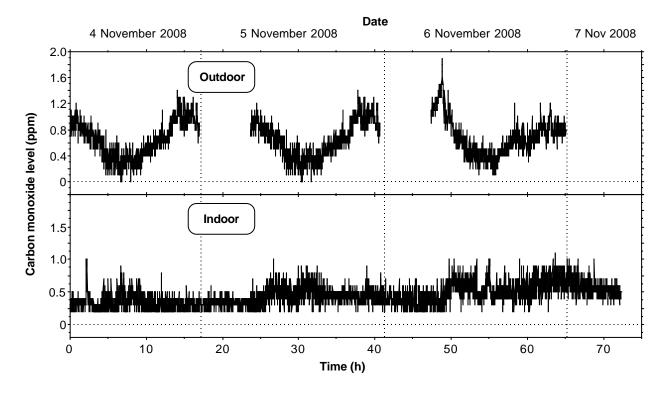


Figure F.44. Time series of carbon monoxide levels for observational monitoring at site S4.

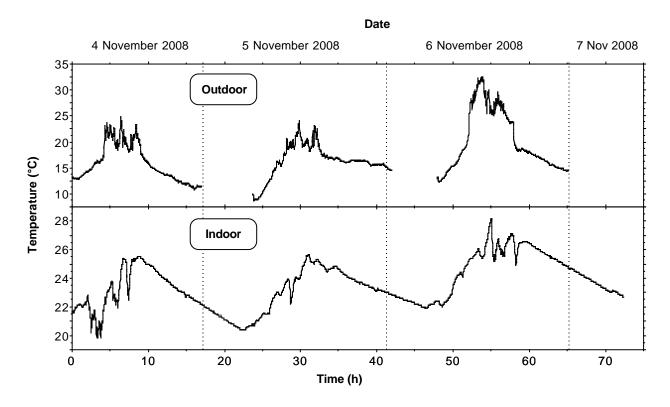


Figure F.45. Time series of air temperatures for observational monitoring at site S4.

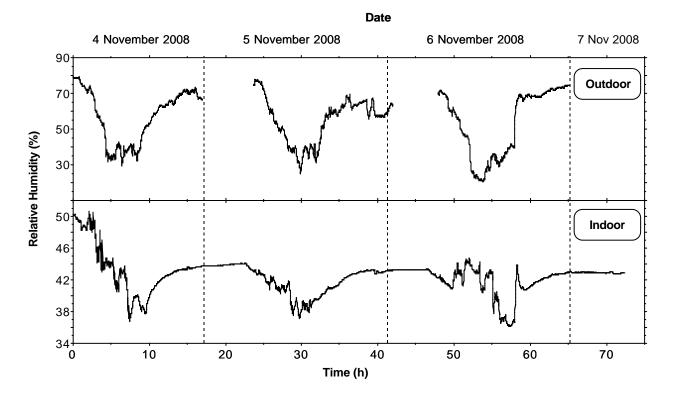


Figure F.46. Time series of relative humidities for observational monitoring at site S4.

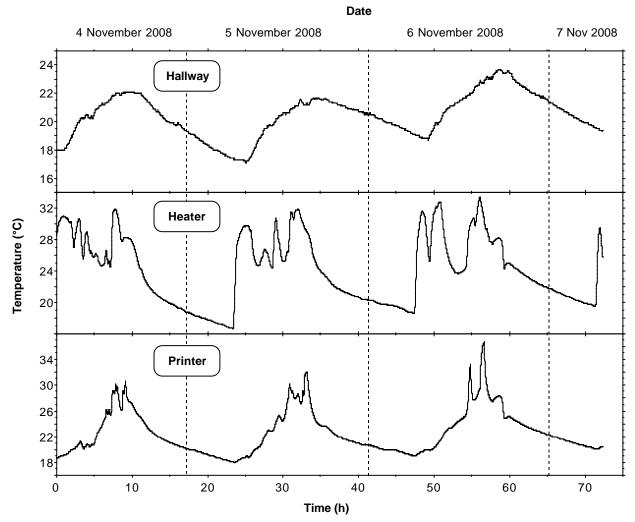
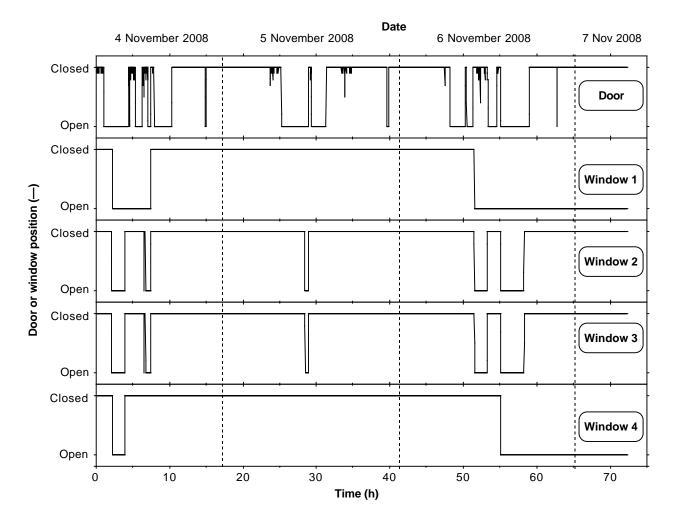
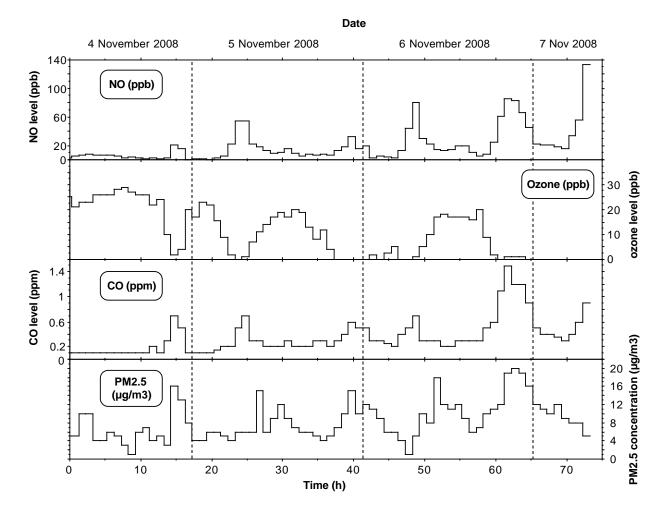


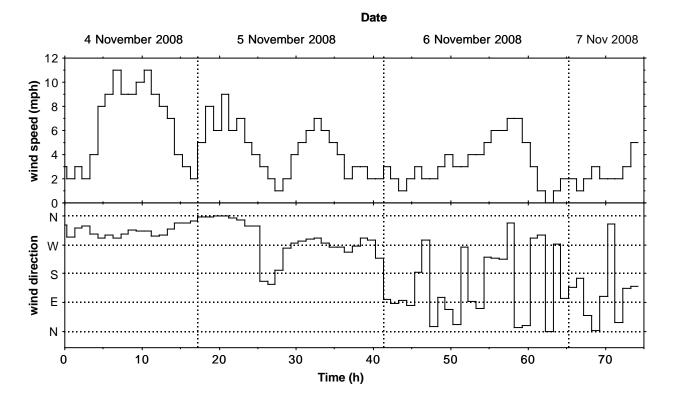
Figure F.47. Time series of sensor temperatures for observational monitoring at site S4.



**Figure F.48.** Time series of door and window positions for observational monitoring at site S4. Vertical spikes in the plot denote changes in the open or closed state that persisted for less than a clock minute.



**Figure F.49.** Time series of pollutant concentrations based on central-station monitoring for the period of observational monitoring at classroom site S4.



**Figure F.50.** Time series of wind speed and wind direction based on central-station monitoring for the period of observational monitoring at classroom site S4.

## F.5 Site S5

Observational monitoring was conducted at classroom site S5 for a contiguous 71-hour period commencing at 8:58 on 18 November and ending at 8:02 on 21 November 2008.

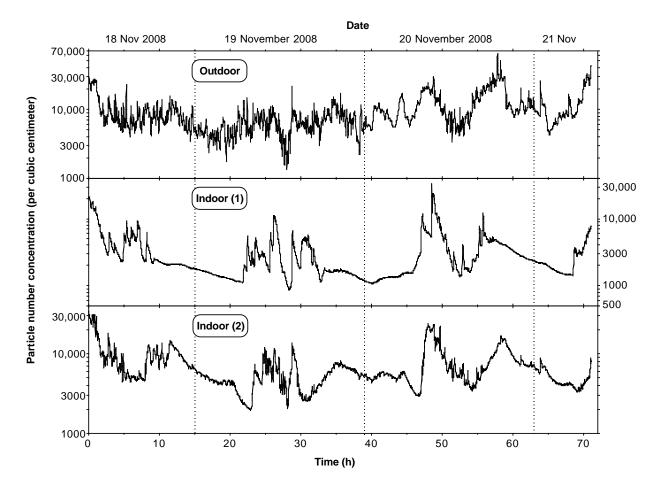
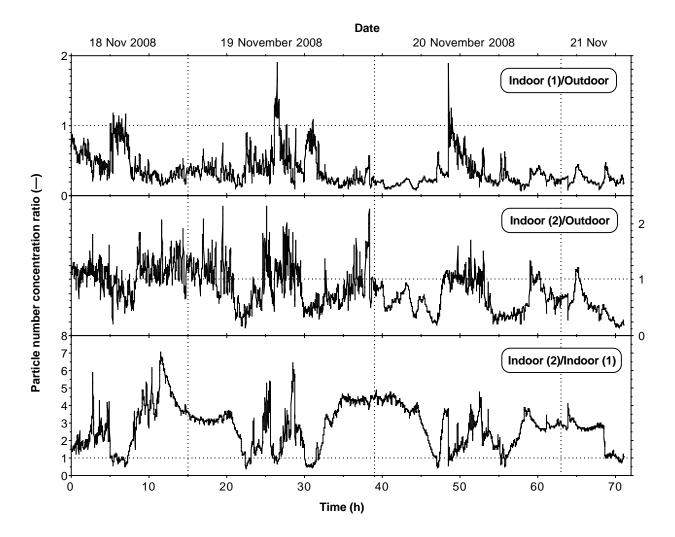


Figure F.51. Time series of particle number concentrations plotted on a logarithmic scale for observational monitoring at site S5.



**Figure F.52.** Time series of ratios of particle number concentrations for observational monitoring at site S5.

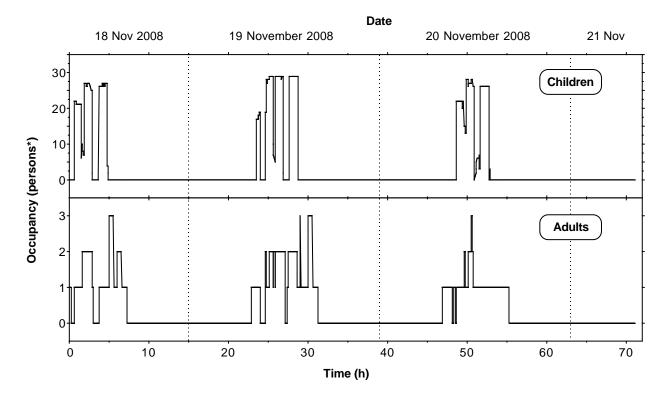


Figure F.53. Time series of occupancy status for observational monitoring at site S5.

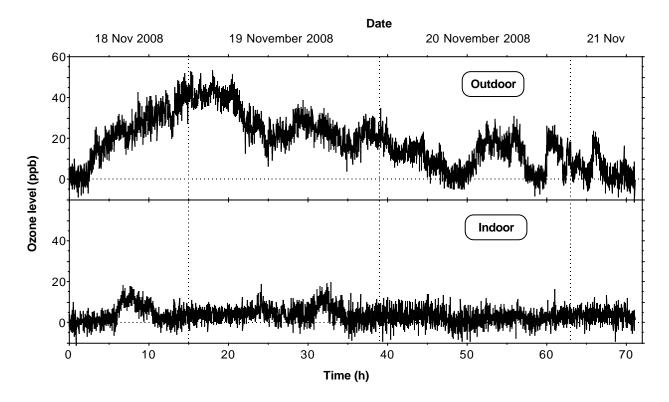


Figure F.54. Time series of ozone levels for observational monitoring at site S5.

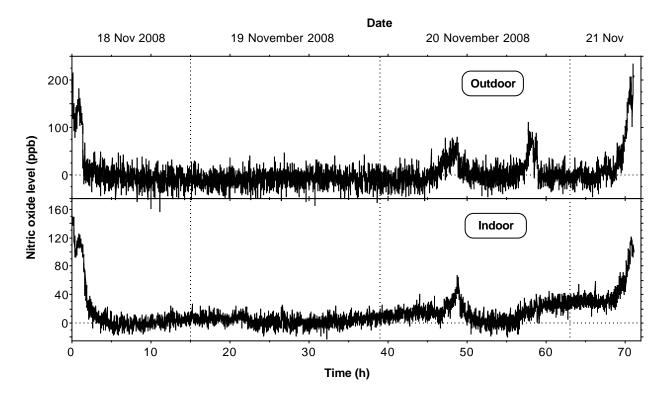


Figure F.55. Time series of nitric oxide levels for observational monitoring at site S5.

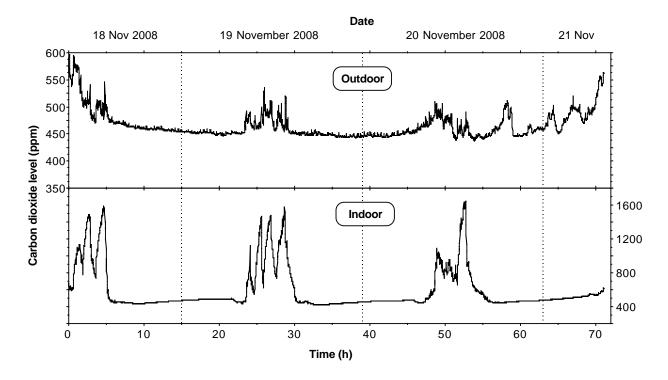


Figure F.56. Time series of carbon dioxide levels for observational monitoring at site S5.

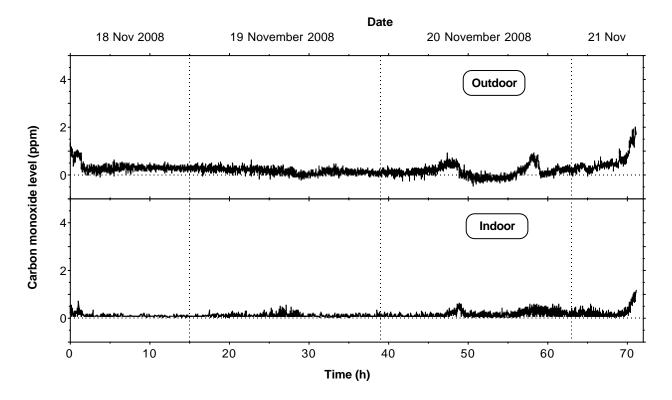


Figure F.57. Time series of carbon monoxide levels for observational monitoring at site S5.

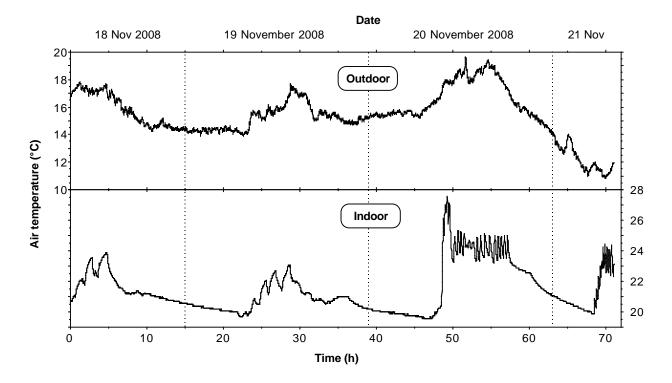


Figure F.58. Time series of air temperatures for observational monitoring at site S5.

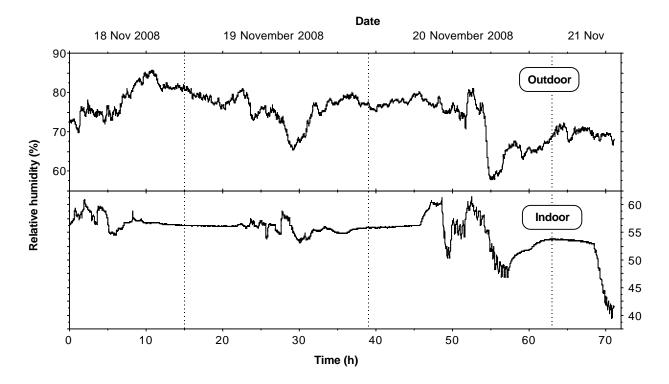


Figure F.59. Time series of relative humidities for observational monitoring at site S5.

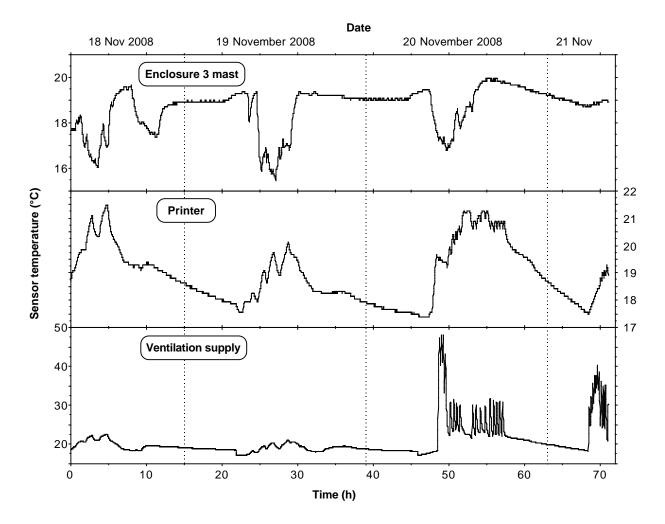
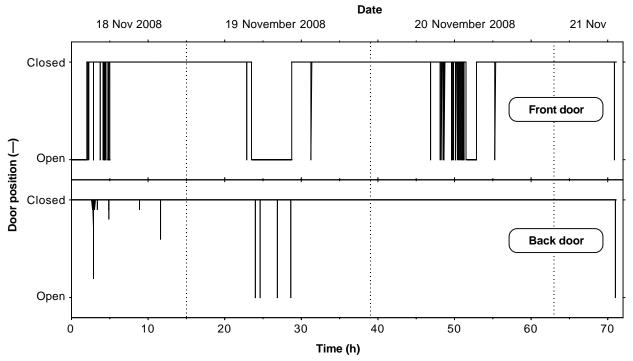
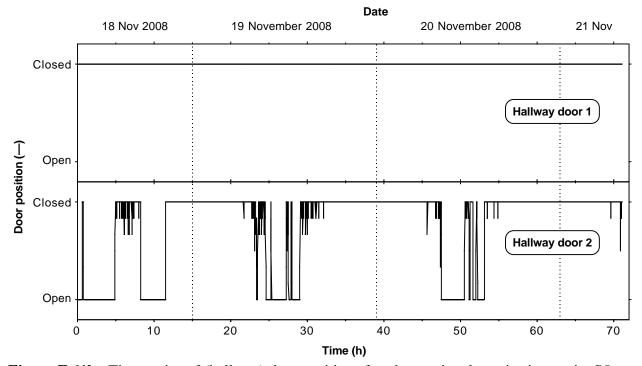


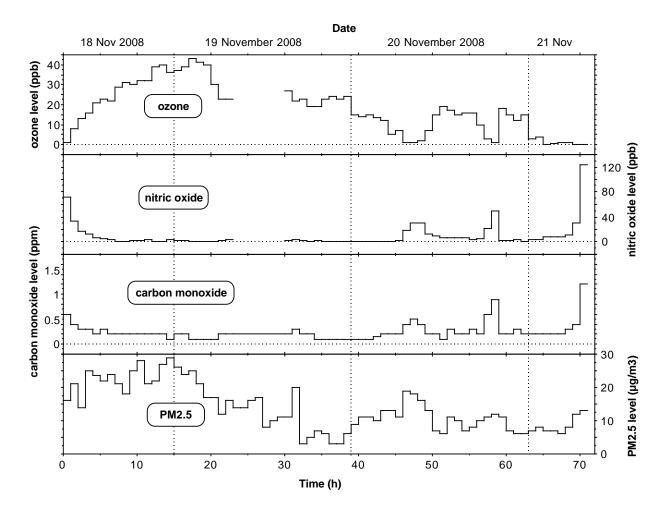
Figure F.60. Time series of sensor temperatures for observational monitoring at site S5.



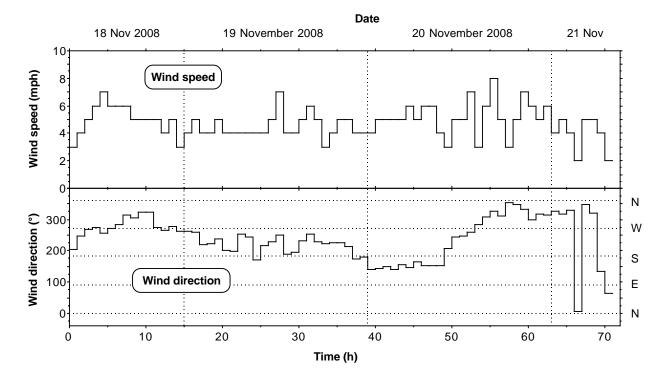
**Figure F.61a.** Time series of (front and back) door positions for observational monitoring at site S5. Vertical spikes in the plot denote changes in the open or closed state that persisted for less than a clock minute.



**Figure F.61b.** Time series of (hallway) door positions for observational monitoring at site S5. Vertical spikes in the plot denote changes in the open or closed state that persisted for less than a clock minute.



**Figure F.62.** Time series of pollutant concentrations based on central-station monitoring for the period of observational monitoring at classroom site S5.



**Figure F.63.** Time series of wind speed and wind direction based on central-station monitoring for the period of observational monitoring at classroom site S5.

## F.6 Site S6

Observational monitoring was conducted at classroom site S6 for a total of 7 hours. Monitoring occurred over four periods in late 2008: (i) from 8:15 on 2 December until 8:36 on 3 December; (ii) from 14:04 on 4 December until 8:30 on 5 December; (iii) from 12:25 on 5 December until 8:30 on 6 December; and (iv) from 8:42 until 15:48 on 8 December.

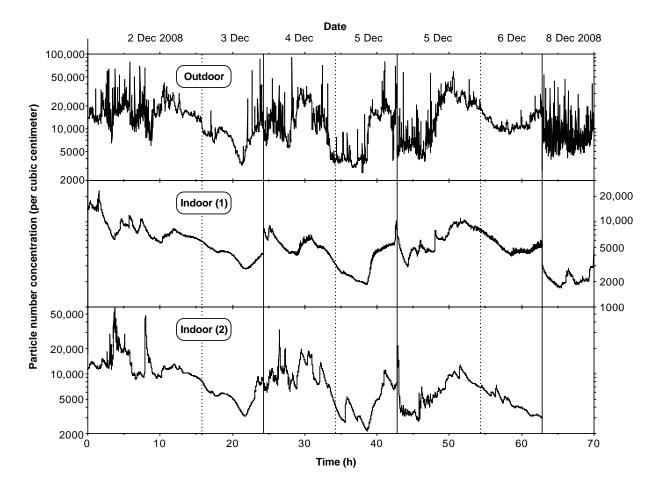
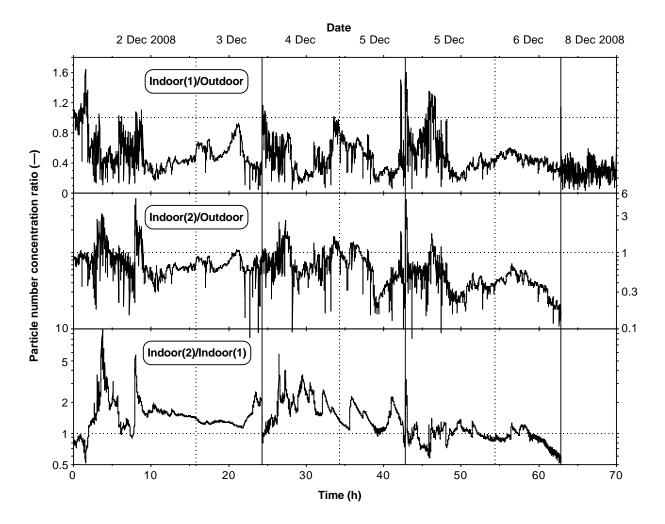


Figure F.64. Time series of particle number concentrations plotted on a logarithmic scale for observational monitoring at site S6.



**Figure F.65.** Time series of ratios of particle number concentrations for observational monitoring at site S6.

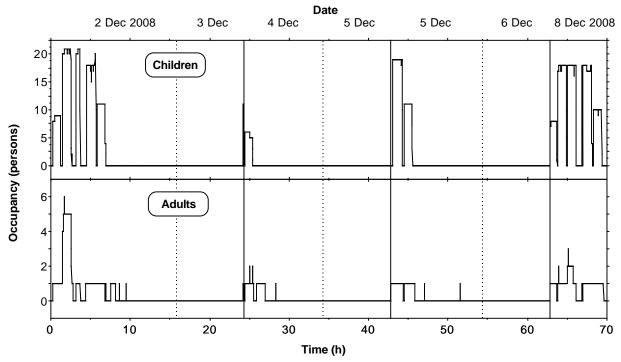


Figure F.66. Time series of occupancy status for observational monitoring at site S6.

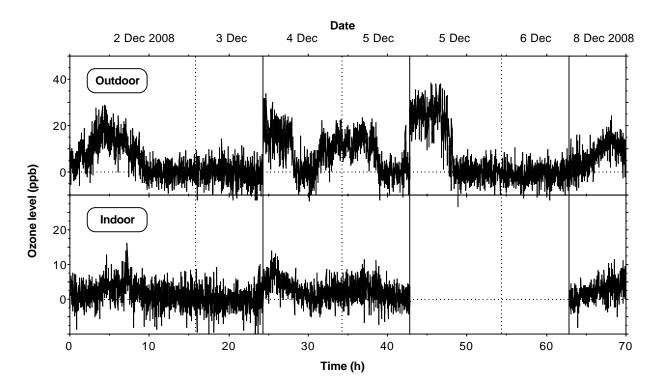


Figure F.67. Time series of ozone levels for observational monitoring at site S6.

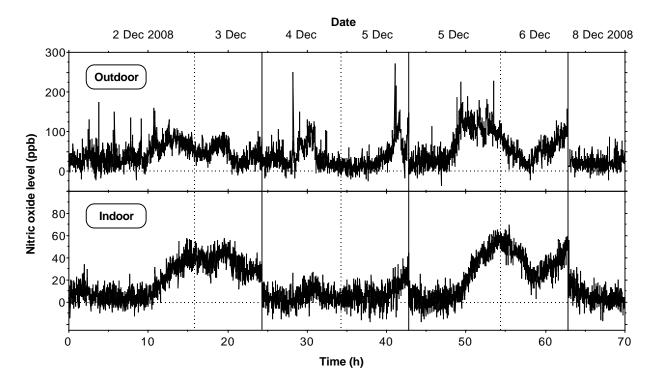


Figure F.68. Time series of nitric oxide levels for observational monitoring at site S6.

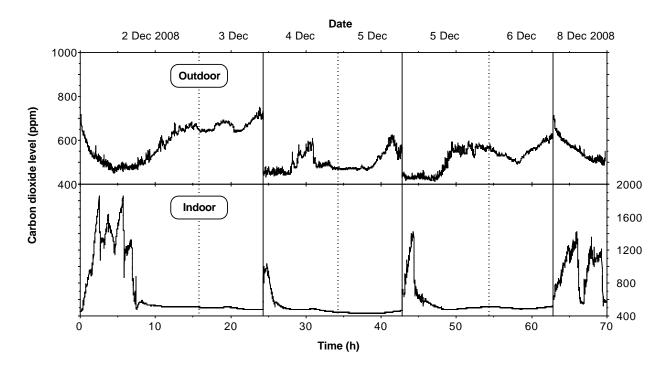


Figure F.69. Time series of carbon dioxide levels for observational monitoring at site S6.

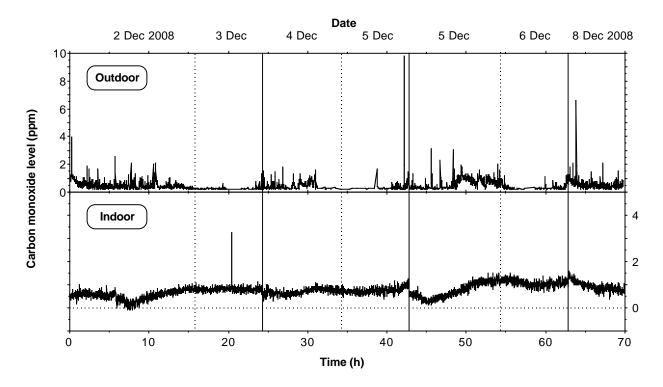


Figure F.70. Time series of carbon monoxide levels for observational monitoring at site S6.

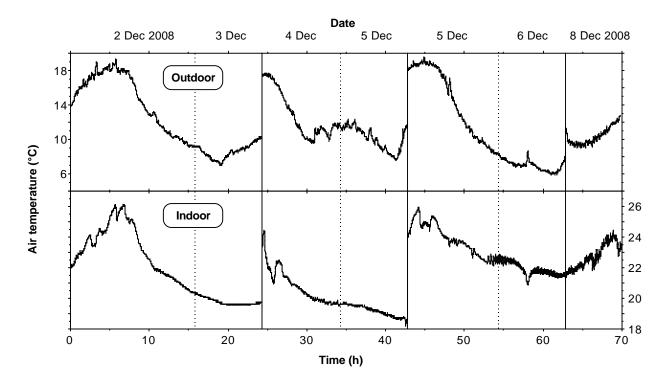


Figure F.71. Time series of air temperatures for observational monitoring at site S6.

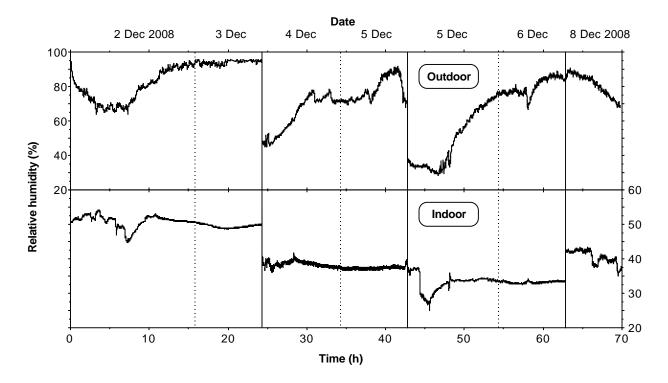


Figure F.72. Time series of relative humidities for observational monitoring at site S6.

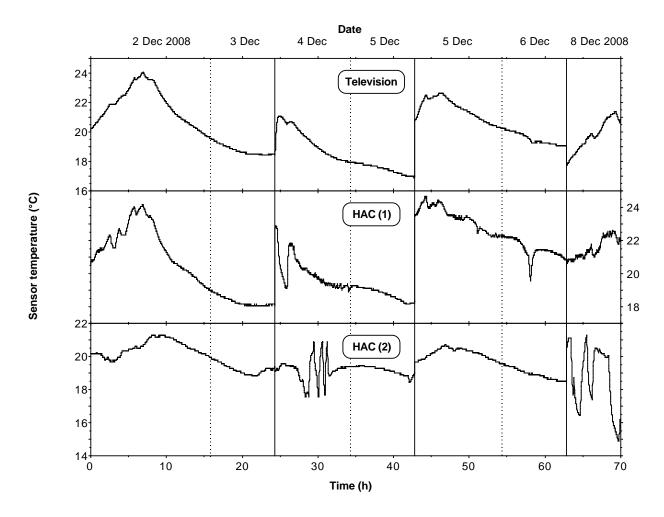
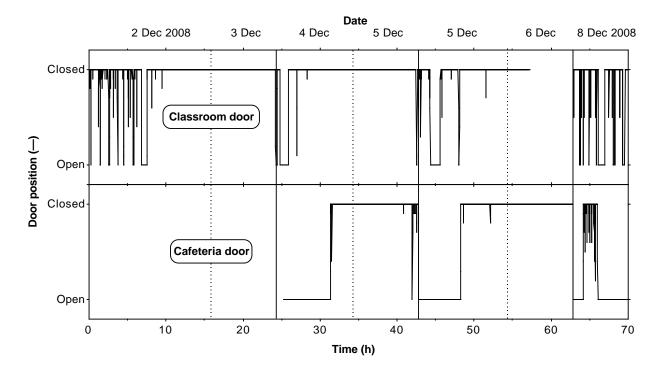
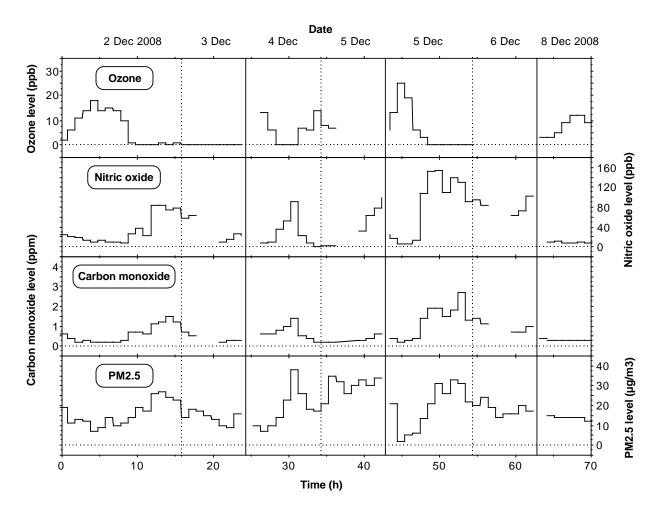


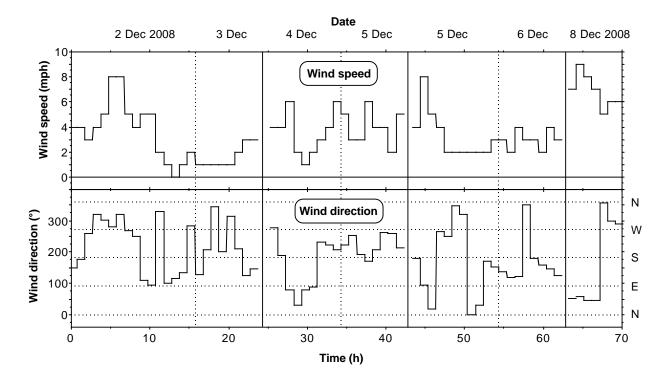
Figure F.73. Time series of sensor temperatures for observational monitoring at site S6.



**Figure F.74.** Time series of door positions for observational monitoring at site S6. Vertical spikes in the plot denote changes in the open or closed state that persisted for less than a clock minute.



**Figure F.75.** Time series of pollutant concentrations based on central-station monitoring for the period of observational monitoring at classroom site S6.



**Figure F.76.** Time series of wind speed and wind direction based on central-station monitoring for the period of observational monitoring at classroom site S6.

## APPENDIX G. PROBABILITY DISTRIBUTIONS OF PARTICLE NUMBER CONCENTRATIONS IN CLASSROOMS

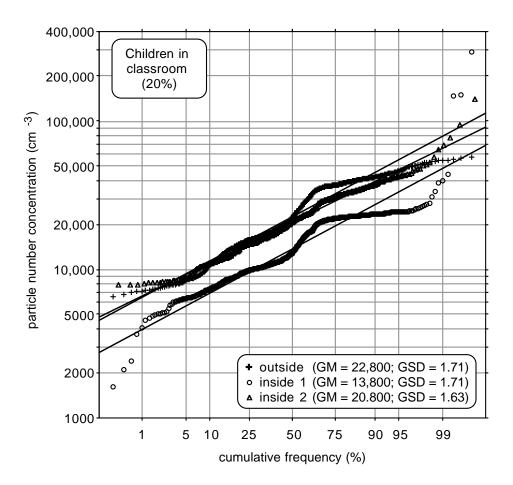
At the school sites, we observed that occupancy correlated with indoor particle levels. This appendix presents cumulative distribution functions of the minute-by-minute record of indoor and outdoor particle number concentrations for each classrom site, sorted according to occupancy status. In general, at each site, three figures are presented. In the first case, at least one student is present in the classroom. For the second figure, at least one adult is present in the classroom, which is a superset of the first condition. In the third figure, the classroom is vacant.

To produce these plots, the occupancy status was used to sort the minute-by-minute readings of PN concentrations from the full observational monitoring period. Once the data were grouped, results from each monitor were sorted in rank order for plotting the distribution. The geometric mean (GM) and geometric standard deviation (GSD) were computed for each monitor and each group, and lognormal distributions were plotted from these parameters on the cumulative distribution plots. In this coordinate system, a parameter that conforms to a lognormal distribution will follow a straight line. To the extent that the data do conform to a lognormal distribution considerable economy in reporting the information is gained, since two parameters, the GM and GSD, completely describe the distribution.

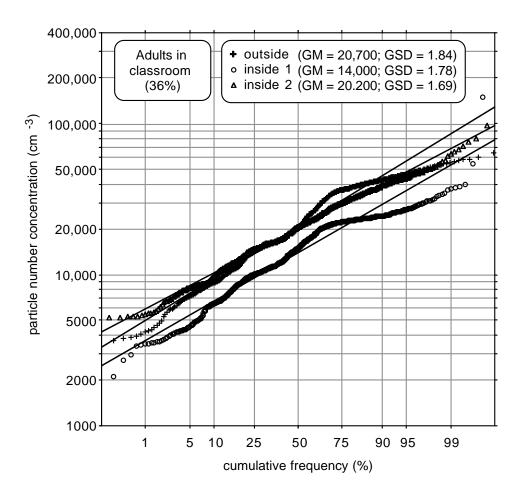
Figures G.1a-G.1c present data for site S1. Figure G.1a shows distributions for times when children are present in the classroom (20% of the monitoring period). The distributions are approximately parallel (the geometric standard deviations are very close to one another). Considering the classroom monitor results in relation to the outdoor monitor values, the ratio of geometric means is 0.61, close to the ratio of arithmetic means, 0.63. The classroom PN concentration distribution exhibits an upward excursion for only a brief proportion of the monitored time of student occupancy, about 1-2%, which corresponds to only 10-20 min during the four days of monitoring. For the outdoor monitor, the central 90% of the distribution spans the range 8500-46,000 cm<sup>-3</sup>.

Figure G.1b shows the cumulative distributions when an adult is in the classroom at site S1. These times represent a superset of the conditions displayed in Figure G.1a, including times when only the teacher (and possibly one or two aides) are in the room as well as times when children are present. The results in Figure G.1b are quite similar to those displayed in Figure G.1a, with GMs and GSDs for each of the three monitoring sites agreeing to within 10% between the two conditions. Figure G.1c shows the cumulative distributions when the classroom is vacant. The overall shape of the distributions is similar to those in Figures G.1a and G.1b, with the absence of the excursion to high indoor concentrations for the top 1-2% of the monitored period. The GSDs for all nine cases (three occupancy conditions  $\times$  three locations monitored) span a narrow range, from 1.63 to 1.84. The most noteworthy difference is the much lower mean concentrations for all three monitored sites when the classroom is vacant: the GMs for the vacant condition are in the range 35-50% of the corresponding values when the classroom is occupied by children.

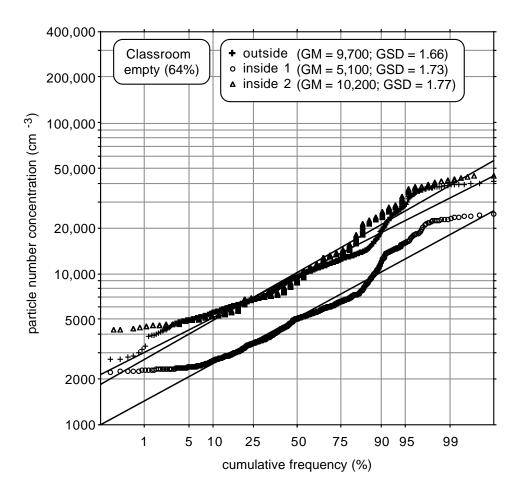
Overall, these distributions substantiate the interpretation that intrusion from the outdoor air is the dominant source of particles in the classroom and in the corridor during observational monitoring.



**Figure G.1a.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S1 when students were present in the classroom (20% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 22,800; classroom GM = 13,800; corridor GM = 20,800 cm<sup>-3</sup>).

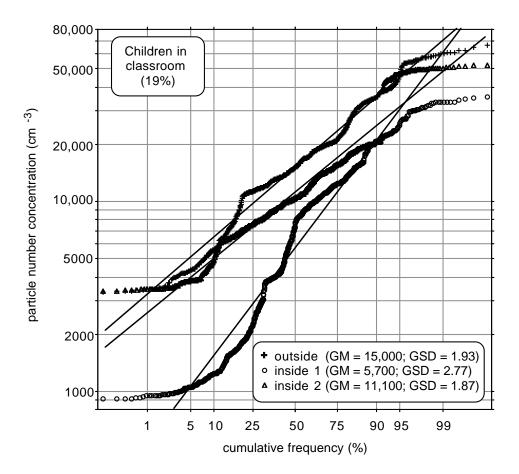


**Figure G.1b.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S1 when adults were present in the classroom (36% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 20,700; classroom GM = 14,000; corridor GM = 20,200 cm<sup>-3</sup>).



**Figure G.1c.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S1 when the classroom was vacant (64% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 9,700; classroom GM = 5,100; corridor GM = 10,200 cm<sup>-3</sup>).

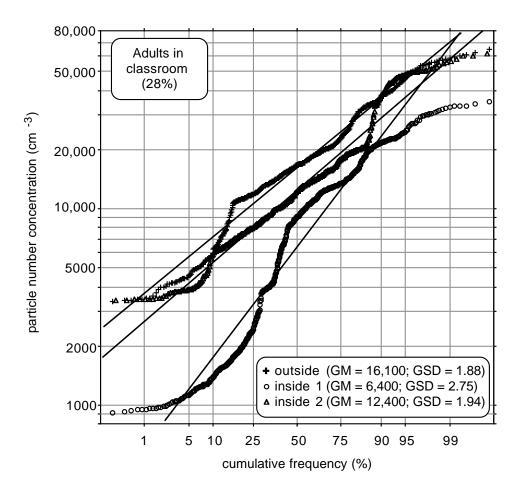
Figures G.2a-G.2c present distributions of particle number concentrations for site S2. Figure G.2a shows distributions for times when children are present in the classroom (19% of the monitoring period); for Figure G.2b, adults are in the classroom (28% of the period); and Figure G.2c represents conditions when the classroom is vacant (72% of the time). In this discussion, we will focus on the outdoor and classroom ("inside 1") monitor results and emphasize the conditions when students are present and when the room is vacant.



**Figure G.2a.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S2 when students were present in the classroom (19% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 15,000; classroom GM = 5,700; cafeteria GM = 11,100 cm<sup>-3</sup>).

The outdoor distributions conform reasonably well to lognormal distributions with fairly consistent dispersion, as indicated by the GSDs being close to 1.9 in all three cases. The geometric mean is roughly twice as high when the classroom is occupied as when it is vacant. When children are present, the central 90% of the outdoor PN distribution spans about an order of magnitude, 4300-44,000 cm<sup>-3</sup>. When children are absent, the central 90% of the outdoor distribution again spans about an order of magnitude, but shifted downward to 2900-27,000 cm<sup>-3</sup>.

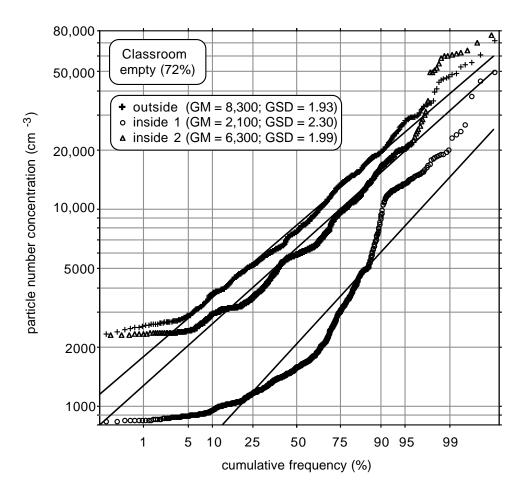
The classroom distributions exhibit a higher level of dispersion and do not conform particularly well to a lognormal distribution, showing a curvilinear pattern on the log-probability coordinates. When the classroom is occupied, the central 90% of the distribution varies from  $1100 \text{ cm}^{-3}$  to ~ 25,000 cm<sup>-3</sup>. When the room is unoccupied, the distribution shifts downward so that the central 90% spans 900 cm<sup>-3</sup> to 13,000 cm<sup>-3</sup>. However, the change in median value is most striking: from 7600 cm<sup>-3</sup> when occupied to 1600 cm<sup>-3</sup> when vacant.



**Figure G.2b.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S2 when adults were present in the classroom (28% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 16,100; classroom GM = 6,400; cafeteria GM = 12,400 cm<sup>-3</sup>).

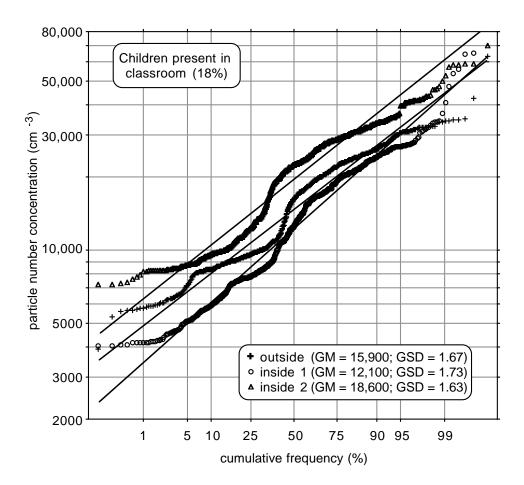
Consistently, the proportional difference between outdoor and indoor concentrations is smaller at the higher ends of the distributions than at the lower end. When students are present, this closer tracking applies across the entire upper 50% of the distributions, whereas when the classroom is vacant, only across the upper 10% of the distributions do indoor and outdoor levels track with the minimal separation distance.

The underlying explanation for these behaviors might be fairly simple. The opening of windows and doors is likely to be more common during afternoons than during the mornings. When the windows and doors are open, we expect the I/O ratio to be higher. The diurnal pattern of outdoor PN levels exhibits the highest levels in the early afternoon. So, it seems that the high outdoor levels and the natural-ventilation rate conditions might be correlated, producing higher I/O ratios at just the time when the outdoor levels are higher than average.



**Figure G.2c.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S2 when the classroom was unoccupied (72% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 8,300; classroom GM = 2,100; cafeteria GM = 6,300 cm<sup>-3</sup>).

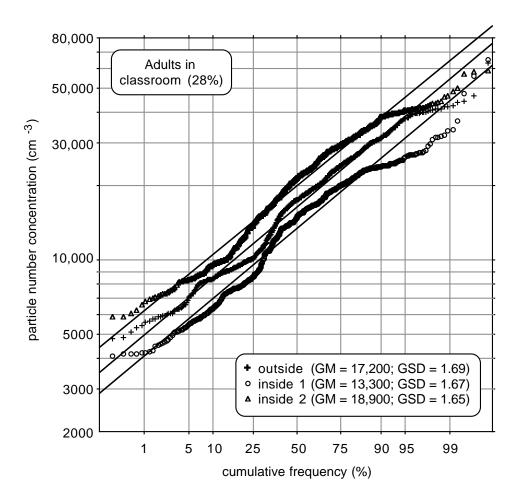
Figures G.3a-G.3c present data on indoor and outdoor PN concentrations sorted according to occupancy status for site S3. Figure G.3a shows distributions for times when children are present in the classroom (18% of the monitoring period); Figure G.3b shows the distribution when one or more adults are in the classroom (28% of the period); and Figure G.3c presents the indoor distributions at times when the classroom is vacant.



**Figure G.3a.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S3 when students were present in the classroom (18% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 15,900; classroom GM = 12,100; cafeteria GM = 18,600 cm<sup>-3</sup>).

The three distributions in the three figures track remarkably well in the sense that they trace approximately parallel trajectories across almost the entire percentile range. One minor deviation is observed when the classroom is occupied. The classroom PN concentration is lower than the outdoor level at all percentiles except for the upper few percent, where the indoor concentration is higher than that outdoors. This behavior reflects conditions in which almost all of the time indoor particles are predominantly of outdoor origin; however for a few percent of the interval (i.e., a few tens of minutes), indoor sources make significant contributions to indoor PN levels. When the classroom is vacant, this crossover behavior is absent: the outdoor PN levels are higher than the indoor levels across the full percentile range.

Another interesting broad observation is that the distribution of PN levels in the cafeteria uniformly exceeds the corresponding level outdoors when the classroom is occupied. Conversely, when the classroom is vacant, the PN level outdoors is consistently higher than that in the cafeteria.

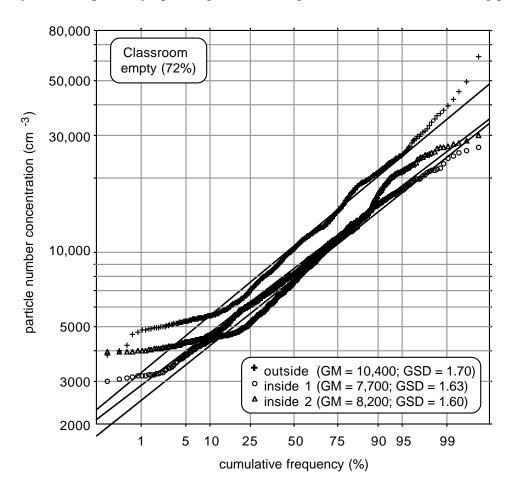


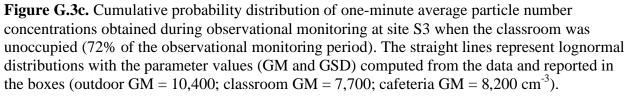
**Figure G.3b.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S3 when adults were present in the classroom (28% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 17,200; classroom GM = 13,300; cafeteria GM = 18,900 cm<sup>-3</sup>).

The variances for all distributions are similar, ranging from 1.60 to 1.73. The consistency in the variances is suggestive of a common main source, which we believe to be the outdoor air. Also reflected in the consistent variances are similar values for the multiplicative dynamic range in each distribution. For example, for the outdoor PN concentration when students are in the classroom, the central 90% of the distribution spans a range of about a factor of 5, from 6800 to 34,000 cm<sup>-3</sup>. The classroom PN concentration when students are present exhibits a similar dynamic range, varying across the central 90% of the distribution by a factor of 5, from 5100 to 27,000 cm<sup>-3</sup>. Consistent GSDs also imply that the ratio of GMs is similar to the ratio of arithmetic means. Again, consider the case when students occupy the classroom. The ratio of the classroom to outdoor arithmetic means (Figure 4.18) is 0.78, similar to the ratio of the GMs, 0.76.

It is interesting and noteworthy that the indoor/outdoor ratio when the classroom is vacant is not too different than the ratio when the classroom is occupied. The ratio of GMs is 0.74 when the classroom is vacant, 0.77 when the teachers occupy the room, and 0.76 when

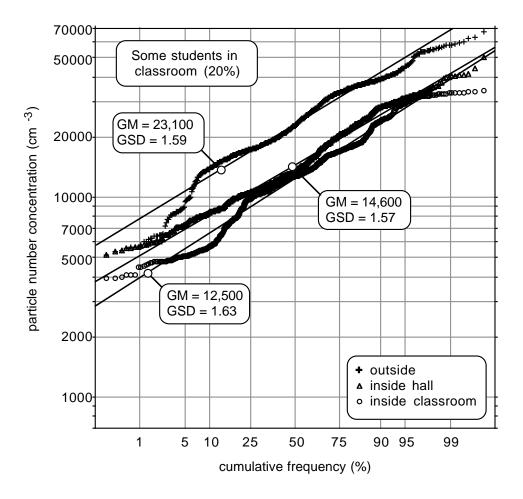
students are present. The similarity of these values — especially when combined with the parallel data traces in the distributional curves — suggests that the dominant factors that govern the indoor to outdoor PN relationship do not vary with occupancy. Not only does this information reinforce the impression that outdoor particles are more important contributors to indoor particles than indoor sources at this site, it also indicates that the mechanical ventilation system was probably operating 24 h/d during the observational monitoring period.



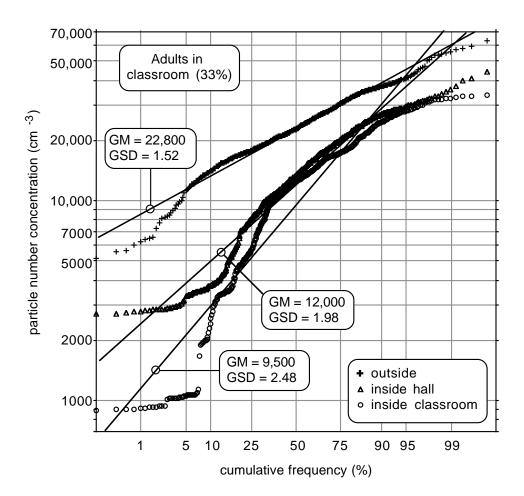


Figures G.4a-G.4d present data on indoor and outdoor PN concentrations sorted according to occupancy status for site S4. Figure G.4a shows distributions for times when children are present in the classroom (20% of the monitoring period); and for Figure G.4b, adults are in the classroom (33% of the period). For Figure G.4c, we present the indoor distributions at times when the classroom is vacant. The outdoor concentration distribution is not plotted in this figure because the overnight data are missing. In Figure G.4d, we present the distributions, both indoor outdoors, for the conditions in which the classroom is vacant *and* the outdoor monitor was operating (39% of the total monitoring period). For all three distributions where indoor and

outdoor data are available, the classroom PN concentrations are lower than the outdoor values across all percentiles.

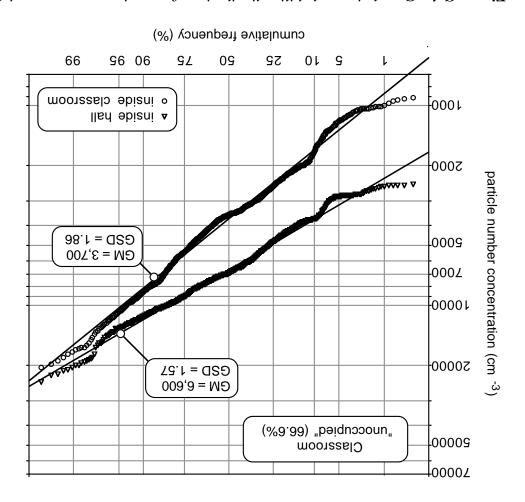


**Figure G.4a.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S4 when students were present in the classroom (20% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 23,100; classroom GM = 12,500; hallway GM = 14,600 cm<sup>-3</sup>).



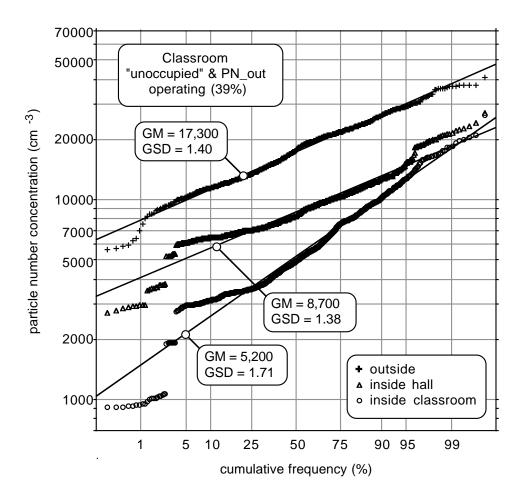
**Figure G.4b.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S4 when adults were present in the classroom (33% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 22,800; classroom GM = 9,500; hallway GM = 12,000 cm<sup>-3</sup>).

When students are in the classroom (Figure G.4a), the outdoor and classroom distributions track in an approximately parallel fashion. The GSDs for both distributions are similar: 1.59 for the outdoor PN concentration and 1.63 for the classroom concentration. Because the distributions are nearly parallel, the ratio of geometric means, 0.54, is almost the same as the ratio of the arithmetic means, 0.55. It also implies that the dynamic range in each distribution spans a similar multiplicative range. For the outdoor PN concentration, the central 90% of the distribution varies from 9700 to 44,000 cm<sup>-3</sup>. For the classroom concentration, the corresponding range is ~ 5100-29,000 cm<sup>-3</sup>. When the classroom is unoccupied (Figure G.4c), the concentration conforms well to a lognormal distribution. Compared with conditions when the classroom is occupied with students, the GM is markedly reduced, from  $12.5 \times 10^3$  cm<sup>-3</sup> to  $3.7 \times 10^3$  cm<sup>-3</sup>. The distribution's range is larger when the classroom is unoccupied as reflected in the larger GSD, 1.86 versus 1.63.



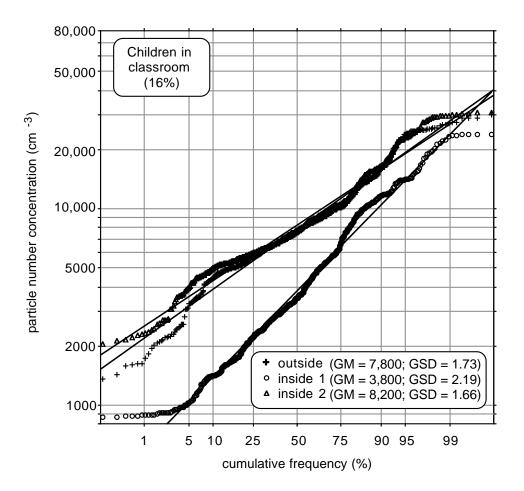
**Figure G.4c.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S4 when the classroom was unoccupied (67% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the data and reported in the boxes (classroom GM = 3,700; hallway GM =  $6,600 \text{ cm}^{-3}$ ).

The consistently higher outdoor than classroom concentrations exhibited and the fairly similar shapes of the distributions reinforce the impression that the dominant source of PN in the classroom at this site is the penetration and persistence of particles from outdoor air.



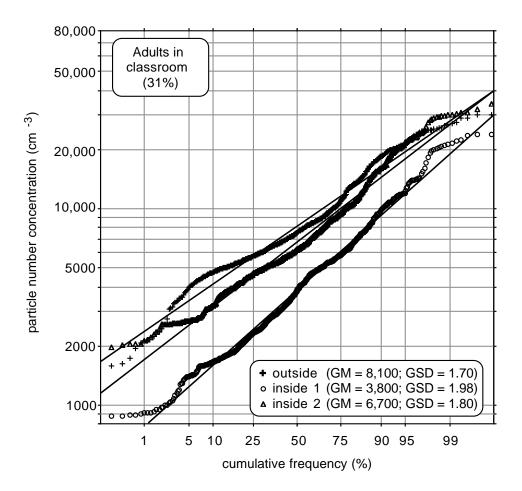
**Figure G.4d.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S4 when the classroom was unoccupied and the outdoor monitor was operating (39% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 17,300; classroom GM = 5,200; hallway GM =8,700 cm<sup>-3</sup>).

Figures G.5a-G.5c present data on indoor and outdoor PN concentrations sorted according to occupancy status for site S5. Figure G.5a shows distributions for times when children are present in the classroom (16% of the monitoring period); Figure G.5b shows the distribution when one or more adults are in the classroom (31% of the period); and Figure G.5c presents the indoor distributions at times when the classroom is vacant.



**Figure G.5a.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S5 when students were present in the classroom (16% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 7,800; classroom GM = 3,800; hallway GM = 8,200 cm<sup>-3</sup>).

When students are present in the classroom, the distributions of PN concentrations are reasonably well described by lognormal distributions. The central 90% of the distribution of outdoor PN concentrations spans the range 3300-24,000 cm<sup>-3</sup>. For the classroom, the corresponding range is shifted downward and proportionately broader, 1,000-14,000 cm<sup>-3</sup>. The distributions are qualitatively similar but differ in detail over the longer period when the teacher is in the classroom. For the outdoor PN distribution with teachers present, the 5<sup>th</sup> percentile is 4000 cm<sup>-3</sup> and the 95<sup>th</sup> percentile is 21,000 cm<sup>-3</sup>. For the classroom, the corresponding values are 1,400 and 12,000 cm<sup>-3</sup>, respectively.



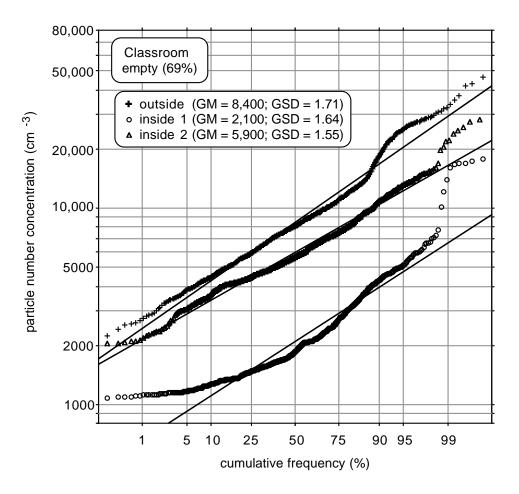
**Figure G.5b.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S5 when adults were present in the classroom (31% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 8.100; classroom GM = 3,800; hallway GM = 6,700 cm<sup>-3</sup>).

It has already been noted that this site is unusual in that the outdoor PN concentrations do not exhibit a strong diurnal pattern. This characteristic can be seen by comparing the parameters of the lognormal distributions for outdoor PN concentrations among the three occupancy conditions. The GSDs, which are indicative of the variances in the distributions, span a narrow range from 1.70-1.73. The GMs also do not differ much, ranging between 7800 and 8400 cm<sup>-3</sup>.

The distributions of concentrations in the classroom show more variability with respect to occupancy than do the outdoor concentrations at this site. Note, for example, that when the classroom is occupied (with or without students included), the classroom PN concentration is less than 2,000 cm<sup>-3</sup> about 20% of the time and greater than 10,000 cm<sup>-3</sup> ~ 10% of the time. However, when the classroom is unoccupied, the PN concentration is less than 2,000 cm<sup>-3</sup> more than half of the time and greater than 10,000 cm<sup>-3</sup> only about 1% of the time.

The conditions at S5 point strongly to the importance of building operation conditions in affecting indoor PN concentrations. Here, more so than at any other site, the particle concentrations outdoors are substantially independent of occupancy. And, as at all school sites

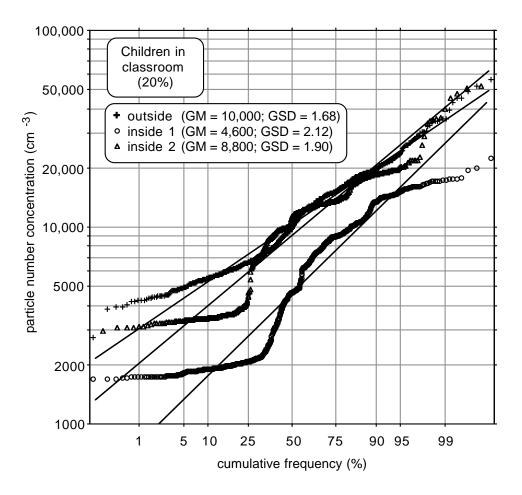
studied, the indoor particles are mainly of outdoor origin. Yet, the indoor concentrations here are markedly different when the classroom is unoccupied than when students or teachers are present. The explanation lies in two main factors here. First, the mechanical ventilation system operates continuously when the classroom is occupied, but only for a portion of the time when the classroom is vacant. Second, the classroom doors are open a substantial fraction of the occupied periods but only a small proportion of the time when the classroom is vacant. In the absence of good filtration on ventilation supply air, lower air-exchange rates will tend to correlate with lower concentrations of particles of outdoor origin.



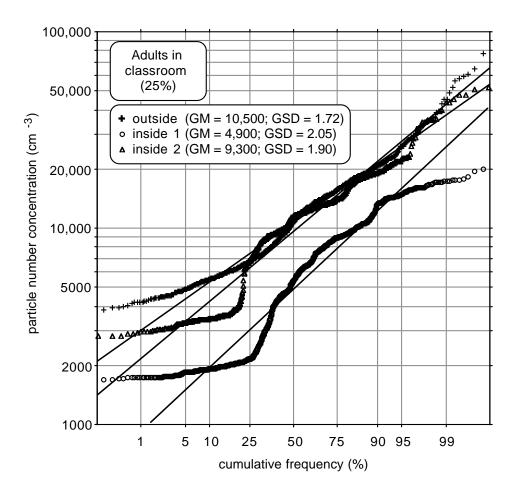
**Figure G.5c.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S5 when the classroom was unoccupied (69% of the observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the data and reported in the boxes (outdoor GM = 8,400; classroom GM = 2,100; hallway GM = 5,900 cm<sup>-3</sup>).

Figures G.6a-G.6c present data on indoor and outdoor PN concentrations sorted according to occupancy status for site S6. Figure G.6a shows distributions for times when children are present in the classroom (20% of the monitoring period); Figure G.6b shows the distribution when one or more adults are in the classroom (25% of the period); and Figure G.6c presents the indoor distributions at times when the classroom is vacant (75% of the time). For all

three distributions, the classroom PN concentrations are lower than the outdoor values across all percentiles.

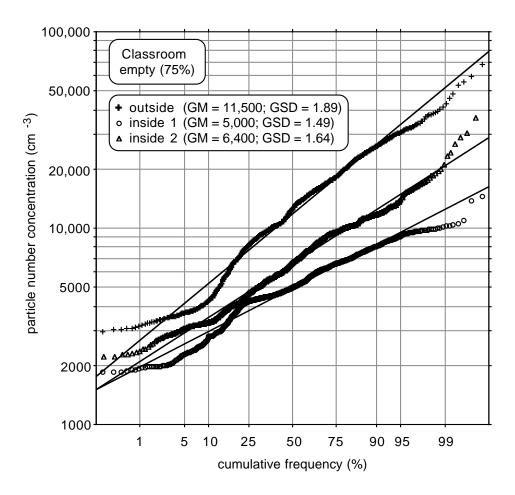


**Figure G.6a.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S6 when students were present in the classroom (20% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 10,000; classroom GM = 4,600; cafeteria GM = 8,800 cm<sup>-3</sup>). The cafeteria data ("inside 2") represent the distributions for the first 63 h of the observational monitoring period.



**Figure G.6b.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S6 when adults were present in the classroom (25% of the total observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the corresponding data and reported in the boxes (outside GM = 10,500; classroom GM = 4,900; cafeteria GM = 9,300 cm<sup>-3</sup>. The cafeteria data ("inside 2") represent the distributions for the first 63 h of the observational monitoring period.

When students are present (Figure G.6a), the outdoor distribution spans a narrower multiplicative range than the classroom. For the outdoor PN concentration, the central 90% of the distribution varies from 4,800 to 22,800 cm<sup>-3</sup>, a factor of ~ 5 from high to low. For the classroom concentration, the corresponding range is 1,800-15,000 cm<sup>-3</sup>, a factor of ~ 8. Interestingly, when the classroom is unoccupied (Figure G.6c), the spread in dynamic range is reversed. The central 90% of the outdoor PN concentration varies 3,600-29,800 cm<sup>-3</sup>, a factor of ~ 8. The corresponding numbers for the classroom PN concentration are 2,300-9,100 cm<sup>-3</sup>, a factor of ~ 4.



**Figure G.6c.** Cumulative probability distribution of one-minute average particle number concentrations obtained during observational monitoring at site S6 when the classroom was unoccupied (75% of the observational monitoring period). The straight lines represent lognormal distributions with the parameter values (GM and GSD) computed from the data and reported in the boxes (outdoor GM = 11,500; classroom GM = 5,000; cafeteria GM = 6,400 cm<sup>-3</sup>). The cafeteria data ("inside 2") represent the distributions for the first 63 h of the observational monitoring period.

The consistently higher outdoor than classroom PN concentrations reinforce the impression that the dominant source of PN in the classroom at this site is the penetration and persistence of particles from outdoor air.