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PILOT INVESTIGATION OF INDOOR-OUTDOOR AND PERSONAL PM10 (THORACIC) AND ASSOCIATED IONIC COMPOUNDS AND MUTAGENIC ACTIVITY

Final Report

Prepared for California Air Resources Board
Contract No. A6 - 129 - 33--California Air Resources Board
27 April 1989

Principal Investigator
Steven D. Colome
Integrated Environmental Services

Co-Investigators
Norman Y. Kado
California Air Resources Board
and
University of California, Davis

Michael Kleinman
University of California, Irvine

Research Staff
Peter Jaques
New York University

Public Policy Research Organization
University of California, Irvine
Irvine, CA 92717

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ABSTRACT

This report describes a laboratory and field pilot study designed to investigate personal and indoor exposures to particulate matter. The primary goal of this research project was to develop needed methods of measurement and characterization of exposures to airborne particles equal to or less than 10 micrometers in diameter (PM10). Using fully developed and tested methods, it should be possible to provide improved dose estimates for PM10 and to relate PM10 and its associated components to health effects. Exposure to particulate matter is currently assessed using fixed-site monitors that may not accurately measure exposure of humans to particles. This study focuses on personal exposure and indoor concentration, since a person typically spends 80-90% of the day indoors.

In the study three sampling devices were used and compared for measurement of PM10 particles: 1) an air pump and sampler specifically designed to collect PM10 particles in indoor environments; 2) an industrial hygiene-type (cyclone) sampler designed to measure personal exposure to inhalable aerosols; and 3) a multistage personal (cascade) sampler designed to quantify concentration for several particle sizes. Filters from these samplers were analyzed for concentration of particles in air, concentration of potentially acid-forming ionic compounds, and mutagenic activity. Ancillary samples were taken for nicotine and biological aeroallergens. Nicotine is a specific marker for tobacco smoke, which has a large impact on indoor particulate matter, and aeroallergen concentration is a necessary control variable when investigating the effects of air pollution on the health of allergic asthmatics (the subjects who took part in this study).

This pilot study was conducted in three phases. First, all methods were tested and background concentrations established in laboratory studies; next, two homes were selected for pretesting; finally, measurements were made in eight homes of asthmatics. This pilot study successfully demonstrated the feasibility of monitoring the mass of particles equal to or less than 10 micrometers diameter inside and outside of residential settings and for measurement of personal exposures.

All of the sampling devices tested in this study reproducibly collected particles both indoors and outdoors. Differences observed between sampler types were expected based upon the designed size cutoffs of the samplers. Indoor particle concentrations

were generally lower than outdoor concentrations in the nonsmoking residences in this study. Further, mass concentrations of indoor particles were correlated with outdoor concentrations which indicated the importance of ambient pollutants on indoor air. Indoor sulfate concentrations were similar to and highly correlated with outdoor concentrations, suggesting again the importance of outdoor sources. Indoor nitrate concentrations, in contrast, were lower than outdoor concentrations and were not well correlated with concurrent outdoor measurements. Several factors led to the conclusion that a portion of the nitrate aerosol is contained in larger particles that do not efficiently penetrate into the home or that settle from indoor air. Mutagenic activity of particles was generally greater outdoors, and associated with the smallest particles; indoor activity correlated with outdoor levels. Aeroallergen concentrations were generally higher outdoors than indoors. Nicotine measurements indoors were generally below detectable limits which confirmed that smoking did not occur in these homes. Personal monitoring results indicated that exposure to particle mass and its components was most directly related to indoor residential measurements.

This pilot study has successfully confirmed that indoor and personal measurements of PM10 mass and its components is feasible and could be extended to large-scale health and exposure-assessment studies.

ACKNOWLEDGEMENTS

This work would not have been possible without the assistance and patience of the asthmatic subjects and their families for allowing us to set up in their homes the extensive sampling system required for this pilot study. Tami Fields and Patty Lee proficiently conducted field and laboratory work that led to high quality measurements. The efforts of Dr. S. Katherine Hammond are appreciated for her analyses of nicotine samples. We acknowledge Dr. Dennis D.P.H. Hsieh for his support and Angela Yee, John Lambert, Hansun Ning, Paul Kuzmicky and Brent Allen for technical support. Final production was admirably handled by Harriet Barshofsky and invaluable administrative support given by Marti Dennis. We thank Dr. Arthur Winer for helpful review of an earlier draft; and we thank Emily, Diana, Kathy, Nathan, Carolyn, George and Amal for support and sustenance in completing this project and report.

This report was submitted in fulfillment of Air Resources Board Contract Number A6-129-33 and "Pilot Investigation of Indoor-Outdoor and Personal PM10 (Thoracic) and Associated Ionic Compounds and Mutagenic Activity" by the University of California, Irvine and the sponsorship of the California Air Resources Board. Work was completed as of April, 1989.

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I. PROJECT SUMMARY

A. Introduction and Statement of the Problem

The state of California currently measures concentrations of particulate matter at fixed-site outdoor sampling locations which are selected to provide representative measurements of pollutant levels in a given geographic area. These fixed-site monitors, however, may not adequately measure exposure of humans to particles. In order to quantify human exposure to airborne particulate matter it is also necessary to monitor indoor concentrations since, 1) activity studies indicate that people spend most of their time (about 80-90%) indoors, 2) building structures modify concentrations of outdoor ambient particles, and 3) consumer products, building materials and appliances, and work and hobby activities are often important sources of indoor particulate matter.

Of greatest health significance are those particles small enough to be inhaled into the deeper parts of the lung. Particles of 10 micrometer diameter or less (PM10) are known to enter and deposit on the trachea, bronchi and the deepest portions of the lung, the alveoli. However, methods to collect these particles indoors (especially with samplers carried by individuals) and methods to measure specific components of the particles indoors have been limited. While appropriate methods exist for measurement in workplace environments, ambient personal sampling methods lag behind. Further, there has been little information reported on the relationship of outdoor to indoor concentrations of PM10. The following points summarize specific research needs in the study of human exposure to PM10.

1. Personal Exposure of Californians to PM10 Should be Determined.

Among the major set of air pollutants, particles have received relatively little attention in indoor-outdoor and personal exposure studies. There are no California-based studies that have measured personal exposure to particulate matter. Outside of California there are very few epidemiological studies of PM10 exposure and those have not yet related personal exposures to health outcomes. Several of the reported studies do not characterize the composition of particles collected on filters, they only measure the period of collection, sampling rate, and the weight of particles on the filter.

These studies indicate that indoor mass concentrations are only weakly correlated with outdoor concentrations and they frequently demonstrate a dominance of indoor mass by cigarette particles (Dockery and Spengler, 1980). Personal exposure to particulate matter is at best weakly correlated with fixed-site ambient measurements of total mass.

In contrast to previous reports on total mass, we have reported on speciated indoor-outdoor particulate filters and have shown that sulfates and certain elements that are associated with sources of industrial combustion have reasonably high indoor-outdoor correlations in other U.S. locations (Colome, et al., 1982; McCarthy et al., 1986). Because of limited measurements, this relationship has not been established in California. Characterization of indoor and outdoor PM10 would help establish the contribution of the indoor environment to total exposure. Also, for epidemiological appraisal and comparative risk assessment of biological effects, accurate exposure estimates are essential; therefore, personal measurements are required because ambient fixed site monitors do not capture most of the variability in exposure.

2. Appropriate Sampling Devices and Methods to Measure Indoor and Personal Exposures to PM10, Especially in Large-scale Field Studies are Needed.

One obstacle to obtaining information on personal and indoor PM10 exposure is that, prior to this pilot study, there were no validated size-selective PM10 sampling devices that could be carried by an individual. Sampling devices used in indoor and personal studies have design requirements that are not of concern in outdoor sampling. For example, personal samplers must be light-weight and battery-operated in order to be carried; and indoor samplers must operate quietly and have sufficiently low flow rates that they do not function like room air cleaners or perturb natural air circulation in a room. As a consequence, high-volume PM10 samplers cannot be used for either application.

There was a need therefore to validate existing personal sampling devices designed for other uses (such as those used in occupational health studies) against established PM10 air sampling devices. The precision and sensitivity of any proposed measurement had to be validated since very little PM10 mass is collected at the low flow rates (about 2 liters/min) at which personal samplers are operated. Further, these sampling devices needed to be evaluated for their suitability for use in larger field

studies, since useful exposure assessments require adequate data from a sufficiently large sample of the population.

3. Improved Indicators are Needed for Potential Adverse Biological Effects of PM10.

Currently, measurement of PM10 mass alone provides only a rough index of potential adverse biological effects. Since the composition and particle size of PM10 can vary widely, a more biologically relevant index for potential adverse effects is needed. Short-term bioassays for carcinogens and mutagens could provide information on potential adverse biological effects, but have in the past lacked the sensitivity to detect activity from samples of limited mass such as particles collected by low-volume samplers. Indoor and outdoor measurements, or personal monitoring, of concentrations of acid-forming ions would also provide a more biologically meaningful measure of potential irritant exposure than does undifferentiated PM10-mass.

4. Supplemental Personal Exposure Measurements Were Needed for the Acidic Atmospheres Study.

In another CARB study (contract No. A4-111-32 entitled "Pulmonary Function and Symptomatic Responses of Asthmatics to Ambient Acidic Atmospheres"), in which the field efforts have been recently completed, a panel of adult asthmatics was followed on a daily basis for changes in physiological function and health symptoms. These changes are currently being related to ambient concentrations of acid sulfates and nitrates as well as precursor gases. All aerometric measurements have been made at the SCAQMD monitoring station maintained in Anaheim.

Since acidic species of concern in outdoor air are thought to originate primarily from outdoor sources, and since acidic particles are considered to exist primarily in the PM10 fraction which will penetrate efficiently into residences, it was presumed that outdoor ambient measurements alone would adequately characterize exposure of asthmatics enrolled in the UCI study. This pilot project is designed to test that important assumption with concurrent indoor and outdoor monitoring of potentially acidic ionic species.

Since aeroallergens will also trigger responses in certain asthmatics, pilot measurements of indoor and outdoor aeroallergens were also made in the homes.

B. Goals and Objectives

The primary goal of this research project was to develop methods that are needed for measurement of indoor and personal exposures to PM10 of asthmatics and other Californians and to examine possible indicators of associated health effects. With fully developed and tested methods it should be possible to: (1) provide improved exposure and dose estimates for PM10; (2) relate PM10 and its associated acid-forming ions to health effects in asthmatics and others; (3) measure mutagenic activity at the personal level as an index of individual exposure to genotoxic compounds in PM10; and (4) measure concentrations of aeroallergens and environmental tobacco smoke to allow for statistical control of these factors in studies of the effects of air pollution on the health of asthmatics and others.

The following are the primary objectives of this study:

1. To investigate the reliability and precision of small-scale PM10 samplers and personal cyclone samplers for measuring PM10 mass and collecting PM10-associated ionic compounds and mutagenic compounds.

2. To conduct a pilot field study of indoor and outdoor levels of PM10 in selected households of asthmatics participating in an acidic atmospheres study entitled: "Pulmonary Function and Symptomatic Responses of Asthmatics to Ambient Acidic Atmospheres" and to investigate the feasibility of conducting larger-scale field studies.

3. To pilot-test measurement of personal, indoor and outdoor exposure to potentially acidic ions and to examine these as possible indicators for associated health effects.

4. To investigate the feasibility of relating PM10 exposures to health symptoms.

5. To conduct a preliminary investigation of PM10-associated mutagenicity indoors and outdoors as an index of exposure to mutagens and potential carcinogens and to compare mutagenic activity with PM10 mass.

6. To determine the feasibility of incorporating aeroallergen and environmental tobacco smoke (ETS) measurements in health studies of asthmatics in order to control for these potentially confounding variables.

C. Overview of Methods

In a pilot study, it is necessary to carefully test all of the methods employed. We compared different types of air samplers including PM10 samplers designed for fixed-site indoor sampling, cyclones used for measuring personal exposure to inhalable aerosols, and personal size-selective cascade samplers. To test this equipment, the research was divided into three major phases (Figure 1), including two phases designed to refine methods prior to monitoring homes of asthmatic volunteers. The three phases were:

Phase I: Laboratory and Chamber Studies
Phase II: Pre-Pilot Test of 2 Homes
Phase III: Pilot Test of 8 Homes

1. Phase I - Laboratory and Chamber Studies

Filter background and detection limits were determined for the low-volume samplers used in Phase III of the study. Preliminary work was conducted in a laboratory to be assured that measurable levels were collected and to define field method protocols. Next, we used a chamber (filtered clean air environment) located at the Air Pollution Health Effects Laboratory (APHEL) at UCI. The APHEL chamber provided a clean air exposure environment for determining background levels of all parameters and a laboratory setting for testing sampling protocols.

a. Laboratory Studies

The PM10 and cyclone sampling devices were initially tested in the laboratory environment to provide experience with filter weighing, handling, and extraction. Samples were analyzed for ionic compounds. Four PM10 air sampling devices were run for 23 to 24 hours in the investigator's laboratory at UCI. This was repeated approximately 6 weeks later in the same location with the identical sampling devices. Outdoor air samples using the four PM10 sampling devices were also

Phase I

Laboratory and Chamber Studies 24 to 72 Hour Sampling Periods
Test background concentrations and detection limits.

Phase II

Pre-Pilot Test 2 Experimental Homes for 6 Sampling Periods of 24 Hours
Initial test of equipment and protocols in realistic environments.

Phase III

Pilot Test 8 Homes of Asthmatics Each for One 24 Hour Sampling Period
Test equipment performance and feasibility for application in large field study.

Figure 1. Major Phases of PM10 Pilot Research Project

obtained from the home of one of the technicians (36 hr sampling time). Ionic compounds were determined by ion chromatography.

b. Chamber Studies

A sampling study using the PM10, cyclone and cascade samplers was conducted in a clean air chamber in order to determine background concentration. We tested 4 PM10, 8 cyclone and 2 cascade impactor samplers for 24 hours in a clean air chamber. Mass, ionic, and mutagenic compounds were measured on particles collected on filters using both PM10 and cyclone samplers. Ionic compounds and mutagenicity were measured on samples from cascade impactors. Mutagenicity of filters and any particles collected in the clean air chamber was determined by extracting the filters in an organic solvent and testing the extract using a microsuspension procedure of the standard Ames *Salmonella* assay.

2. Phase II - Pre-Pilot Test of Two Homes

Two homes located in Orange County were selected for trial runs, including set-up, operation, collection and measurement, using all air sampling devices. Sampling devices in these two homes were located indoors and outdoors and included fixed-site PM10, cyclone, and personal-style cascade samplers. Filters collected from the sampling devices were analyzed for concentrations of ions and mutagenicity. Phase II was designed as a trial to test methods and field protocols and the two homes were selected for convenience and were therefore not drawn from the asthma volunteers.

Environmental tobacco smoke (ETS), which is the aged mixture of sidestream and exhaled tobacco smoke, contains carcinogens and mutagens and is known to be genotoxic and would contribute to activity measured by the Ames assay. It was therefore important to determine the concentration of ETS as a possible confounding source of mutagenic compounds. Airborne concentrations of nicotine have been used as a specific marker for ETS. We therefore measured airborne nicotine in the homes in Phase II.

3. Phase III - Pilot Test of Asthmatics' Homes

Eight homes of volunteers with asthma were selected from a CARB-funded study of acidic atmospheres (No. A4-111-32) and were sampled for airborne particle mass,

ionic compounds, mutagen concentrations and aeroallergens. Sampling was conducted from October-December 1987 in Orange County, California with homes centered around cities of Anaheim and Orange. For this phase of work, three PM10 samplers were placed indoors and three placed outdoors. As duplicates, two indoor and two outdoor PM10 samplers were used to sample for ionic concentrations at the 2nd, 4th, 6th and 8th homes, while one indoor and one outdoor sampler obtained samples for mutagenicity testing at those homes. At the 1st, 3rd, 5th and 7th homes, the pattern was reversed and two of the indoor and two of the outdoor PM10 samplers were assigned to mutagenicity testing and one indoor and one outdoor PM10 sampler were assigned to ionic compound determinations. At all homes, pairs of cyclone samplers were matched as indoor and outdoor sets, which were then analyzed for either ionic composition or mutagenicity. One-half of the indoor-outdoor sample pairs were dedicated to analysis of ionic compounds, while the remaining half were used for mutagenicity testing.

Aeroallergens (airborne pollen and mold spores, for example) are known to trigger or exacerbate lung problems in asthmatics and are therefore important components of the individuals' indoor environments. The feasibility of measuring aeroallergens in field studies was tested by monitoring in seven homes during Phase III.

D. **Summary of Results and Conclusions**

This pilot study successfully demonstrated the feasibility of monitoring the mass of particles equal to or less than 10 micrometers diameter (PM10) inside and outside of residential settings. We also characterized the ionic species and mutagenic activity from PM10 particles and found that it is possible to monitor daily personal exposure of asthmatic volunteers to respirable particulate matter by using portable air samplers. Further, this study showed that mass collected with portable sampling devices correlates well with PM10 mass. Since results from the field sampling and laboratory analyses were more reliable than anticipated, we were able to observe a number of strong patterns and relationships in this sample of ten homes, including eight homes with asthmatics. The observed patterns and relationships form the basis of several important findings that go beyond the original objectives of this pilot study. However, while certain patterns emerged in this sample, there are too few homes in the pilot study to generalize the findings to other California residences. Further, homes with asthmatics may be different than homes from the nonasthmatic general population The

approaches taken in this study should be extended to a larger sample of residences in order to construct an exposure model and conduct a risk assessment for PM10.

Findings of the study can be conveniently grouped according to particulate mass, ionic speciation, mutagenic activity, correlations between mass and ionic speciation or mutagenic activity, and ancillary measurements. These results are summarized below:

1. Particulate Mass

- Mass, as determined from samplers designed to collect particles 10 micrometers and less in diameter (PM10), was reproducibly measured both indoors and outdoors. Mass was also reliably measured using a personal (cyclone) sampler designed to collect particles 7 micrometers and less in diameter. Values from both types of samplers were highly correlated.

- Airborne concentrations of mass collected using a cyclone sampler, designed to mimic the collection characteristics of the upper respiratory system in humans, was found to correlate well with mass collected with PM10 samplers. This relationship held up both inside and outside of homes. Concentrations of PM10 mass were slightly greater than cyclone mass indicating a small but significant contribution of the 7 to 10 μm particle range to total mass. Specifically, the sampling efficiency for particles collected by the PM10 sampler is essentially 0% for particles with an aerodynamic diameter 10 micrometers or greater while the cyclone sampler collection efficiency is approximately 0% for 7 micrometer or larger particles and about 50% for particles of 5μm diameter.

- In the sample of homes with asthmatics, mass concentration was consistently lower inside the homes than immediately outside. This result was observed for both PM10 and cyclone samples, but was more pronounced with the PM10 samples. It is likely that the indoor-outdoor ratios for PM10 samples were reduced relative to those for cyclones due to lower penetration efficiency for the larger ambient particles and to settling of larger particles inside the home. It is important to note that all homes participating in this study were occupied by nonsmokers. Cigarette smoke is a known source of fine particles. Wood stoves, fireplaces and unvented kerosene heaters (which are all potential indoor sources of particles) were also not used by the subjects

during this pilot study. Additionally, 8 of the homes studied may generally be "cleaner" than average California homes since asthmatics and their families often take precautionary measures (such as removal of dust sources, frequent cleaning, and no pets) to minimize possible triggers of asthma attacks.

- Even though concentrations of indoor mass were lower than concentrations of outdoor mass, the indoor concentration was moderately correlated with the concentration measured outside the home. This finding indicates that variation of indoor concentrations in this sample of homes was driven by the variation in ambient concentration. Therefore, in this sample of nonsmoker homes, it appears that outdoor particle concentrations determined indoor exposure.

- Some protection from higher outdoor concentrations is afforded by shelter if smokers and other particulate sources are not present. This observation was also confirmed with the personal samplers worn by the asthma volunteers.

2. Ionic Species

- Sulfate concentrations inside all ten homes were strongly correlated with sulfate concentrations measured outside the home. The homes provided minor protection from outdoor concentrations since indoor levels were only slightly lower than outdoor levels of sulfate. Total concentrations and indoor-to-outdoor correlations of sulfate were similar for PM10 and cyclone samplers, indicating that most of the sulfate was found on particles smaller than 7μm. Based upon the very strong indoor to outdoor correlation for sulfates, there was no measurable evidence of major sources of indoor sulfate.

- Nitrate concentrations inside all ten pilot study homes were only weakly correlated with outdoor nitrate concentrations. For PM10 samplers, indoor nitrate concentrations were lower than simultaneously measured outdoor concentrations; however, for cyclone samplers, indoor and outdoor concentrations were comparable. Since the two samplers have different size cutoffs, this finding is consistent with a substantial fraction of nitrate aerosol associated with particles in the 5 to 10 μm range. It is also possible that there were minor indoor sources for fine nitrate particles.

- For sulfate, indoor and outdoor measurements using the PM10 samplers had values similar to matched indoor and outdoor measurements using cyclone samplers. This finding gave further evidence that the majority of sulfate mass was found in the 7μm and lower particle size ranges.

- In contrast, indoor and outdoor measurements of nitrate from PM10 samplers were higher than those obtained with cyclone samplers operated in parallel. This finding adds to the evidence that a substantial fraction of nitrate is found in the larger size region not collected by the cyclone samplers.

3. Mutagenic Activity

- Outdoor air in the sample of eight asthmatics has higher mutagenic activity (expressed as revertants per cubic meter of air sampled) than simultaneously measured indoor air.

- Expressed on the basis of revertants per microgram of particulate mass collected, the mutagenic activity of particles in indoor air was nearly identical to that of particles in outdoor air. This finding holds for particles collected by both the PM10 and cyclone samplers. This suggests that the specific activity of indoor air was dependent on the specific activity of outdoor air and would support the hypothesis that most mutagenic activity was of outdoor origin. Therefore, in this sample of homes indoor mutagenic activity associated with particles was likely to be due to mutagenic components penetrating into the home from outdoor air. This observation would not be expected to hold where cigarette smoking is present or in the presence of particles from other indoor combustion sources, such as indoor use of unvented kerosene heaters.

- At first inspection, this finding may appear to contradict findings of the EPA TEAM study that measured much higher concentrations of volatile organic compounds inside residences compared to outdoor community measurements. However, the EPA studies measured *gaseous* compounds (many of which are known or suspected carcinogens) that come from consumer products and building materials, while we measured the mutagenicity of *particles* which come from entirely different sources. We suspect that gaseous indoor compounds do not contribute significantly to particulate mutagenicity.

- The mutagenicity per weight of particles (known as the specific mutagenic activity of collected aerosol) changed markedly from day to day, most likely because of day-to-day variation in the composition of particles. Identical concentrations of mass may have divergent mutagenic activity.

- Mutagenic activity measured by cyclone samplers was similar to activity measured by PM10 samplers. This suggests that most activity is contained on particles less than 7μm.

- Three personal samples were collected for mutagenicity testing. The specific activity of these samples was correlated with the activity measured by the indoor fixed-location monitors.

- The microsuspension assay had the sensitivity to detect mutagenic activity from low volume air samples.

4. Relationships Between Particle Mass and Other Parameters Measured

- Sulfate and nitrate ions correlated well with mass concentration. The correlation between nitrates and mass was greater than between sulfates and mass. These relationships were observed with PM10 and cyclone samplers and for indoor and outdoor samples. This finding indicates that the factors influencing variation in PM10 mass also influence variation in concentration of sulfate and nitrate ions.

- In contrast, PM10 as well as cyclone mass correlated only weakly with mutagenic activity. This suggests that factors affecting variation in the specific mutagenic activity of particles are different from factors influencing variation in PM10 mass.

5. Ancillary Studies

- As anticipated, nicotine measurements were near or below detectable limits for this sample of residences without smokers. These measurements also helped to confirm that there were no visiting smokers during the period of sampling. Nicotine can serve as a direct indicator of exposure to environmental tobacco smoke and as a control variable when investigating the mutagenicity of air samples.

- Aeroallergens were measured inside and outside of all homes. Concentrations of pollens and molds were higher outdoors than inside. This project has demonstrated that it is feasible to monitor for indoor or outdoor aerollergens in larger-scale field studies. These measurements are important control variables for investigating the effect of air pollutants on allergic asthmatics.

- A limited number of size-specific cascade impactor samplers, utilized in Phase II, indicated that most of the sulfate mass was found on particles 1μm or less in diameter while nitrate was observed with larger particles. This finding further explains why nitrate concentrations collected with cyclone samplers are lower than nitrate concentrations collected with PM10 samplers.

E. Recommendations for Future Research

Using careful laboratory and field techniques it is clearly possible to collect and analyze personal and indoor samples of PM10 particles. The techniques utilized in this pilot study may be extended to larger-scale surveys of human exposure and microenvironmental characterization of concentrations of PM10 and its associated compounds. These techniques could also be incorporated into the design of epidemiological studies in order to measure directly the concentration to which the study subjects are exposed.

More data from personal air samples would assist in determining the range of integrated exposures to PM10 mass, ionic species, and mutagens while at home, during commute and at work. Samples from a variety of homes with known sources of particulate matter would provide exposure information concerning these indoor sources. Measurements should also be taken from other locations in California which have different concentrations of mass, ionic compounds, or mutagenicity.

This pilot study focused on a selected sample of asthmatic subjects which is expected to take actions that will minimize exposures to substances that might trigger symptoms. As a consequence, this group may use certain consumer products differently, or avoid their use entirely. For example, none of the particpants in this study had a cigarette smoker in the household. Because of the selected nature of the sample and its small size, the results should be generalized with caution. However, since the data has excellent precision and reproducibility, we believe the relationships

are real for the sampled group. A primary recommendation is that a larger scale PM10 exposure and microenvironmental concentration study should be conducted in California.

In this sample of nonsmoking households it is clear that variation of indoor residential PM10 concentrations is controlled by outdoor particle levels. In homes with smokers or other sources of particles, the pattern is expected to be more complicated; but we still expect that indoor PM10 concentrations will be influenced by and vary with ambient conditions. As sources of indoor particle pollution are added it becomes more difficult statistically to identify the role of ambient pollution on indoor exposures. Therefore, future studies should carefully characterize potential sources of indoor particluate matter and stratify the sample, according to indoor source characteristics.

Measurement of PM10 mass alone does not adequately characterize exposure. Using the low volume samplers that are employed for personal or indoor sampling, there is a moderate level of inherent error in measurement of collected mass. This error will tend to obscure the effect of factors that influence PM10 concentration. The measurement of certain constituent compounds contained within PM10 samples is more repeatable; for instance, in this study we were able to measure sulfate and nitrate ion and mutagenic activity with more repeatability than we could measure mass. For this reason, it is advisable to continue to characterize PM10 by more factors than its mass alone.

For measurement of personal exposures, we used available samplers that are employed in occupational settings. The particle size cut for these samplers is somewhat different from the size cut of PM10 samplers. Since sulfate ion and mutagenic activity appeared to be concentrated on the smallest particles, PM10 and occupational (cyclone) samplers measured the same concentrations for these two constituents. For these measurements, available occupational cyclone samplers can be used to evaluate personal exposure to sulfate and mutagens associated with particles collected by PM10 samplers.

For mass concentration and nitrate ion, however, the PM10 and cyclone samplers would often indicate different concentrations. For certain samples, the concentration measured by a PM10 sampler would exceed the concentration measured by a cyclone sampler. This difference indicated the presence of larger particles and large-particle

nitrate. Due to potential differences in biological activity at various size cuts, future studies should classify sizes into multiple groupings.

At the time this study was initiated, cyclone samplers provided the only comparable size cuts to PM10 that were available for personal monitoring. During this study, a personal PM10 sampler has been developed and should be tested in future expanded studies. Due to differences in chemical composition and biological activity by particle size, we would recommend that a size cut smaller than 10 micrometers be maintained, even if the new PM10 personal samplers are proven in field applications.

Since two samples with the same weight may have divergent ionic composition and mutagenic activity, the mix and concentration of aerosol components changes from one air mass to another. Quantitative differences in concentrations of ionic species and mutagenicity have been reported in California. For example, Atkinson et al. (1988) report locational differences in polycyclic aromatic hydrocarbons and airborne mutagenicity. This suggests that certain adverse biological effects could vary considerably for the same mass of PM10 particles. Therefore, mutagenicity and ionic composition must be measured directly and cannot be inferred from mass concentration. The concentration of specific compounds and the level of mutagenic activity could be used to help set priorities for further investigation of adverse health effects from PM10 mass.

There are several recommendations that can be made relative to laboratory elements of this type of study. We can not overstress the importance of good quality control and quality assurance procedures. Extensive quality control efforts increased confidence in the present study results. There are several areas that warrant additional efforts.

Determination of mass on filters is the least reliable of all measurements. Although repeatability was actually quite good on an absolute basis, we believe that it might be improved. Our protocol called for weighing under a restricted range of environmental conditions. Additional work should explore whether weight measurements for this type of sample can be improved under more restrictive environmental conditions within climate-controlled weighing rooms. Our ion samples were extracted through sonication. Extraction efficiency by sonication should be compared with shaking, which is the most commonly used extraction method.

For field efforts there are several recommendations to be derived from this study. First, we have found that day to day variability in ambient concentrations of PM10 may totally obscure factors of interest such as personal exposure to cooking aerosols and particles from cleaning activities and hobbies. A study to investigate factors such as these should involve multiple days of measurement at individual locations so that a range of ambient concentrations can be observed at each site.

Second, when investigating a range of housing types and climatic regions in California, it is advisable to add measurements of ventilation rate. It may turn out the range of actual ventilation rates in residences is small, but until that issue is resolved, particle penetration and settling are known to be influenced by the level of ventilation.

Third, siting criteria for location of monitors at residential sites should be tightened. It is desirable with outdoor monitors to characterize the general ambient environment surrounding the home. The potential influence of isolated activities and shielding by plants and other obstructions should be avoided.

Fourth, a data file manager should be utilized during the early phases of the study in order to organize the data for concurrent quality control checks and future analyses. This is a task that is often overlooked in large field studies but it will substantially improve efficiency at the analysis stages.

II. INTRODUCTION AND BACKGROUND

A. Particulate Matter of 10 Micrometer Diameter (PM10)

The following points reflect the initial scientific needs regarding PM10 exposure.

There has been little information on exposure to indoor levels of particulate matter of a specific and physiologically relevant size; nor has much indoor exposure data been reported on concentrations of absorbed compounds. Further, information on exposure, especially at the personal or individual level in California has been very limited. The specific particle size range of interest in regards to health is 10 micrometers and less in diameter, usually referred to as PM10. Particulate matter in this size range is known to deposit in the tracheal, bronchial and alveolar regions of the lung. The current California ambient 24 hour standard for PM10 (based on high-volume air samplers) is 50 $\mu g/m^3$.

The current pilot project had multiple components which attempted to address for the first time important questions on PM10 exposure indoors. The project met those initial goals and exceeded expectations by discovering several interesting relationships between outdoor and indoor PM10 mass and PM10-associated ions and genotoxic compounds.

B. Ionic Speciation

Under a separate contract with CARB, the Principal Investigator is completing a study on the relationship between acidic sulfates, nitrates and precursor gases with symptoms in asthmatics. In the study, a group of adult asthmatics was followed on a daily basis for physiological effects (peak expiratory flow), symptomatic outcomes (diary reports of airway obstruction), and changes in medication use. Health effects are related in the study to aerometric measurements made at the Anaheim monitoring station operated by the SCQMD.

A presumption in the acidic atmospheres study is that personal exposure to acidic particulate species is well characterized over a wide geographic area by one or two outdoor ambient monitors. Unfortunately, very few measurements exist to support this assumption.

This pilot study was needed to supplement the aerometric measurements made in the asthma study. It is presumed in the asthma study that outdoor ambient measurements of fine sulfates and nitrates adequately characterize indoor exposures. This assumption is based on: 1) previous measurement experience of the investigators; 2) the small size of sulfate and nitrate particles that will therefore pass easily into the indoor environment and have long residence times; and 3) the fact that there are few indoor residential sources of particulate sulfate and nitrate. The personal and indoor measurements made in this study help to establish the relationships between indoor, outdoor and personal exposure to acidic (ionic) species on PM10 particles. Asthmatics may be especially sensitive to general PM10 exposure in addition to acidic constituents.

C. **Aeroallergens**

Aeroallergens are also known to trigger attacks that reduce peak expiratory flow and increase symptoms of airway obstruction in asthmatics. We are currently accounting for outdoor pollen counts in our study; but the relationship between outdoor pollen counts measured at one location and personal exposure to pollen spores has not been established. Due to the large size of many pollen spores, indoor and outdoor values could differ substantially. Furthermore, pollen sources are often localized and therefore their effect would not be captured by a single outdoor monitor located in the community. Smaller fungal spores may also trigger asthma attacks and these often have indoor sources. The relationship between indoor, outdoor and personal exposure to pollen and fungal spores should be identified in order to control for these factors.

Since asthmatics may have attacks triggered by residential aeroallergens and local sources of particulate matter, the measurement and characterization of PM10 and size speciated particles add an important factor in the asthma study. Failure to account for these factors in studies of asthmatics could obscure relationships that exist between asthma symptoms and air pollution exposure. One community study of asthmatics by Perry, et al. , 1983, was found in the latest addendum to the EPA Criteria Document for PM10 and SO_2 to be of limited usefulness because aerometric data came from only two outdoor monitoring locations in the study area of Denver.

D. **Airborne Mutagenicity**

Measurement of airborne mutagenicity associated with particulate matter has generally been reported for particles collected outdoors and with high volume air sampling devices. The mutagenicity, primarily based on the *Salmonella/* microsome test (Ames et al, 1975), has generally been used as an index of exposure to complex mixture of carcinogens absorbed onto the collected particulate matter (Tokiwa et al, 1977; Talcott and Wei, 1977; Pitts et al, 1977; Moller and Alfheim, 1980; Chrisp and Fisher, 1980; Flessel et al, 1984; Lewtas et al, 1986). Because the sample mass collected using low volume type sampling devices has in the past been too small for mutagenicity testing, the measurement of such activity has been limited.

We have previously reported a simple and sensitive modification of the *Salmonella/* microsome test (microsuspension assay) for detecting mutagenicity in complex environmental mixtures, including indoor air samples collected using low volume sampling pumps (Kado et al, 1983, 1986, 1988). The microsuspension assay is approximately 10 times more sensitive than the standard plate incorporation test of Ames et al (1975) based on absolute amounts of mutagen required for a specific mutagenic response. Recently, a number of investigators have used the assay for measuring the mutagenic activity of environmental tobacco smoke (ETS) indoors (Ling et al, 1987). Also, mutagenicity of size-segregated particulate matter has been measured (Kado et al, 1986).

Some of the advantages of using the microsuspension procedure for airborne particulate matter include: 1) less sample mass is required for testing and therefore the potential for detecting mutagenic activity from personal low volume (2 liters/min) sampling devices is possible and, 2) the information on mutagenicity can be compared to information from standard mutagens within the assay and the information can be compared to mutagenicity from the plate incorporation test.

Currently, the standards for PM10 are based solely on collected mass. However, an identical mass of material could potentially have diverse biological activity. Therefore, a more biologically relevant index associated with the PM10 or particulate matter of respirable size is needed. Further, the levels and biological activity of PM10 in indoor environments could be different compared to PM10 found outdoors. The use of a genotoxic (damage to genetic material) endpoint could initially serve as a biological endpoint associated with particles.

III. MATERIALS AND METHODS

A. Air Sampling

1. Description of Air Sampling Equipment

a. PM10 Sampler

Following promulgation of CARB and U.S. EPA standards for PM10, there was a clear need for samplers that could reliably collect particles smaller than 10 micrometers. High volume samplers designed to eliminate particles larger than 10 micrometers (Wedding and Weigand, 1985; Wedding et al., 1985), and dichotomous samplers which collect a fine (<2.5 μm diameter) and coarse (>2.5 μm) particles have been used to collect PM10 particles in the outdoor ambient environment. Neither of these samplers is appropriate for sampling in the indoor environment due to their size, noise and high flow rates. In fact, the high volume sampler can behave like a vacuum cleaner or air purifier for airborne particles when operated in a residential-sized structure.

Recently, a four liter per minute (L/m) sampler has been developed to collect PM10 and PM 2.5 particles in the indoor environment (Marple, et al., 1987). A recent intercomparison has been reported with side-by-side monitoring using this PM10 sampler and the dichotomous sampler with fiberglass filters (Lioy et al., 1988). The investigators used the two sampling instruments and reported consistent results; any inconsistencies were believed to be due to a problem of filter loss associated with using fiberglass filters in the sample holders. The authors did not believe that this problem would occur for Teflon™ filters.

For this study we utilized the Marple sampler set up for PM10 collection. Figure 2 provides a schematic diagram of the sampling head and an illustration of the assembled sampling device. Twin impaction plates provide a sharp 10μm particle cut size. Figure 3 shows the theoretical and experimental collection efficiencies of the sampler with the PM 2.5 or PM10 impaction inserts. The sampling inlet produces a sharp size cut.

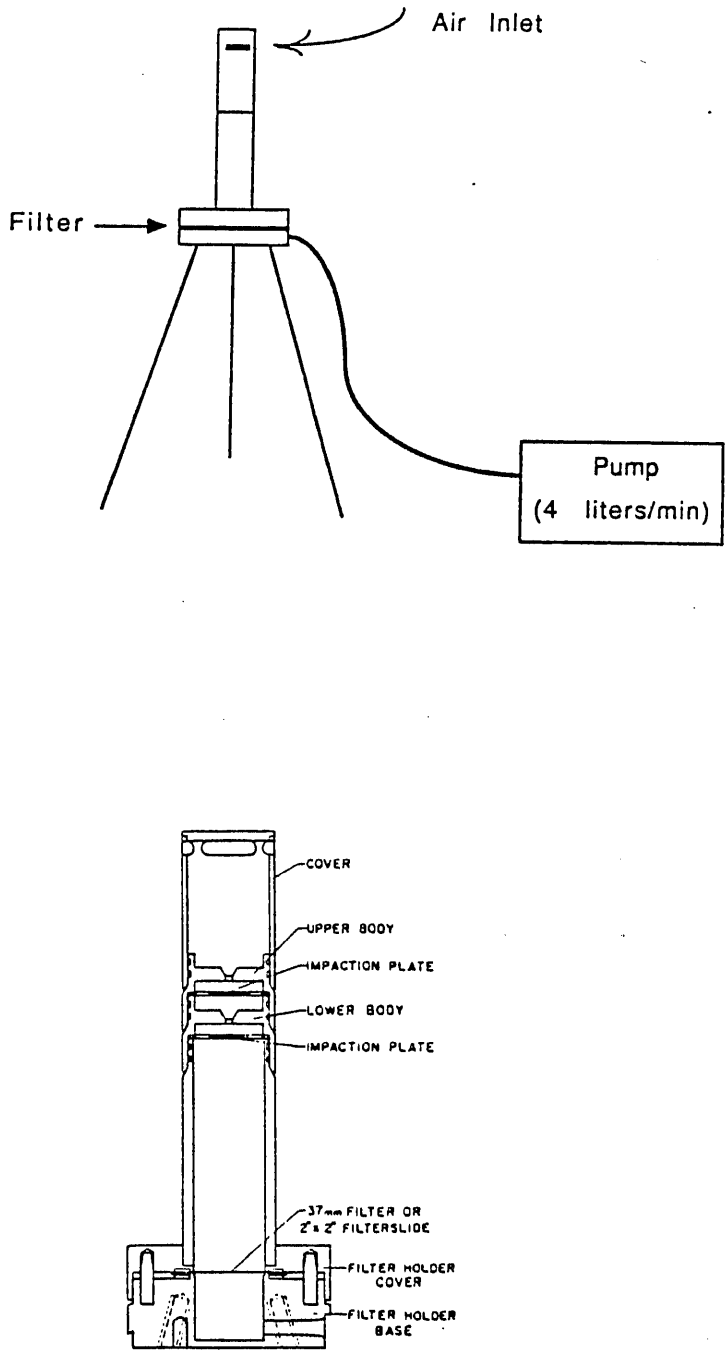


Figure 2. PM10 sampling assembly. A) Sampling device in field setting. B) Schematic of sampling head (from Marple et al. 1987).

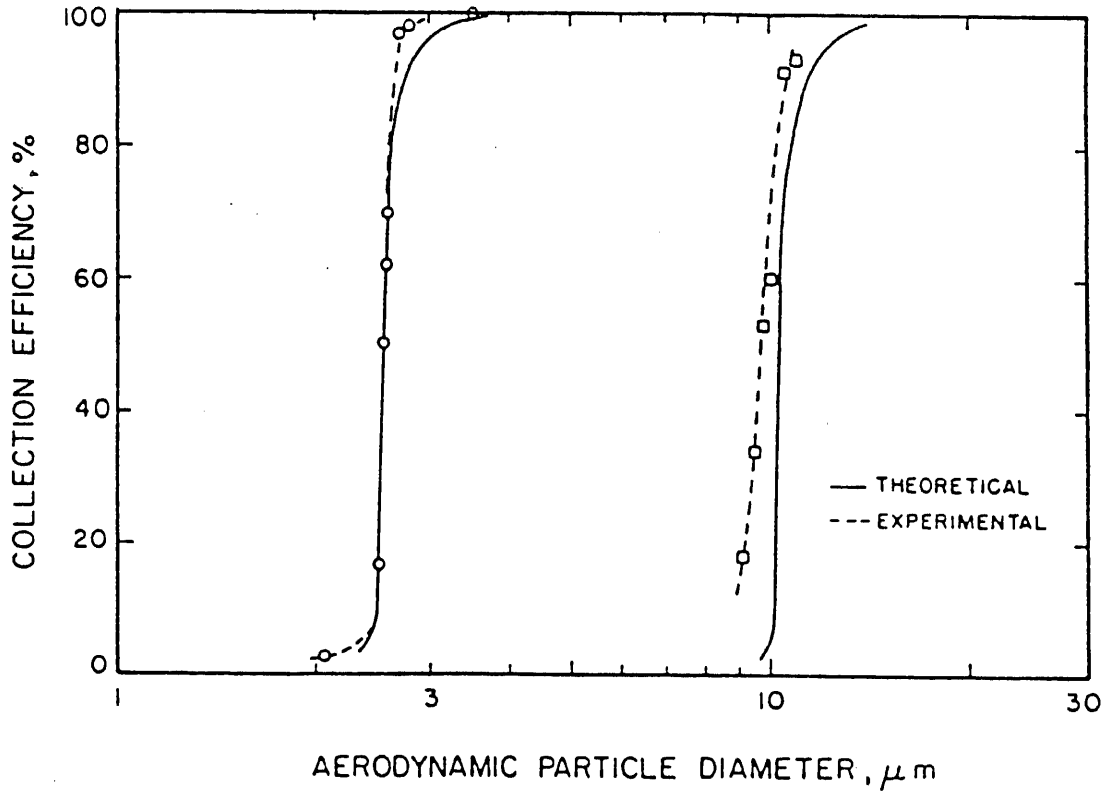


Figure 3. Theoretical and actual collection efficiencies of PM10 and PM2.5 sampling heads. PM10 sampling heads used in this study (from Marple et al. 1987).

Two impactor stages were utilized, each with the same size cutoff, in order to minimize problems of particle bounce. These stages were coated with approximately 100 µl of light mineral oil in order to further reduce any particle bounce, which would cause particles larger than 10µm to be collected. Plastic filter holders were fitted with Teflo™ filters (Gelman Filter Co.) and flows calibrated in the field with a custom-fitted cap and calibrated rotameter. Rotameters were calibrated in the laboratory using a bubble flow meter. Teflo™ filters are Teflon membrane filters which are supported by a plastic ring to keep the membrane in place.

Field sampling was performed with a vacuum pump connected to a flow controller and timer. Flows were recorded before and after each sampling period.

b. Cyclone Sampler

There were no available samplers capable of collecting personal or portable PM10 particles at the time that this study was initiated. One objective of this study was to determine the feasibility of directly monitoring personal exposure to particles using portable monitors that are carried by human volunteers as they go about their regular activities. In the absence of portable PM10 monitors, we utilized portable sampling devices (cyclone samplers) that have been developed for monitoring personal particulate exposure in occupational environments. A schematic of the cyclone sampler is provided in Figure 4.

For this pilot study, we used cyclone samplers that block all particles larger than 7µm in diameter and have a 50% cutoff at approximately 5µm. The curve of particle collection efficiency for the aluminum cyclone used in this project is displayed in Figure 5. The cyclone sampler maintains its collection efficiency independent of its orientation while being carried by an individual. The cyclone is connected to a portable constant-flow battery-operated sampling pump. The pump and cyclone were operated at 2 L/m. The pump and cyclone were supplied by SKC, Inc. (Fullerton, California).

When used as personal samplers, the cyclone is attached on a shirt or blouse lapel near the breathing zone. Air is drawn by the pump through the cyclone and then through a filter mounted in an acrylic cassette. The pump is either attached to the subject's belt or worn on a shoulder strap.

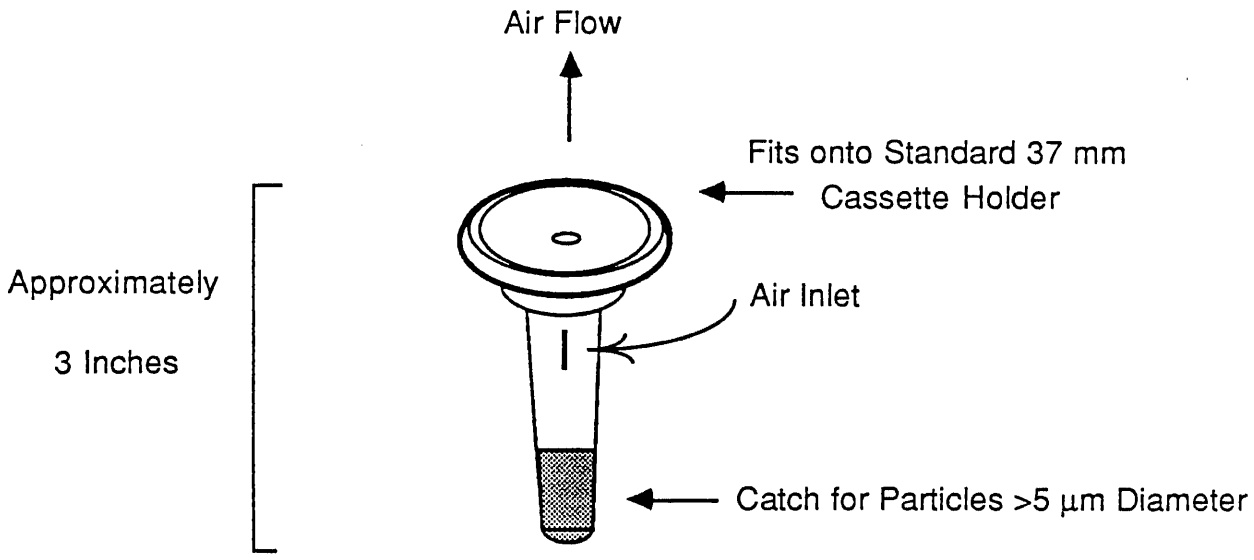


Figure 4. Cyclone Sampler

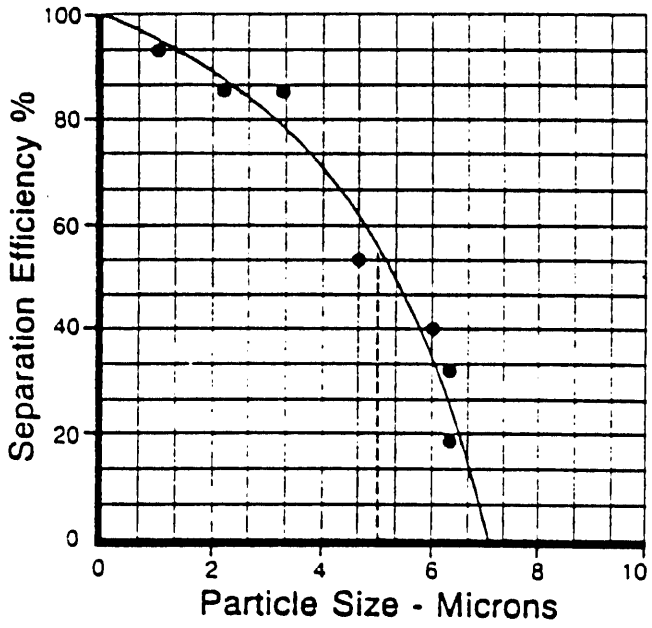


Figure 5. Collection efficiency of cyclone preselectors. The preselector passes particles under the curve to the filter for collection.

c. Cascade Sampler

Since the size collection characteristics of the PM10 and cyclone samplers are not the same, it was decided to conduct limited sampling for ionic compounds using a portable cascade impactor. The eight-stage cascade impactor was designed by Marple and supplied by Sierra Instruments (the impactors are presently available through SKC).

Each stage of the cascade impactors captures sequentially smaller particles, from a cutpoint of approximately 20μm to 0.5μm. A backup filter at stage 9 collects particles smaller than 0.5μm. The sampler and its cutpoints are illustrated in Figure 6 (Rubow, Marple, and Olin, 1988). The same portable sampling pump as used with the cyclone sampler was used for the cascade samplers.

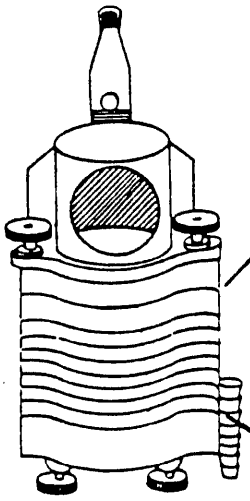
2. Filter Material

a. Filter Selection

For this project there was a need to optimize filters for three different types of samplers (PM10, cyclone, and cascade) and for the major analyses that were conducted (gravimetric determination, ion chromatography, and microsuspension mutagenicity assay). The major issues were: (1) compatibility of filter type with sampler, (2) background concentration of each filter for species of individual analyses, and (3) filter collection efficiency.

For the PM10 samplers, filters had to be placed into a 2" x 2" plastic slide; the slide was then placed between rubber gaskets in the sampler body. The Teflo™ filters have little intrinsic support and each are manufactured with a polymethylpentene (PMP) support ring. The filter with support ring drops conveniently into the plastic slides that go into the body of the PM10 samplers.

Because of their low background concentration for trace materials, high reported collection efficiency for small particles, and fit into the 2" x 2" PM10 slide holders, 41 mm diameter Teflo™ filters were used in this project for all PM10 sampling. The filters are composed of Teflon, have a nominal pore size of 0.2μm and were used in the PM10 samplers for collection of particles for measuring mass, ionic speciation, and mutagenicity.



Stage No.	Cut Point Diameter (μm) @ 2 liters/min
1	21.3
2	14.8
3	9.8
4	6.0
5	3.5
6	1.6
7	0.9
8	0.5
9	Filter Trap

Figure 6. Cutpoints for personal cascade impactor.

Ideally, filters of the same composition would have been used for the other samplers. However, it was not convenient to use the Teflo™ filters for mutagenicity testing since the filters had to be extracted with organic solvents that could dissolve the PMP support ring on the Teflo™ filters. While Teflo™ filters had to be used for mutagenicity testing in the PM10 samplers, it was necessary for those samples to manually cut off the PMP ring. Removal of the ring was tedious. For that reason, cyclone samples destined for mutageniticy testing were fitted with Zefluor™ filters.

Zefluor™ filters are composed of Teflon™ and also have a nominal pore size of 0.2µm. They have a thin Teflon sampling membrane overlayed onto a matrix of Teflon for support. Zeflour™ filters were also used for the cascade impactor sampler studies and were individually handcut to fit on top of the eight regular stages. The cutout involved a spoke-and-wheel design in the filters to match the aluminum stages and to accommodate airflow between stages.

3. Filter Preparation

a. Handling

All filters were handled with precleaned Teflon™ coated tools. For cutting, pre- and postweighing, extractions, and analyses, the filters were handled in a HEPA filtered laminar flow hood. Filters were conditioned to room temperature and humidity for 24 hours before weighing. The filters were passed over a polonium source to eliminate static charge on the filter immediately before weighing.

Filters assigned to mutagenicity testing were, after each 24-hour sampling period, wrapped in foil, placed in dry ice and then taken back to the laboratory. Once at the laboratory, the filters were allowed to equilibrate to room temperature for at least 1 hour, reweighed and then placed in a -20°C freezer. Filters within their cassettes were shipped in dry ice to UC Davis by overnight courier for subsequent extraction with organic solvent and testing in the bioassay. All filters were coded and all handling was done under subdued light to minimize potential photo-oxidation to any adsorbed mutagens.

b. Cutting

For use in the cascade samplers, we individually cut the Zefluor™ filters to fit the dimensions of its eight different cascade impaction plates. Blades used for cutting were pre-cleaned with dichloromethane.

c. Filter Pre-cleaning

Teflo™ filters were used for ion chromatographic analyses without any special preparation. For mutagenicity studies, Teflo™ and Zefluor™ filters were precleaned using 1:1 solution of dichloromethane and methanol.

4. Sampler Flow Calibrations and Flow Controller Adjustments

We monitored and adjusted the flow of each sampler immediately before and after each sampling period. If the flow changed more than 0.20 ml/min the sample was considered invalid. All sample lines were made of tygon tubing and were placed downstream from the filter.

A custom-made flow chamber was used to calibrate flow rates for the personal samplers (cyclone and cascade). The flow chamber consisted of a tightly sealed plastic container with one port leading in and another leading out. The port leading in was connected to a rotometer. The port leading out was connected to a pump. Between the rotometer and the inlet port we connected a filter to prevent contamination of the sample filter. The external air first passed through the rotometer; second, the preventive filter; third, into the container; fourth, through the sample head and sample filter; and finally, through the sample pump.

For the PM10 sampler, the following configuration was used for sampling the air: air first entered the rotometer, then the in-line prefilter, the PM10 sampling head with nozzle, and finally the PM10 pump. The PM10 pump was equipped with a cannister filter attached to the exhaust line. We also calibrated the rotometer against a 500 ml SKC bubble meter. The rotometer was accurate to the second decimal place.

Prior to any of the air sampling measurements, we tested the effectiveness of the mass flow controller of each pump. This was done by decreasing the pressure of the pump on its inlet side using a pressure regulator on the pump. The purpose of this was to simulate a "worst-case" condition where there was sufficient trapping of

particulate matter to cause a pressure drop in the pump and cause the pump to decrease its flow. The pumps could sustain up to 15 inches of pressure drop and still maintain the desired flow rate of 2 liters/min to within 0.10 ml/min.

B. **Analytical**

1. Gravimetric Analysis

a. All weighing was conducted under a laminar flow hood. The climatic conditions were measured with a Short & Mason, chart No. 37 hygrothermograph, which was calibrated with a psychrometer. Weighings were only taken when the room temperature was between 70 to 74 degrees Fahrenheit, and the relative humidity was between 35 and 45 %.

b. On a daily basis, we cleaned all surfaces in the weighing area with deionized water. Weekly, forceps were washed by sonication in detergent and rinsed in deionized water.

c. We routinely calibrated the automatic electrobalance (CAHN, model 29) prior to weighing a batch of filters. We also maintained a filter batch blank and weighed it before and after each sample period; if there was greater than a seven microgram difference between the two measurements, the entire batch was reweighed.

2. Ionic Compounds

a. Extraction Protocol and Chemical Analysis

A laboratory test (Phase I) was conducted to measure the precision of our collection, extraction, and analysis of particle mass and particle-associated nitrate and sulfate. Air in the laboratory was sampled for 36 hr using six personal-cyclone samplers and four stationary-PM10 samplers. Half of the personal cyclone samplers were loaded with 37mm Teflo™ filters, while the other three were collected on 37 mm Zefluor™ filters. The four PM10 samplers were loaded with 41 mm Teflo™ filters. All filters were analyzed for ion content. Once the precision was established the following extraction protocol was finalized:

Teflo™ and Zefluor™ filters were treated identically. The filters were extracted in 60 ml polyethylene containers. Ion chromatograph eluent ($NaHCO_3/Na_2CO_3$ in deionized water) was used as a solvent.

The filters were placed face down in the container to maximize the contact between the particle impaction side and the solvent. With a class A volumetric pipette, we dropped 0.24 ml (+/- 0.04 ml) of 95% ethanol over the entire filter to form a less hydrophobic state between the filter and the solvent. A blank filter in this solvent mixture contained no measurable nitrate or sulfate. This was followed by five injections of the solvent (4.0076 +/- 0.0008 ml per injection), resulting in a total volume of $20.28 \pm .04$ ml.

The final extract was sonicated with a Cole-Parmer Ultrasonic cleaner for fifteen minutes. Since there was slight heat formed during this process, we allowed it to cool to room temperature prior to ion analysis. Samples that were not immediately analyzed were stored in a lab refrigerator at 4°C.

The anions in our extract were separated and analyzed by ion column chromatography (Dionex, model 2000i) and the electronic output was integrated with a Spectro Physics integrater (model 4270).

3. Mutagenicity

a. Chemicals

Benzo(a)pyrene (BaP), 2-nitrofluorene (2-NF) and dimethyl sulfoxide (DMSO; spectrophotometric grade) were purchased from Aldrich Chemical Co. (Milwaukee, Wisconsin)and used without further purification. Dichloromethane (DCM; resi-analyzed grade) and methanol (HPLC grade) were purchased from Baker Chemical Co., Phillipsberg, New Jersey.

b. Extraction

Filters were placed in pre-cleaned screwcapped bottles or flasks and 5 ml of dichloromethane (DCM) was added. The container was shaken for 15 minutes followed by sonication for 15 minutes and the solvent decanted into a 15 ml pre-cleaned scintillation vial. The extraction procedure was repeated once more with DCM and then by methanol, and the extraction solvents pooled into a single pre-

washed vial. The solvents were subsequently evaporated under a steady stream of nitrogen to about 100-200 µl of solvent, transferred to a mini-vial and since solvent exchange to DMSO was impractical for the very small volumes of DMSO we wanted to add, the solvents were allowed to evaporate to dryness. The extract was then tested for mutagenic activity.

c. Mutagenicity Assay

A microsuspension procedure previously reported (Kado et al, 1983, 1986), which is a simple modification of the *Salmonella*/microsome test, was used throughout. The assay is approximately 10 times more sensitive than the Ames *Salmonella* procedure based on absolute amounts of material added per determination. The assay has been used to measure mutagenic activity of airborne particles of 2.5 µm diameter or less (Kado et al, 1986) and of particles collected from indoor environments where environmental tobacco smoke was present (Ling et al, 1987; Kado et al, 1988).

d. Procedure

Tester strains TA 98 and TA 100 were kindly provided by Dr. B.N. Ames, Berkeley, CA. For the modification, bacteria were grown overnight in Oxoid Nutrient Broth No. 2 (Oxoid Ltd., Hants, England) to approximately 1-2 x 10^9 cells/ml and harvested by centrifugation (5,000 x g, 4 °C, 10 minutes). Cells were resuspended in ice-cold phosphate-buffered saline (PBS, 0.15M, PH7.4) to a concentration of approximately 1 x 10^{10} cell/ml.

The S9 and S9 mix were prepared according to the procedure of Ames et al. (1975). The S9 from Aroclor 1254 pretreated male Sprague-Dawley rats contained 52.4 mg protein/ml as determined using the modified Biuret method of Ohnishi and Bar (1978) was used throughout. The extract from the particulate matter was resuspended in enough DMSO to give approximately 0.5 cubic meter equivalents per 5 µl of the extract mixture. For the extracts from the cyclone filters, enough material was available for duplicate determination at a single dose.

For the microsuspension assay, the following ingredients were added, in order, to 12 x 75 mm sterile glass culture tubes on ice: 0.1 ml S9 mix, 0.005 ml extract in DMSO, and 0.1 ml concentrated bacteria in PBS (1 x 10^{10} / ml PBS). The mixture was incubated in the dark at 37°C with rapid shaking. After 90 minutes the tubes

were placed in an ice bath and taken out one at a time immediately before adding 2 ml molten top agar (Ames et al., 1975) containing 90 nmoles of histidine and biotin. The combined solutions were vortex-mixed and poured onto minimal glucose plates. Plates were incubated at 37°C in the dark for 48 hours. Strain markers were routinely determined for each experiment.

All procedures were carried out in a room fitted with yellow fluorescent lights (G.E. F40Go) to minimize potential photo-oxidation.

4. Airborne Nicotine

The collection of airborne nicotine as a marker for environmental tobacco smoke (ETS) followed the procedure of Hammond et al (1987). The analysis of nicotine was conducted under contract by S.K. Hammond, Worcester, Massachusetts. Environmental tobacco smoke is the aged sidestream and exhaled smoke from tobacco products known to contain a number of mutagenic and carcinogenic compounds (National Research Council, 1987; Surgeon General, 1987).

Collection of indoor and outdoor airborne nicotine was primarily from two experimental homes measured during Phase II and from the Phase I experimental chamber studies at APHEL. Collection from an office where the employee was a smoker was also done to serve as a positive field control. For the collection, vapor phase nicotine was collected downstream of the particulate filter on a 37 mm Teflon™ coated glass fiber filter (Enfab TX40 H120WW, Pallflex Corp., Putnam, CT.) which had been pre-treated with sodium bisulfate. The filter for nicotine was separated from the particulate trap filter by a stainless steel support screen and Teflon™ O- ring. For the analysis, the bisulfate-treated filters were desorbed with 2 ml water and 0.1 ml of ethanol and vortex mixed for 1 min. Two ml of 10 N sodium hydroxide was added , the tube vortex mixed and the nicotine concentrated by liquid/liquid extraction into heptane (0.25 ml ammoniated heptane). A 3 µl sample of the heptane was analyzed immediately in a Shimadzu GC-7A gas chromatograph equipped with nitrogen selective detector. A 6-ft long, 1/8th inch diameter stainless steel column of Chromosorb W coated with 10% Apiezon L containing 3% KOH was used at a isothermal temperature of 170°C.

5. Aeroallergen

Short-term aeroallergen samples were collected inside and outside of all homes using a volumetric sampler that deposits pollens and mold spores directly onto a glass slide for microscopic speciation. A portable volumetric sampler manufactured by the Burkard Manufacturing Company (Rickmansworth, Herts, England) was selected since the impaction principle used in the sampler is capable of collecting particles down to approximately 1 micrometer in diameter. The sampler is designed for portable sampling, weighing only 590 grams and can be operated on battery or line power. Aeroallergens were analyzed by Janet Gallop, an aerobiologist who maintains a laboratory that specialized in measuring ambient concentrations of mold and pollen and is operated by a private allergy clinic.

A glass microscope slide coated with a thin layer of petroleum jelly was inserted into the Burkhard sampler which was run for ten to fifteen minutes at a nominal flow rate of 10 lpm. All samples in this experiment were taken in the battery mode using fully charged batteries. Start and stop times were recorded to insure an accurate representation of the volume sampled. At the time equipment was set up at the homes, the field technicians simultaneously started one outdoor and one indoor aeroallergen sampler to provide short-term paired samples.

The aeroallergen sampling was the only air sampling conducted for a relatively short duration (10-15 min). If concentrations of pollens or molds were variable over short time periods, indoor and outdoor concentrations would not be in equilibrium. The typical air exchange between indoor and outdoor air, which is approximately 0.5 to 1.0 air changes/hour in Southern California (Wilson et al, 1986), necessitates several hours of sampling to obtain stable average concentrations.

Microscopic slides were labeled and quantitatively evaluated using an optical microscope.

C. Experimental Procedures

1. Phase I - Laboratory and Chamber Studies

Prior to Phase II pre-pilot and Phase III pilot studies, we conducted laboratory and chamber tests to establish background levels of mass, ions and mutagenic activity as well as develop a field monitoring protocol. Sampling was conducted in our air quality lab, at one of the lab technician's home, and at the UC Irvine Air Pollution Health Effects Laboratory (APHEL). We evaluated the background of the filters and samplers, the precision of the samplers and our analysis methods, and we twice monitored in the field as a trial performance. A summary of the experimental design of Phase I (Laboratory and Chamber) studies is illustrated in Tables 1 and 2.

a. Laboratory Studies

Blank filters were analyzed for mass, nitrates and sulfates or mutagenic activity as outlined in Table 1. Four 41 mm PM10 Teflo™ filters and three 37 mm cyclone Teflo™ filters were assessed for nitrate and sulfate background. Three cleaned 37 mm Zefluor™ filters and three 37 mm Zefluor™ filters were assessed for revertant background. We did not find measurable ions or mutagenic activity at the analysis range that was used for field sample filter extracts.

b. Chamber Study

At APHEL, we operated the samplers in a zero-air (pre-filtered air) chamber to analyze background of the sample heads, pumps, and adjacent pumps. The experimental design is summarized in Table 2. Four PM10 samplers were used; two filter samples were analyzed for mass, nitrates and sulfates, and two for mass and mutagenic activity. Eight personal (cyclone) samplers were used; three filters were dedicated for ion analysis and five for mutagenicity testing. Also, two eight-stage cascade samplers were tested.

2. Phase II - Pre-Pilot Homes

Two homes were selected for trial runs, including set-up, operation, collection and measurement using all air sampling devices. Also determined were concentrations of ions and mutagenicity. The experimental design was set up to accommodate both the ionic compounds and mutagenicity. One home (designated SCOL) was re-visited for

Table 1. Experimental Design of Phase I Studies (Laboratory)

Pre-Selector	Location	Indoor or Outdoor	N	Filter Type	Sampling Period (Hr)
PM10	Lab	Indoor	4	Teflo™ [a]	24
	Lab	Indoor	4	Teflo	23
	Home	Outdoor	4	Teflo	36
Cyclone	Lab	Indoor	4	Teflo	24
	Lab	Indoor	4	Zefluor™ [b]	23

a Teflo™ filters are Teflon membrane filters.

b Zefluor™ filters are Teflon membrane filters overlayed onto a support matrix of Teflon.

Table 2. Experimental Design of Phase I Study (Chamber).

Pre-Selector	N	Filter Type	Sample
PM10	2	Teflo	IC [a]
	2	Teflo	Mut [b]
Cyclone	3	Teflo	IC
	3	Teflo	Mut
	2	Zefluor	Mut
Cascade	1	Zefluor	IC
	1	Zefluor	Mut

[a] IC = Ionic Compounds

[b] Mut = Mutagenic Activity

a second set of two 24-hr sampling periods. A summary of the experimental design of Phase II (Pre-Pilot) studies is illustrated in Table 3.

Environmental tobacco smoke (ETS) which is the aged mixture of sidestream and exhaled tobacco smoke, contains carcinogens and mutagens and is known to be genotoxic. It was therefore important to determine the concentration of ETS as a possible confounding source of mutagenic compounds. Airborne concentrations of nicotine have been used as a specific marker for ETS. We therefore measured airborne nicotine in the homes in Phase II using the methods developed by S.K. Hammond et al (1986). The filter samples for nicotine were analyzed by S.K. Hammond.

3. Phase III - Pilot Test of Asthmatics' Homes

Eight homes of volunteers selected from a CARB-funded study of acidic atmospheres (No. A4-111-32) had PM10 and cyclone samples taken for airborne particle mass concentrations, ionic compound and mutagen concentrations. All homes were located in Orange County, centered about the cities of Anaheim and Orange. Sampling took place during a ten week period from October to December 1987. For this phase of work, three PM10 samplers were placed indoors and three placed outdoors. Two of the three PM10 samplers located indoors and two outdoors, were dedicated to ionic concentrations for the 2nd, 4th, 6th and 8th home, while one sampler each was dedicated for mutagenicity testing indoors and outdoors on those days. For the 1st, 3rd, 5th and 7th home, two of the three PM10 samplers (located indoors and outdoors) were dedicated toward mutagencity testing and one PM10 sampler each (located indoors and outdoors) was dedicated for ionic compound determinations. For all homes cyclone samplers were matched with sets indoors as well as outdoors. One-half of the indoor and one-half of the outdoor samples were dedicated to ionic compounds, while the other half were dedicated for mutagenicity testing. The experimental design of Phase III (Pilot) studies is summarized in Table 4.

Aeroallergens (airborne pollen and mold spores, for example) are known to trigger or exacerbate lung problems in asthmatics and are therefore important components of the individuals' indoor environment. Measurements of aeroallergens were also determined in every home in Phase III. Aeroallergen samplers were run for a ten to fifteen minute period which was timed by stopwatch and recorded on the data sheet.

Table 3. Experimental Design Per Home-Phase II (Pre-Pilot Study)

Pre-Selector	Indoor No. Samples Day 1	Day 2	Outdoor No. Samples Day 1	Day 2	Sample
PM10	2	0	2	0	IC[a]
PM10	0	2	0	2	Mut[b]
Cyclone	2	2	2	2	IC
Cyclone	2	2	2	2	Mut
Cascade	1	0	1	0	IC
Cascade	1	0	1	0	Mut
Nicotine	0	2	0	2	-
Aeroallergen	0	1	0	1	-

[a] IC = Ionic Compounds

[b] Mut = Mutagenic Activity

Table 4. Experimental Design Per Home - Phase III (Pilot Study)

Pre-Selector	Indoor No. Samples	Outdoor No. Samples	Sample Type
PM10	2[a]	2[a]	IC
PM10	1[b]	1[b]	Mut
Cyclone, Stationary	2	2	IC
Cyclone, Stationary	2	2	Mut
PM10	2[a]	2	IC
PM10	1	1	Mut
Cyclone, Stationary[b]	2	2	IC
Cyclone, Stationary	2	2	Mut
Aeroallergen	2	2	-

[a] The number of mutagenicity and ion chromatography samples alternated from home to home. At alternative home sites, a total of four PM10 samples were assigned for mutagenicity testing and two PM10 samples assigned for ion chromatography.

[b] One personal cyclone sample was scheduled to be collected from each home. Samples were alternatively assigned for mutagenicity or ionic testing.

a. Subject Recruitment

The subjects were chosen from a pool of asthma subjects participating in an acid-atmosphere study, in which they had been interviewed and could be expected to cooperate during the study. We made preliminary calls to their homes, informing them of our plan, and arranged for a convenient sampling day. Approximately two weeks prior to field sampling, each participant was sent a letter which detailed the experimental protocols, the space requirements for the project and their participation in the study.

b. Sampling Plan

All samplers were placed as sets either indoors or outdoors for ionic compounds and mutagenicity. They were always placed adjacent to each other. We placed an equal amount of samplers indoors and outdoors. Since there were only two Burkard aeroallergen samplers, we located one indoors and one outdoors. The detailed experimental design is summarized in Table 4. Briefly, six PM10 samplers were used (3 indoors, 3 outdoors; 2 each for ionic compounds for indoor or outdoor and 1 each for mutagenicity testing for the 1st, 3rd, 5th and 7th home). There were also eight personal (cyclone) samplers available per home and were divided equally for the ionic compound or mutagenicity determinations. All samplers were run for 24 hours.

In summary, for each sampling day for Phase II and III, we had a total of approximately fourteen filters for both ionic compound as well as mutagenicity analyses. During Phase II, we had an additional 18 filters from cascade impactors. For the entire study, approximately 300 individual filters were analyzed.

The following samples were collected for each of the locations: 1) indoor and outdoor fixed-site PM10 samples; 2) indoor and outdoor fixed-site cyclone samples; and 3) personal cyclone samples. At selected sites, indoor and outdoor samples of aeroallergens and environmental tobacco smoke were also collected.

4. Personal Monitoring

One objective of this study was to determine the feasibility of collecting personal particle samples from subjects as they go about normal daily activities. This requires equipping subjects with portable personal samplers. Since there were no PM10 personal samplers available at the time this project was initiated, we utilized size

selective cyclone samplers that were designed for measuring personal respirable particle exposure in occupational settings during an eight hour period.

We wanted to collect personal samples during the same 24-hour period that fixed-site PM10 and cyclone samples were operated, therefore it was necessary to use a portable sampling pump with a large battery capacity. Most sampling pumps designed for occupational monitoring will only hold a charge for twelve to fifteen hours. For this project we utilized a moderately heavy personal pump, with sufficient battery capacity to extend the sampling period, that could be operated on either line or battery power (SKC, Fullerton, CA).

Subjects were instructed to wear the monitors or keep the monitors near themselves during the entire 24 hour period. The subjects were instructed that during sleep or any extended stationary period they were to plug the pump into a nearby wall outlet. When they moved from that location, the subjects were told to unplug the monitor from line power and continue to wear the monitor in the battery mode. This procedure was designed to insure that there would be sufficient battery charge to sample an entire 24-hour period.

5. Time, Location and Health Records

To investigate the possibility of using time diaries to supplement personal particle measurements, we used a time-and-location diary displayed in Appendix A. The form was developed by our group for use in personal exposure studies of nitrogen dioxide. Use of the diary permits application of the 'indirect' method of Duan for determining contributions of various microenvironments to total personal exposure (Duan, 1982).

For this pilot study the location diaries were used only to assess the efficacy of using diaries in conjunction with the personal sampling pumps. Since there was a small sample of asthmatic subjects utilized in this pilot study, it would not be appropriate to apply Duan's method. In this pilot study there were too few degrees of freedom to attempt to statistically disassociate the influence of microenvironments on personal particulate exposure.

Similarly, health diaries were used in this pilot study to test volunteer response and acceptance of multiple questionnaires. Multiple forms can become burdensome for

volunteers and could result in a decrease in participation rates and compliance. The health diaries used in the study were designed to assess symptom and pulmonary function changes in asthmatics.

An example of the health diary is shown in Appendix 2. The subjects in the pilot study were all participating in an assessment of the health responses of asthmatics to acidic aerosols, therefore they had been previously instructed on the use of the diaries. The diaries are designed to measure the *changes* in health status and pulmonary function over time in individual subjects. The diaries are not designed to assess differences in response among a group of subjects. Since the diaries are designed for *time series* analysis, and not *cross sectional* analysis, it is not possible to interpret the specific entries for the eight subjects in this study. Rather, the health diaries were evaluated for the feasibility of combining exposure assessments with health evaluations in a single study.

D. **Quality Assurance**

1. Blank Filters

Throughout the course of the study we used blank filters as an internal check for ionic compound and mutagen content. For each filter type there was associated a blank filter - indoors and out. The filters were inserted into a filter holder and placed adjacent to the field monitors during the entire sampling period. The handling, extraction and analysis of blank filters were identical to procedures used for field samples.

2. Paired Samples

Samplers were paired for all sites visited - indoors and out. For all filter types there were two filters. The pairings consisted of the following: PM10 samplers for ion analysis; PM10 samplers for mutagen analysis; cyclone samplers for ion analysis; cyclone samplers for mutagen analysis.

Due to the number of PM10 samplers available for use during a single 24-hour period, pairings for ion and mutagen analysis occurred on alternate sampling days in homes of asthmatics (Phase III only).

3. Analytical

As an internal check on the performance of the microbalance, reference blank filters representing each filter type were weighed repeatedly throughout the study. These filters were labeled "balance blanks". This procedure checks for drift of the microbalance and for the influence of environmental factors such as temperature or humidity on the weighing day. Balance blanks were weighed before, during and after measurement of filters collected from field sites. As a convention, if balance blanks changed by more than 10% the filter samples would be reweighed.

The ion chromatograph was calibrated before each series of determinations with known sulfate and nitrate standards.

E. Data Analysis

This study was designed as a pilot effort to test methods and procedures. Therefore, we did not design an elaborate sampling and analysis plan that would be more appropriate to a large-scale population survey of personal and indoor exposure to PM10 mass, sulfate, nitrate and mutagens. Specifically, we set out to determine the efficacy of using the selected sampling and analysis techniques in a population survey or epidemiological study. To meet that objective, the design was straightforward and the analysis matched the design.

Data were entered on computer from laboratory notebooks and managed using SAS on the IBM mainframe and Excell on the Apple Macintosh computers. All statistical analyses were performed on the Macintosh using StatView. Three groups of analyses were performed to support the objectives of the study. Quality assurance analyses were conducted to determine the reproducibility of results. Data from the homes were presented descriptively to present information on the distribution of concentrations for air pollution species and mutagenic activity. Finally, inferential methods were used to compare indoor and outdoor concentrations and to evaluate correlations that were not visually obvious.

1. Quality Assurance

Samplers were paired as often as permitted by the total number of sampling units. This was done to allow comparison of the samples from the stage of preparation through sampling and laboratory analysis. Paired samples were presented as scatterplots to facilitate visual inspection of the relationships. Highly reproducible methods appear as nearly straight lines along the 1:1 line. Paired data are summarized in terms of paired differences, correlation coefficients, and the coefficients of variation* of the paired difference .

2. Descriptive Data

Concentrations were presented using arithmetic means, standard deviations, medians, mean ratios, median ratios, interquartile ranges and by scatterplots. Since air quality distributions are often skewed to the right, medians are presented along with arithmetic means. Indoor/outdoor concentration ratios were presented in terms of mean ratios and median ratios. Interquartile ranges and scatterplots present information on the distribution of observations.

3. Inferential Methods

Indoor/ outdoor differences are evaluated in terms of a nonparametric method (Mann Whitney U statistic) due to the skewed nature of the concentrations. When it was uncertain whether correlations were significant, the nonparametric Spearman rank order correlation coefficient was used.

* This is the standard deviation of the paired differences divided by the average of the difference, expressed as percent.

IV. RESULTS

The following sections contain the major results of this pilot study. It has been demonstrated in the course of this study that aerosol mass, ionic compounds, mutagenic activity, aeroallergens, and nicotine can be reliably and accurately collected and analyzed from residential samples. Personal sampling for mass, ionic compounds, and mutagenic activity has also been demonstrated to be feasible. This effort went well beyond the objectives of the original pilot study to generate interesting and provocative results on indoor-outdoor relationships and on interrelationships among particulate species.

This section is divided into divisions describing results from analysis of Phase I (Laboratory and chamber studies), Phase II (Pre-Pilot) and Phase III (Pilot Study) for particulate matter, ionic compounds, determination of mutagenic activity, analysis of nicotine and aeroallergens, and review of quality assurance activities.

A. Phase I - Laboratory and Chamber Studies

Phase I efforts, with a laboratory test and clean chamber studies, were designed to assure that high quality results could be obtained from sampling and analytical results that were being adapted for application in residential and indoor monitoring.

1. Laboratory Studies

The results of the laboratory tests, presented on Table 5 indicate that highly precise measurements were made from the start of Phase I. The air inside the laboratory is relatively clean since the building has central air cleaning and conditioning equipment. Therefore, the results represent a difficult test of the sampling and analysis of air particulates from an environment which has low concentrations.

There are several notable trends apparent on Table 5. First, the coefficient of variation for measuring mass, sulfate and nitrate is very low. The coefficient of variation is the standard deviation of a sample divided by the sample mean and expressed as a percent. For the low concentration samples collected in the laboratory, the standard deviation of the measures is generally less than 10% of the mean concentration. It is

Table 5. Phase I - Laboratory Test. Precision of Indoor and Outdoor Samples[a]

Site	In/Out	Date	Sample	N	Mean Conc (µg/m3)	C.V. (%)	Sampling Duration (hrs)
UCI Lab	Indoor	6/25/87	Mass	4	12.8	8.0	24
		6/25/87	Nitrate	4	0.3	4.5	24
		6/25/87	Sulfate	4	5.7	3.9	24
UCI Lab	Indoor	8/10/87	Mass	4	11.4	9.0	23
		8/10/87	Nitrate	4	0.5	6.1	23
		8/10/87	Sulfate	4	3.2	2.9	23
UCI Lab	Indoor[b]	8/21/87	Mass	4	10.1	8.5	72
		8/21/87	Mass[c]	4	11.6	17.4	72
		8/21/87	Nitrate	4	1.3	2.5	72
		8/21/87	Sulfate	4	2.9	0.7	72
Technician's Home (Outdoor)		8/26/87	Mass	4	14.5	7.7	36
		8/26/87	Nitrate	4	.62	7.7	36
		8/26/87	Sulfate	4	5.3	5.3	36

[a] Unless otherwise indicated, PM10 air samplers used (flow rate = 4 liters/min) with Teflo™ filters.

[b] Cyclone pre-selectors and personal air samplers operated at 2 liters/min.

[c] Zefluor™ filter.

expected that the coefficient of variation would be even lower for samples typical of outdoor and residential settings.

Second, the coefficient of variation of mass is greater than that of the constituent anionic species. This is due to the inherent variability associated with measuring mass. The lighter PM10 samples will have collected a total mass of approximately 50μg of particulate matter after 24 hours of sampling. It is very difficult to reliably measure such a small change in mass on the filters. Nonetheless, our weighing protocol led to precise weighings. From Table 5, it is clear that individual chemical constituents of particulate matter, in this case sulfates and nitrates, can be measured more precisely than the particulate mass of which they are a part.

Third, there is some indication that Teflo™ filters are better than Zefluor™ filters for gravimetric determination of mass. On the August 21 laboratory test, four cyclone samplers were fitted with each filter type. During Phase II and III, the Zelfuor™ filters were used only for mutagenicity testing in the cyclone samplers and were not used in the PM10 samplers or in the cyclone samplers used for ionic speciation. Teflo™ filters were used exclusively in all PM10 samplers and in all cyclone samples used for ion speciation.

Fourth, outdoor measurements made at the home of one of the laboratory technicians (also on a clean ambient day) indicated that precision obtained in the laboratory is also possible to obtain in the field. Together, these results clearly indicate that highly precise data are obtainable using portable particulate sampling equipment and careful laboratory procedures.

2. Chamber Study

Following the trial measurements at the technician's home, we tested the sampling equipment inside an APHEL exposure chamber set to deliver clean air. These results are presented in Table 6 and indicate that background levels are very low. The small residual amounts of mass, sulfate, nitrate and mutagenicity measured are probably from residue in the air delivery system. Due to inherent measurement error, which is proportionately exaggerated at low concentrations, the coefficients of variation in this test will be higher than for the field and open laboratory measurements. Nonetheless,

Table 6. Phase I - Chamber Studies (24-hr Exposures)

Pre-Selector	Sample	N	Mean Concentration		SD
PM10	Mass	4	2.6	µg/m3	1.0
	Nitrate	2	0.2	µg/m3	NA*
	Sulfate	2	1.5	µg/m3	NA
	Mutagenicity	2	6.1	rev/m3	NA
Cyclone	Mass	8	3.5	µg/m3	2.0
	Nitrate	3	0.0	µg/m3	0.0
	Sulfate	3	0.2	µg/m3	0.3
	Mutagenicity	3	2.4	rev/m3	5.3

* NA = Not compatible with two samples.

these results are extremely tight and provide a further indication of the precision and accuracy of the measurement methods.

3. Blank Filters

Filter blanks were evaluated for sulfate, nitrate, mutagenicity and mass. Field blanks were weighed before and after field visits. In order to compare the blank values on the same scale as samples, the measured mass and particulate species were divided by the nominal volume for a 24-hour sampling period. These data are presented on Table 7.

The important result of this effort is that the filter backgrounds are very low. All means are within at least two standard deviations of zero, indicating that any background is too low to be measured.

B. Phase II (Pre-Pilot) and Phase III (Pilot) Studies

The Phase II pre-pilot studies were conducted in the homes of (2) nonasthmatic volunteers to practice all protocols. This phase was planned so that if problems were encountered at any level of analysis we would not use the data and would be able to adjust the protocol before entering Phase III.

The Phase II field and laboratory efforts went smoothly and it was decided to merge the Phase II results with the Phase III results and present the combined residential data in this section. Since the total sample of homes was relatively small, this increases the sample statistics and helps to reinforce the trends that are apparent from the data.

1. PM10 Mass Studies (Data from PM10 Samplers Only)

a. Summary Statistics

All PM10 mass observations were combined for the prepilot and pilot (experimental) homes to produce Table 8. The distributions for these mass data were skewed toward higher values, which is typical of pollutant concentrations. For this reason, the median concentrations are presented in Table 8 along with the arithmetic averages. The outdoor median as well as average concentrations exceed the corresponding values observed indoors in this sample of homes. An indication

Table 7. Blank Filter Results

Sample	N	Mass (µg/m3) Mean (±S.D.)	Nitrate (µg/m3) Mean (±S.D.)	Sulfate (µg/m3) Mean (±S.D.)	Mutagenic Activity (Revertants/m3) Mean (±S.D.)
PM10 Ion [b]	12	-0.67 (2.24)	0.10 (0.08)	0.26 (0.23)	NA [a]
Cyclone Ion [b]	10	2.05 (3.91)	0.07 (0.07)	0.44 (0.40)	NA
PM10 Mutagen [b]	14	-0.09 (1.28)	NA	NA	6.00 (6.57)
Cyclone Mutagen [c]	10	1.87 (1.43)	NA	NA	7.77 (4.95)

[a] NA = Not applicable: analysis not performed on filter

[b] Teflo™ filter (Teflon membrane filter)

[c] Zefluor™ filter (Teflon membrane with Teflon support)

Table 8. PM10 Mass Concentration for All Homes Combined (Filters used for Ionic and Mutagenesis Analyses)

Location	n	Mean Mass µg/m3 (± SE)	Median Mass (µg/m3)	No. Observations > 50µg/m3 (%)[a]
Indoor	35	42.5 (3.7)	35.7	10 (29%)
Outdoor	34	60.8 (4.7)	50.9	18 (53%)

[a] 50µg/m3 of PM10 is the California State Ambient Air Quality Standard (24 hour average).

of the average difference between indoor and outdoor PM10 is computed as the indoor-to-outdoor ratios of the average and median concentrations:

$\bar{I} / \bar{O} = 0.70$; indoor-outdoor ratio of averages

$I_{50\%} / O_{50\%} = 0.70$; indoor-outdoor ratio of medians

Another notable observation from these summary data is the large number of PM10 measurements that exceed the 24-hour California state standard. Over 50% of all outdoor measurements exceeded the state standard and nearly 30% of indoor concentrations exceeded the standard. The late fall and early winter period when these measurements were made is typically the most stagnant meteorological time in Southern California. If ambient PM10 data had been taken on a daily basis near the homes, it would have been possible to compare our monitoring results with those from the compliance network. It may still be possible to identify area-wide PM10 monitoring with which to compare the monitoring days in this study.

In Figure 7 we present the paired indoor and outdoor average PM10 concentrations by monitoring site and day. The observations are ranked by indoor concentration, with the associated outdoor concentration shown adjacent to the indoor concentration. Each bar represents the average of one to four simultaneous measurements.

Several features are noted in this figure. First, over half of the outdoor day-site observations are above the state PM10 standard. Second, individual indoor concentrations are generally lower than corresponding outdoor levels. The four instances when the indoor levels exceeded outdoor concentrations come from four days of sampling in the home of a nonasthmatic participating in the prepilot study. The highest indoor concentration was recorded in the home of another nonasthmatic from the prepilot study, although the outdoor level was also high on that sampling day. Finally, the home with the lowest indoor concentration was the residence of an asthmatic who used a central air cleaning system. This residence also had the lowest indoor/outdoor concentration ratio.

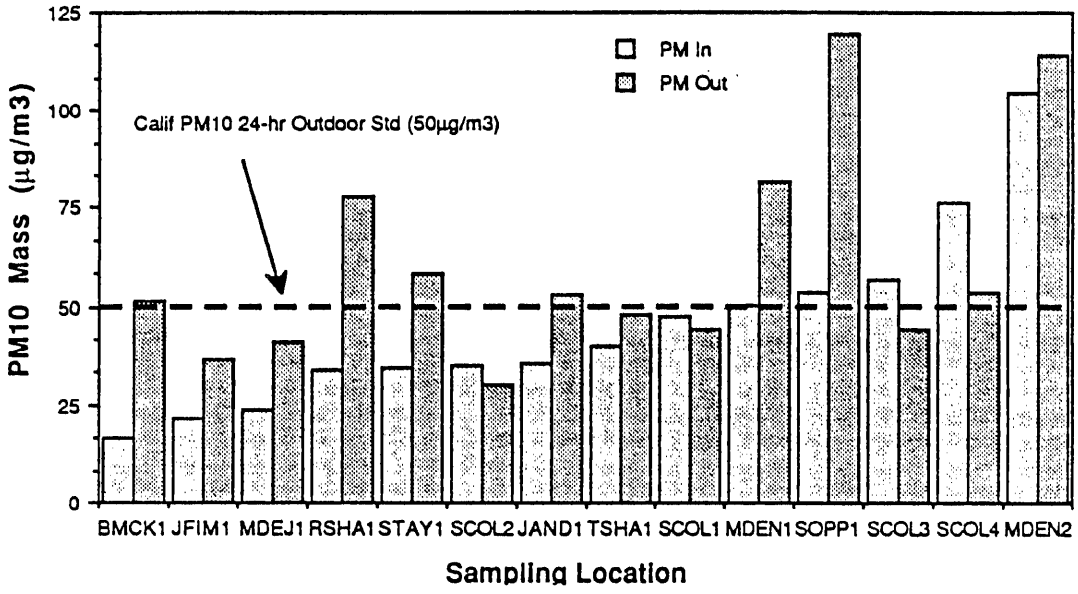


Figure 7. Indoor and outdoor PM10 measurements by home. Observations ranked from lowest to highest indoor concentration.

b. Indoor and Outdoor Comparisons

Indoor-outdoor paired PM10 concentrations were tested to determine whether the indoor or outdoor concentrations were significantly higher. The higher outdoor average concentration noted in the previous section was found to be significantly higher than the corresponding indoor values ($p<0.05$). Since the concentrations are not normally distributed, indoor-outdoor pairs were tested using the nonparametric Wilcoxon-Signed Rank test for paired data.

In Figure 8 the individual indoor and outdoor pairs are presented in a scatterplot. This plot displays the results for cyclone samplers along with the PM10 data pairs. Table 9 contains the regression results that correspond to these data pairs. It is clear from the scatterplot that most indoor concentrations fall below or near the outdoor concentrations.

The results from the regressions indicate that indoor concentrations are moderately correlated with outdoor concentrations; the outdoor concentrations accounting for between 34 and 56% of the variation of indoor concentrations. Recall from Section IV. A.1 that mass is the one of the least reliable measures from paired samples. The inherent difficulty of measuring mass represents some of the unaccounted variability. Indoor sources of particulate matter and modification by the building envelope of outdoor concentrations penetrating to the indoor account for additional unexplained variation. Homes from this sample of nonsmokers, with (8) residences (asthmatic homes), tend to have indoor concentrations that are lower than those measured outdoors.

c. PM10 Sampler and Cyclone Sampler Comparisons

In Table 10 we presented the overall average concentration for indoor and outdoor cyclone samplers from all homes, including the prepilot residences. Since there were additional duplicate samplers available for cyclone sampling, the number of cyclone samples exceeds the number of PM10 samples presented in Table 8. As with the PM10 samples, the outdoor average concentration exceeds the corresponding indoor average. Similar to the case with PM10, the cyclone mass concentrations are skewed to the right, which is the reason that median concentrations are lower than average concentrations.

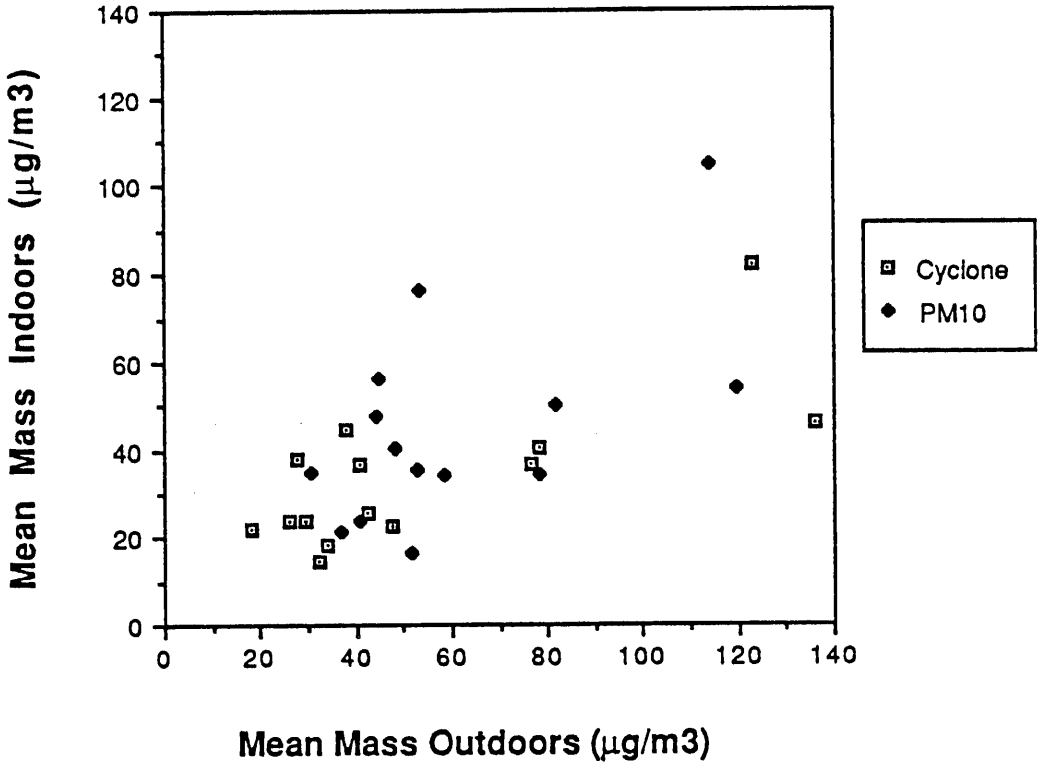


Figure 8. Scatterplot of paired indoor and outdoor mean particulate mass concentrations.

Table 9. Regression of Indoor Mass on Outdoor Mass
(Outdoor Values used to Predict Indoor)

Pre-Selector	DF[a]	R^2	Slope	S.E. Slope[b]	Intercept	p
Cyclone + PM10	27	0.41	0.42	0.10	15.7	<0.01
Cyclone	13	0.56	0.35	0.09	15.3	<0.01
PM10	13	0.34	0.49	0.20	15.0	<0.05

a Degrees of freedom (number of indoor/outdoor pairs used in regression minus 1).

b Standard error of the slope.

Table 10. Cyclone Mass Concentration for All Homes Combined
(Filters used for Ionic and Mutagenesis Analyses)

Location	n	Mean Mass µg/m3 (± SE)	Median Mass (µg/m3)	No. Observations > 50µg/m3 (%)
Indoor	54	32.7 (2.2)	30.4	4 (7%)
Outdoor	54	51.5 (4.9)	38.5	16 (29%)

The indoor-to-outdoor ratios for the cyclone samples are computed in the same manner as they were computed for the PM10 samplers:

$\bar{I} / \bar{O}$ = 0.64; indoor-outdoor ratio of averages

$I_{50\%}/O_{50\%}$ = 0.79; indoor-outdoor ratio of medians

Since the *outdoor* cyclone concentration distribution is strongly skewed, having a few high measurements, the mean and median ratios differ. Within experimental error the cyclone ratios are the same as the PM10 ratios. Since the cyclone excludes particles larger than 7μm and has a 50% cutpoint at approximately 5μm, the concentrations on the PM10 filters is expected to be greater than those measured on cyclone filters. Since the cyclone concentrations are lower, fewer observations exceed the state standard of 50μg/m^3. While the smaller size distribution sampled by the cyclone does not justify a direct comparison with PM10 concentrations, it is interesting to note that a substantial number of cyclone observations still exceed the state standard.

Figure 9 shows the scatterplot of paired PM10 to cyclone observations, indoors and outdoors. If there was more than one monitor type in the indoor or outdoor location, the values were averaged. As seen from the figure, there is good agreement between the cyclone and PM10. Table 11 gives the regression relationship between the monitoring types. Nearly 90% of the variation in PM10 mass is explained by cyclone mass measurements. In spite of the high correlation, the intercept value is still higher than expected and the regression coefficient lower than anticipated. This is due to a combination of measurement error, which will bias the slope and intercept, and to two individual high outdoor cyclone measurements for which PM10 measurements were lower.

Table 12 provides information on the average difference between PM10 and cyclone measurements. The PM10 values, as expected, are generally greater than cyclone measurements.

A Wilcoxon Signed rank test indicates that the PM10 concentrations are significantly greater than the paired cyclone concentrations ($p<0.01$). Table 12

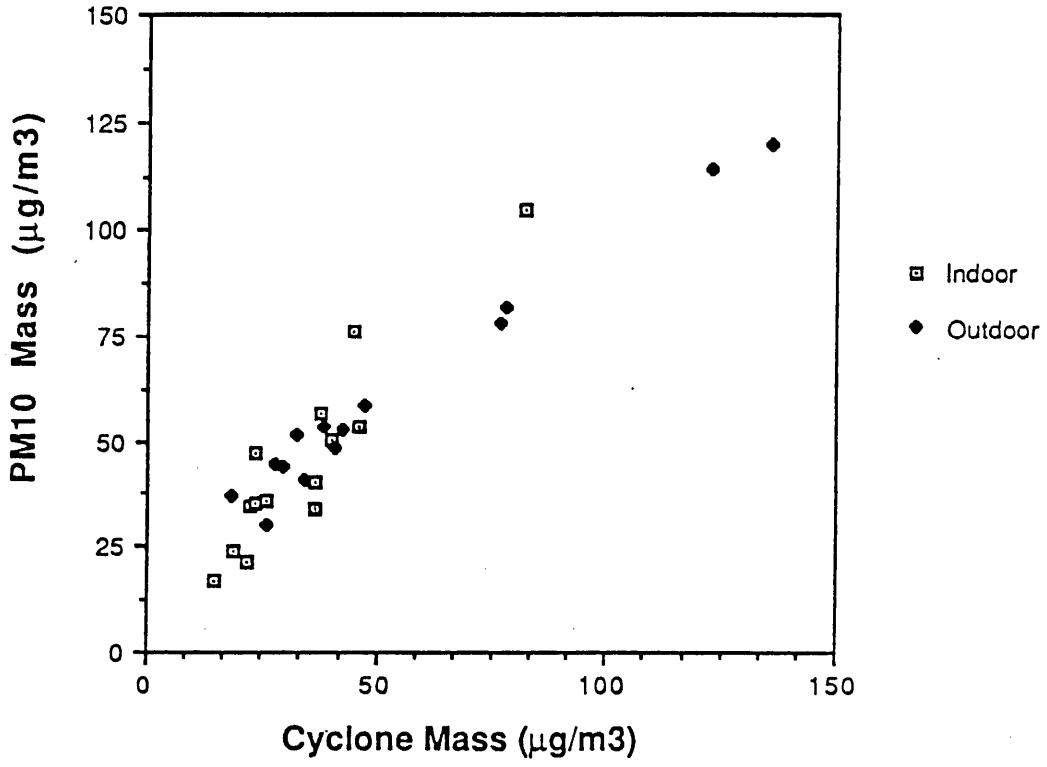


Figure 9. Scatterplot of PM10 mass to cyclone sampler mass.

Table 11. Regression Analysis of PM10 Mass on Cyclone Mass[a] (Cyclone Values used to Predict PM10)

DF[b]	R^2	Slope[c]	S.E. Slope	Intercept	p
27	0.89	0.83	0.58	16.6	<0.01

[a] Indoor and outdoor observations combined.
[b] Degrees of freedom (1 minus number of indoor/outdoor pairs used in regression.)
[c] Standard error of the slope

Table 12. PM10 vs Cyclone Mass (Indoor Observations combined with Outdoor)

Pre-Selector	Mean	S.E. Mean	Median	Mean Ratio	Median Ratio
PM10	53.1	5.0	48.0	0.90	0.84
Cyclone	43.8	5.6	36.8		

also indicates that an average cyclone mass is approximately 85-95% of PM10 mass. The high proportional mass explains why there is a good correlation between PM10 and cyclone mass. In general, these observations imply that for the sampling period and location, cyclone mass is a good predictor of PM10 mass.

d. Personal Sampling Measurements

Figure 10 shows the personal particulate exposure data from three out of the seven asthmatic subjects that carried the portable (cyclone) sampler. For comparison, the figure also displays fixed-site indoor and outdoor concentrations of the corresponding PM10 and cyclone samplers. The personal samples consistently compare better with the indoor samples than they do with the outdoor samples. From these data it appears that monitoring indoor residential air, using either a PM10 or cyclone sampler, closely represents personal exposure to particulate matter in this sample of asthmatic subjects.

Asthmatic subjects might be expected to behaviorally avoid particulate exposure, especially from cigarette smoke (a major source of personal exposure to particulate matter).

2. Ionic Compound Studies

a. Summary Statistics

Sulfate and nitrate concentrations were combined for the prepilot and pilot homes and are presented in Table 13. As in the case of the PM10 mass samples, the distributions of concentrations for the constituent ionic species is skewed to the right. The median concentrations, which are uniformly lower than the arithmetic mean concentrations, are a better indicator of the central or most common concentrations.

As an indication of the difference between indoor and outdoor concentrations, the indoor-to-outdoor ratios are shown on Table 14 for the average and median concentrations. From this table it is clear that indoor and outdoor concentrations are similar for sulfate (with average indoor concentrations ranging from 70-96% of the average outdoor concentrations); and dissimilar for nitrate with average indoor concentrations ranging from only 21% up to 57% of average outdoor concentrations.

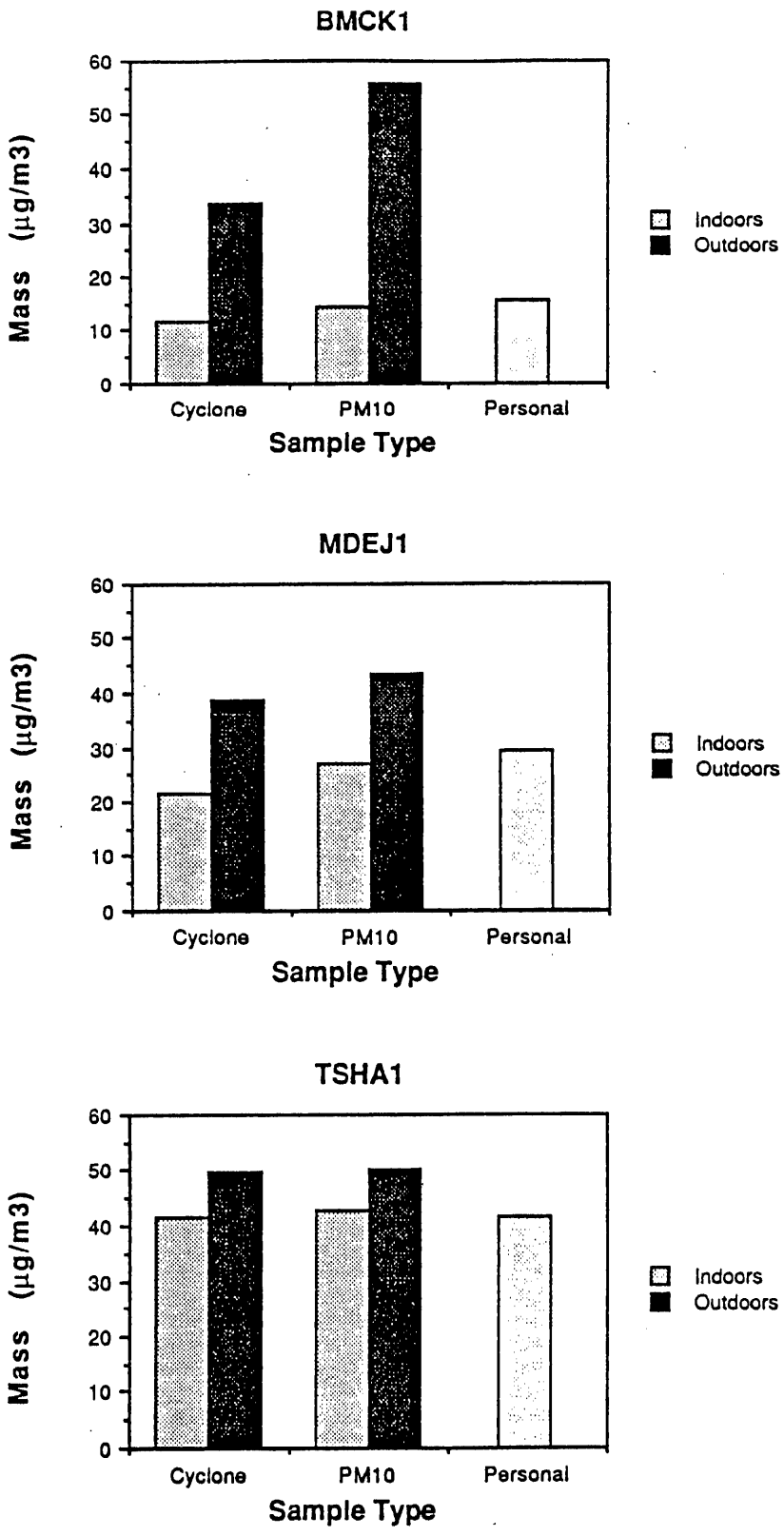


Figure 10. Bar graph of personal cyclone mass with indoor and outdoor PM10 and cyclone samples from Phase III Pilot Studies.

Table 13. Descriptive Summary of Indoor and Outdoor Ions

		Indoor			Outdoor		
		Mean Concentration µg/m3 (S.E.)	Median Concentration µg/m3	n	Mean Concentration µg/m3 (S.E.)	Median Concentration µg/m3	n
PM10	SO4	4.3 (0.9)	3.5	17	6.1 (1.2)	4.0	17
	NO3	2.3 (0.5)	1.4	17	11.1 (3.2)	5.4	15
Cyclone	SO4	5.0 (0.9)	3.9	26	5.2 (0.8)	4.6	24
	NO3	1.1 (0.3)	0.8	22	3.1 (1.7)	1.4	24

Table 14. Ratios of Indoor to Outdoor Average Ionic Concentrations

		Indoor/Outdoor Ratios *	
		Mean Ratio $(\overline{I}/\overline{O})$	Median Ratio $(I_{50\%}/O_{50\%})$
PM10	SO_4	0.70	0.88
	NO_3	0.21	0.26
Cyclone	SO_4	0.96	0.85
	NO_3	0.35	0.57

*Derived from means presented in Table 13.

b. Indoor and Outdoor Comparisons

i. Sulfate. Results from paired indoor and outdoor samples were pooled to investigate the relationship between outdoor and indoor sulfate. The values from regression analyses (see Table 15) indicate that indoor sulfate concentrations are highly correlated with concurrently measured outdoor concentrations of this species. Approximately 90% of the variation of indoor concentration of sulfate is explained by levels measured outside the home.

Similar to what was presented in section IV.B.1 above, the regression coefficient indicates that approximately 70% of the outdoor concentration of sulfate is observed inside the home. The low values for the regression intercept and the high correlation coefficient together make the 70% net penetration factor a plausible number.

A scatterplot of indoor to outdoor sulfate levels is shown in Figure 11a. From the figure it can be seen that both sampling devices span the range of observed concentration and that there is no particular pattern for either the PM10 or cyclone samplers with regard to scatter or clustering at high or low concentrations. This is consistent with most sulfate particles being found in the submicrometer size range.

ii. Nitrate. As shown in Table 15, indoor and outdoor concentrations of nitrate are not well correlated. The nonsignificant regression explains only 4% of the variation in indoor levels of nitrate.

Figure 11b illustrates that outdoor concentrations (note that outdoor nitrate is displayed on the log scale) of nitrate span a much greater range than indoor concentrations and that cyclone samplers tend to collect generally lower concentrations of nitrate. This finding is consistent with a substantial proportion of the nitrate mass being found on larger particle nitrate. This is in contrast to the behavior of sulfate which tend to be found on submicrometer particles.

c. Cascade Impactor Studies

Cascade impactors for ionic speciation were located inside and outside of two homes during the Phase II pre-pilot studies. The results are best shown as histograms of the concentration found on each of the impactor stages. These limited cascade results indicate that stage 7 or 8 is the modal size for both indoor and

Table 15: Indoor to Outdoor Regression Results from Phase II & III Homes (Cyclone results combined with PM10 results)

Dependent Variable*	Independent Variable*	R^2	R	S.E.	P	Intercept	Slope
$Sulfate_i$	$Sulfate_o$	0.89	0.94	1.08	< 0.01	0.07	0.70
$Nitrate_i$	$Nitrate_o$	0.04	0.20	1.66	p =0.40	1.8	-0.03
$Mass_i$	$Mass_o$	0.39	0.62	9.25	< 0.01	19.8	0.25

* Subscript (i) used to represent an indoor sample and subscript (o) represents an outdoor sample.

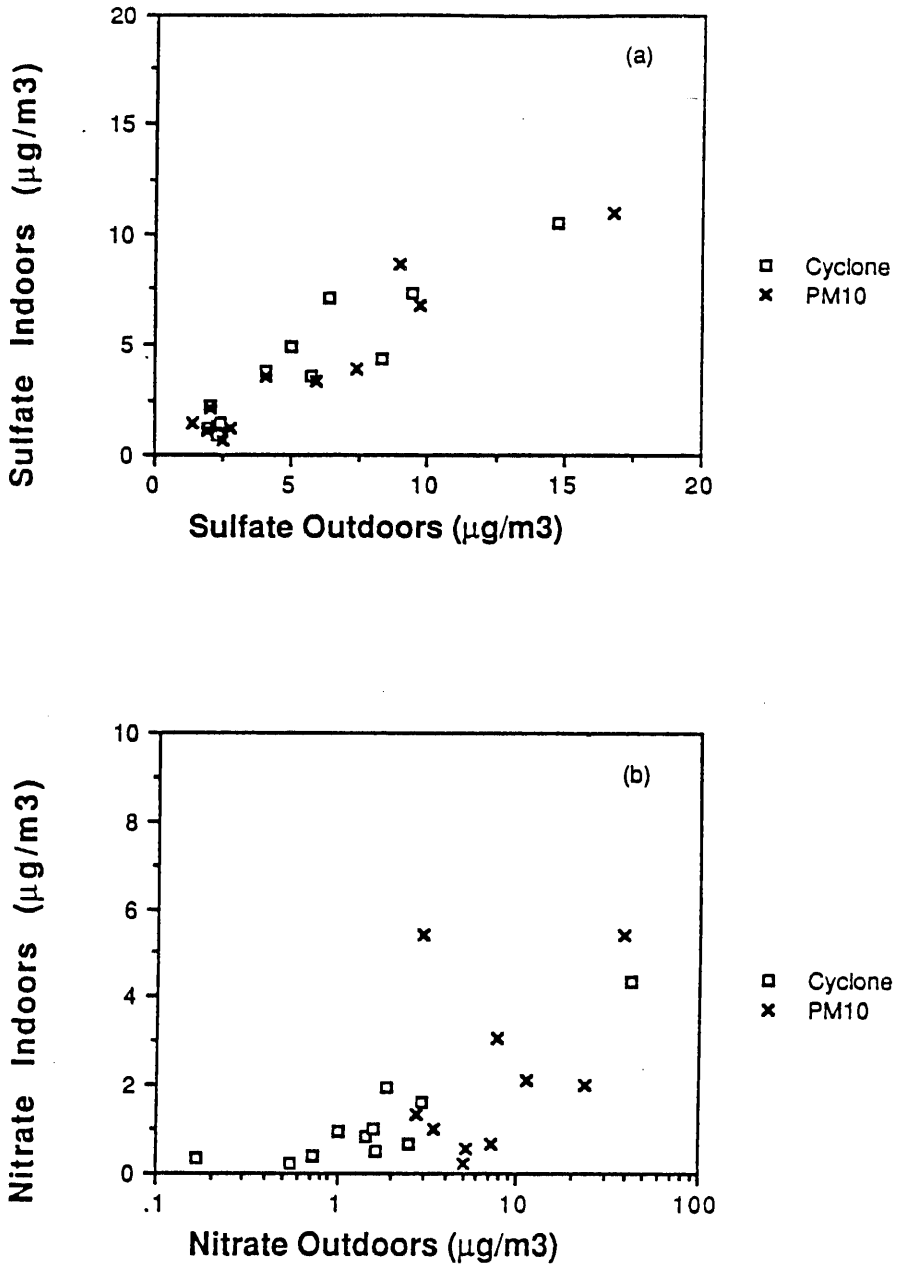


Figure 11. Scatterplots of indoor versus outdoor ionic compounds; a)sulfates, b) nitrates.

outdoor sulfate. Very little sulfate is observed indoor and outdoor on stages 1 through 5. An example is presented from a single home in Figure 12.

In contrast, stage 6 is most commonly the modal stage for nitrate and considerable nitrate can be found on stages 3 through 5. There are too few data to generalize these results, but they are consistent with the differences found in the indoor-outdoor relationships between sulfate and nitrate. The results are also consistent with discrepancies observed for nitrate between the PM10 and cyclone samplers. The cyclone samplers, with a 50% cutoff at 5 μm, tend to collect less nitrate than the PM10 samplers that have a sharp cutoff at 10 μm.

d. Cyclone and PM10 Samplers

i. Sulfate. Figure 13 and Table 16 contain results relating sulfate concentrations collected on cyclone and PM10 samplers. The very high coefficients of variation between PM10 and cyclone-collected sulfate ($R^2 = 0.97$) is as good as can often be expected for paired samples using a single type of sampler. This tight relationship is apparent in Figure 13a and is not systematically dependent on indoor or outdoor sampling.

ii. Nitrate. In contrast to sulfate, nitrate particles show a sampler-dependent relationship. Cyclone samples alone explain 68% of the variation of PM10 nitrate. Another apparent pattern observed in Figure 13b is that higher concentrations for both the cyclone and PM10 samplers are observed outside the house. This indicates that the home may act to lower concentrations of large-particle nitrate observed outside.

e. Ions and Particulate Mass

In this section we relate mass concentration to different combinations of ionic mass. It is important here to note that the nitrate and sulfate concentrations are expressed for the anion alone. We do not assume the mass of a corresponding salt. Therefore, sulfate and nitrate are associated with a higher fraction of the mass than indicated in these results.

i. Sulfate. Figure 14 and Table 17 indicate that sulfate is moderately related to both PM10 and cyclone mass. With the cyclone, approximately 40% of its mass is explained by measurement of sulfate alone. The results for the PM10 sampler are

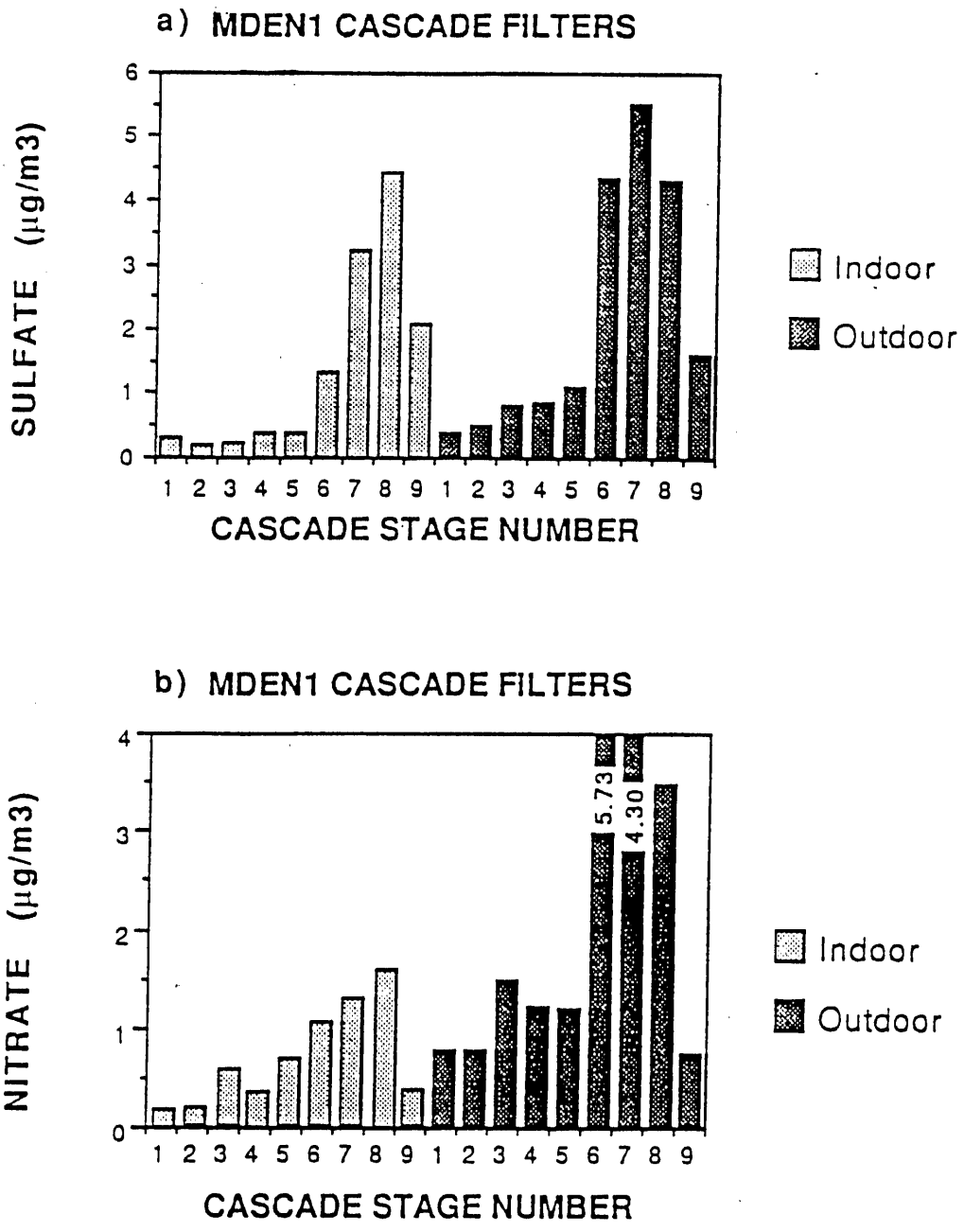


Figure 12. Bar graph of cascade impactor concentrations from home of one subject; a) sulfates, b) nitrates. The nominal 50% size cuts for each stage are as follows in µm: Stage 1 = 21.3; Stage 2 = 14.8; Stage 3 = 9.8; Stage 4 = 6.0; Stage 5 = 3.5; Stage 6 = 1.6; Stage 7 = 0.9; Stage 8 = 0.5; Stage 9 = filter trap (See Figure 6 for details).

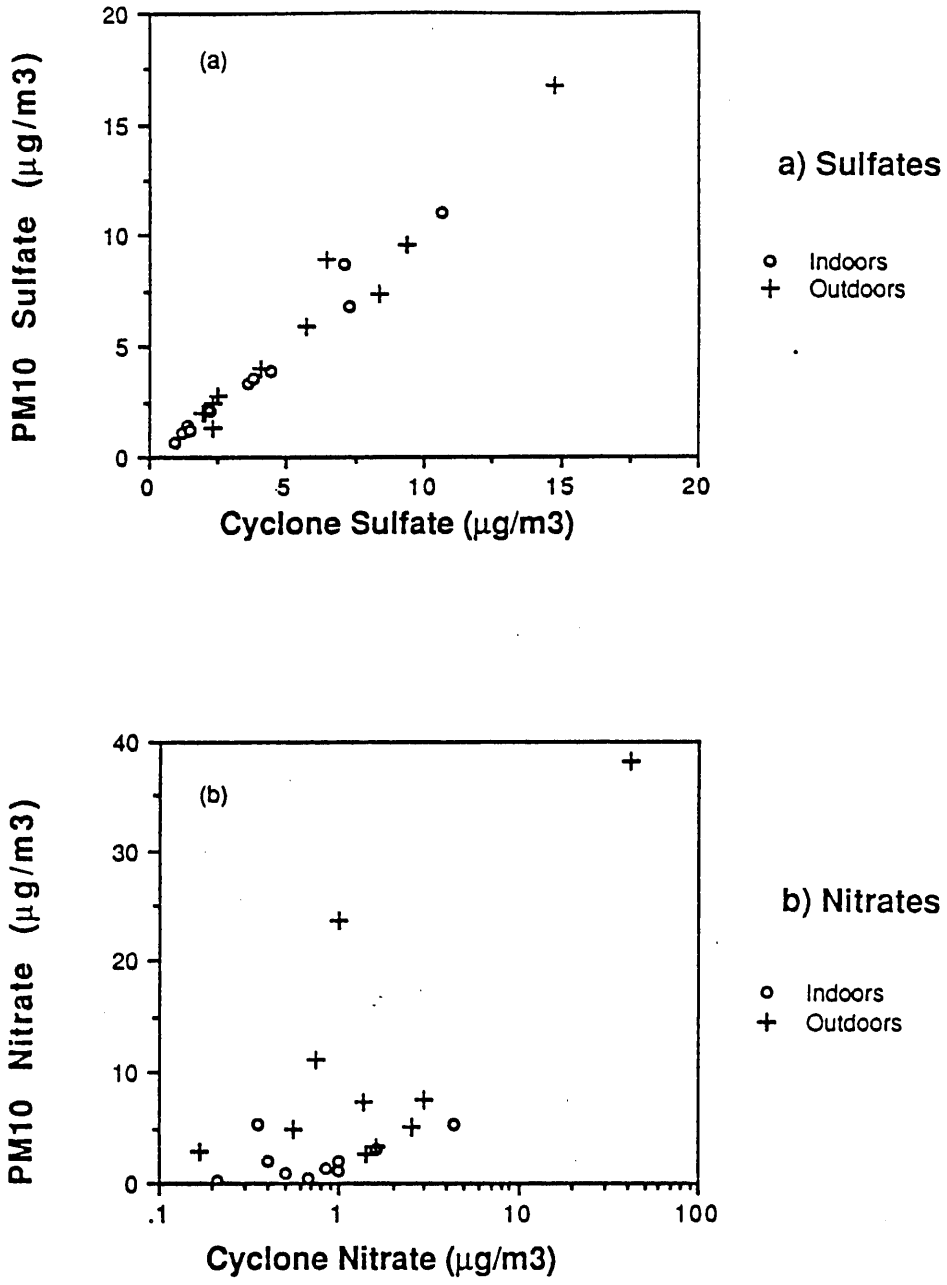


Figure 13. Scatterplots of cyclone versus PM10 concentration.

Table 16: Regression Relationship Between PM10 and Cyclone Samplers

Dependent Variable	Independent Variable	R2	R	S.E.	P	Intercept	Slope
$Sulfate_{PM10}$	$Sulfate_{cyclone}$	0.97	0.98	0.8	<0.01	-0.4	1.1
$Nitrate_{PM10}$	$Nitrate_{cyclone}$	0.68	0.82	5.3	<0.01	3.8	0.82

Table 17: Regression Relationship Between Mass Concentration and Ionic Species, by Sampler Type*

Dependent Variable	Independent Variable	R2	R	S.E.	P	Intercept	Slope
$Mass_{cyclone}$	$SO_{4cyclone}$	0.42	0.65	15.0	<0.01	19.1	3.3
$Mass_{PM10}$	SO_{4PM10}	0.30	0.55	20.8	<0.01	32.8	3.1
$Mass_{cyclone}$	$NO_{3cyclone}$	0.57	0.76	12.1	<0.01	28.9	2.4
$Mass_{PM10}$	NO_{3PM10}	0.82	0.91	10.7	<0.01	32.9	2.4

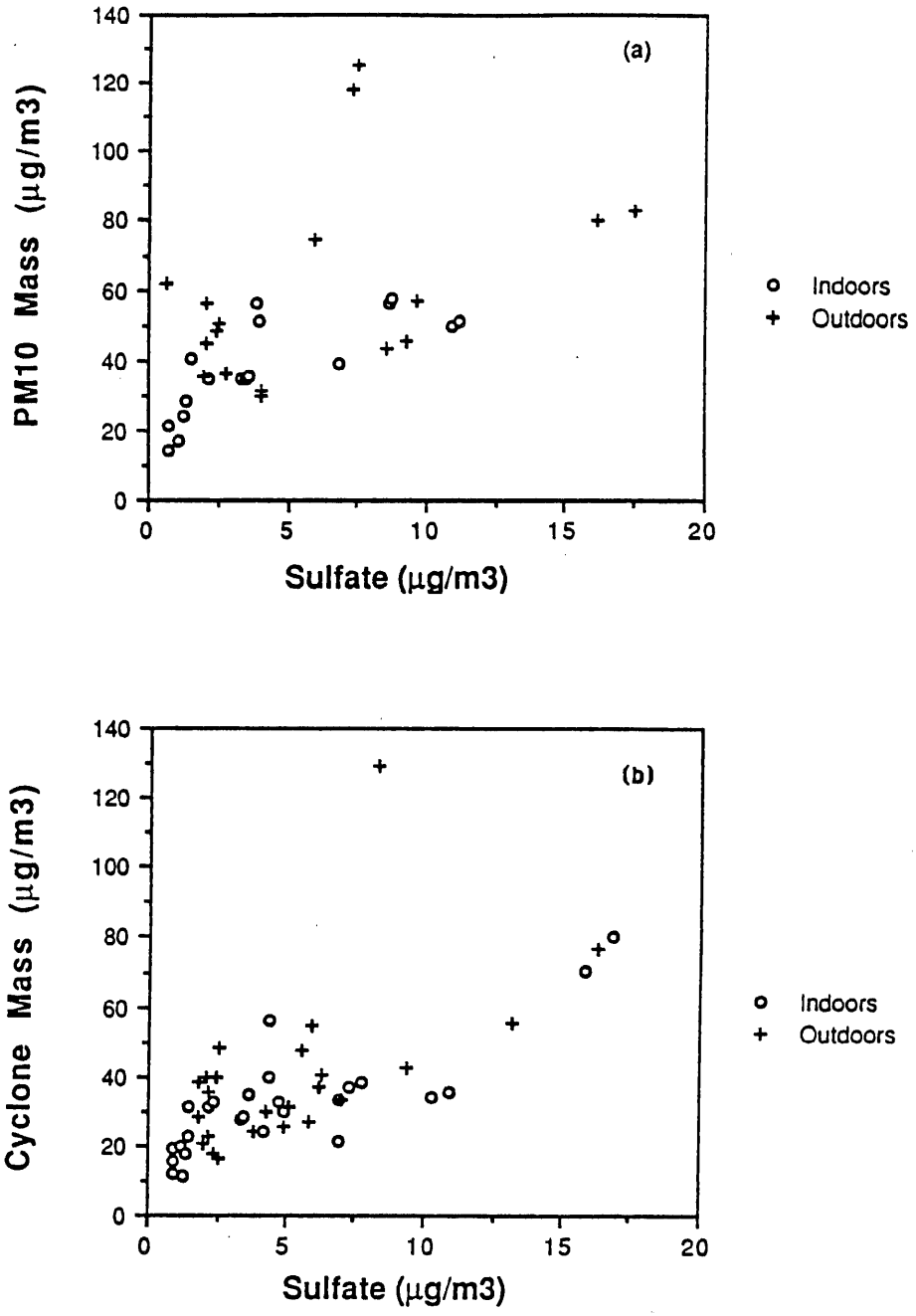


Figure 14. Scatterplots of mass concentration versus sulfate; a) PM10, b) cyclone.

not as strong, with approximately 30% of the variation in mass explained by sulfate measurement. From inspection of Figure 14, it can be seen that scatter is created by one or more occasions when sulfate levels were moderate while the highest outdoor mass concentrations were observed.

ii. Nitrate. Figure 15 and Table 17 demonstrate that nitrate is correlated with mass concentration. For the cyclone, nearly 60% of the variation in mass is explained by the nitrate measurement. For the PM10- sampler an even higher 80% of the mass variation is explained by the nitrate levels. For the cyclone samples it is clear that a single high outdoor sample drives the relationship and provides most of the association; and, in fact, the nonparametric Spearman correlation is not significant. The PM10 sampler demonstrates a more consistent relationship over the range of mass and nitrate concentrations. It should be noted in Figure 15a that indoor concentrations of mass and nitrate are consistently lower than outdoor concentrations (note the cluster of circles in the lower left portion of the figure).

iii. Sulfate and Nitrate Correlations. In the following section, we explore the extent of any relationship between sulfate and nitrate concentrations. Table 18 indicates that sulfate and nitrate concentrations are not significantly associated for either the PM10 nor the cyclone samplers. This lack of association is apparent in Figure 16 and is not explained by the presence of outliers nor a pattern between indoor and outdoor samples.

f. Personal Samplers

The results for ionic concentrations from the limited personal sampling is shown in Figures 17 and 18 for sulfate and nitrate, respectively. Figure 17 shows a clear pattern in the four homes with personal monitoring, where indoor concentrations of sulfate are found to be lower than outdoor concentrations; and, personal levels appear to reflect the indoor concentrations. PM10 and cyclone samplers give consistent results for sulfate.

Results for nitrate, shown in Figure 18, similarly indicate that higher concentrations are generally observed outside and personal samples reflect the lower indoor concentrations. This result is consistent with personal samples of particulate mass. On several of these sampling efforts, the outside PM10 samples collect more nitrate than outdoor cyclones or than indoor concentrations from any sampler.

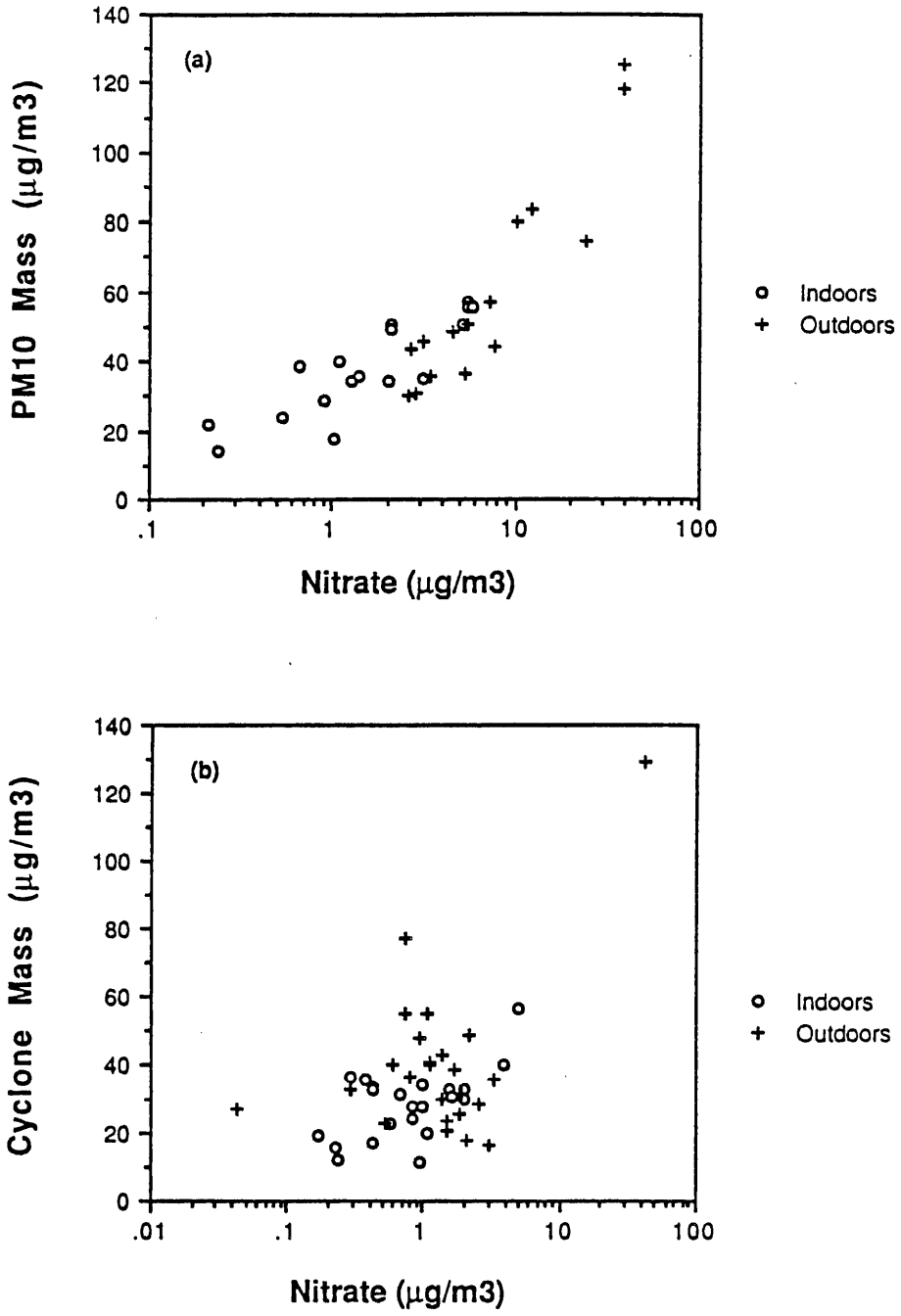


Figure 15. Scatterplots of mass concentration versus nitrate; a) PM10, b) cyclone.

Table 18: Relationship Between Nitrate and Sulfate Concentration

Dependent Variable	Independent Variable	R^2	R	S.E.	P	Intercept	Slope
$Nitrate_{cyclone}$	$Sulfate_{cyclone}$	0.04	0.20	8.3	0.34	0.67	0.49
$Nitrate_{PM10}$	$Sulfate_{PM10}$	0.15	0.39	3.8	0.08	2.77	0.18

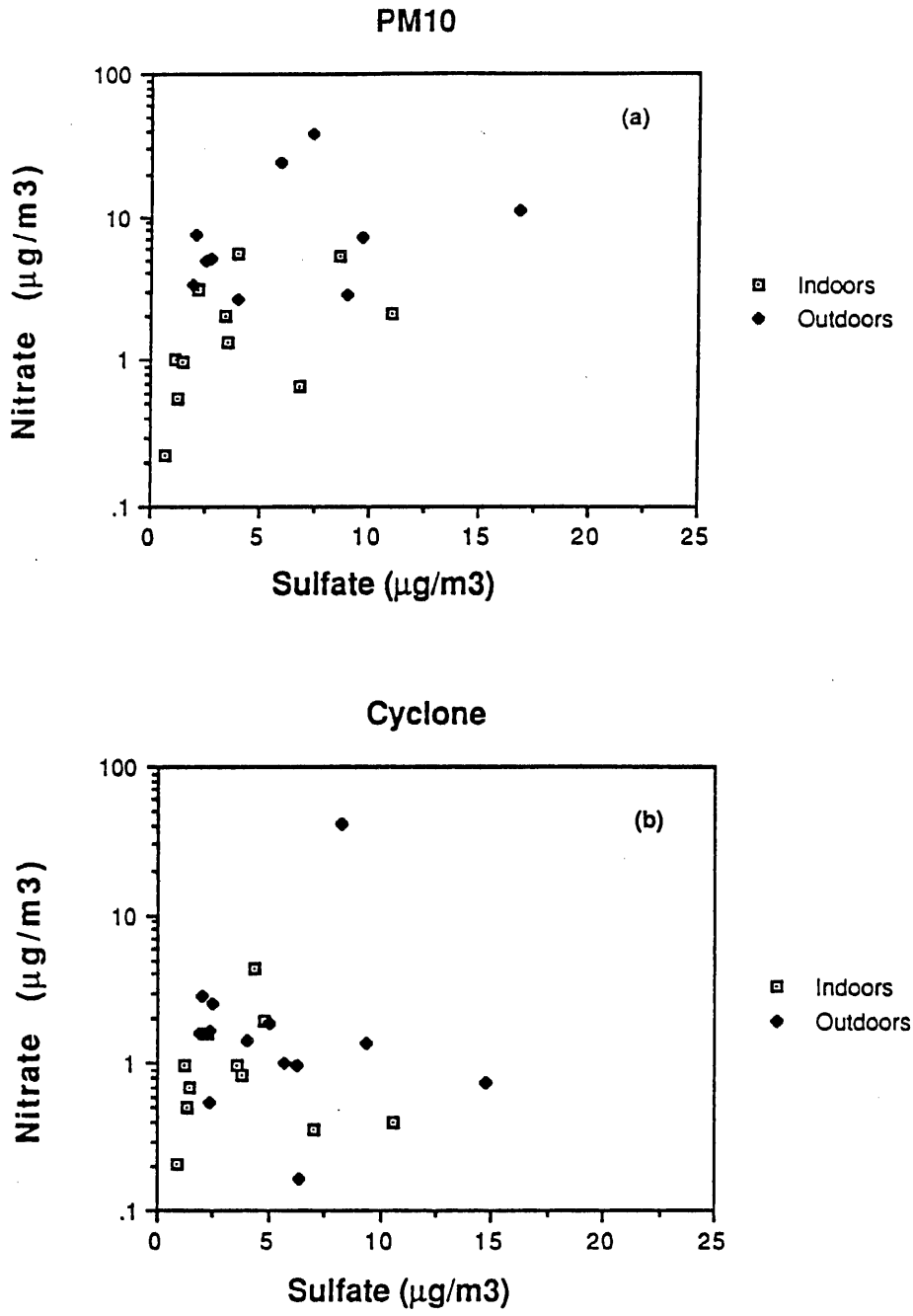


Figure 16. Scatterplots of sulfate versus nitrate concentrations; a) PM10, b) cyclone.

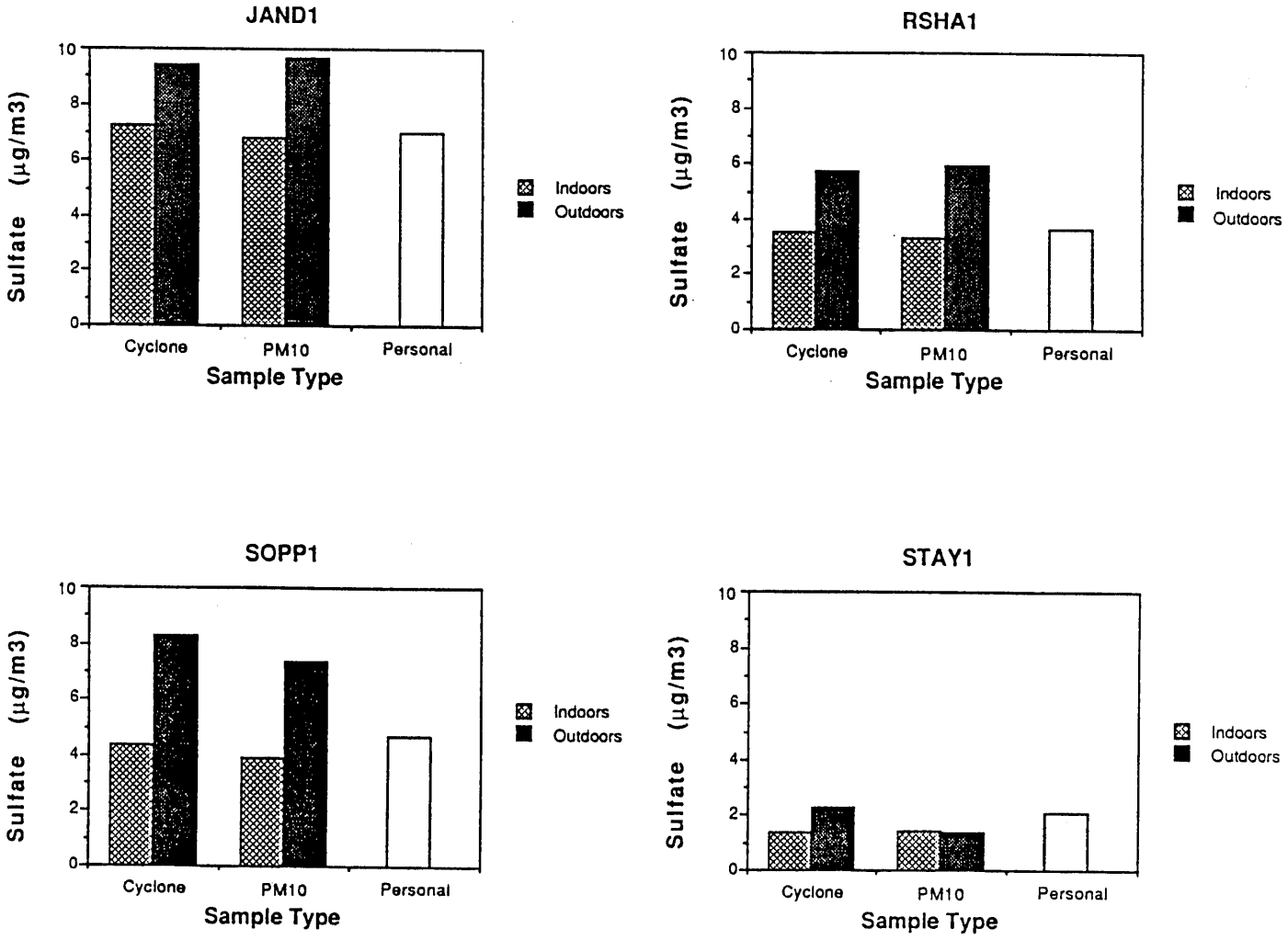


Figure 17. Bar graph of personal cyclone sulfate with indoor and outdoor PM10 and cyclone samples.

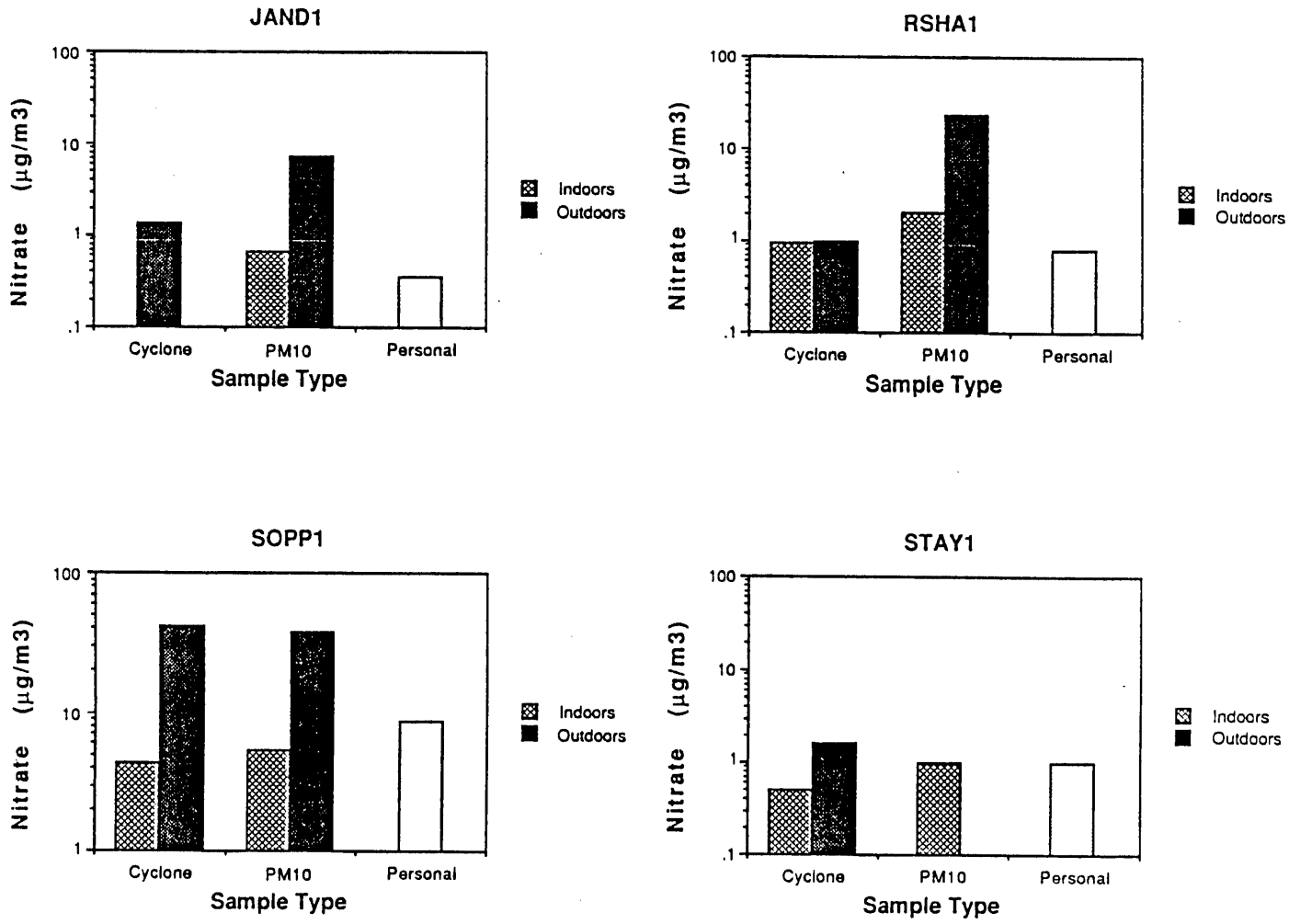


Figure 18. Bar graph of personal cyclone nitrate with indoor and outdoor PM10 and cyclone samples.

C. **Mutagenicity**

Mutagenic activity from the *Salmonella* bacteria is expressed as "revertants" which is directly related to the number of bacteria which have mutated when exposed to compounds which damage their genetic material (DNA). The number of revertants is directly proportional to the dose of the compound or complex mixture being tested. Organic solvents were used to extract the particulate matter captured on the filters from PM10 and cyclone-fitted air samplers; and this extract, which contains a very complex mixture of toxic compounds, was subsequently tested in the bioassay described in the Methods section.

The mutagenic activity of the filter extract is expressed in units of revertants per cubic meter of air sampled. The activity is also expressed per unit of particulate matter collected on the specific filter which was extracted for testing in the bioassay.

1. Summary Statistics

The airborne concentrations of genotoxic agents, based on mutagenic activity (rev/m^3) for cyclone or PM10 samplers is summarized in Table 19. The first observation is that mutagenic activity was detected on all sample filters extracted and tested. The blank filters were generally at or near background activity.

The concentrations were significantly higher outdoors compared to indoors ($p<0.01$ for cyclones; $p<0.01$ for PM10 samplers; Wilcoxon Signed-Rank Test). The average mutagenic activity measured for PM10 samples indoors was 175 revertants/m^3 (rev/m^3), while outdoors, the PM10 associated mutagenicity was 338 rev/m^3. The filters from cyclone samplers had an average indoor mutagenic activity of 178 rev/m^3, while the outdoor average activity was 342 rev/m^3. This relationship holds when the median mutagenic activities are examined. The median mutagenic activity indoors was 163 rev/m^3 (cyclone) and 177 rev/m^3 (PM10), and the median mutagenic activity outdoors was 252 rev/m^3 (cyclone) and 325 rev/m^3 (PM10). The average as well as median activity indoors was very similar between the cyclone and PM10 samplers, whereas in the outdoor activity, the median concentrations from the cyclone appeared slightly lower than samples from PM10 collection. However, this difference was not statistically significant (Mann Whitney U statistic, $p<0.43$).

Table 19. Summary Statistics for Particle Associated Mutagenic Activity. Airborne Concentrations (Rev/m3)

	Mutagenic Activity (Rev/m3) [a]					
	INDOOR			OUTDOOR		
Pre-Selector	Mean (±S.E.)	Median	n	Mean (±S.E.)	Median	n
PM10	175.0 (21.1)	177.5	18	337.7 (49.4)	325.4	17
Cyclone	177.6 (23.4)	163.1	27	341.6 (49.7)	252.4	27

[a] Net TA98 Revertants (+S9) per cubic meter air sampled.

In contrast to the airborne concentrations of particulate matter as well as the airborne mutagenic activity per cubic meter of air, the mutagenic activity calculated on a per mass basis of particulate matter were not statistically different (Wilcoxon Signed-Rank Test) between indoors and outdoors (Table 20). There also were no differences between the cyclone and PM10 samplers. This uniformity of mutagenic activity per unit mass outdoors and indoors can be further evaluated by the correlation of indoor and outdoor specific mutagenic activity based on mass. If the correlation is highly linear, it would suggest that indoor and outdoor particulate matter had similar chemical composition or were derived from similar sources.

2. Mutagenic Activity Indoors and Outdoors

Another specific aim of the project was to examine the mutagenic activity outdoors in parallel with indoor measurements. The comparisons can be done in at least two ways. First, mutagenic activity can be represented as airborne concentrations (revertants/m3), and we could examine the resulting association between indoor and outdoor concentrations. We would examine whether or not indoor concentration of mutagens have systematic relationships with outdoor concentrations. Second, the mutagenic activity per mass of particulate matter (specific mutagenic activity) will indicate an important relationship between outdoor and indoor air. If the correlation between indoor and outdoor activity per mass is randomly scattered, then different sources of mutagens indoors are suspected compared to outdoor sources. If, however, the correlation is statistically significant, then outdoor sources are potentially significant contributors to indoor concentrations of the mutgens.

The results for the correlation of indoor and outdoor mutagenic activity based on airborne concentrations (rev/m^3) is presented in Figure 19. Each data point represents matched arithmetic mean indoor and outdoor airborne mutagenicity from each sampling site for cyclone or PM10 samplers. If duplicate indoor or outdoor samples were not available, the single value for mutagenic activity is used in the matched pair. The correlation based on the Spearman rank between indoor and outdoor mutagenicity (rev/m^3) was $r_S = .65$ (n=21) and was statistically significant ($p<0.001$). The estimated slope for the correlation was 1.5 which indicated that concentrations of mutagenic activity outdoors were approximately 50% higher than concentrations indoors. The concentrations indoors appear to be related linearly with the concentrations outdoors,

Table 20. Summary Statistics for Particle Associated Mutagenic Activity. Specific Activity per µg of Particulate Matter.

	Mutagenic Activity (Rev/µg) [a]							
	INDOOR				OUTDOOR			
Pre-Selector	Mean	(±S.E.)	Median	n	Mean	(±S.E.)	Median	n
PM10	4.8	(0.6)	5.5	18	6.1	(0.9)	6.9	17
Cyclone	5.9	(0.7)	5.1	27	7.2	(1.2)	5.6	26

[a] Net TA98 Revertants (+S9) per µg particulate matter sampled.

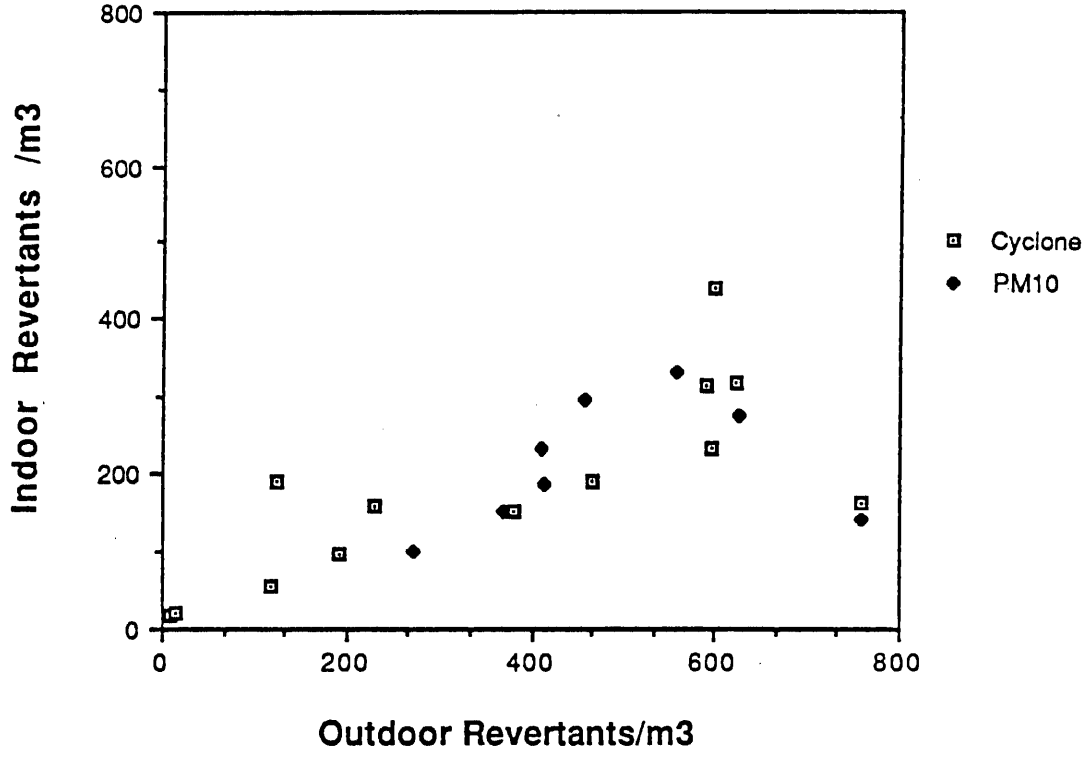


Figure 19. Scatterplot of indoor versus outdoor mutagenic activity per cubic meter of air.

which suggests that the mutagens located outdoors could find their way indoors. However, caution should be taken in reporting this increase based on slope since outliers in the data could highly influence the slope. A further test of the hypothesis that outdoor particles were migrating into the homes was to examine the correlation of mutagenic activity per unit (μg) of particulate mass.

The correlation of mutagenic activity based on revertants per unit particulate mass (rev/μg) are presented for matched pairs of samplers indoors and outdoors in Figure 20. Each data point represents matched arithmetic mean indoor and outdoor airborne mutagencity from each sampling site for cyclone or PM10 samplers. The correlation between indoor and outdoor values is highly significant ($p<0.001$) with a Spearman rank coefficient of $rs = 0.85$ ($n=22$). The standard Pearson correlation coefficient was $r=0.89$. The slope of the best fit line was 1.36, indicating that mutagenicity associated per mass of particles outdoors had slightly higher activity per mass of particles than the mutagenicity found on particles indoors. This is consistent with the hypothesis that the particulate matter outdoors migrates indoors with its associated mutagens.

3. Airborne Particulate Matter Mass and Mutagenicity

Results of the correlation between particle mass concentration (PM10) and airborne mutagenicity are illustrated in Figure 21. The correlation for the cyclone samplers is illustrated in Figure 21b; and the correlation for the PM10 samplers is illustrated in Figure 21a. Both sets of graphs represent indoor and outdoor samples. The relationship for the cyclone samplers is significantly correlated with airborne particle concentrations ($p<0.001$; $n=54$) with a Spearman rank $r_S=0.51$ (Figure 21b). However, the particles collected with PM10 Samplers were not significantly correlated (Spearman rank $r_S =0.212$; $p<0.10$, $n=35$). The range of mutagenic activity was large at any particle concentration for both types of samplers; and based on Figure 21, the concentrations of particles and mutagenic activity indoors was observed to be lower than the concentrations outdoors.

4. Mutagenic Activity of PM10 and Cyclone Derived Samples

An initial aim of this pilot project was to evaluate the correlation of mutagenic activity from available cyclone samplers compared to PM10 samplers. Data from all houses sampled for mutagens are summarized in Table 21. The data represent the

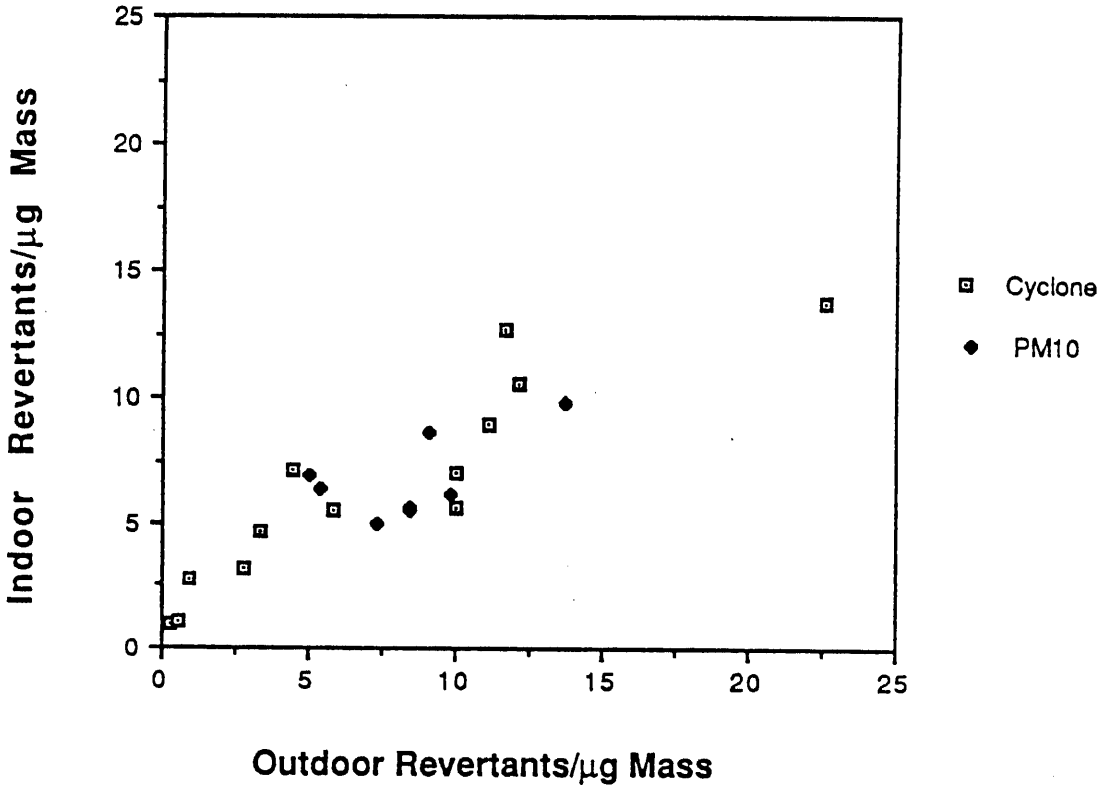


Figure 20. Scatterplot of indoor versus outdoor mutagenic activity per microgram of collected particulate mass.

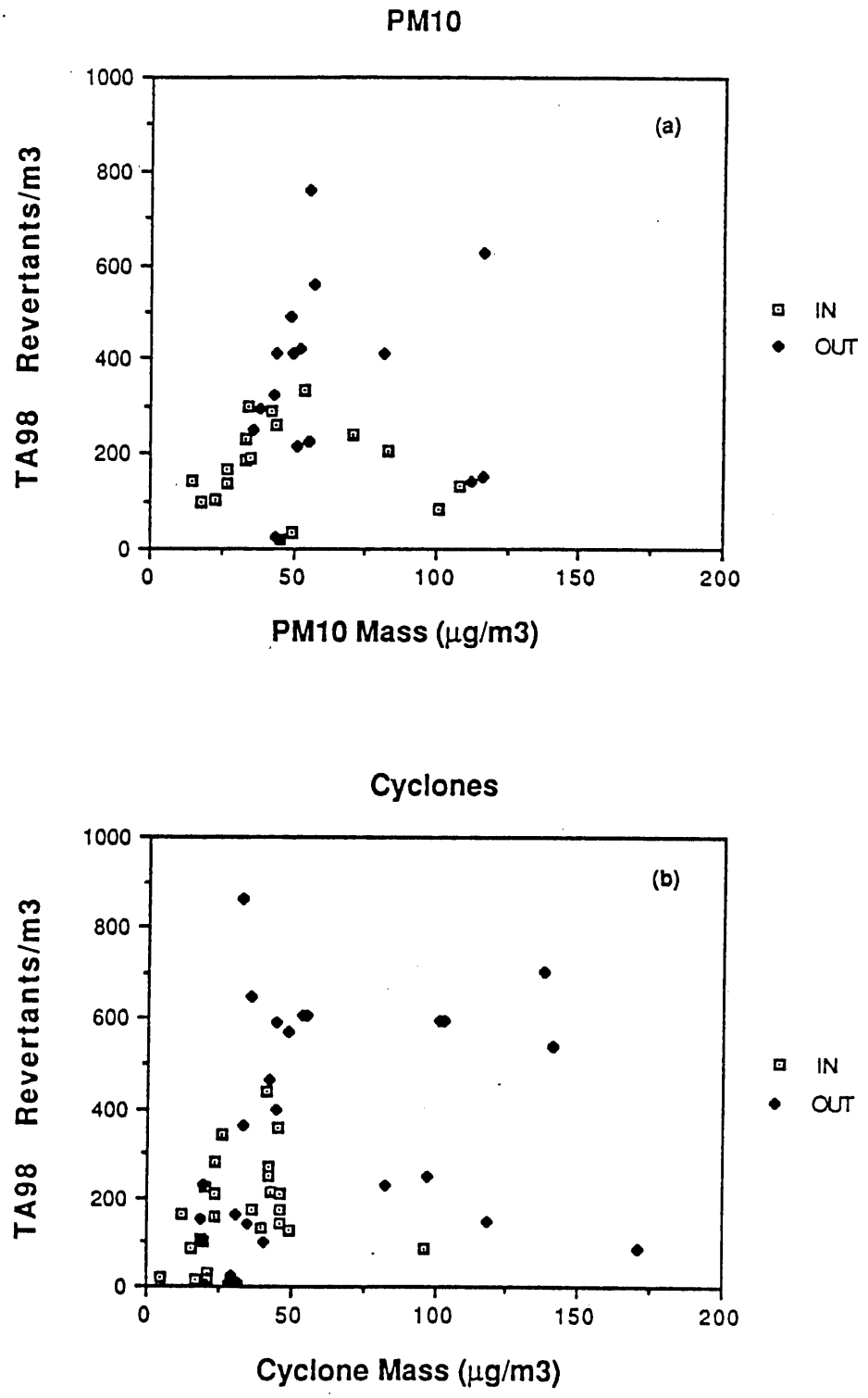


Figure 21. Scatterplots of particulate matter mass versus mutagenic activity; a) PM10, b) cyclone.

Table 21. Regression Analysis of PM10 and Cyclone Mutagenicity (Cyclone Values Used to Predict PM10 Activity).[a]

DF[b]	R^2	Slope	S.E. Slope[c]	Intercept	p
21	0.90	0.81	0.06	45.71	<0.001

[a] Indoor and outdoor observations combined.

[b] Degrees of freedom (1 minus the number of indoor/outdoor pairs).

[c] Standard error of the slope

mutagenicity at a subject's house for the matched cyclone and PM10 pair (located indoors or outdoors). Based on regression analysis, mutagenicity from cyclones is highly correlated with mutagenicity from PM10 samplers (r^2=0.90; p<0.001). Ninety percent of the variation of the PM10-associated mutagenicity is explained by the mutagenicity associated with cyclone samples. The slope of the best fit line is 0.81, which indicates that based on revertants per cubic meter of air, the filter with the cyclone sampler has approximately 20% higher mutagenic activity. The matched samples from indoors generally had mutagenic activity in the lower portion of the regression line (around the range of 0 to 400 rev/m^3), while the matched samples from outdoors were in the upper portion of the line (400 to 800 rev/m^3).

5. Mutagenicity from Personal Sampling

The results of the personal sampling for PM10 associated mutagenicity are illustrated in Figure 22. The personal sampling apparatus was carried by three of the eight asthmatic subjects during Phase II of the project. One of the four initially scheduled to wear a pump did not carry the portable air sampling apparatus. Three subjects are represented by three separate bar graphs, where concurrent cyclone and PM10 sample measurements are presented as mean values of at least two samples. The indoor and outdoor measurements are also presented. The mutagenicity for the personal sample is from a single sample. A consistent pattern seen in Figure 22 is that the personal samples have very similar mutagenic activity as the filter sample taken indoors (from either cyclone or PM10 sampler). This is consistent with the results of PM10 mass as well as ion determinations. The personal sample from the subject from home 7 appears to be similar to the indoor cyclone samples and the outdoor PM10 samples. The results from these personal samplers indicate that the individuals studied probably stayed indoors during this samplers and probably stayed in an indoor environment similar to where the "fixed-site" samplers were located.

C. **Airborne Nicotine**

Airborne concentrations of nicotine as a marker for environmental tobacco smoke (ETS) exposures are summarized in Table 22. All indoor and outdoor measurements from the two experimental houses in Phase II were below or very near the limit of detection. Blanks indicate samples below the limit of detection. The positive controls from the office of a smoker carried detectable amounts of nicotine of 0.689 μg/m^3; the

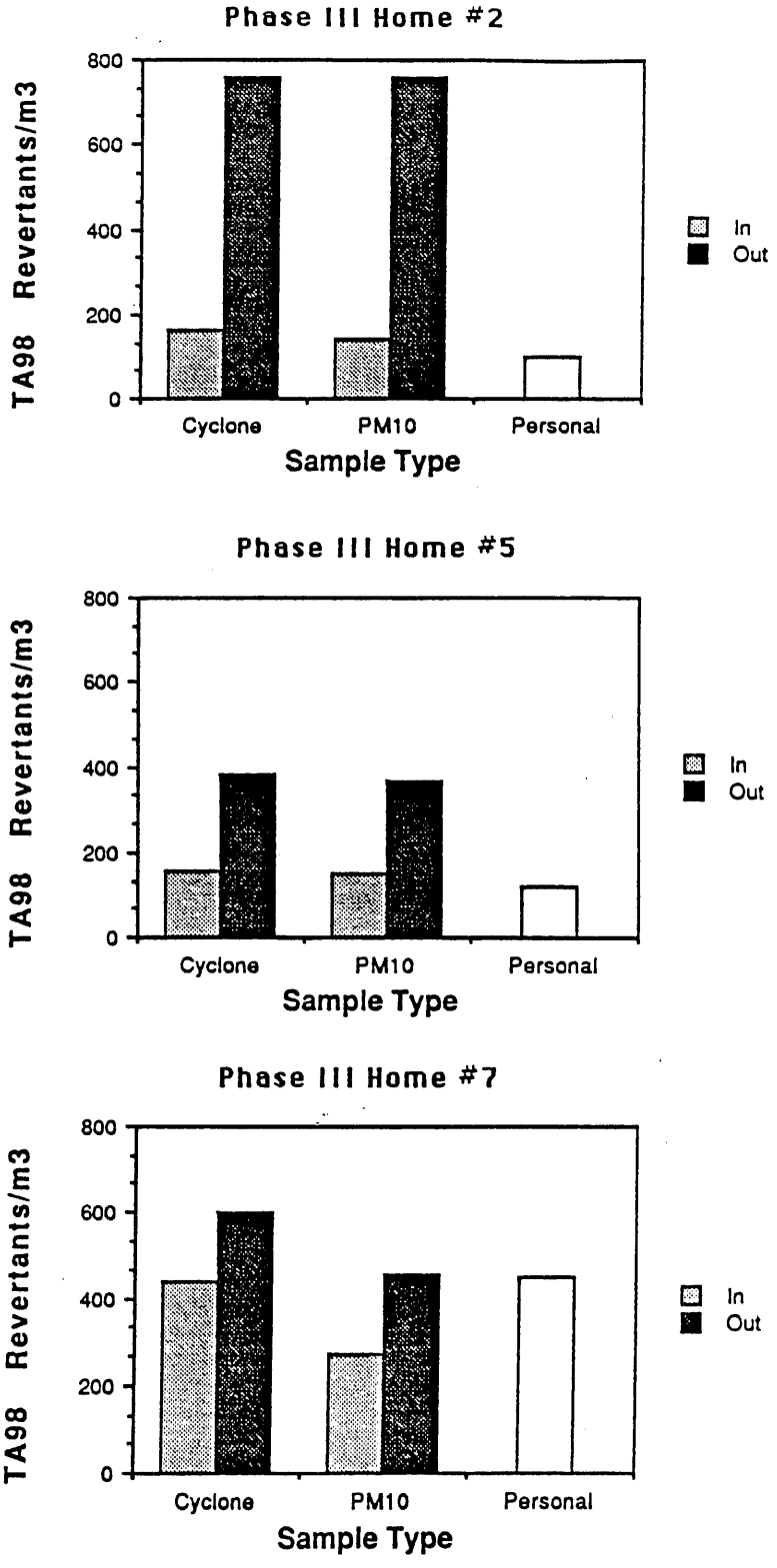


Figure 22. Bar graph of personal cyclone mutagenicity with indoor and outdoor PM10 and cyclone samples.

Table 22. Airborne Nicotine Concentrations

Location	Indoor/Outdoor	Airborne Nicotine (µg/m3)
SCOL3 [a]	Indoor	LD-0.007 [b]
SCOL3	Outdoor	LD-0.007
MDEN 2 [a]	Indoor	LD
MDEN 2	Outdoor	LD
Smoker's Office	Indoor	0.70
Non-smoker Office	Indoor	0.02-0.03
Casino (Lake Tahoe)		8.02 [c]

[a] Phase II-Pre-Pilot Homes.
[b] LD = Limit of Detection (<0.01 ug quantitated by GC)
[c] Values represent actual range of concentrations
Median value of six samplers.

concentration was the same for both filters in the duplicate pair. Samples taken from the main office located approximately 20 feet from the smoker's office also had detectable amounts of nicotine (0.031 and 0.024 $\mu g/m^3$).

D. **Aeroallergens**

From the microscopic analysis of aeroallergen samples the two major classes of materials found were pollens and molds. Consistent with the time of year in which the sampling was conducted, pollens were from trees and were inclusively comprised of Eucalyptus, Elm, Alder, Cedar, Pine, Ash and Cenopod pollens. Of the fourteen indoor pollen samples taken, only one had a minimally detectable concentration of pollen. Pollens were detected in five of the fourteen outdoor samples. Because of the low pollen counts, it was not possible to compute indoor/outdoor pollen ratios.

Mold spores were detected and speciated in all 28 samples. There was no consistent pattern among mold species from house-to-house or between the inside and outside of the home. Therefore, mold counts were totaled for each sample and descriptively presented here. The average indoor/outdoor ratio was 0.94 (s.d.= 1.2). The distribution of I/O ratios was skewed to the right and the median ratio of 0.69 is more reflective of the central tendency. The distribution of I/O ratios is displayed in Figure 23.

For three sampling data points the I/O ratio exceeded 1.0. Two of those points represent one home on two different days, including one high I/O ratio of 4.7. It is likely that this home has an indoor source of molds as has been seen previously in Southern California by Kozak (Kozak et al., 1980a).

We have shown that it is feasible to use a portable volumetric sampler to collect samples of aeroallergens. We have also demonstrated that it is feasible to reliably determine on a spot basis the relationship between indoor and outdoor concentrations of aeroallergens. These data could be collected in conjunction with an epidemiological study to determine the possibility that aeroallergens are confounding the results of air pollution studies in atopic individuals. This is especially important since certain homes appear to have an indoor source for allergenic mold spores.

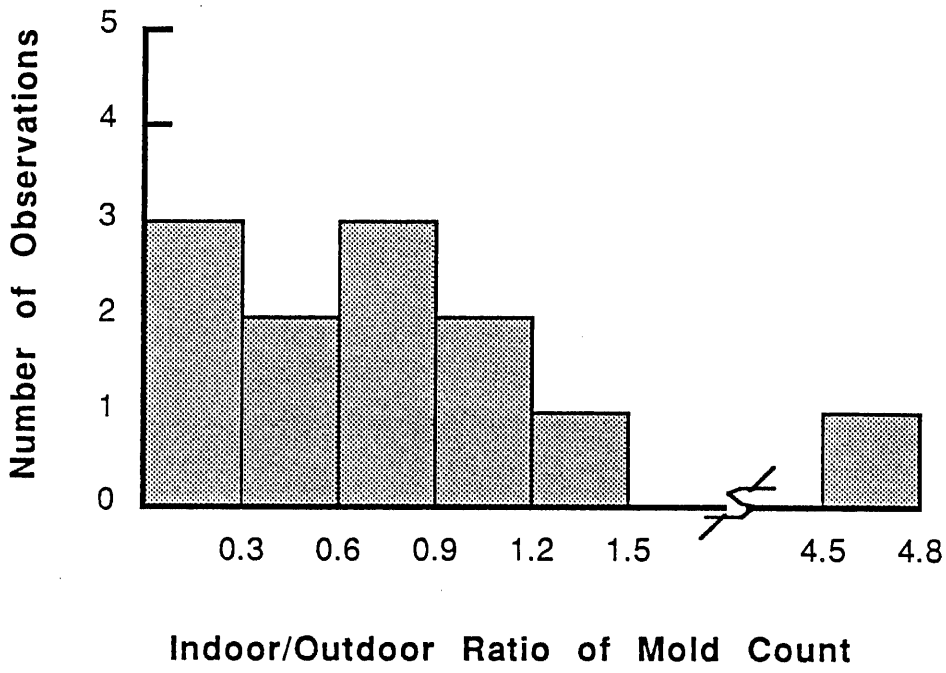


Figure 23. Indoor/Outdoor ratio of mold count from pre-pilot and pilot homes.

E. Time, Location and Health Diaries

Subjects in Phase III in this study completed two data sheets in order to evaluate the possibility of coupling personal or residential air pollution monitoring with assessment of health outcomes. The first data form was a location diary that indicated time spent in certain key microenvironments (Appendix 1). The second form is a daily diary of symptoms associated with asthmatic attack (Appendix 2).

The time-location diary was designed by our group for assessing microenvironmental contributions to personal exposure from nitrogen dioxide. The diary is a simple checklist in closed form which requires only minimal effort from the volunteer. We have had extensive experience with this form, having administered it to over 700 individuals. With careful instruction of volunteers, reliable data can be obtained. It is necessary that the field technician review the form with the subject and demonstrate how to complete the entries. We have found it helpful to use a practice sheet and have the subject recall his or her activities of the previous day with the field technician. With this training, the subject can more easily maintain a concurrent diary on the assigned sampling day.

Since only eight asthmatic subjects participated in this pilot study, the sample is too small to analyze or interpret the time-location diaries. In general, the subjects were able to maintain good diaries. We did not stress this portion of the study to our technicians or subjects, and it is our impression that the diaries were not as carefully completed as in a previous study in which we stressed the central importance of maintaining good diaries.

It is clearly feasible to incorporate time, location and activity diaries with air pollution exposure and health studies. In going from a pilot study to a major exposure study, it would be necessary to design the diary to meet the specific objectives of the research. In the case of a PM10 study, it would be necessary to account for exposure to particle-producing sources such as cigarette smoke and wood fires. Activity levels would be accounted for in order to relate exposure concentrations to dose of air pollution inhaled by the subject.

The health and pulmonary function diaries were similarly used to evaluate the feasibility of assessing health outcomes in the context of a personal or residential exposure study. The diaries, described and presented in the methods section, are intended for use in a study with a time series design. That is, they are designed to follow the

changes in health status for an individual as environmental conditions change over time. These asthmatic subjects were participating in a long-term study of the effect of acid-forming particles on pulmonary function and health symptoms. Using the diary format, the health effects of environmental factors can only be distinguished after many days of changing conditions.

Since we only have one or two days of observation per subject, it is not possible in the present pilot study to investigate the role of residential or personal PM10 exposure in provoking an asthmatic attack or reducing pulmonary function. The diaries also cannot be used to investigate the differences among individuals, known as a cross-sectional study, when the sample of persons is small.

Therefore, we have been able to use the diary forms to verify the feasibility of incorporating self-administered components into exposure and health studies. It is not possible to stress too strongly the importance of careful instructions to subject on use of any diary forms. It is also necessary to carefully follow the progress of subjects completing diaries to insure that they continue to maintain good records. Our experience is that direct contact must be maintained on at least a weekly to biweekly basis.

F. Quality Assurance

Since this was a pilot study, considerable effort was devoted to quality control and quality assurance activities. Phase I demonstrated that mass, sulfate, nitrate, and mutagenicity samples and analyses were reproducible and precise in the laboratory and in a trial field measurement. During Phase I we also demonstrated that filter blanks were low for all parameters measured.

In the Phase II and III efforts, in addition to the quality assurance procedures incorporated into the protocols, a large number of paired samples were routinely taken to evaluate quality control. The paired samples were separated in the field and analyzed blindly by the laboratory technicians. The results of these paired analyses are presented in this section.

Table 23 presents the mean differences among paired samples for mass, sulfate, and nitrate, and the standard deviation of the mean differences. In all cases, the abso

Table 23. Precision of Paired Mass, Ions and Mutagen Samples.

Pre-Selector	Paired Sample	Mean Difference[a]	Mean Diff[b] Std.Dev	N Pairs
PM10	Mass[c]	4.09	5.32	12
	Nitrate	0.49	0.83	11
	Sulfate	0.24	0.40	12
	Mut[d]	29.82	36.33	13
Cyclone	Mass	7.77	9.77	25
	Nitrate	0.25	0.43	23
	Sulfate	0.45	0.76	25
	Mut	49.16	67.26	26

a Calculated as the average of the absolute value of the paired concentration differences.

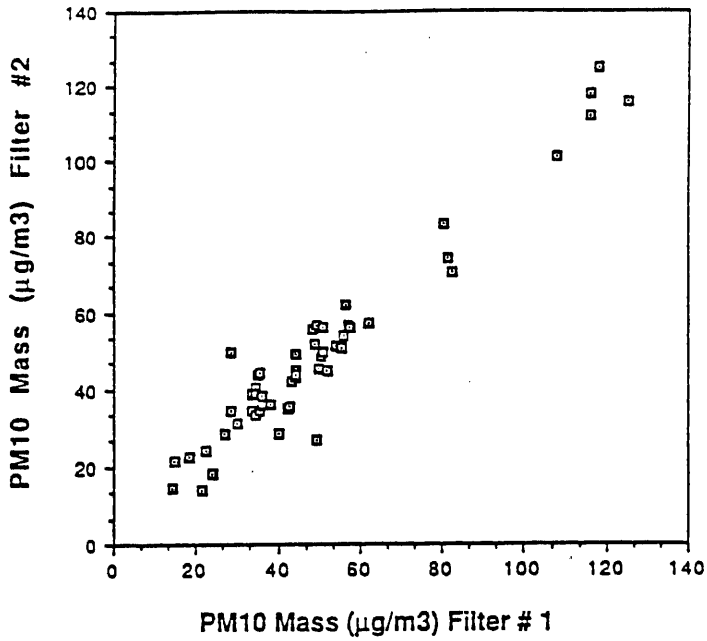
b. Calculated as the standard deviation of the actual paired concentration differences.

c Mass results are from PM10 and cyclone samples assigned to ionic speciation

d Mut = mutagenicity values expressed as TA98 revertants/m3.

lute value of the paired differences is quite low and is less than the standard deviation of the paired differences. This indicates that the paired samples give similar values.

The high quality of the paired data are represented graphically in Figures 24 through 27. From visual inspection, the precision of the entire protocol, from sample preparation through field sampling and chemical analysis, is readily apparent. The coefficients of variation for the paired samples ranges from a low of 73% for cyclone mass to nearly 100% for PM10 sulfate and nitrate. In general, the cyclone results are more variable. This is at least partially due to the lower mass collected by the cyclone sampler and may also be due to the configuration of the sampling head. It is also notable that duplicate mass measurements are more variable than duplicate nitrate or sulfate determinations.



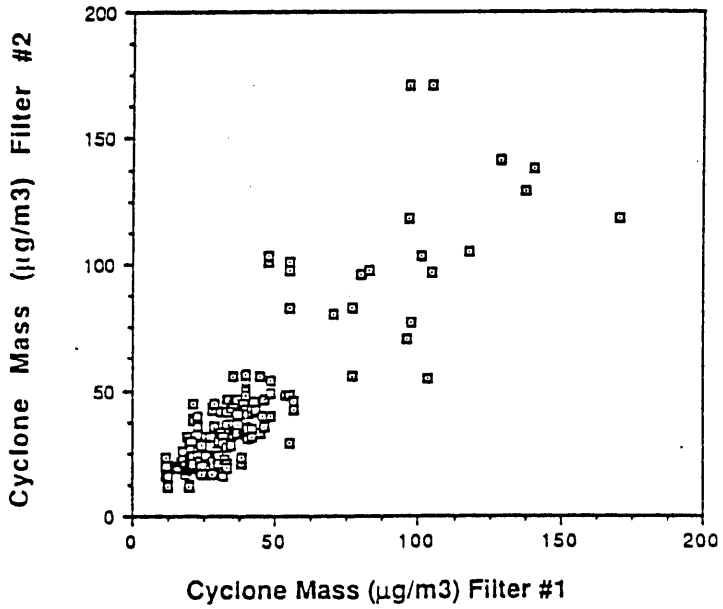


Figure 24. Scatterplot of duplicate mass filters for combined indoor and outdoor paired samples; a) PM10 pairs; $R^2 = 0.93$, and b) cyclone pairs; $R^2 = 0.73$.

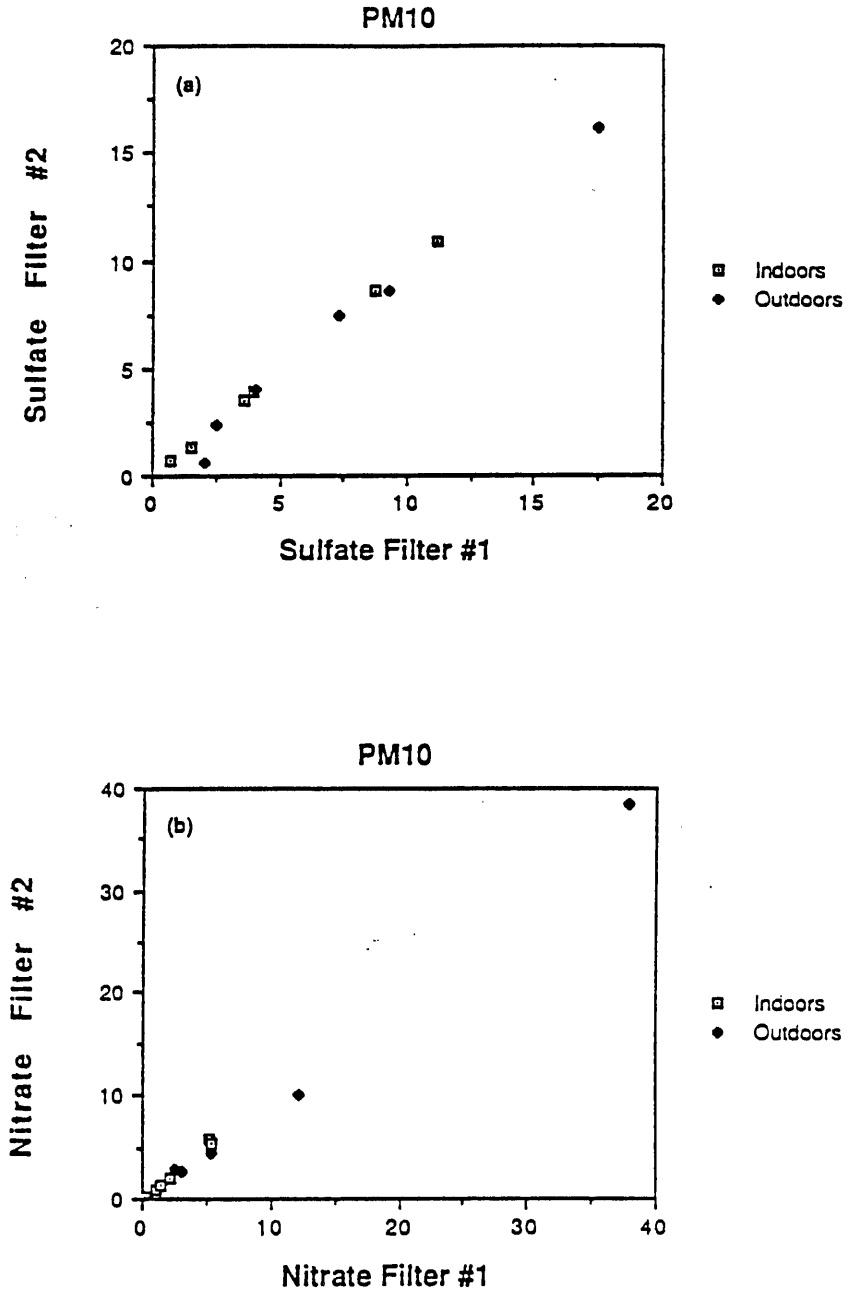


Figure 25. Scatterplot of duplicate PM10 filters for combined indoor and outdoor paired samples, units in $\mu g/m^3$; a) sulfate pairs; $R^2 = 0.97$, and b) nitrate pairs; $R^2 = 0.84$.

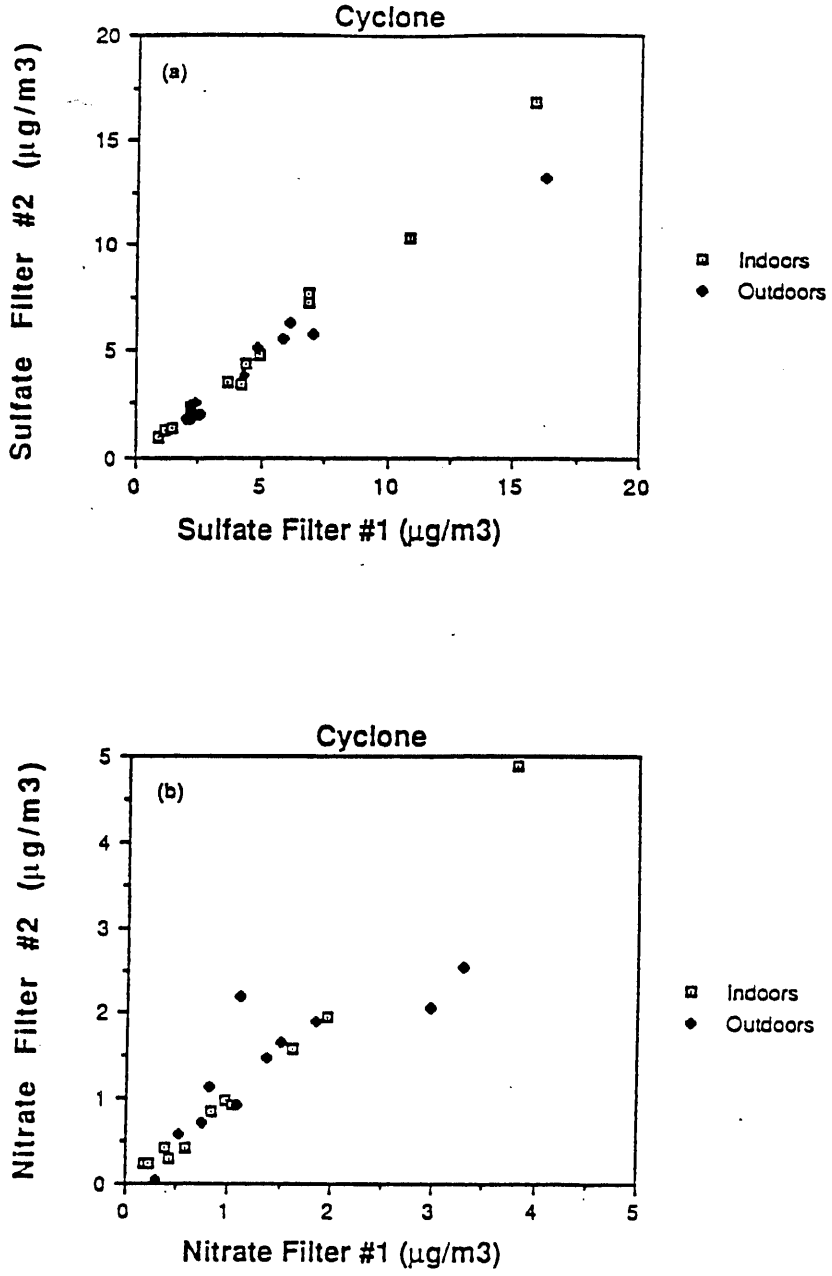


Figure 26. Scatterplot of duplicate cyclone filters for combined indoor and outdoor paired samples; a) sulfate pairs; $R^2 = 0.97$, and b) nitrate pairs; $R^2 = 0.84$.

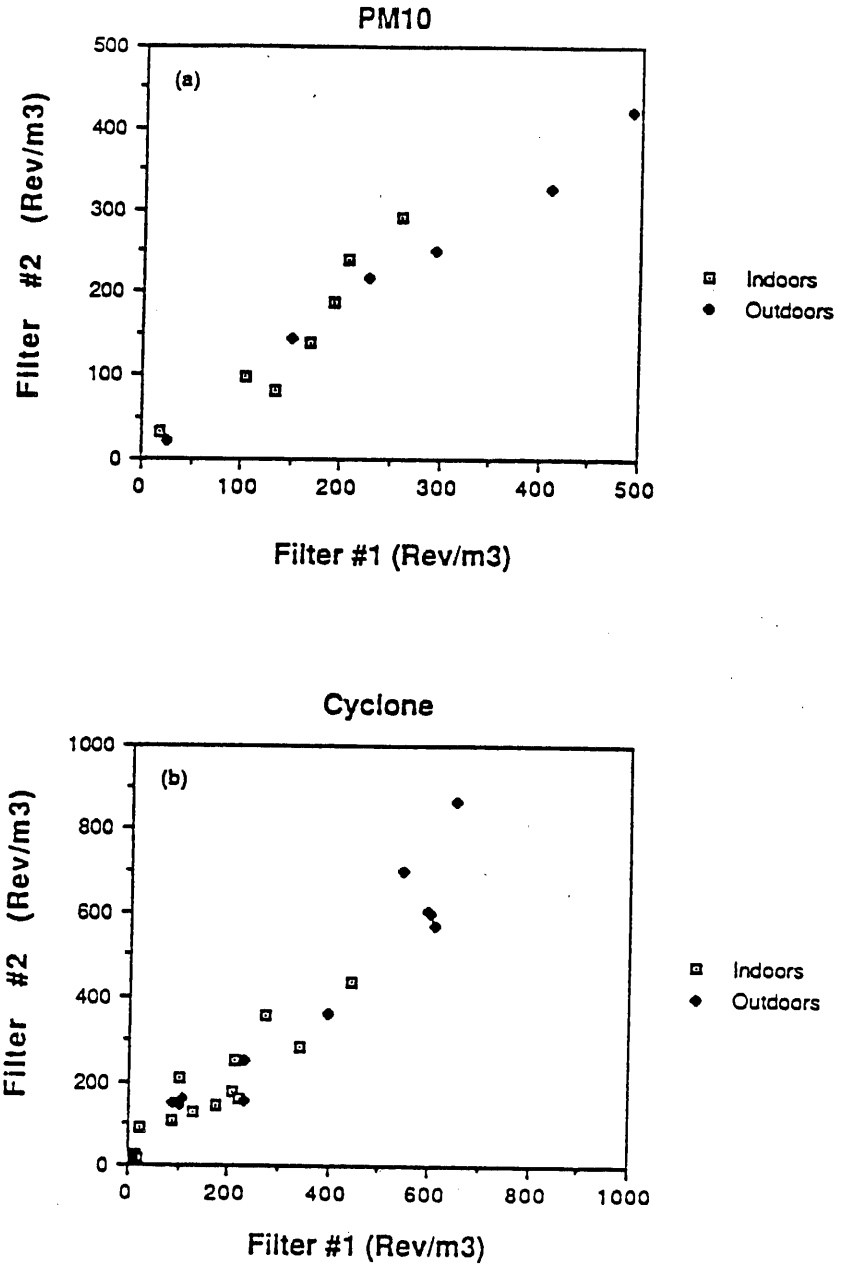


Figure 27. Scatterplot of duplicate mutagenicity for combined indoor and outdoor paired samples; a) PM10 pairs; $R^2 = 0.939$, and b) cyclone pairs; $R^2 = 0.917$.

V. DISCUSSION AND CONCLUSIONS

This pilot study successfully demonstrated the feasibility of monitoring the mass of particles equal to or less than 10 micrometers diameter (PM10) inside and outside of residential settings. We have characterized the ionic species and mutagenic activity from PM10 particles and have found that it is possible to monitor daily personal exposure of volunteers to respirable particulate matter by using portable air samplers.

Further, this study showed that mass collected with portable sampling devices correlated well with PM10 mass. Since results from the field sampling and laboratory analyses were more reliable than anticipated, we were able to observe a number of strong patterns and relationships in this sample of ten homes, including eight homes with asthmatics. The patterns and relationships form the basis of several important findings that go beyond the original objectives of this pilot study. While certain patterns are clear in this sample, there are too few homes in the pilot study to generalize the findings to other California residences. Mass, as determined from samplers with a sampler designed to collect particles less than 10 micrometers in diameter (PM10), was reproducibly measured both indoors and outdoors. Mass was also reliably measured using a personal sampler designed to collect particles less than 7 micrometers in diameter. The approaches taken in this study could be extended to a larger sample of residences in order to construct an exposure model and conduct a risk assessment for PM10. Further, this sample may not represent other California residents because asthmatics take precautionary measures to reduce exposures to potential triggers of asthma attacks (no pets, no smoking, special cleaning, etc.).

Airborne concentrations of mass collected using a cyclone sampler, designed to mimic the collection characteristics of the upper respiratory system in humans, was found to correlate well with mass collected with PM10 samplers. This relationship held up both inside and outside of homes. As expected, concentrations of PM10 mass were slightly greater than cyclone mass since the PM10 sampler is designed to collect slightly larger particles than those collected by the cyclone sampler. Specifically, the largest particle collected by the PM10 sampler is 10 micrometers while the largest particle collected by the cyclone is approximately 7 micrometers (with very close to 0% efficiency of collection at this particle size; about 50% efficiency at 5μm diameter).

In the sample of homes with asthmatics, mass concentration was consistently lower inside the homes than immediately outside. This result was observed for both PM10 and cyclone samples, but was more pronounced with the PM10 samples. It is likely that indoor PM10 samples were reduced relative to outdoor samples due to lower penetration efficiency for larger ambient particles and settling of larger particles inside the home. It is important to note that all homes participating in this study were occupied by nonsmokers. Cigarette smoke is a known source of fine particles smaller than PM10. Wood stoves, fireplaces and unvented kerosene heaters (which are all potential indoor sources of particles) were not used by the subjects during this pilot study.

Even though concentrations of indoor mass were lower than concentrations of outdoor mass, the indoor concentration was moderately correlated with the concentration measured outside the home. This finding indicates that variation of indoor concentrations in this sample of homes was driven by the variation in ambient concentration. Therefore, in this sample of nonsmoker homes, it appears that outdoor particles determined indoor exposure. Furthermore, some protection from higher outdoor concentrations is afforded by shelter if smokers and other particulate sources are not present. This observation was confirmed with the personal samplers worn by the asthma volunteers.

Sulfate concentrations inside all ten homes were strongly correlated with sulfate concentrations measured outside the home. The homes provided minor protection from outdoor concentrations since indoor levels were slightly lower than outdoor levels of sulfate. Concentrations and indoor-to-outdoor correlations were similar for PM10 and cyclone samplers, indicating that most of the sulfate was found on smaller particles. There was no measurable evidence in the homes of major sources of indoor sulfate.

Nitrate concentrations inside all ten pilot study homes were only weakly correlated with outdoor nitrate concentrations. For PM10 samplers, indoor nitrate concentrations were lower than simultaneously measured outdoor concentrations; however, for cyclone samplers, indoor and outdoor concentrations were comparable. Since the two samplers have different size cutoffs (PM10 sampler collects larger particles than are collected by cyclone samplers), this finding is consistent with a nitrate aerosol size distribution that is shifted toward the upper PM10 size range. It is also possible that there were minor indoor sources for fine nitrate particles.

For sulfate, indoor and outdoor measurements using the PM10 samplers had values similar to matched indoor and outdoor measurements using cyclone samplers. This finding gave further evidence that the majority of sulfate mass was found in the fine particle fraction.

In contrast, for nitrate the indoor and outdoor measurements obtained with the PM10 samplers were higher than those obtained with cyclone samplers operated in parallel. This finding adds to the evidence that a substantial fraction of nitrate is found in the larger size region not collected by the cyclone samplers.

For the houses measured in the pilot study, the airborne concentration of mutagens associated with collected particulate matter was measureable using the microsuspension assay. Mutagenic activity was statistically higher outdoors compared to indoors and within the indoor or outdoor environment, there were no differences in the mutagencity (rev/m^3) associated with the particles collected by cyclone or PM10 samplers. This is consistent with reports that mutagens associated with airborne particulate matter are found primarily on particles less than 1 μm in aerodynamic diameter (Talcott and Harger, 1980, Sorenson et al, 1982).

The results of indoor and outdoor mutagenic activity based on a per mass of particulate matter (rev/μg particulate matter) were not statistically different. These results suggest that the mutagens associated with particles indoors are similar to the mutagens associated with particles found outdoors.

There have been a few reports measuring indoor and outdoor mutagenicity simultaneously and over at least a 12-24 hr period. For example, preliminary results of diurnal measurements of indoor and outdoor mutagenicity have been investigated in an office environment where there were no smokers present (Wesolowski et al, 1986, extended abstract). The investigators collected parallel indoor and outdoor fine particles (<2.5μm) at 8 hour intervals at a flow rate of 50 liters/min. The filters were extracted and tested in the microsuspension assay used in the current work reported here. The results showed that indoor levels followed the diurnal pattern of the outdoor mutagenic activity. The mutagenic activity indoors was about an average of 70% that of the outdoor activity.

There was a statistically significant correlation between airborne concentrations of mutagens (based on rev/m^3 indoors and outdoors. The Spearman rank correlation coefficient was 0.65 (n=21) and the slope of the best-fit line (based on linear regression) was 1.5 (outdoor:indoor). However, the mutagencity based on a per cubic meter of air could be expected to be different indoors compared to outdoors due to possible differences in indoor sources as well as filtration of particles moving into the indoor environment. Further, since there were different individual homes measured on different days and locations, a relationship of mutagenicity based on airborne concentrations indoors and outdoors would be at first, expected not to correlate well unless there was a constant percentage of outdoor particles which move indoors at all locations. All sites except one appeared to correlate well for both PM10 and cyclone samplers, suggesting that a constant percentage of outdoor air moved indoors at the homes tested. Also, since there were no significant statistical differences for the PM10 and cyclone collected particles with respect to mutagenicity per cubic meter of air, the mutagenicity appeared to be associated with smaller (<1μm) particles collected on the filters with cyclone samplers.

Although the correlation of the particulate mass and mutagenicity concentrations for the cyclone samplers was significant, the scatter of the data at a given mass (for example 50 $\mu g/m^3$), indicates that there are indeed divergent biological activities. The mass of particles from the PM10 samplers did not significantly correlate with the concentration of mutagenic activity from those particles and here too, there was a great divergence of mutagenic activity at a specific particulate mass. We conclude that particles can have the same mass but can have completely different mutagenic activities.

The mutagenic activity measured from personal sampling demonstrated to us that personal sampling for mutagens is possible. Also, when the mutagenic activities for the three individuals were analyzed and compared to concurrent measurements from cyclone and PM10 samplers, the personal samples closely reflected the mutagenic activity from the indoor mutagenicity values. Since the individuals measured are asthmatics, their personal preference would probably be to favor indoors compared to outdoors and this was reflected in the results.

The airborne nicotine measurements showed us that of the two houses measured, there was little environmental tobacco smoke indoors and outdoors. Therefore, the mutagenicity measured was not due to ETS exposure, but most probably to other

sources. The mutagenicity of airborne particulate matter has been demonstrated by a number of investigators and specific toxic compounds have been identified from extracts of the particles (Pitts et al, 1983, Ramdahl et al, 1986; Atkinson et al, 1988). Further, the main source of atmospheric particulate matter of the size where most of the mutagenic activity is found is thought to come from vehicular sources (Talcott and Wei, 1977; Pitts et al, 1983; Flessel et al, 1984; Kado et al, 1986).

Aeroallergens were measured inside and outside of all homes. Concentrations of pollens and molds were higher outdoors than inside. This project has demonstrated that it is feasible to monitor for indoor or outdoor aerollergens in larger-scale field studies. These measurements are important control variables for investigating the effect of air pollutants on allergic asthmatics.

A limited number of size-specific cascade sampler samples, utilized in Phase II, indicated that most of the sulfate mass was found on fine particles while nitrate mass was observed over a wider size range. This finding supports the observation that nitrate collected with cyclone samplers is lower than nitrate collected with PM10 samplers. Most of the sulfate was contained on those cascade stages which collect particles less than 1 micrometer in diameter.

We also investigated the relationships among measurements made on each sample. Sulfate and nitrate ions correlated well with mass concentration. The correlation between nitrates and mass was greater than between sulfates and mass. These relationships were observed with PM10 and cyclone samplers and for indoor and outdoor samples. This finding indicates that the factors influencing variation in PM10 mass also influence variation in concentration of sulfate and nitrate ions.

In contrast, PM10 or cyclone mass correlated only weakly with mutagenic activity. This suggests that factors affecting variation in the specific mutagenic activity of particles are different from factors influencing variation in PM10 mass.

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VII. APPENDICES

APPENDIX 1

Time-Location Diary

START DATE: _____ TIME: _____ AM PM END DATE: _____ TIME: _____ AM PM

Circle A.M. or P.M.	TIME		HOME				TRAVEL			WORK/SCHOOL		ALL OTHER LOCATIONS		ADDITIONAL COMMENTS
			IN KITCHEN WITH STOVE		IN ANY OTHER ROOM	OUT-DOORS/ YARD	NEAR MAJOR ROADS		BY WHAT MEANS?	IN-DOORS	OUT-DOORS	IN-DOORS	OUT-DOORS	
	BEGIN	END	ON	OFF			YES	NO						
AM PM	:	:												
AM PM	:	:												
AM PM	:	:												
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REMINDER. Please be sure there is only one check mark (✓) for each line

APPENDIX 2

Health Symptom Diary

DAILY DIARY

Monday's Date _____
Day Month Year

Using the symptom codes from the next page, in the past twenty-four hours, have you had	MON	TUES	WED	THUR	FRI	SAT	SUN
Red, itchy, or watery eyes?							
A stuffy or runny nose?							
A sore throat?							
Cough?							
Wheezing or whistling in the chest?							
An attack of shortness of breath with wheezing or asthma?							
A chest cold?							
Fever or a feverish feeling?							
A headache?							
Unusual fatigue or tiredness?							
A general achy feeling?							
CHECK HERE IF YOU HAD NONE OF THE ABOVE SYMPTOMS							

Put a check in the box IF in the past twenty-four hours:

You were unable to do your usual activities							
Your symptoms caused you to start or change current medications*							
Your symptoms caused you to contact a doctor**							
You spent most of the day outside							
You had wheezing, chest tightness or shortness of breath last night							
You had a cough last night							

Record highest of two efforts at each designated time period:

Peak flow (on rising in the morning)							
Peak flow (before dinner)							
Peak flow (before retiring in the evening)							

*If you changed your medications or started new medications, please make a notation on the next page

**What did the doctor say was your problem? _____

Symptom Codes

0 = None

1 = Trivial or doubtful

2 = Mild, clearly present, but causing little or no discomfort

3 = Annoying, but not causing marked discomfort

4 = Moderately severe, causing marked discomfort

5 = Severe, some interference with sleep or activities but not incapacitating

6 = Incapacitating

In the following section please identify any changes from your regular weekly pattern. For example, if you are out of town please list the location; if you change medications, record the new medication; if your physical activity is more or less than customary, identify that activity; and list unusual passive exposure to cigarette smoke. Record any other notable symptoms or events.

Date	Comments