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PART I

**DEVELOPMENT OF AN EXPOSURE FACILITY TO CONDUCT INHALATION
STUDIES TO AMBIENT AEROSOLS**

Final Report

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California Air Resources Board

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ABSTRACT

Particles in the atmosphere are a complex and heterogeneous mixture that have been difficult to reproduce in the laboratory. As a result, scientists have not been able to conduct toxicological and clinical experiments that replicate realistic conditions in the environment. Investigators have typically generated synthetic atmospheres that differ in significant ways from the true environment. This has made it difficult to address unanswered questions about the true nature and mechanisms of action of atmospheric particles (PM) on human health.

To study the health effect of PM in a realistic setting we designed, built and evaluated a Versatile Aerosol Concentration System (VACES) for testing the toxicological significance of concentrated atmospheric aerosols in animals. The system has been designed for conducting animal exposure studies, but it can be readily scaled-up for human exposures. This report describes the development and bench-testing of a VACES capable of simultaneously concentrating ambient particles of the coarse, fine and ultrafine size fractions for conducting *in vivo* and *in vitro* exposure studies to “real” ambient aerosols over a wide dynamic range of concentrations. The VACES consists of three parallel sampling lines (concentrators), each operating at an intake flow rate of 110 LPM. Coarse particles are concentrated using a single round nozzle virtual impactor. Concentration enrichment of PM_{2.5} and ultrafine particles is accomplished by first drawing air samples through two parallel lines, having 2.5 and 0.18 μm cutpoint pre-impactors, respectively, to remove particles larger than these sizes from the air sample. Both of the smaller PM fractions are drawn through a saturation-condensation system that grows particles to 2-3 μm droplets, which are subsequently concentrated by virtual impaction. A diffusion dryer is used in the fine and ultrafine concentrators to remove excess vapor and return the concentrated particles to their original size, prior to supplying them for *in vivo* exposures. The VACES can also provide highly concentrated liquid suspensions of particles of these three modes for *in vitro* toxicity studies. This is accomplished by connecting the concentrated output (minor) flows of each of the VACES parallel concentrators to a liquid impinger (BioSampler), used in a modified configuration, to collect particles under near-ambient pressure.

Detailed laboratory characterization of the individual components of the VACES are presented in this paper, including evaluation of its ability to preserve particle mass, number, and chemical species during the concentration enrichment process. The experimental characterization of the VACES demonstrated that concentration enrichment is accomplished with very high efficiency, minimal particle losses and without any dependence on particle size or chemical composition.

During the field evaluation of the VACES, the enrichment and preservation of ambient ultrafine, fine and coarse particles by size and chemical composition is determined by comparisons made between the VACES and a co-located multistage MOUDI impactor, used as a reference sampler. Furthermore, preservation of the ultrafine fraction is measured by the enrichment based on ultrafine particle numbers, morphological characteristics as well as their elemental carbon (EC) content. The results suggest that the concentration enrichment process of the VACES does not differentially affect the particle size or chemical composition of ambient PM. The following fractions: 1) mass (coarse and fine PM); 2) number (ultrafine PM); 3) sulfate (fine PM); 4) nitrate (fine PM, after correcting for nitrate losses within the MOUDI); 5) EC (ultrafine PM);

and 6) selected trace elements and metals (coarse and fine PM), are concentrated very close to the “ideal” enrichment value of 22 – thereby indicating a near 100% concentration efficiency for the VACES. The field results also suggest that volatile species, such as ammonium nitrate, are also preserved throughout the supersaturation and concentration-enrichment processes. Furthermore, ultrafine particles are concentrated without substantial changes in their compactness or denseness, as measured by fractal dimension analysis.

EXECUTIVE SUMMARY

The goal of this investigation was to design, build and install a mobile particle concentrator for testing the toxicological significance of concentrated atmospheric aerosols. This system, which we have named Versatile Aerosol Concentration Enrichment System (VACES) is the first capable of concentrating ultrafine, fine and coarse particles. Particle concentration enrichment is accomplished by means of virtual impaction, using well characterized, and single-nozzle virtual impactors. The portable concentrators were designed for use primarily in animal inhalation studies since these systems are compact. In addition, their modular design makes them readily adaptable to accommodate higher output flow rates that are required for human exposures.

Extensive proof-of-concept testing was conducted during the period covered by this report in order to determine any influence of the process and system of concentrators on the physical or chemical properties of ambient aerosols. These proof-of-concept studies were conducted at UCLA and USC. Particle size and chemical composition of the concentrated aerosols was compared to ambient aerosol at the concentrator inlet to ensure that no substantial distortion in the physico-chemical characteristics of PM occurs during the concentration enrichment process. The characterization of the coarse, fine and ultrafine concentrators for animal exposures has been completed ahead of schedule allowing us to initiate toxicological studies, starting in late June 2000, prior to the second year contract. The particle concentrator completed under this contract can be transported to various locations to take advantage of regional variation in particulate air quality, populations of interest, and to coordinate with ongoing field studies of air quality. At these sites extensive animal toxicology and human clinical studies will be undertaken to provide further understanding of the relationship and mechanism between adverse health effects and exposure to airborne particulate matter.

We are currently ahead of schedule defining *in vitro* and *in vivo* toxicological experiments using the concentrator. The advanced schedule has been facilitated by rapid development and validation of a new generation of more portable and versatile concentrators. These new concentrators can also be scaled up to accommodate the higher output flows that are desirable in conducting human exposure studies. This scale-up is easily achieved by placing several of the single-nozzle virtual impactors in parallel. While the design and construction of these larger systems is not part of our activities covered by this contract, this will be pursued in future research.

In our original scope of work, we proposed to enclose the entire concentrator facility, including the exposure chambers, in a 4 m x 4 m x 5 m shipping container, so that it could be transported to different locations within the greater Los Angeles Basin. We substantially revised that plan and decided to build the mobile concentrator facility in a trailer, as this design makes it even easier to transport to other locations, with minimum installation and/or dismantling time.

The Versatile Aerosol Concentration Enrichment Systems VACES that we developed are substantial technological improvements over the originally proposed Harvard Ambient Fine Particle Concentrators. The VACES portable concentrators are capable of enriching the concentration of particles in the entire range of 0-10 μm by a factor up to 40, depending on the output flow rate. These systems are very compact in size and modular in design.

There are several advantages to using the new, portable concentrators compared to the older version of Harvard concentrators. First, Harvard concentrators focus mainly on concentrating the accumulation mode of ambient PM, e.g., $\text{PM}_{2.5}$ without its ultrafine or coarse PM component. These concentrators are bulky and not easily transportable as they require placement inside a large trailer. They require a considerable amount of electric power; and the blower that drives the major flows of the virtual impactors requires a three-phase, 30-amp current. The concentration enrichment depends on particle size, with larger particles in the accumulation mode being concentrated in general more effectively than smaller particles. Under certain conditions, the performance of the Harvard concentrators becomes unstable during operation. Typical indications of instabilities are abrupt increases in pressure drop across the slit nozzles of the virtual impactors, followed by a sharp decrease in the concentration enrichment factor. These problems have been observed under conditions of high particle concentration and/or when operating these systems in days with high humidity and temperature.

The VACES consists of three parallel sampling lines (concentrators) that separately sample ambient coarse, fine and ultrafine aerosols, each at 110 LPM. The fine and ultrafine fractions are separated from the air sample and drawn through a supersaturation and condensational growth system. All fractions (i.e., size-selected and enlarged fine and ultrafine, and ambient coarse) are subsequently concentrated with a virtual impactor. The number/mass concentration may be enriched by a factor as great as 33, which is, ideally, determined as a function of the ambient inlet flow rate to the minor flow-rate of the virtual impactor (typically between 3.3 and 10 LPM, depending on the desirable configuration). In the experiments described in this field study, the minor flow of each concentration-enrichment sampling line of the VACES was set at 5 LPM, thereby resulting in an ideal concentration enrichment factor of 22 for coarse, fine and ultrafine aerosols.

The VACES has been designed to simultaneously conduct *in vivo* and *in vitro* exposures to concentration-enriched ambient particles of either one, or all of the coarse, fine and ultrafine size fractions of PM. A system was developed for particle collection for *in vitro* analysis. This collection is accomplished by connecting a modified liquid impinger (BioSampler™) to each of the minor flows of the coarse, fine and ultrafine portable concentrators, respectively. Highly concentrated aqueous suspensions can thus be obtained, which can be readily used for exposing cell cultures to ambient particles of all three modes. This direct particle collection also eliminates uncertainties related to incomplete extraction from filter media and preserves the biologically active components of the collected PM.

The ability of the VACES to concentrate particles was first tested in laboratory experiments using different type of particles in the size range of 0.05-1.9 μm and at three minor flow rates of two 7, 10, and 20 LPM with the total intake flow rate of 220 LPM. The enrichment factors based on number concentrations were close to the ideal values. Hygroscopic aerosols, such as

ammonium sulfate and ammonium nitrate were concentrated as effectively as hydrophobic PSL particles.

The experimental characterization of the VACES demonstrated the concentration enrichment does not depend on particle size or chemical composition. Volatile species such as ammonium nitrate are preserved through the concentration enrichment process under the laboratory conditions used in this study.

Field characterization of the VACES was conducted outdoors in the facilities of Rancho Los Amigos National Rehabilitation Center in south-central Los Angeles. The coarse, fine and ultrafine particle concentrations of the VACES were compared to direct concurrent measurements made with a co-located MOUDI. Comparisons between the VACES and MOUDI for coarse and fine PM are based on particle mass, sulfate, nitrate and selected trace element and metal concentrations. For ultrafine PM (aerodynamic diameter smaller than a 0.18 μm), the VACES number concentrations is compared to those of a co-located Condensation Particle Counter, whereas the preservation and concentration enrichment of the elemental carbon (EC) content is determined by comparing VACES concentrations to those of the MOUDI within this size-fraction.

Results from the field study indicated that concentration enrichment is differentially affected by particle size or chemical composition. For either coarse or fine particles, the concentration enrichment factors based on mass, sulfate, nitrate after correcting for nitrate losses of the MOUDI, and selected trace elements and metals were very close to the ideal enrichment value of 22. The experiments, additionally, indicated that volatile species such as ammonium nitrate are preserved throughout the concentration enrichment process. Furthermore, concentration enrichment obtained for ultrafine particle counts suggests that no particle coagulation occurs during the enrichment process. Finally, ultrafine EC concentrations obtained with the VACES were about 22 times higher than those obtained with the MOUDI, thereby indicating that ultrafine PM are concentrated without loss, with a nearly 100% collection efficiency by this system. In addition, detailed morphological examination of ambient and concentrated ultrafine particles indicated that ultrafine particles are concentrated without substantial changes in their compactness or denseness.

The ability of the VACES to enrich the concentrations of all particles in the fine mode including its ultrafine particle component enables inhalation toxicologists to conduct exposures to any selected sub-range of PM_{2.5}. For example, previous studies in California showed the presence of two sub-modes within the accumulation mode of ambient PM; one mode peaks at around 0.2 μm consisting mainly of gas-to-particle reaction products, such as carbonaceous PM and the other peaks at about 0.7 μm mainly associated with hygroscopic PM such as ammonium sulfate and nitrate. These observations have been confirmed by our recent yearlong measurements at the facility of Rancho Los Amigos in Downey. By placing a conventional impactor (with a 0.4 μm cutpoint) upstream of the fine concentrator of the VACES, inhalation studies could be conducted to ultrafine PM plus the elemental and organic carbon content of the accumulation mode, but without the majority of the larger sulfate and nitrate constituents.

In addition to the animal and human exposures, we will also use the newly developed versatile concentrators for direct PM collection for *in vitro* tests. Collection and chemical characterization of coarse, fine and ultrafine particles for *in vitro* tests using the combined concentrator/BioSampler method will be conducted at the PIU locations as well as at the sites where animal exposures to freeway-originated aerosols are planned. We have already initiated these studies, by collecting PM samples using the VACES at UCLA and at Rancho Los Amigos. In our current sampling scheme, outdoor particles are collected concurrently with human exposure studies. In addition to the biological content of ambient or indoor PM, we also monitor the following parameters over the 6 hours of the experiment: particle number concentration (continuously) and particle mass concentration (time-integrated).

We have made considerable progress in using the VACES for animal inhalation studies in two different locations in the Los Angeles Basin. These studies were not part of the originally proposed scope of work but have been made possible by the rapid development of VACES. The studies were conducted jointly by investigators from UCLA, University of Southern California, UC Irvine and UC Davis. Healthy rats (and in a later summer study, sensitized mice) were exposed to fine and ultrafine PM, concentrated by a factor of 22, harvested at UCI in June 2000 and UCLA in July 2000 in west Los Angeles. Preliminary measurements in the later location have indicated an unusually high number concentration of both ambient as well as indoor PM on the order of 10,000-50,000 particles/cm³, roughly 5-10 times higher than levels typically encountered in urban areas of the East Coast of the U.S., which makes these experiments of particular interest. The same particle sampling protocol used for the *in vitro* tests was also used to monitor the physico-chemical PM characteristics during the animal exposure studies: monitoring of particle mass, number concentration, elemental composition and selected PAH. Biological analysis of the animals exposed in these studies is currently under way.

1. Introduction

The goal of this investigation was to design, build and install a mobile particle concentrator exposure facility at UCLA for testing the toxicological significance of concentrated atmospheric aerosols. This facility is the first capable of concentrating ultrafine, fine and coarse particles. Extensive proof of concept testing was conducted during the period covered in this report to determine any influence of the process and system of concentrators on the physical or chemical properties of ambient aerosol. Particle size and chemical composition of the concentrated aerosols was compared to ambient aerosols at the inlet to ensure that no substantial distortion in the physico-chemical characteristics of PM occurs during the concentration enrichment process. The characterization of the coarse, fine and ultrafine concentrators for animal exposures has been completed ahead of schedule and we have initiated toxicological studies, which began in June 2000. The completed particle concentrator is transportable to take advantage of regional variation in particulate air quality, populations of interest, and to coordinate with ongoing field studies of air quality. Extensive animal toxicology and human clinical studies will be undertaken to provide further understanding of the relationship and mechanisms of adverse health effects associated with exposure to PM. We are ahead of schedule defining *in vitro* and *in vivo* toxicological experiments using the concentrator. The advanced schedule has been facilitated by rapid development and validation of a new generation of more portable and versatile concentrators.

This report addresses the period between June 1, 1999 and August 31, 2000. During this period we were awarded two additional grants from EPA which led to creation of the Southern California Particle Center and Supersite (SCPCS). The support from the California Air Resources Board has been crucial in our successful competition for federal funds to develop a major particle center in Southern California, and we are wholly dependent on ARB support for the successful development of the range of activities in SCPCS.

2. Construction of Concentrator Trailer and Set-Up of Related Aerosol Instrumentation

In our original scope of work, we proposed to enclose the entire concentrator facility, including the exposure chambers, in a 4 m x 4 m x 5 m shipping container, so that it can be transported to different locations within the greater Los Angeles Basin. We revised that plan to house the mobile concentrator facility housed in a trailer, since this design would make it easier to transport to other locations with minimum installation and/or dismantling time. A trailer would not require special transportation permits that are required for shipping and transporting containers.

Two trailer laboratories were constructed that can be towed with a 3/4-ton pickup truck to various sites. The two trailers were ordered from Wells-Cargo, Inc. for February 1, 2000 delivery. One trailer is 32' X 8' for the concentrator and the other is 20' X 8' for a Particle Instrumentation Unit (PIU) to be used for PM physicochemical characterization. The latter trailer has been purchased and equipped through funding from the SCPCS.

The concentrator trailer has two separate compartments, partitioned by a plywood bulkhead; one compartment is 12' X 8' and will be used for all concentrator-related apparatus. The other compartment will be used for equipment related to the animal studies, including nose only and whole-body animal exposure chambers, as well as for an animal vivarium to store the animals for the exposures that will be conducted at locations other than UCLA. Both compartments of the exposure trailer are air-conditioned.

We have received and calibrated all major pieces of sampling equipment and direct reading instruments being used to characterize the concentrated aerosols. These include a Tisch Environmental Hi-Vol sampler with a PM-10 inlet, a TSI Aerodynamic Particle Sizer (APS), three rotating-versions of the MSP ten-stage Microorifice cascade impactors (MOUDI), and a MIE DataRAM. In addition, the TSI Scanning Mobility Particle Spectrometer (SMPS) has been received. These instruments were purchased through the SCPCS. They have been used to evaluate the performance of the concentrators and will be also used to provide data on ambient and concentrated aerosol characteristics during human and animal exposures.

We have also completed the construction of the whole-body exposure chambers for human inhalation studies. The single-person exposure chamber is a plywood-and-Plexiglas whole-body plethysmograph modified by extending the lower front wall to form a foot well, in which a small cycle ergometer can be placed. The straight 7.5-cm stainless steel outlet pipe from the particle concentrator enters the chamber at the chest height of a seated subject. The inlet pipe is interrupted by a demountable butt joint to permit disassembly of the system for cleaning. The inlet and outlet ports of the concentrated aerosol are designed such that the exposure atmosphere exits the chamber through multiple ports above and behind the subject's head. Further details of the whole-body human exposure chamber are given in Gong et al. (1999).

We have also received sampling equipment and direct reading instruments for gaseous pollutants. These instruments were provided in-kind by the Biological Effects Research Section of the California Air Resources Board. These monitors include a Continuous Chemiluminescence Analyzer (Monitor Labs Model 8840) for nitrogen oxides measurement, Thermo Environmental Inc. Model 48C trace level carbon monoxide analyzer and a UV photometer (Dasibi Model 1003 AH) for measurement of ozone. The instruments were installed in the PIU trailer and calibrated.

3. Development and Characterization of the Coarse, Fine and Ultrafine Particle Concentrator for Animal Exposure and *In Vitro* Studies

3.1. Operating Principle of Particle Concentrators

Concentration enrichment of particles larger than a critical size (herein referred to as the cutpoint of a concentrator) is accomplished by means of virtual impaction. Particles are drawn through a nozzle gradually decreasing diameter and become accelerated to a high velocity, the magnitude of which depends on the cutpoint of the virtual impactor (higher velocities are required for smaller cutpoints). Immediately downstream of the acceleration nozzle, the majority of the airflow (herein referred to as the "major" flow) is deflected around a probe placed within few mm from the exit of the acceleration nozzle and in perfect alignment with the acceleration

nozzle. A small portion of the original total air volume (typically 3-10%, also called “minor” flow) is diverted into the collection probe, and along with it particles that have acquired sufficient momentum to cross the deflected air streamlines. These particles are concentrated ideally by the ratio of the total-to-minor flow rates. Thus diverting the particles into a minor flow 5% of the total flow entering the virtual impactor would ideally concentrate particles larger than the cutpoint by a factor of 20. The concentration enrichment of a virtual impactor can be adjusted by adjusting the minor-to-total flow ratio.

A major advantage of virtual impactors is that they accomplish particle concentration enrichment while keeping the particles airborne, i.e. without collecting the particle on a filter or any other substrate. These concentrated and airborne particles could subsequently be supplied to exposure chambers for human or animal inhalation studies with minimum distortions in their physical, chemical and morphological characteristics and their gaseous copollutants equilibrium.

3.2 Justification for Changing the Design of the Concentrators

We had originally proposed to install Harvard Ambient Fine Particle Concentrators in the first year of this program (Sioutas et al., 1995; Sioutas et al., 1997). Our original plan was to develop improved coarse and ultrafine particle concentrators in subsequent years. We had initially projected installation of the fine particle concentrator around the second week of December 1999. That installation was postponed to the second or third week of March 2000. In a subsequent communication with Dr. Petros Koutrakis (Harvard School of Public Health), we were informed because of construction problems relating to quality control difficulties in the machining and alignment processes of the slit-nozzle virtual impactors, Harvard could not commit to any delivery time prior to late June 2000. Harvard would not provide any assurances that even this late delivery would be met.

This delivery time was unacceptable, since it substantially delayed our proposed health studies to concentrated PM, which are major foci of our ARB as well as our PM Center programs. We therefore requested ARB’s approval to a change in our research direction. We decided not to proceed with the Harvard fine particle concentrator for this program. Instead, we used the new and improved portable concentrators (described in section 3.2) that we have developed over the past two years. These portable concentrators are based on technologies already developed and published (Sioutas et al., 1999; Kim et al., 2000a) by the Aerosol Laboratory of the University of Southern California, and are capable of enriching the concentration of particles in the entire range of 0-10 μm by a factor up to 40, depending on the output flow rate. These systems are very compact in size and modular in design. They can thus be readily adaptable to accommodate higher output flow rates that are desirable in conducting human exposure studies. Over the past year, scaled-up versions of the coarse, fine and ultrafine concentrators were developed through this program and their laboratory and field evaluation is described in greater detail by Kim et al (2000b; 2000c).

Unique features of the new generation of portable concentrators:

1. The virtual impactors of these systems employ round acceleration and collection nozzles, compared to the rectangular geometry designs of older version of concentrators (described

by Sioutas, C., Koutrakis, P., and Burton, R.M. "A technique to expose animals to concentrated fine ambient aerosols." *Environmental Health Perspectives*, 103:172-177, 1995). Due to intrinsic design characteristics associated with the three-dimensional flow of round nozzles (compared to the axisymmetric flow of slit-nozzle impactors), higher particle efficiency and lower losses are achieved. Thus, a single-stage system can concentrate particles up to a factor of 40, without altering the size distribution and chemical composition of the sampled and concentrated aerosols.

2. The high-efficiency, single-stage design makes the entire system very small and portable. This is exceedingly important, as it makes it possible to place these concentrators in light-duty trailers and transport them at various sites with distinctly different chemical and physical characteristics of PM.
3. These concentrators are capable of concentrating particles of all three discrete size groups concurrently. These groups are:
 - Ultrafine Particles ($<0.1\mu\text{m}$), which are freshly generated particles, such as those generated by combustion,
 - Fine Particles (including their ultrafine mode) of any size sub-range between $0\text{-}2\mu\text{m}$ and;
 - Coarse ($>2.5\mu\text{m}$) particles.
4. Concurrent concentration of all of three PM modes allows specific size ranges and chemical characteristics of concentrated ambient PM to serve as test aerosol to conduct specific hypotheses-driven toxicity studies.
5. Short-term health impacts of real-life PM associated with different size ranges and sources can be evaluated.
6. Because of their high concentration efficiency, operation of these systems requires very low power. For example, for a 9-nozzle system that provides 100 LPM of concentrated PM for human exposures, all flows can be driven by three Gast 2067 pumps. Each of these pumps consumes 0.7 kW (total of 2.1 kW). These pumps are single-phase, 110 Volts, and can be readily plugged into any standard power outlet. Compared to these systems, the previously developed Harvard concentrators employ a three-phase blower, consuming 9 kW. This blower requires three-phase power installation, generates 120 dB of noise, and hence requires some type of enclosure for noise reduction, which in turn requires some means of ventilating the generated heat by the blower. The volume and power requirement of the Harvard concentrators makes them impractical for transportation and field use. None of these problems are encountered in the use of the new, portable particle concentrators.
7. A unique feature of these systems is also the ability to provide concentrated ultrafine particle to an exposure chamber with a very low-pressure drop (less than 5 inches of H_2O). Concentrated coarse and fine particles with their ultrafine component can be provided to an inhalation chamber under a negative pressure of less than 1 inch of H_2O ,

almost atmospheric pressure. By comparison, the older version of Harvard Concentrators delivers the aerosol under 15-20 inches of water negative pressure.

8. Due to the larger size of the round nozzles (0.4-0.6 cm) compared to the width of the previously developed Harvard concentrators (0.03 cm), the new systems do not suffer from clogging and performance instabilities associated with the rapid increase in pressure drop, followed by a sharp decrease in the concentration enrichment factor. These problems have been observed under conditions of high particle concentration or when operating these systems in days with high humidity and temperature (personal communications; F.R. Cassee, RIVM, Netherlands, C.S. Kim, U.S. EPA, J.J. Godleski, Harvard University, D. Costa, U.S. EPA, J.R. Brook, Health Canada). A paper investigating the effects of parameters such as ambient relative humidity, dew point temperature, ambient PM_{2.5} mass concentration, ambient PM_{2.5} mass median diameter (MMD), and total pressure drop per unit time across the Concentrator on the overall concentration enrichment achieved by the Harvard Fine Particle Concentrator has been just accepted for publication (Kim et al., 2000).
9. Another unique feature of the portable concentrators is their ability to be used in conjunction with a liquid impinger (BioSampler™, SKC West Inc., Fullerton, CA) to collect directly large volumes of outdoor and indoor particles on a cell culture solution or any other liquid solution. Traditionally, particle collection for *in vitro* tests has been conducted by using collection substrates such as filters or impactors. The collected particles are subsequently extracted from the substrates and administered into the *in vitro* culture either directly or after lyophilization of the solvent. This process suffers from several shortcomings, including imperfect particle extraction from the substrate but most importantly, this mechanism for particle collection does not preserve biologically active agents of airborne PM. Direct impingement of these particles onto the cell culture solution will substantially improve the *in vitro* evaluation of toxic effects of PM. The collection efficiency of the BioSampler is close to 100% for particles larger than about 1.5 μm, and operating at a flow rate of 12.5 l/min. For particles less than 1.0-micron diameter the collection efficiency decreases sharply to less than 50% for particles at 0.5 μm. Operating in conjunction with our prototype ultrafine, fine or coarse particle concentrators, the BioSampler can collect any of the PM size ranges with 100% efficiency and at sampling flow rates that are 20-30 times higher than its nominal operating flow rate. Thus, the condensation growth of even ultrafine PM to super-micrometer particles enables effective trapping of these particles by the impinger and allows us to “concentrate” large volumes of ambient PM into a very small solution on the order of 5-10 ml. The resulting particle concentration in the *in vitro* solution is on the order of 50-400 μg/ml, depending on ambient PM number and mass concentrations.
10. The ability to collect large volumes of particles directly into a small volume of any solution is a particularly attractive feature when intratracheal instillation is used as the method to conduct particle toxicity tests.

3.3 Description of the Portable Coarse, Fine and Ultrafine PM Concentrators

Figures 1a and 1 b show a schematic of two different configurations of the new concentrators, which we have named Versatile Aerosol Concentration Enrichment Systems (VACES). The VACES incorporate the following features:

1. The ability of concentrating ultrafine particles only, and supplying them to an exposure chamber at virtually atmospheric pressure (0.99 atmospheres).
2. The ability to allow concurrent animal exposures to coarse, fine and ultrafine particles.
3. When exposures to one PM mode are desirable, this technology can concentrate up to 330 LPM of ambient PM to a flow rate as low as 10 LPM. This feature makes it possible to use more animals in an inhalation study, hence increase the confidence level in the observed outcomes.
4. The capability of collecting concurrently very high quantities of coarse, fine and ultrafine PM in a small liquid volume (4-10 ml). The resulting highly concentrated suspensions can be used for *in vitro* tests to evaluate the relative toxicity of ambient PM, collected simultaneously in a given location.

Figure 1a shows the configuration used for *in vivo* inhalation exposures, whereas Figure 1b shows the version of the same system for *in vitro* toxicity studies.

The VACES consists of three parallel sampling lines. In each line, ambient coarse, fine and ultrafine aerosols are drawn at 110 LPM. Coarse PM is drawn through a round nozzle, single-stage virtual impactor, having a 50% cutpoint at 1.5 μm . The performance of these virtual impactors is described in greater detail by Kim et al (2000a). Coarse particle in this sampling line can be concentrated by as much as a factor of 35, and supplied to the exposure chamber at a flow rate ranging from 3.3-11 LPM.

The other two sampling lines of the VACES consist of identical components, with the only exception of the cutpoints of the impactors through which the samples are drawn prior to passing through the saturator. In the line concentrating fine plus ultrafine PM, air samples are first drawn through a single slit nozzle impactor, having a 50% cutpoint at 2.5 μm at a flow rate of 110 LPM. The impactor's acceleration nozzle is 0.2 cm wide and 5 cm long. At a sampling flow rate of 110 LPM, particles are accelerated to a velocity of 1834 cm/s, and the corresponding pressure drop across the impactor is 1.5 inches of H_2O .

In order to remove all but the ultrafine PM, particles in the third sampling line of the VACES are drawn through a multi-nozzle, high volume conventional impactor with a design 0.15 μm cut-off size at a flow rate of 110 LPM. Separation of these particles is accomplished under a very low-pressure drop (i.e., 7-8 inches of H_2O). This is a very important feature of these new concentrators, since inhalation studies cannot be conducted under a substantial vacuum. The impactor consists of 5 slit-shaped nozzles in parallel, each 5 cm long and 0.015 cm wide. At a flow rate of 110 LPM, the resulting velocity through each rectangular jet is approximately 4200

cm/s and the corresponding pressure drop across the impactor is 7.5 inches of water (or 0.019 bar). A 5 x 0.2 cm quartz fiber strip is placed underneath each acceleration nozzle, at a distance of 0.04 cm. The strips are coated with mineral oil and serve as bounce-free impaction substrates for collecting particles above 0.16 μm in aerodynamic diameter. It should be noted that concentration of ultrafine particles is optional. Without the use of the 0.15 μm impactor, the VACES can also deliver fine and ultrafine PM at 10 LPM, enriched in concentration by a theoretical factor of 22.

After the impactor pre-separators, the aerosol in both the fine and ultrafine lines is drawn through a stainless steel container used as the aerosol saturator. The container has a capacity of 10 liters and is used to mix the aerosol with warm, distilled deionized vapor at a temperature of about 30 (± 2) degrees C. The stainless steel container is placed inside a heating bath (VWR Scientific, Model 1024), with a heating power of 0.5 kW.

The saturated aerosol is drawn through a cooler, which is an icebath with two aluminum tubes (2.2 cm in diameter and 80 cm long) through it. In each cooler, the saturated and warm air is cooled by about 9-10 degrees C. The produced supersaturation in the cooling causes all particles to grow to about 2.5-2.6 μm droplets.

The grown droplets are subsequently drawn through two identical virtual impactors. Each virtual impactor separates particles into two different size ranges, approximately above and below 1.5 μm . These virtual impactors are also identical in design to those used for concentration of coarse ambient particles. The virtual impactors are made of anodized aluminum. The grown fine and ultrafine particles are drawn into the minor flow of virtual impactor (which can be as small as 3 LPM), and thereby become concentrated by a factor up to 40.

Concentrated droplets are drawn through a Diffusion Dryer (TSI Model 3062, TSI Inc., St. Paul, MN), placed immediately downstream of the collection nozzle of each virtual impactor. The diffusion dryer is used to remove the excess moisture around the particles and return these grown particles to their original size. Operating at a maximum flow rate of 10 LPM, each diffusion dryer reduces the relative humidity of the incoming aerosol from 100% to 50%, thereby returning the grown particles to their original size.

All three major flows of the parallel virtual impactors are drawn by a single rotary vane pump (Gast model 2067, Gast Manufacturing, Cerritos, CA). This pump is capable of drawing up to 360 LPM under a vacuum of 150 inches of water, while consuming only 0.5 kW at 110 V. The pump is light (20 lb), takes up very little space, and does not require any special power installation.

In vitro sampling:

Figure 1b shows the alternative configuration of the VACES, used for simultaneous coarse, fine and ultrafine PM collection for *in vitro* toxicology experiments. For *in vitro* collections the concentrated coarse, fine and ultrafine particles in each parallel sampling line are drawn through a liquid impinger instead of passing through a diffusion dryer (BioSampler, SKC West Inc.,

Fullerton, CA). The performance of this device is described in greater detail by Willeke *et al.* (1998). Unlike conventional impingers in which the aerosol is impacted into a reservoir filled with liquid, particles in the BioSampler are injected into a swirling flow for collection by a combination of inertial and centrifugal forces onto the surface over which the air flow swirls.

Traditionally, particle collection for *in vitro* tests has been conducted by using collection substrates such as filters or impactors. Particles collected on filters are subsequently extracted from the substrates and administered into *in vitro* culture media, either directly or after lyophilization of the solvent. This process suffers from several shortcomings, including inefficient particle extraction from the substrate, and variable losses of potentially toxic semi-volatile PM constituents, and of biologically active components of airborne PM. In addition, a recent study by Dick *et al.* (2000) showed that components of filters used to collect particles could contaminate the preparation and interfere with biological investigations.

Particle collection using liquid impingers has been shown to be advantageous over the traditional filtration or impaction methods for collection of airborne particles, because impingers are not easily overloaded (Willeke *et al.*, 1998), and impingement eliminates the need for elaborate extraction procedures (Zucker *et al.*, 2000). Under normal operating conditions at its nominal flow rate of 12.5 LPM, the BioSampler has collection efficiency close to 100% for particles larger than about 1.5 μm . For particles smaller than 1.0 μm in aerodynamic diameter, the collection efficiency decreases sharply to less than 50% (Willeke *et al.*, 1998). Operating in conjunction with the VACES, however, the BioSampler can collect any of the PM size ranges with 100% efficiency and at sampling flow rate that is at least 10-fold higher than its nominal operating flow rate. Thus, the supersaturational growth of even ultrafine PM to super-micrometer particles enables effective trapping of these particles by the impinger and allows us to “concentrate” large volumes of ambient PM into a very small solution on the order of 5-10 ml. The ability to collect large volumes of particles directly into a small volume of any solution is a particularly attractive feature when intratracheal instillation is used as the method to conduct particle toxicity tests.

3.4 Experimental Characterization of the VACES Components

3.4.1 Characterization of the 2.5 μm and 0.15 μm Low Pressure-Drop Slit Impactors

The collection efficiency of the 2.5 μm cutpoint slit impactor was determined using monodisperse aerosols generated by atomizing suspensions of PSL particles (size range: 0.5-10 μm ; Bangs Laboratories Inc.) in a constant output Nebulizer (HEART, VORTRAN Medical Technology, Inc., Sacramento, CA). The generated aerosols passed through Po-210 static charge neutralizers and were mixed with filtered air prior to passing through the slit impactor. The mass concentrations of the monodisperse aerosols upstream and downstream of the impactor were measured by means of a nephelometer (DataRAM, MIE, Inc., Billerica, MA). For each test, repeated measurements of the concentrations upstream and downstream of the impactor were taken. The concentrations of the generated aerosols were in the range of 100-400 $\mu\text{g}/\text{m}^3$, thus several orders of magnitude higher than the limit of detection of the DataRAM which is about 1-5 $\mu\text{g}/\text{m}^3$. As a nephelometer, the response of the DataRAM is dependent on particle size (Sioutas *et al.*, 2000). Particle collection efficiency as a function of aerodynamic diameter is

shown in Figure 2. The results confirm that the cutpoint of the impactor is at about 2.5 μm in aerodynamic diameter.

The collection efficiency of the multi-slit 0.15 μm cutpoint impactor was estimated using ambient air as the test aerosol. For particles in the size range of 0.015 to 0.5 μm , penetration was determined by measuring the aerosol concentrations upstream and downstream of the impactor by means of the Scanning Mobility Particle Sizer (SMPS Model 3096, TSI Inc., St. Paul, MN). The SMPS sampled 0.2 LPM of the total flow rate of 110 LPM through the impactor. Number concentration of ambient aerosols was measured with and without the block holding the acceleration slit nozzles of the impactor to account for possible diffusional losses of ultrafine particles through the sampling lines connecting to the SMPS. Particle size was selected in the interval of 0.02-0.5 μm by adjusting manually the voltage to the Differential Mobility Analyzer of SMPS and measuring the particle counts upstream and downstream of the 0.15 μm cutpoint impactor.

In addition to the SMPS, the DataRAM was used to evaluate the collection efficiency of the multi-slit impactor for particles in the 0.2 to 1.0 μm range, using artificially generated monodisperse PSL particles as described above. The DataRAM could not be used to monitor particles less than 0.2 μm because the sensitivity of the instrument decreases sharply below this particle size.

Finally, limited field tests were conducted in which the ambient aerosol concentrations measured by the 0.15 μm cutpoint impactor was compared to that measured by means of the Microorifice Uniform Deposition Impactor (MOUDI, MSP Corp., Minneapolis, MN), which was used as a reference sampler. A 4.7 cm Teflon filter (2 μm pore, Gelman Science, Ann Arbor, MI) was placed immediately downstream of the multi-slit impactor, which was operated at a flow rate of 110 LPM. The MOUDI was placed at a distance of 1 m from the impactor and sampled at 30 LPM. Ambient particles smaller than 0.18 μm in aerodynamic diameter were collected on a 3.7 cm Teflon filter following the last impaction stage of the MOUDI. Both MOUDI and multi-slit impactor Teflon filters were weighed before and after each test on a Mettler Microbalance (MT5, Mettler-Toledo, Inc., Highstown, NJ) under the controlled relative humidity (40-45%) and temperature (22-24 $^{\circ}\text{C}$) conditions in order to determine the mass concentrations.

Figure 3 shows the pressure drop across the multi-slit impactor as a function of flow rate. The pressure drop across the multi-slit impactor is about 7 inches of H_2O at the standard flow rate of 110 LPM. The ability of this impactor to remove all but ultrafine particles with a very low pressure drop is a very important feature of the VACES, since inhalation health studies cannot be conducted under a substantial vacuum.

The collection efficiency of multi-slit impactor, determined from the decrease of both number (SMPS) and mass (DataRAM) concentrations measured downstream of the impactor, is plotted as a function of particle aerodynamic diameter in Figure 4. Error bars represent the standard deviation of the experimental results.

The particle collection efficiency curve obtained from data using the SMPS increases sharply starting at 0.1 μm and reaches the value of about 90% at particles larger than 0.3 μm in

aerodynamic diameter. The collection efficiency values obtained by means of the DataRAM are in a good agreement with those obtained by SMPS for the overlapping particle size range between 0.2 and 0.5 μm . The data shown in Figure 4 indicate that the 50% cutpoint of the multi-slit nozzle impactor has a mobility diameter of 0.18 μm .

The comparison between the mass concentrations measured by multi-slit impactor and the reference MOUDI is shown in Table 1. Despite the small number of data points, the mass concentrations of ultrafine particles obtained with the two samplers are in excellent agreement, with the average slit impactor-to-MOUDI ultrafine particle concentration being 1.07 (± 0.15). The agreement between the two samplers is remarkable because even a small cutpoint difference in the 0.1-0.2 μm range might result in substantial differences in the amounts of particles collected by two different impactors. Mass-based concentration of ambient PM-2.5 decreases sharply for particle sizes smaller than 0.2 μm (Whitby and Svendrup, 1980) and a small entrainment of accumulation mode particles into the ultrafine mode resulting from a small disparity in the impactor cutpoints would result in a significantly higher mass concentration measured by the sampler having the largest cutpoint impactor. The low cut point of the high volume multi-slit impactor with the low pressure drop makes it possible for toxicologists to conduct health study on the ambient particles containing only ultrafine mode.

3.4.2. Characterization of the BioSampler

At the standard operation flow rate of 12.5 LPM, the pressure drop across the BioSampler is close to 0.5 atm, which has been shown to cause excessive evaporation of liquid collection media such as water. It is also expected that under these sampling conditions, excessive losses of semi-volatile components of ambient particles would occur. In order to reduce the pressure drop across the BioSampler used in conjunction with the VACES virtual impactors, a flow rate of 5 LPM was used instead. The decrease in flow rate was expected to increase the cutpoint of the BioSampler. However, as most of fine and ultrafine PM is grown to 2.5-2.7 μm via supersaturation in the VACES, our primary concern was to ensure that particles in that size range are efficiently collected by the modified BioSampler.

Another modification of the BioSamplers used in conjunction with the VACES was the amount of water used in its reservoir to collect the impinging particles. In its nominal configuration, 20 ml of liquid are required in the BioSampler reservoir. However, from the standpoint of toxicological studies, it is highly desirable to maximize the concentration of the collected ambient particles in the liquid medium of the BioSampler. We thus investigated the effect of different volumes of water on the collection efficiency of BioSampler at the reduced flow rate of 5 LPM. We specifically tested the BioSampler using water volumes of 2, 4, 10 and 20 ml, respectively. For each liquid volume, the collection efficiency for particles in the range of 0.5-5 μm was determined by measuring the upstream and downstream BioSampler monodisperse aerosol concentrations using the DataRAM, as described above. At 5 LPM, the pressure drop across the BioSampler was approximately 17 inches of H_2O . The exhaust of the DataRAM pump was returned downstream of the BioSampler in order to avoid sampling biases, which might occur when this instruments samples under a vacuum. This sampling strategy is recommended by the manufacturer.

Figure 5 shows the pressure drop across the BioSampler as a function of flow rate. The pressure drop at 5 LPM is 17 in. H₂O (0.035 atm), which is substantially lower value than the 145 in.H₂O at the standard flow rate of 12.5 LPM. As a result of this small pressure drop, less than 0.5 ml of water volatilized after 6 hours of sampling ambient concentrated air at relative humidities ranging from 45 to 65%. By comparison, 80 % or more of 20 ml of water normally evaporates within 2 hours under reduced pressure at 12.5 LPM (Willeke *et al.*, 1998). The small pressure drop is essential in preserving labile semi-volatile species such as ammonium nitrate and a host of organic compounds would be more pronounced under the high pressure drop across the sampler (Zhang and McMurry, 1987).

The collection efficiency of BioSampler at 5 LPM is shown in Figure 6 as a function of particle size for various amounts of water in the BioSampler reservoir. Error bars represent the standard deviation of repeated tests. Data shown in Figure 6 indicate, for any particle size, there is no significant dependence of the collection efficiency of the BioSampler on the amount of water in its reservoir, at least for the range of 4-20 ml. Based on these results, even 4-5 ml in the BioSampler reservoir would ensure high particle collection efficiency, while maximizing the particle concentration in the aqueous suspension to be used for *in vitro* tests. Five ml is also sufficient to ensure complete wetting of the bottom of the BioSampler reservoir, a feature that ensures effective particle capture by the instrument.

The collection efficiency of the BioSampler is close to 100% for particles larger than 2 μm at a flow rate of 5 LPM, regardless of liquid volume in the reservoir. For particles less than 1 μm in aerodynamic diameter, the collection efficiency decreases sharply to about 50% at 0.5 μm . Any significant decrease in the collection efficiency due to particle bounce was not observed up to about 5 μm of aerodynamic diameter. Figure 6 also shows that the BioSampler collects fine and ultrafine particles that were grown to water droplets more efficiently than dry PSL particles of similar size.

3.4.3 Laboratory Characterization of the Fine and Ultrafine Concentrators of the VACES

The coarse particle concentrator component of the VACES had already been developed and described elsewhere (Kim *et al.*, 2000); laboratory tests focused on the experimental characterization of the fine and ultrafine concentrators of the VACES. It should be noted that the use of the 0.15 μm impactor to remove all but ultrafine particles is optional. The VACES can also be used to concentrate fine PM including the ultrafine fraction from 220 LPM to a flow as small as 7 LPM. Thus experiments were conducted at a sampling flow of 220 LPM as a worse case scenario, since this flow rate represent the most challenging configuration for the saturator and the cooler of the VACES.

The experimental characterization of the VACES was conducted using laboratory monodisperse particles as well as real-life ambient particles as the test aerosols. Monodisperse aerosols were generated by atomizing suspensions of ultrafine and fine particles using a constant output HEART Nebulizer (VORTRAN Medical, Inc., Sacramento, CA). Different types of suspensions were used, including monodisperse PSL fluorescent latex particles (size range 0.05 – 2 μm ; Polysciences, Inc., Warrington, PA) as well as monodisperse silica bead (0.36 μm ; Bangs

Laboratories, Inc., Carmel, IN). In addition, aqueous solutions of ammonium sulfate and ammonium nitrate were atomized. Finally, indoor aerosol was also used as test aerosol. The size distributions of the polydisperse aerosols were determined using SMPS.

The nebulizer generated aerosols were dried, neutralized and were then drawn to the saturator at 220 LPM. The aerosol was mixed and saturated with water vapor at about 30-32 °C, and drawn through two condenser tubes at 110 LPM each. The temperature of the aerosol exiting the condenser was about 23 (\pm 1)°C.

The grown droplets were subsequently drawn through the two virtual impactors. Three different minor flow rates were tested, 7, 10, and 21 LPM, respectively (corresponding to theoretical enrichment factors of 30, 22, and 10.5, respectively). The TSI Condensation Particle Counter (CPC 3022, TSI, Inc., St. Paul, MN) was connected immediately upstream of the saturator and downstream of the diffusion drier (as shown in Figures 1a and 1b) to measure the number concentrations of the original and concentrated aerosols. For each particle size, concentration enrichment was defined as the ratio of the concentration measured downstream of the diffusion dryer to that measured upstream of the saturator.

Results from the laboratory evaluation of the VACES at three different minor flow rates are summarized in Figure 7. In all three minor flow configurations, the major flow rate is adjusted to yield a total intake flow of 220 LPM. Hence, the maximum obtainable concentration enrichment factors for each configuration are 31, 22, and 10.5, respectively. The concentration enrichment factors as a function of particle size, shown in Figure 7, have been obtained using monodisperse aerosols in the size range of 0.05 – 1.9 μm , except for the data corresponding to 0.025, 0.31, and 0.32 μm particles. The number mean diameter (NMD) of polydisperse aerosols were obtained from the count-based size distributions of ammonium sulfate, ammonium nitrate and indoor aerosols using the SMPS.

The enrichment factors at minor flow rates of 7 LPM, 10 LPM, and 20 LPM are 30.1, 20.4, and 9.6, respectively, which are very close to the ideal values. In addition, hygroscopic ammonium sulfate and ammonium nitrate aerosols did not show any observable difference in the enrichment factors compared to the hydrophobic PSL particles.

3.4.4. Field Evaluation of the VACES

The performance of the VACES was evaluated in a field study, conducted outdoors in the facilities of Rancho Los Amigos National Rehabilitation Center in Downey, CA. Situated near the Los Angeles “Alameda corridor”, Downey has some of the highest inhalable particle concentrations (PM_{10}) in the US, very often exceeding the National Ambient Air Quality Standard of 150 $\mu\text{g}/\text{m}^3$. The 25-mile long Alameda corridor is named after Alameda Street, which joins the coastal area of Long Beach (where a major port, large number of industrial plants, and oil refineries are currently operating) to downtown Los Angeles.

The main goal of the field study was to confirm that the physical or chemical properties of ambient aerosol are preserved during the process of concentration enrichment using the VACES. Measurements of concentration-enriched coarse, fine and ultrafine aerosol fractions were

compared to direct ambient measurements made with a co-located MOUDI which was used as a reference sampler. In part, the MOUDI was used because of its high sampling flow rate which allows for sufficient sample collection for comparisons with the VACES in relatively short time periods. Because each of the sampling lines of the VACES sample at 110 LPM, the analytical sensitivity and quantity of particle mass by the VACES, itself, was of less concern. It should be noted that the MOUDI is not a reference sampler for labile species, such as ammonium nitrate and semivolatile organic compounds, and losses of these compounds may occur under conditions of high temperature and low relative humidity (Chang et al, 2000).

Instead of using all of the stages of the MOUDI, only those stages having cut-points of 10, 2.5 and 0.18 μm were used. The first MOUDI stage (2.5-10 μm) was used as reference sampler for coarse ambient particles, the second stage (0.18-2.5 μm) for the ambient PM accumulation mode, and the last stage (i.e., the after-filter) to determine ambient ultrafine particle concentrations. The MOUDI and VACES coarse and fine (accumulation plus ultrafine) PM concentrations were compared by mass, nitrate, sulfate, trace elements and metals. For these analyses, concentration enriched aerosols were collected on 4.7 cm Teflon filters (Gelman Science, 2 μm pore), which were placed immediately downstream of the diffusion dryer of the VACES fine and ultrafine particle concentrators, and directly downstream of the minor flow of its coarse concentrator. For direct ambient measurements, the same type of filters was placed in each MOUDI stage and its after-filter.

Ultrafine concentrations obtained by means of the VACES and MOUDI were compared based on mass and elemental carbon (EC) concentrations, as EC has been shown to be a predominant ultrafine PM constituent at this ambient site (Sioutas et al., 2000). For this analysis, quartz filters (Pallflex Corp., Putnam, CT) were placed downstream of the diffusion dryer of the VACES ultrafine concentrator and of the co-located MOUDI after-filter. Organic carbon (OC) may also be a significant constituent of ultrafine PM mass, however positive artifacts due to adsorption of organic gases on the MOUDI's quartz after-filter (Eatough et al., 1993; McMurry and Zhang, 1987) may introduce significant bias in the MOUDI-VACES comparisons. As the minor flow rate of the VACES (containing virtually all of the ambient particles) is 5 LPM compared to 30 LPM of the MOUDI, gas-phase adsorption on the VACES filter would be theoretically 1/6 of the MOUDI, thus less severe.

In this study, comparisons were based only on the EC fraction of particle-associated carbon, as the organic carbon fraction may consist of several volatile or semi volatile compounds. Data based on the EC fraction better reflect performance of the concentrators. In our first pilot study, the performance of our smaller scale ultrafine and fine concentrators (Kim et al., 2000), the OC comparisons conducted indoors between the concentrators and the MOUDI showed excellent agreement (within $\pm 10\%$) between the two systems.

In order to evaluate whether the chemical composition of concentration-enriched ambient particles are effected by using the *in vitro*/ BioSampler version of the VACES (Kim, et. al., 2000b), measurements were compared to those made directly onto filters. For the samples collected by means of the BioSampler, only the inorganic ion (i.e., sulfate and nitrate) content of the concentrated aerosols were determined, because of the logistical difficulties associated with weighing (for mass) the BioSampler or analyzing its aqueous extract for EC or OC.

For mass measurements, Teflon filters were weighed before and after each field tests using a Mettler 5 Microbalance (MT 5, Mettler-Toledo Inc., Highstown, NJ), under controlled relative humidity and temperature conditions. Filters were weighed immediately at the end of each experiment as well as after a 24-hour equilibration time period. Laboratory and field blanks were used for quality assurance. Filters and filter blanks were weighed twice in order to increase precision. In case of a difference of more than 3 μg between consecutive weightings, a filter was weighed a third time. The Teflon filters were then analyzed by means of ion chromatography to determine the concentrations of particulate sulfate and nitrate. Trace element and metal concentrations for ambient and concentrated PM were determined by analyzing the MOUDI and VACES Teflon filters by means of inductively coupled plasma mass spectroscopy (ICP/MS). This analysis was conducted by the Monitoring and Laboratory Division of the California Air Resources Board.

The EC concentrations were determined by thermo-analysis. A slice of approximately 0.2 cm^2 from each filter was placed in a platinum boat containing MnO_2 . The sample was acidified with an aliquot of HCl and heated to 115 $^\circ\text{C}$ to dehydrate the sample, and form CO_2 as an index of particle-associated carbon. The boat was then inserted into a dual zone furnace, where MnO_2 oxidized Organic Carbon at 550 $^\circ\text{C}$ and Elemental Carbon at 850 $^\circ\text{C}$. A Flame Ionization Detector (FID) converted the CO_2 combustion product to CH_4 for detection. This analytical method is more elaborately described by Fung (1990).

Effect of Condensation and Evaporation in the VACES on Agglomerate Structure:

A significant fraction of the ultrafine particles in the Los Angeles atmosphere are agglomerate structures, mainly emitted from diesel and other high temperature sources. Agglomerate structures have higher surface areas than spherical particles with the same equivalent diameter; agglomerate transport properties in both gas and liquid phases differ from spherical particles as well (Friedlander, 2000). These differences in surface area and transport properties may influence the biochemical effects of inhaled ultrafine particles. For these reasons, it is important to know whether condensation and evaporation that precedes aerosol concentration in the VACES is likely to affect the morphological properties of the ultrafine particles.

Atmospheric ultrafine particles and those concentrated by the VACES were sampled using a low-pressure impactor (LPI) on the UCLA campus, in west Los Angeles. Concentrated ultrafine aerosols generated by the VACES were sampled after they were dried by diffusion. The LPI is an eight-stage single jet impactor equipped with a critical orifice that maintains a flow rate of 1 LPM under the appropriate pressure drop (Hering *et al.*, 1978 and Hering *et al.*, 1979). The stages have 50% efficiency cutoffs in aerodynamic diameter of 4.0, 2.0, 1.0, 0.5, 0.26, 0.11, 0.075, and 0.05 μm for stages one to eight, respectively. The particles were collected on a nickel grid. To minimize the effects of particle bounce, only one stage at a time had a grid attached for sampling; the grid is secured at the center of a 25 mm diameter glass stage, while the other glass stages are coated with apiezon grease. Air is drawn through the impactor by a vacuum pump for 5 minutes per stage. Analysis was done for changes in structural characteristics of the agglomerate fraction. These agglomerates were collected on stages 7 and 8, which have particle aerodynamic diameter ranges of 0.075 - 0.11 μm and 0.05 - 0.075 μm , respectively. Transmission Electron Microscope

(TEM) photomicrographs of the grids were taken using a JEOL 100CX and 2000FX TEM at a magnification of 10^5 . The morphology of ultrafine particles ($d_p < 0.10 \mu\text{m}$) was characterized using the fractal concept applied to TEM micrographs. More details on fractal analysis can be found in Xiong (2000).

Experiments and simulations have shown that the fractal concept can be applied to aggregates of nanometer primary particles (Forrest and Witten, 1979; Witten and Sander, 1981). In applying the fractal concept, the fractal dimension and the prefactor for both ambient and concentrated particles were calculated. The fractal dimension (D_f) is a measure of the stringiness of the agglomerate and the prefactor (A) is a measure of denseness of the agglomerate. An agglomerate with the same fractal dimension as another may have a higher prefactor if it contains a higher primary particle overlap. Agglomerates produced by computer simulation algorithms help in the understanding of structure and fractal dimension (Friedlander, 2000). Figure 8 illustrates two examples for diffusion limited aggregation. An agglomerate with a chain-like structure has a lower fractal dimension than a more compact, spheroidal one. The structure of an agglomerate with a D_f value of 1 is a linear chain of primary particles. For a D_f value of 2 the agglomerate structure is a two-dimensional arrangement of closely packed primary particles with six nearest neighbors; and the structure for a D_f value of 3 is a three-dimensional closely packed sphere. The agglomerate fractal dimension and prefactor arise from the following relationship (Weber et. al., 1995):

$$N_p = A \left(\frac{R_g}{R_o} \right)^{D_f} \quad (1)$$

where D_f is the fractal dimension, N_p is the number of primary particles in the aggregate, A is the fractal pre-factor or structural coefficient, R_o is the average primary particle radius, R_g is the radius of gyration. The radius of gyration can be calculated using the relation: $[(1/M)\Sigma(m_i r_i^2)]^{1/2}$, where m_i is the mass of the i^{th} primary particle, M is the total mass given as Σm_i , and r_i is the distance of the i^{th} primary particle from the center of mass. The fractal dimension and prefactor of randomly sampled ambient and concentrated particles were obtained by plotting the number of primary particles positioned radially from the center of mass to the radius of the gyration of the agglomerate. The fractal dimension was determined from the slope and the prefactor was determined from the inverse log of the intercept of the least squares fit line.

Results and Discussion of the Field Study:

In each of the sampling lines of the VACES, coarse, fine and ultrafine particles were concentrated from an intake flow of 110 to a minor flow of $5 \text{ l}\cdot\text{min}^{-1}$. Thus the ideal concentration enrichment factor for any chemical PM species is expected to be 22. Results from these field tests are summarized in Tables 1-3 and in Figures 9-14. In each figure, the concentrations determined by the VACES are compared to those determined by the MOUDI; the coordinates are fit by a linear regression and the tightness-of-fit by correlation coefficients. The slopes of the regression lines thus provide an average estimate of the overall concentration enrichment factor obtained by means of the VACES for a given PM fraction and species.

Table 2 presents the sulfate and Table 3 the nitrate content of the coarse fraction of ambient (MOUDI) and concentration-enriched ambient aerosol (VACES). The corresponding

enrichment factor (defined as the ratio of the VACES coarse aerosol concentration to that of the MOUDI) is presented for each of 5 samples. Figure 9 presents paired measurements of ambient coarse aerosol mass concentrations obtained by the MOUDI versus those concentrated by the VACES.

As indicated by the slope of the regression line in Figure 9 the average concentration enrichment of the VACES is $22.5 (\pm 3.8)$, thus, very close to the ideal value of 22. The rather limited data obtained for coarse particle sulfate and nitrate (Tables 1a and 1b, respectively) also indicate a close agreement between the VACES and MOUDI, with the concentration enrichment factors for sulfate and nitrate of $22.1 (\pm 4.9)$ and $19.9 (\pm 2.6)$, respectively. The limited data for these inorganic ion measurements are due to the very low nitrate and sulfate content within the coarse fraction of PM in the specific Los Angeles location. Thus, a greater level of uncertainty exists in the measurements made with the MOUDI (which samples at about one fourth of the flow rate of each sampling line of the VACES). Nevertheless, the overall agreement between the VACES and the MOUDI for coarse particle mass, sulfate, and nitrate is near “ideal”.

Figure 10 shows the $PM_{2.5}$ mass concentrations measured by the MOUDI and VACES. The overall concentration enrichment obtained for the fine PM mode is slightly higher (25.6 ± 3.7) than the ideal value of 22, as indicated by the slope of the regression line. As further discussed below, this may be due, in part, to losses of volatile species, such as ammonium nitrate, from the MOUDI substrates in the lower stages. Evidence of this phenomenon was not the case for the coarse PM collected in the upper stage of MOUDI, where the pressure drop is much lower than the smaller cutpoint stages. Moreover, coarse particulate nitrate in south and western Los Angeles (i.e., areas closer to the coast) is mostly associated with stable sodium nitrate (Noble and Prather, 1996).

These experiments were conducted during the months of May and June 2000, with temperatures averaging $32 (\pm 3) ^\circ\text{C}$ and low relative humidity values (i.e., about 35% or less). These conditions have been shown to favor loss of ammonium nitrate from impactor samplers (Chang *et al.*, 2000; Zhang and McMurry, 1987) due to the higher values of the dissociation constant of ammonium nitrate. For this temperature and humidity range, the study by Chang and colleagues (2000), which was conducted at the same site, presented that the total losses of nitrate from the MOUDI averaged between 40-60%. Furthermore, a previous study by Kim *et al.* (2000) showed that concentration enrichment through a smaller-scale portable fine PM concentrator, which has similar design parameters to that of the VACES (in terms of aerosol saturation and cooling temperatures), occurs without any measurable loss of particulate nitrate, despite heating and saturation of the aerosol to about $35 ^\circ\text{C}$. In that earlier study, ambient nitrate concentrations were determined by means of the Harvard/EPA Annular Denuder System (HEADS;), used as the reference sampler. The HEADS measures total particulate nitrate without losses (Koutrakis *et al.*, 1988). Thus, any bias in the enrichment factor of ammonium nitrate above the “ideal” value (i.e., inlet-flow divided by minor-flow), may be due to its losses in the MOUDI, and concerns of a negative bias, due to potential ammonium nitrate losses in the saturation-condensation segment of the VACES, which may be masked by this effect, is discussed below.

Ammonium nitrate dissociates to ammonia and nitric acid, with its dissociation constant increasing exponentially with temperature. However, the dissociation constant decreases sharply

as the relative humidity (RH) exceeds 90-95% (Stelson and Seinfeld, 1982). For example, at 50°C and at RH=95%, the dissociation constant of ammonium nitrate is approximately 7 ppb, which is also the value of the dissociation constant at 18°C, and RH = 50%. Therefore, despite the increase in the aerosol temperature (which would have increased, exponentially, the value of the dissociation constant), aerosol exposure to high water vapor conditions in the VACES seems to prevent nitrate losses due to volatilization.

These conclusions are further supported by the results shown in Figure 11, where the PM_{2.5} nitrate concentrations measured by means of the MOUDI are compared to those measured by the VACES. The average concentration enrichment based on nitrate is 43.8 (± 20.3), roughly twice the value of the ideal concentration enrichment. Given that nitrate losses depend significantly on several parameters such as temperature, humidity and overall particle concentration, the MOUDI-to-VACES agreement should be highly variable, which is indicated by the somewhat lower correlation coefficient (R²=0.66) of the VACES vs. MOUDI data.

By comparison, the concentration enrichment obtained for the non-volatile fine particulate sulfate (shown in Figure 12) was 19.8 (± 4.3) and thus in very good agreement to the ideal value of 22. The above results confirm that the disparity between the ideal and actual concentration enrichment factors based on nitrate is due to sampling artifacts of the MOUDI.

The results plotted in Figure 12 also show that there is no significant difference (p=0.38) in the sulfate-based concentration enrichment values obtained with the *in vivo* version of the VACES (in which concentrated particles are dried by diffusion and collected on filters) and the *in vitro* version (in which particles are collected by the BioSampler). The concentration enrichment obtained by means of the BioSampler was 21.2 (± 3.5), compared to 18.9 (±2.5) obtained using the diffusion-dried concentrated particles collected on Teflon filters. Given the high values and random nature (due to meteorological factors) of nitrate losses within the MOUDI during the sampling period, a similar comparison of the *in vivo* and *in vitro* versions of the VACES based on fine particulate nitrate would be difficult, if not meaningless.

The MOUDI fine PM mass concentrations were corrected for nitrate losses as follows:

$$PM_{corr} = PM_{MOUDI} + 1.29 \left(\frac{NO_{3,VACES}}{22} - NO_{3,MOUDI} \right) \quad (2)$$

where NO_{3,VACES} and NO_{3,MOUDI} are the nitrate concentrations measured by the VACES and MOUDI, respectively, and PM_{MOUDI} is the total MOUDI fine PM mass concentration determined gravimetrically. The above equation assumes that all nitrate found in the fine particulate mode is associated with ammonium nitrate. The corrected values of the MOUDI mass concentrations are also shown in Figure 10, along with the adjusted concentration enrichment factor. The nitrate-adjusted concentration enrichment factor becomes 22.8 (±3.4), thus very close to the ideal enrichment value of 22. These results imply that the discrepancy between the PM_{2.5} mass concentrations between VACES and MOUDI can be entirely attributed to the difference in the nitrate concentrations measured by these two systems.

The results of Figure 11 also indicate that the overall impact of nitrate losses from the MOUDI substrates on the mass concentration determined by the MOUDI is rather small. This is because ammonium nitrate accounts on the average for 30 - 40% of the total PM_{2.5} mass concentration at Downey, CA (Sioutas *et al.*, 2000). Thus, even if nitrate losses are as high as 50%, the overall difference between the uncorrected and nitrate-adjusted mass concentrations is not substantial, as indicated by the data presented in Figure 10.

Results from the concentration enrichment obtained for selected trace elements and metals are shown in Figure 13. Due to the low ambient concentrations of trace elements and metals measured by the MOUDI, quantifiable concentration enrichment values were obtained only for the following metals: Mg (coarse PM only), Al, K, Ca, and two iron isotopes (i.e., Fe⁵⁶ and Fe⁵⁷). Measurable amounts of Zn, Cu, Ni and Mn were also identified in the filters connected to the fine concentrator of the VACES, but not in the corresponding MOUDI stages. The average and the standard deviation values of concentration enrichment shown in Figure 13 correspond to seven (of ten) field experiments. In the remaining four field tests, the ambient concentrations of the aforementioned metals were either comparable to the blank content of the Teflon filters or lower than the ICP/MS limit of detection (defined as three times the standard deviation of the laboratory blank filters).

The data in Figure 13 indicate that the Al, K, Ca, Fe⁵⁶ and Fe⁵⁷ content of fine and ultrafine PM is enriched by a factor of 21.2 (± 4), 19.4 (± 3.3), 22.1 (± 3.8), 24.3 (± 3.1) and 22.4 (± 3.4), respectively. Similarly, the Mg, Al, K, Ca, Fe⁵⁶ and Fe⁵⁷ content of coarse PM is enriched by a factor of 18.6 (± 4.2), 20.4 (± 3.3), 19.3 (± 3.8), 18.3 (± 4.2), 22.1 (± 3.4) and 21.6 (± 3.5), respectively. These concentration enrichment values are also close to the ideal enrichment value of 22, thereby indicating that the concentration enrichment process preserves the concentrations of these elements and trace metals in both coarse and fine PM.

Table 4 shows the concentration enrichment achieved by the ultrafine concentrator of the VACES based on particle counts, using a condensation particle counter (3022 CPC; TSI Inc., St. Paul, MN). The first column of Table 4 shows the ambient concentration based on particle counts; the second column shows that the number concentration, measured immediately downstream of the 0.18 μm cut-point impactor; and the third column corresponds to the particle number concentrations measured immediately downstream of the diffusion dryer of the ultrafine concentrator of the VACES. The fourth column shows the ratio of particle counts downstream to that upstream of the 0.18 μm impactor, indicating that about 84% of ambient particle counts are associated with particles smaller than that size. The final column of Table 4 shows the concentration enrichment obtained for ultrafine particles, defined as the ratio of the count-based concentration downstream of the VACES to that downstream of the 0.18 μm impactor. The overall concentration enrichment for ultrafine particles was 20.8 (± 1.4), thereby indicating that ultrafine particles are concentrated with very high efficiency by the VACES.

Earlier investigations of the size distribution of ambient elemental carbon (EC) in Los Angeles (Venkataraman and Friedlander, 1994), showed that EC displays a bimodal size distribution, with peaks within the 0.05 - 0.12 μm (mode I) and 0.5 - 1.0 μm (mode II) size ranges. Mode I was attributed to primary emissions from combustion sources while mode II was attributed to the accumulation of secondary reaction products on primary aerosol particles. Mode I contained 75

– 85 % of EC, by mass, in the Los Angeles air basin during the summer season. Therefore, the performance of the ultrafine particle concentrator of the VACES was characterized by further comparing EC concentrations obtained with the VACES to those measured in the afterfilter of the MOUDI (collecting 0- 0.18 μm particles).

Results from these field comparisons are shown in Figure 14. Similar to the results based on particle count and mass concentrations, a high level of comparability resulted between the VACES and MOUDI EC concentrations, with the average concentration enrichment factor being 22.2 (± 2.3). Ultrafine particle EC concentrations obtained by means of the MOUDI and VACES are also very highly correlated ($R^2 = 0.94$).

It should be noted that the ability of the VACES to enrich the concentrations of all particles in the fine mode (including its ultrafine component) is a particularly important feature of this technology, as it enables inhalation toxicologists to conduct exposures to any selected sub-range of $\text{PM}_{2.5}$. For example, previous studies in California presented the presence of two sub-modes within the accumulation mode of ambient PM (Hering et al., 1997; John et al., 1990). One mode peaks at around 0.2 μm , consisting mainly of gas-to-particle reaction products, such as carbonaceous PM and the other peaks at about 0.7 μm , mainly associated with hygroscopic PM species, such as ammonium sulfate and ammonium nitrate. These observations have been confirmed by our recent yearlong measurements at the facility of Rancho Los Amigos in Downey (Sioutas et al, 2000). By thus placing a conventional impactor upstream of the fine concentrator of the VACES, having for example a 0.35 μm cutpoint, inhalation studies could be conducted to ultrafine PM plus the elemental and organic carbon content of the accumulation mode, however, without the majority of its sulfate and nitrate constituents.

Effect of Condensation and Evaporation in the VACES on Agglomerate Structure:

Changes in agglomerate structure were investigated by comparing the fractal dimension and the prefactor of concentrated ultrafine particles from the VACES to ambient particles. Our results, shown in Figures 15 and 16, indicate that the concentrated and ambient particles show very similar morphology. The fractal dimension and prefactor values were determined for a total of 38 ambient and 39 concentrated ultrafine particles. Figures 15 and 16 show the fractal dimension value distributions for concentrated and ambient aerosols, respectively. The count median fractal dimension is very similar (between 1.6 and 1.8) for both concentrated and ambient particles. Furthermore, the average prefactor for the particles collected from the VACES is 2.73 and for the atmospheric is 2.83. Higher prefactor values are typically associated with denser agglomerates, but similar to what was found with the fractal dimension, the difference between the concentrated and atmospheric aggregate prefactor is not significant.

Previous research suggests that chain agglomerates may become more compact when subjected to condensation and evaporation processes (Colbeck *et al.*, 1990; Hallet *et al.*, 1989; Wells *et al.*, 1976). A study shows that for diesel chain-agglomerate particles the fractal dimension increased from 1.56 to 1.76 and 1.40 to 1.54 for mid and low sulfur fuel after condensation and evaporation processes (Huang *et al.*, 1994). However, in our study the average fractal dimension showed practically no change in value following condensation and evaporation in the VACES. An explanation is that in the study by Huang et al., the particles underwent up to 3 cycles of

condensation and evaporation while in our study the particles only went through 1 cycle. We can therefore conclude that the condensation and evaporation process used with the VACES is effective in concentrating the sampled ultrafine particles but causes little change in the compactness or denseness of the particles, as measured by the fractal dimension and prefactor. However, both the sources of the fractal-like structures and associated trace gases may affect this phenomenon. Since the measurements were made for one sampling site, more experiments will need to be made in different sites to make these conclusions generalizable

Finally, Figure 17 shows the concentration enrichment as a function of particle size obtained by measuring the size distributions of ambient aerosols upstream of the VACES and immediately downstream of the diffusion dryer of the VACES line sampling fine PM by means of the SMPS. These experiments were conducted at a minor flow rate of 20 LPM (thus the ideal concentration enrichment is by a factor of 11). Each experiment started by first measuring the ambient particle number concentration by means of the TSI 3022 Condensation Particle Counter for 5 minutes. Subsequently, the concentration immediately downstream of the 0.18 μm impactor was measured for an additional 5 minutes, followed by a concentration measurement downstream of the ultrafine VACES concentrator for 5 minutes. The above cycle was repeated three times in each experiment.

It should be noted that the lowest particle size that could be detected with the specific SMPS configuration was 17 nm. Due to the very low concentration of ambient particles below that size, ambient readings for particle smaller than about 20 nm are somewhat unreliable. Overall, the results of Figure 17 show categorically that there is absolutely no distortion in the size distributions between ambient and concentrated aerosols, as the number median diameters (41 nm) and geometric standard deviation (1.7) of the concentrated and ambient aerosols are virtually identical. These results confirm that drying by diffusion returns the concentrated droplets to their original size with minimal distortion.

3.5 Conclusions for the Laboratory and Field Evaluations of the VACES.

The experimental characterization of the versatile coarse, fine and ultrafine concentrators demonstrated that concentration enrichment does not depend on particle size or chemical composition. Volatile species such as ammonium nitrate are preserved through the concentration enrichment process under the laboratory conditions used in this study. Furthermore, the concentration enrichment based on particle counts showed clearly that no particle coagulation occurs during the enrichment process, for any of the three minor-to-total flow configurations tested.

The ability of the VACES to enrich the concentrations of all particles in the fine mode including its ultrafine particle component enables inhalation toxicologists to conduct exposures to any selected sub-range of PM_{2.5}. For example, previous studies in California showed the presence of two sub-modes within the accumulation mode of ambient PM (Hering et al., 1997; John et al., 1990); one mode peaks at around 0.2 μm consisting mainly of gas-to-particle reaction products, such as carbonaceous PM and the other peaks at about 0.7 μm mainly associated with hygroscopic PM such as ammonium sulfate and nitrate. These observations have been confirmed by our recent yearlong measurements at the facility of Rancho Los Amigos in

Downey. By thus placing a conventional impactor upstream of the fine concentrator of the VACES, having a 0.4 μm cutpoint, inhalation studies could be conducted to ultrafine PM plus the elemental and organic carbon content of the accumulation mode, but without the majority of its sulfate and nitrate constituents.

4. *In vitro* Experiments at UCLA using the Portable Particle Concentrators

In addition to the animal and human exposures, we are currently using the newly developed versatile concentrators for direct PM collection for *in vitro* tests. Collection and chemical characterization of coarse, fine and ultrafine particles for *in vitro* tests using the combined concentrator/BioSampler method will be conducted in the primary SCPCS locations as well as in the sites where animal exposures to freeway originated aerosol. We have already initiated these studies, by collecting PM samples using the VACES at UCLA and at Rancho Los Amigos. In our current sampling scheme, outdoor particles are collected concurrently with human exposure studies for approximately 5-6 hours using autoclaved BioSamplers. The BioSamplers are connected immediately downstream of the ultrafine plus fine particle concentrator and the coarse concentrators of the VACES. Given that particle growth is based on mixing and saturation with warm water vapor, it is imperative that no bacterial growth occurs during the saturation process. Preliminary analysis of the BioSampler extracts has shown that no bacterial growth occurs during the saturation process. In addition to the biological content of ambient or indoor PM, we also monitor the following parameters over the 6 hours of the experiment: particle number concentration (continuously) and particle mass concentration (time-integrated).

5. Current *In Vivo* Experiments using the Portable Particle Concentrators

In addition to the *in vitro* tests described in the previous paragraph, we conducted our first series of animal exposures to ultrafine and fine particles. These studies were conducted jointly by investigators from UCLA, University of Southern California, UC Irvine and UC Davis. Healthy rats were exposed to fine and ultrafine PM, concentrated by a factor of 22, harvested at UCI (in June 2000) and UCLA (in July 2000) in west Los Angeles. Preliminary measurements in the later location have indicated an unusually high number concentration of both ambient as well as indoor PM on the order of 10,000-50,000 particles/cm³, roughly 5-10 times higher than levels typically encountered in urban areas of the East Coast of the U.S., which makes these experiments of particular interest. The same particle sampling protocol, currently followed for the *in vitro* tests, was used to monitor the physico-chemical PM characteristics during the animal exposure studies, monitoring of particle mass, number concentration, elemental composition and selected PAH. Biological analyses from these exposure studies are currently under way.

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Table 1. Comparison of ultrafine mass concentration after the multi-slit impactor of the VACES and MOUDI

Ambient ultrafine mass concentration ($\mu\text{g}/\text{m}^3$)	Multi-Slit Impactor ultrafine mass concentration ($\mu\text{g}/\text{m}^3$)	Ratio of mass concentrations between Multi-slit impactor and MOUDI ^b
1.89	2.48	1.31
2.78	2.47	0.89
3.28	3.16	0.96
3.81	4.13	1.08
4.23	5.05	1.19
<i>Average</i>		1.07
<i>Standard Deviation</i>		0.15

- a. Determined by reference MOUDI sampler.
b. MOUDI Collected particles in the size below 0.18 μm .

Table 2. Coarse Ambient Particle Sulfate Concentrations Determined with the MOUDI and the VACES

Ambient ($\mu\text{g}/\text{m}^3$)	VACES ($\mu\text{g}/\text{m}^3$)	Enrichment Factor
1.0	20.8	21.6
1.4	22.0	15.2
0.8	16.6	20.8
1.4	34.0	24.3
2.1	58.5	28.5
average		22.1
SD		4.9

Table 3. Coarse Ambient Particle Nitrate Concentrations Determined with the MOUDI and the VACES

Ambient ($\mu\text{g}/\text{m}^3$)	VACES ($\mu\text{g}/\text{m}^3$)	Enrichment Factor
4.68	83.85	17.91
6.43	135.87	21.13
3.71	57.80	15.58
6.78	155.77	22.98
5.41	118.28	21.86
	Average	19.90
	SD	2.6

Table 4. Ultrafine PM Number Concentrations Upstream and Downstream of the 0.18 μm Cutpoint Impactor and Downstream of the Ultrafine Concentrator of the VACES. All concentrations are averaged over 30 minutes sampling time.

VACES Particle Number Concentration (particles/ cm^3)	Particle Number Concentration Downstream of the 0.18 μm Impactor (particles/ cm^3)	Ambient Particle Number Concentration (particles/ cm^3)	Ratio of Downstream-to-Upstream the 0.18 μm impactor Concentration (particles/ cm^3)	Concentration Enrichment
551429	26714	32185	83%	20.7
801429	35000	43166	86%	22.9
420000	23000	31750	74%	18.3
600000	29857	33666	85%	20.1
648571	30428	35714	85%	21.3
795714	38285	45142	84%	20.8
574286	26880	31523	86%	21.4
		Average	0.83	20.8
		SD	0.042	1.41

FIGURE LIST

Figure 1a. Versatile Aerosol Concentration Enrichment System (VACES) for concurrent in vivo studies to coarse, fine and ultrafine PM

Figure 1b. Versatile Aerosol Concentration Enrichment System (VACES) for in vitro studies

Figure 2. Particle Collection Efficiency of the 2.5 μm Cutpoint Slit Nozzle Impactor. Flow Rate; 110 LPM.

Figure 3. Pressure drop across the 0.18 μm cutpoint, multi-slit impactor as a function of flow rate

Figure 4. Removal efficiency of multi-slit low-pressure drop impactor as function of particle diameter

Figure 5. Pressure drop across the BioSampler nozzle as a function of flow rate

Figure 6. Particle collection efficiency of BioSampler as a function of particle aerodynamic diameter. Sampling flow rate: 5 LPM.

Figure 7. Characterization of the Versatile Aerosol Concentration Enrichment System for three minor flows. Total intake flow: 220 LPM. Transparent data labels correspond to indoor air (NMD=0.028 μm) ammonium sulfate (NMD=0.16 μm) and ammonium nitrate (NMD=0.36 μm) particles. Solid data labels correspond to PSL particles.

Figure 8. Structure and fractal dimension of agglomerates produced by two computer simulation algorithms (after Schaefer, 1988). Diffusion-limited aggregation was simulated for two subcases, (a) particle-cluster aggregation and (b) cluster-cluster aggregation. Particle-cluster aggregation refers to the release of single particles, which attach to a growing cluster by Brownian diffusion. In cluster-cluster aggregation, agglomerates of primary particles are released and collide by Brownian motion.

Figure 9. Plot of ambient (MOUDI) and VACES Coarse Particle Concentrations

Figure 10. Plot of Ambient (MOUDI) and VACES PM-2.5 Mass Concentrations

Figure 11. Plot of Ambient (MOUDI) and VACES PM-2.5 Sulfate Concentrations

Figure 12. Plot of Ambient (MOUDI) and VACES PM-2.5 Nitrate Concentrations

Figure 13. Concentration Enrichment of Selected Trace Elements and Metals in coarse and fine ambient particles. Average and standard deviation values correspond to seven field experiments.

Figure 14. Plot of ambient (MOUDI) and VACES ultrafine elemental carbon (EC) concentrations

Figure 15. Fractal dimension distribution for agglomerates from the VACES. The count mean D_f value was found to be between 1.6 and 1.8. Samples were taken at the Center for Health Sciences at UCLA using a Low-Pressure Impactor (LPI)

Figure 16. Fractal dimension distribution for agglomerates sampled from the ambient aerosol. The count mean D_f value was found to be between 1.6 and 1.8. Samples were taken at the Center for Health Sciences at UCLA using a Low-Pressure Impactor (LPI).

Figure 17. Size distribution of ambient aerosols before and after the VACES measured by SMPS

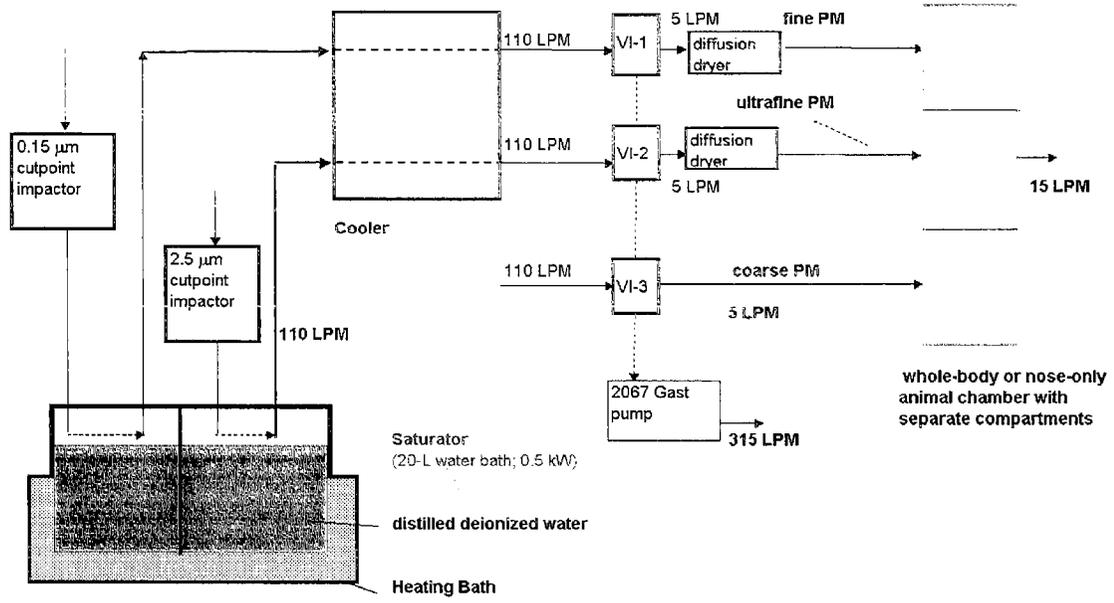


Figure 1a. Versatile Aerosol Concentration Enrichment System (VACES) for concurrent in vivo studies to coarse, fine and ultrafine PM

VI = virtual impactors

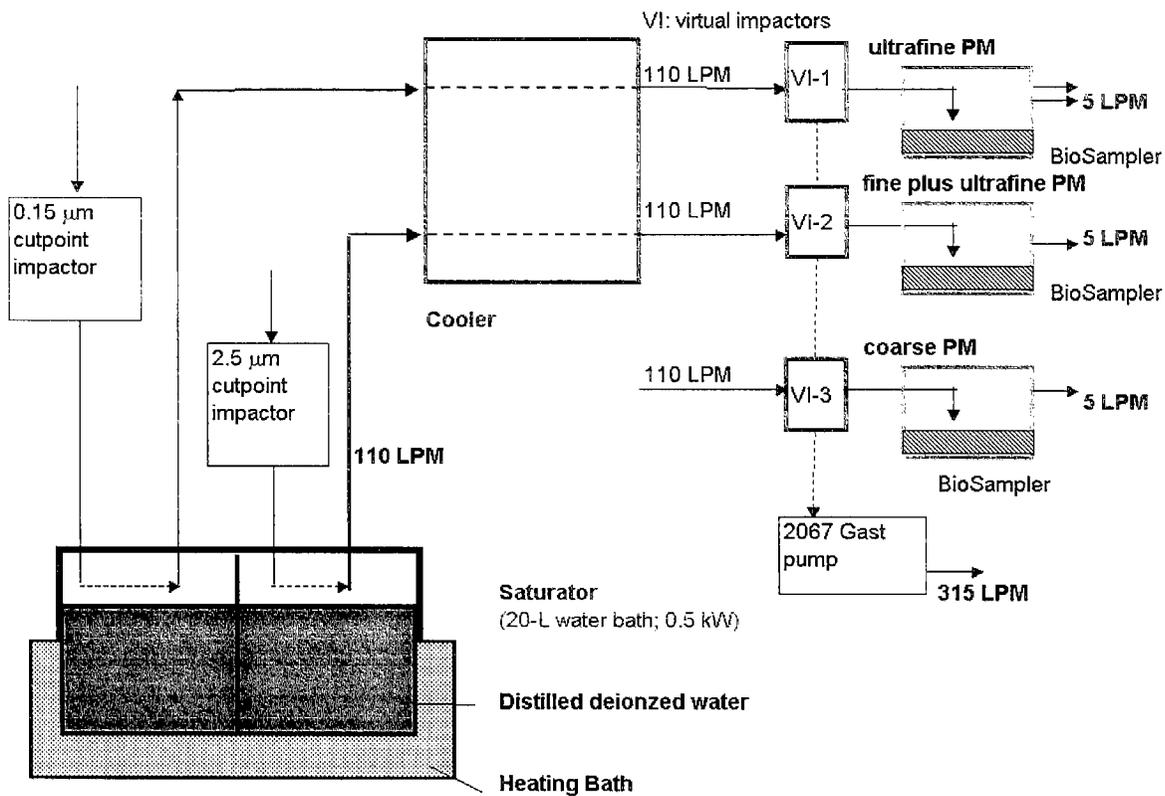


Figure 1b. Versatile Aerosol Concentration Enrichment System (VACES) for in vitro studies

Figure 2. Particle Collection Efficiency of the 2.5 μm Cutpoint Slit Nozzle Impactor. Flow Rate; 110 LPM

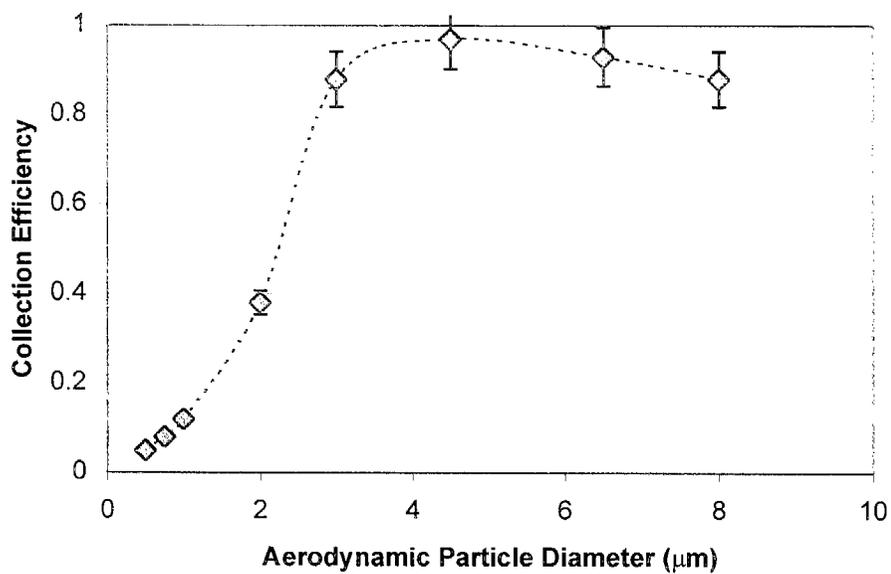


Figure 3. Pressure drop across the 0.18 μm cutpoint, multi-slit impactor as a function of flow rate

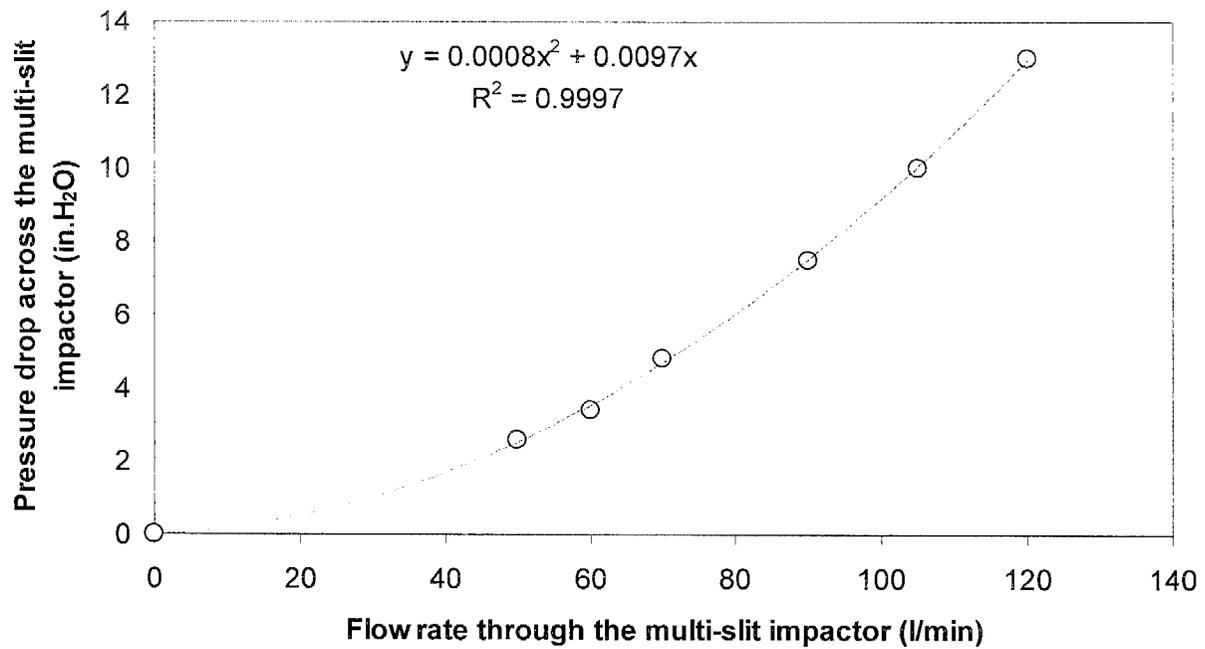


Figure 4. Removal efficiency of multi-slit low pressure drop impactor as function of particle diameter.

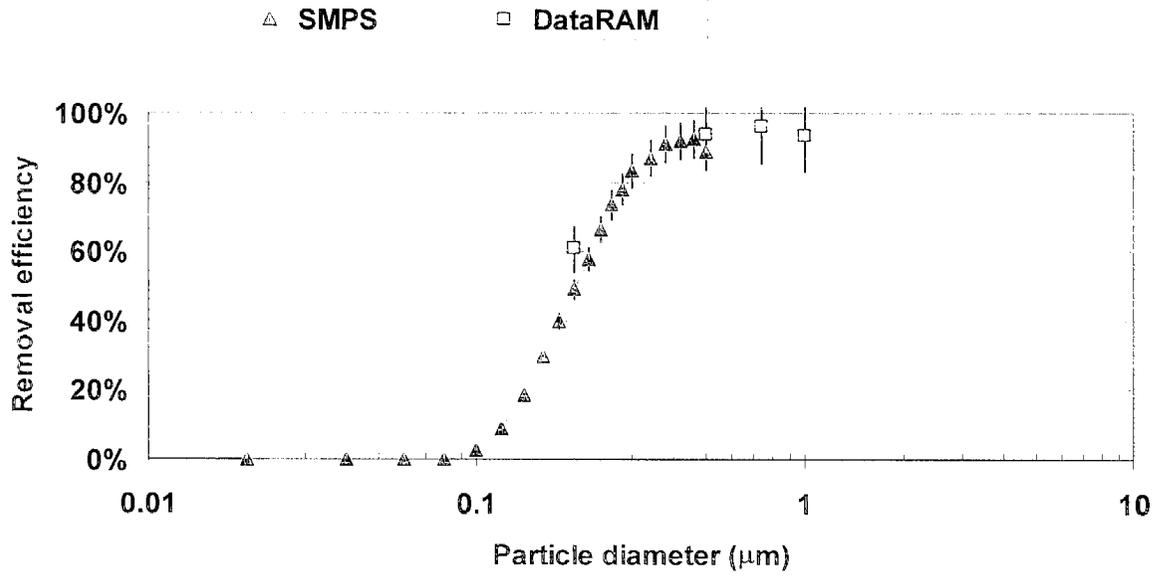


Figure 5. Pressure drop across the BioSampler nozzle as a function of flow rate

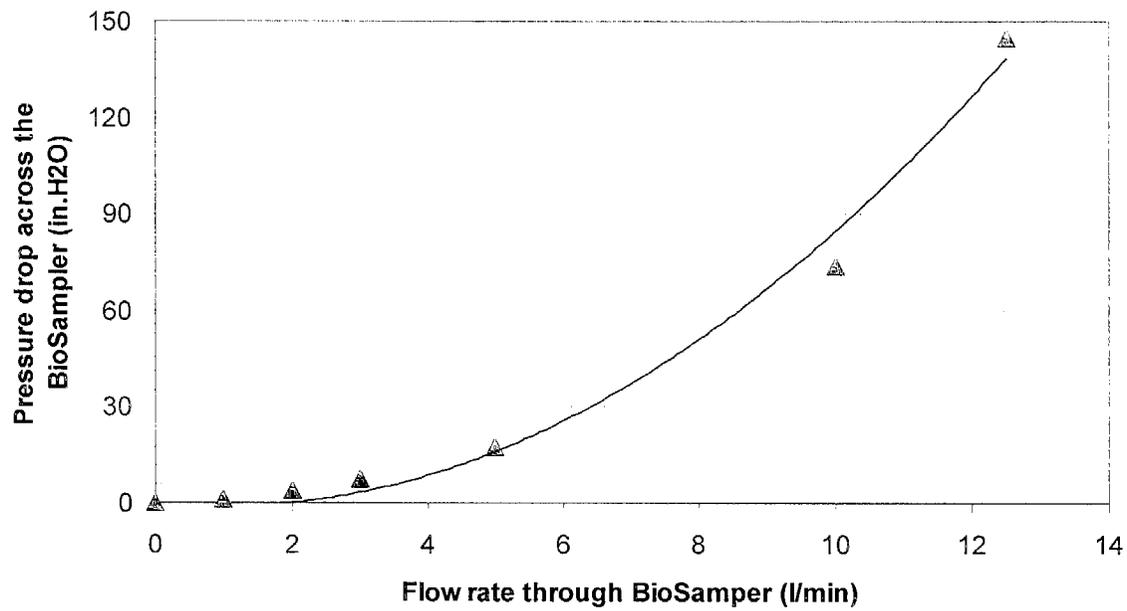


Figure 6. Particle collection efficiency of BioSampler as a function of particle aerodynamic diameter. Sampling flow rate: 5 LPM

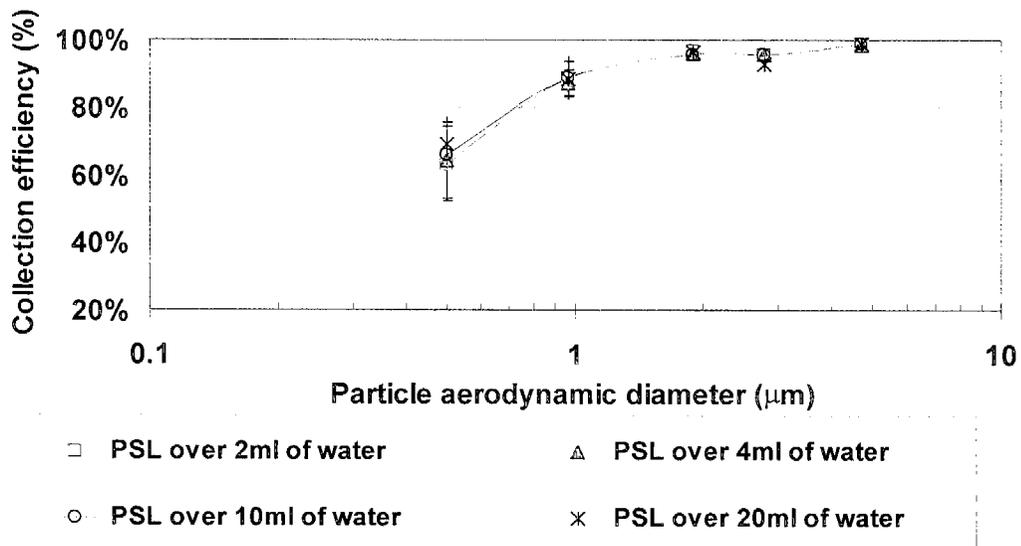


Figure 7. Chracterization of the Versatile Aerosol Concentration Enrichment System for three minor flows. Total intake flow: 220 lmin^{-1}

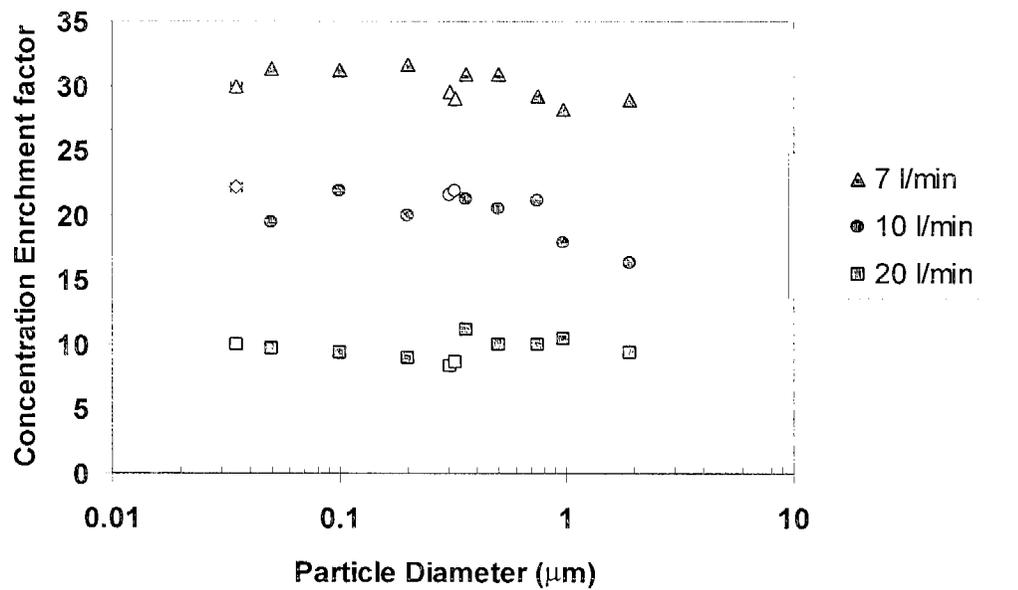
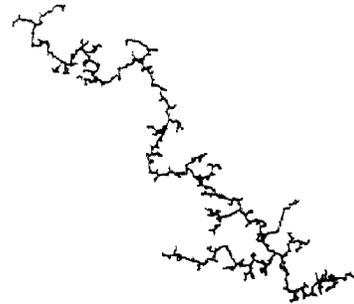


Figure 8. Structure and fractal dimension of agglomerates produced by two computer simulation algorithms (after Schaefer, 1988). Diffusion-limited aggregation was simulated for two subcases, (a) particle-cluster aggregation and (b) cluster-cluster aggregation. Particle-cluster aggregation refers to the release of single particles which attach to a growing cluster by Brownian diffusion. In cluster-cluster aggregation, agglomerates of primary particles are released and collide by Brownian motion.



$$D_f = 2.50$$

(a) particle-cluster aggregation



$$D_f = 1.80$$

(b) cluster-cluster aggregation

Figure 9. Ambient (MOUDI) and VACES Coarse Particle Concentrations

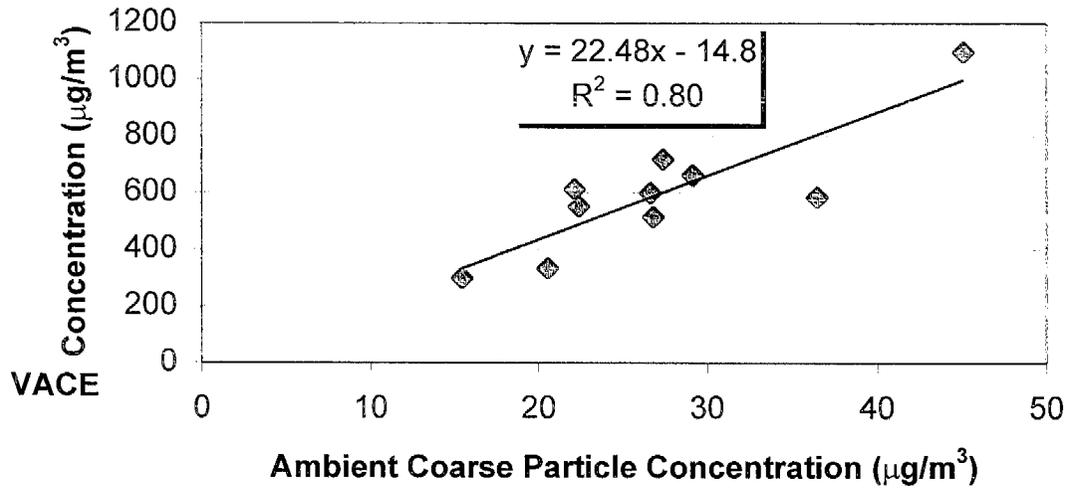


Figure 10. Ambient (MOUDI) and VACES PM_{2.5} Mass Concentrations

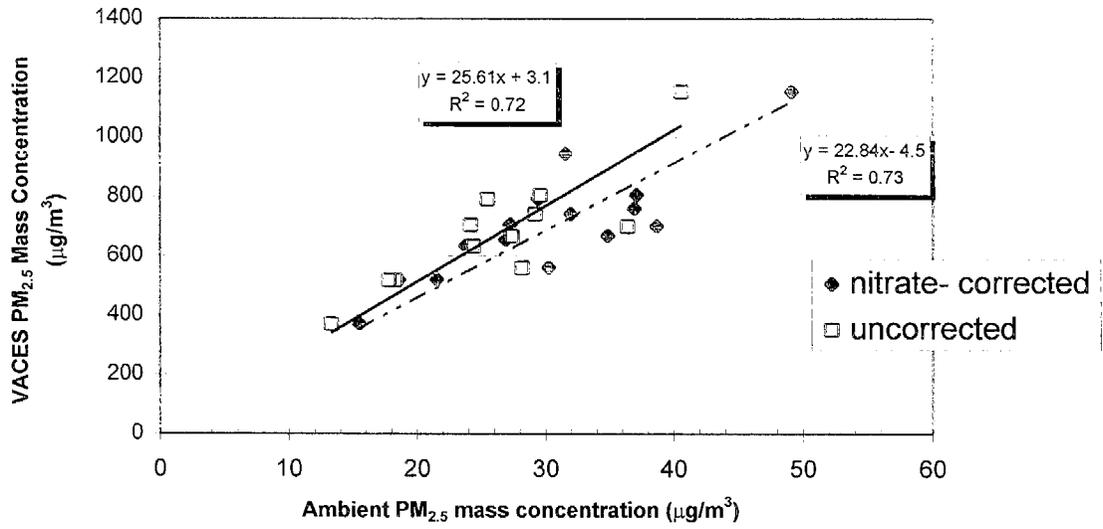


Figure 11. Ambient (MOUDI) and VACES PM2.5 Sulfate Concentrations

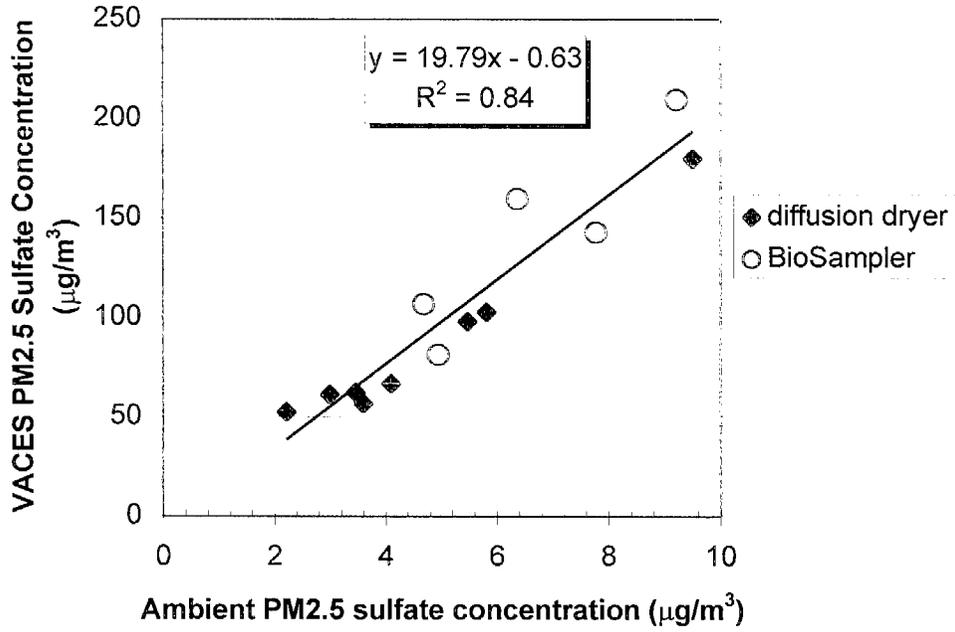


Figure 12. Ambient (MOUDI) and VACES PM2.5 Nitrate Concentrations

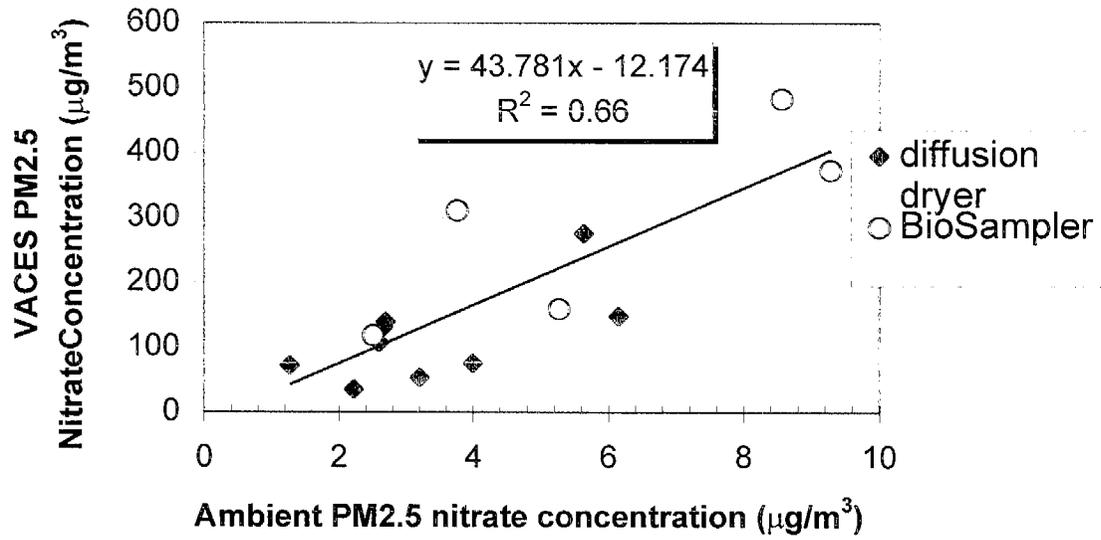


Figure 13

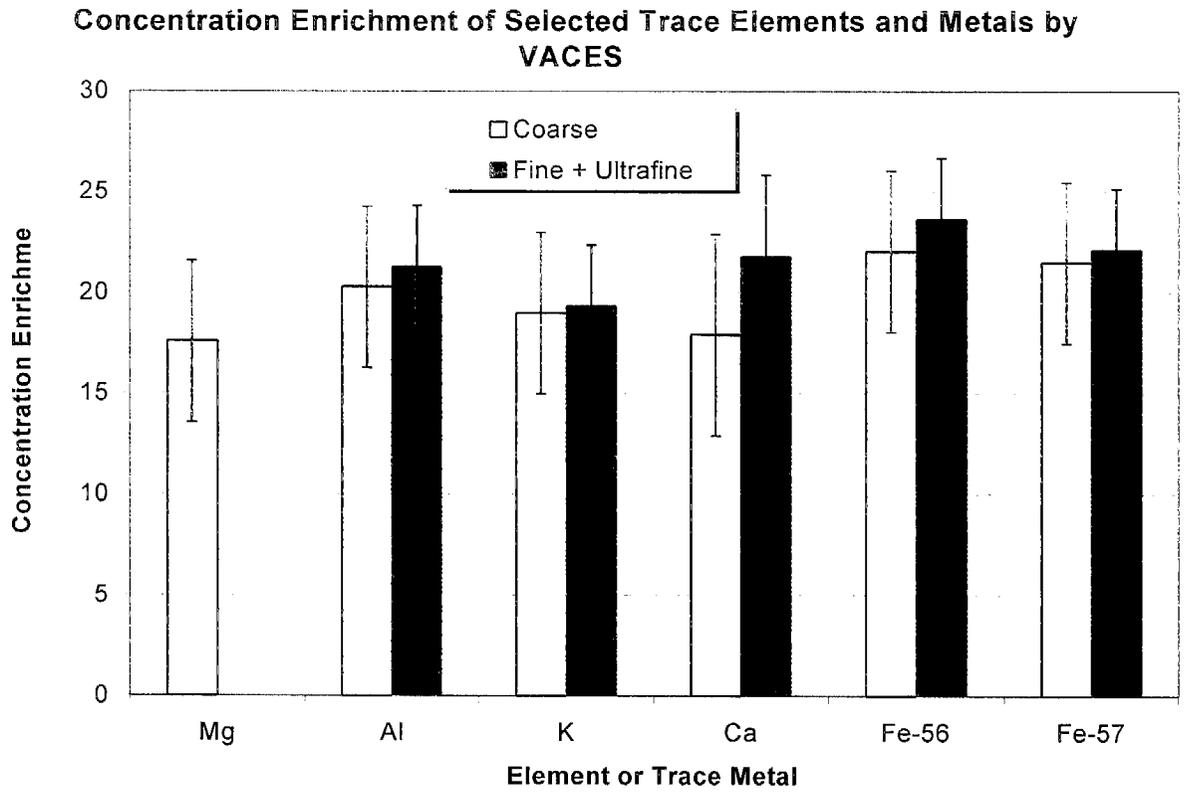


Figure 14. Concentration enrichment of ambient ultrafine particle Elemental Carbon (EC) by VACES

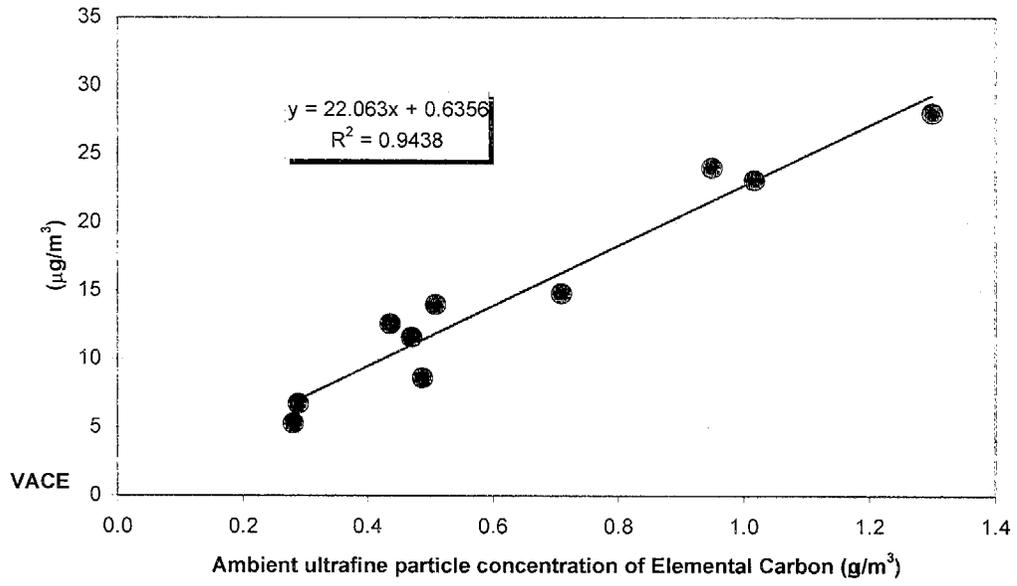


Figure 15. Fractal dimension distribution for agglomerates from the VACES. The count mean D_f value was found to be between 1.6 and 1.8. Samples were taken at the Center for Health Sciences at UCLA using a LPI.

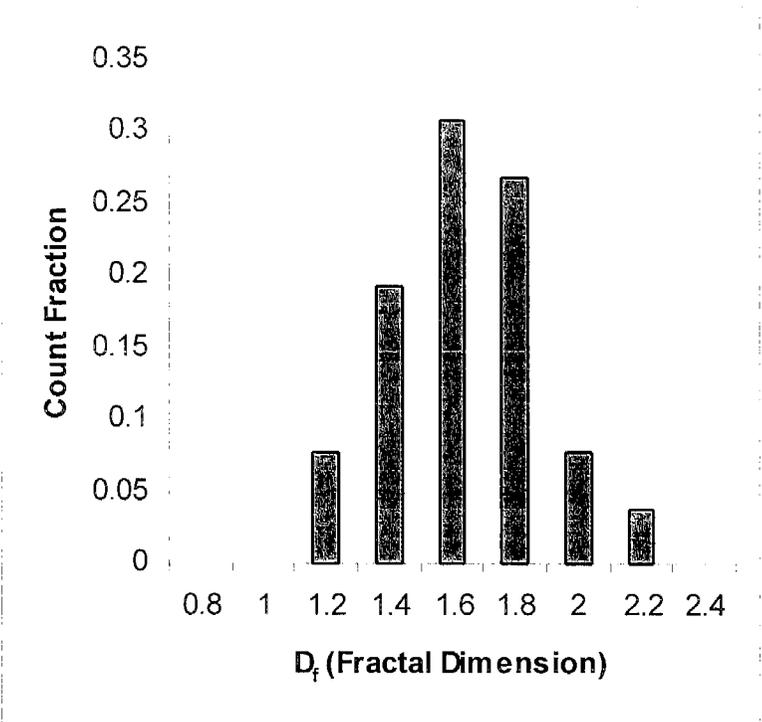


Figure 16. Fractal dimension distribution for agglomerates sampled from the ambient aerosol. The count mean D_f value was found to be between 1.6 and 1.8. Samples were taken at the Center for Health Sciences at UCLA on 8/22/00 using a LPI.

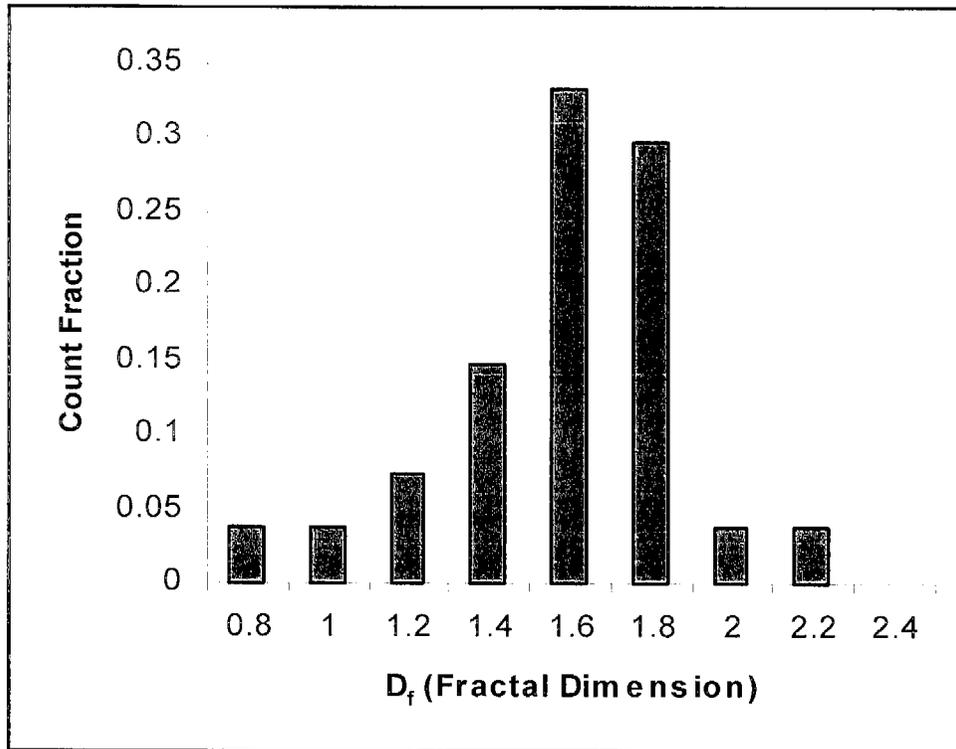
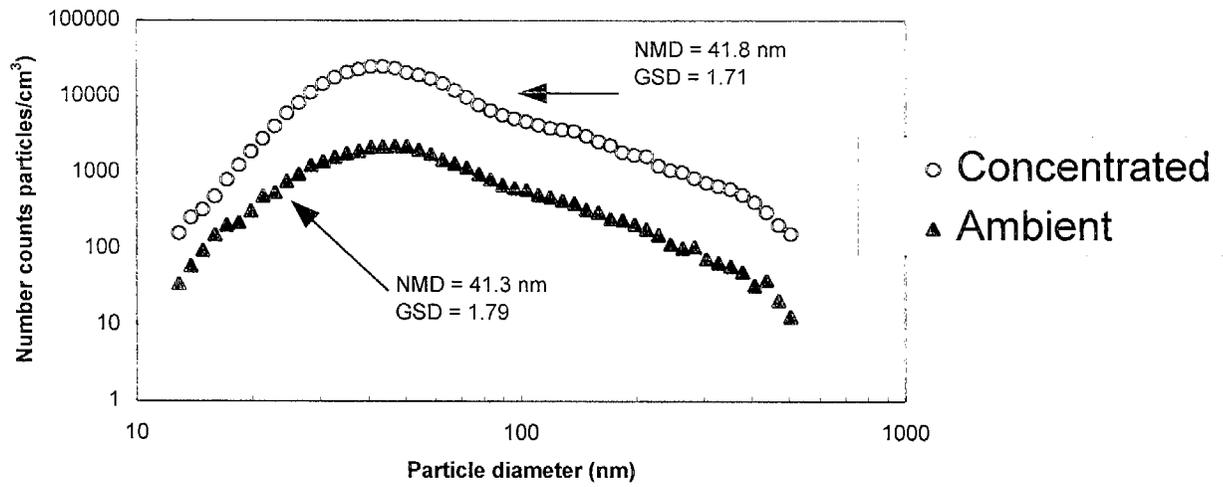


Figure 17. Size distribution of ambient aerosols before and after the VACES measured by SMPS



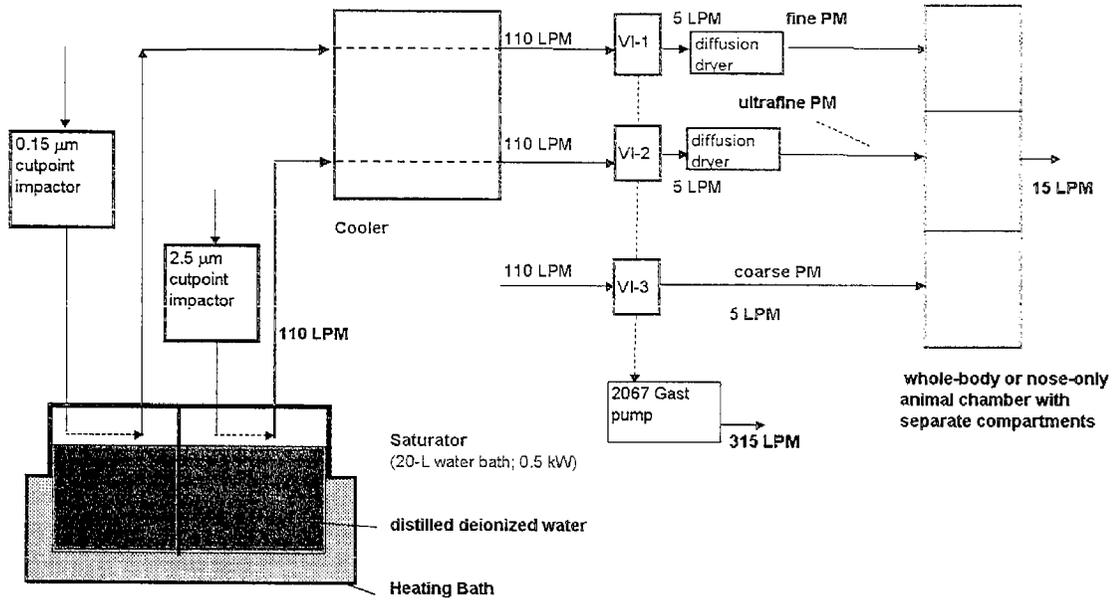


Figure 1a. Versatile Aerosol Concentration Enrichment System (VACES) for concurrent in vivo studies to coarse, fine and ultrafine PM

VI = virtual impactors

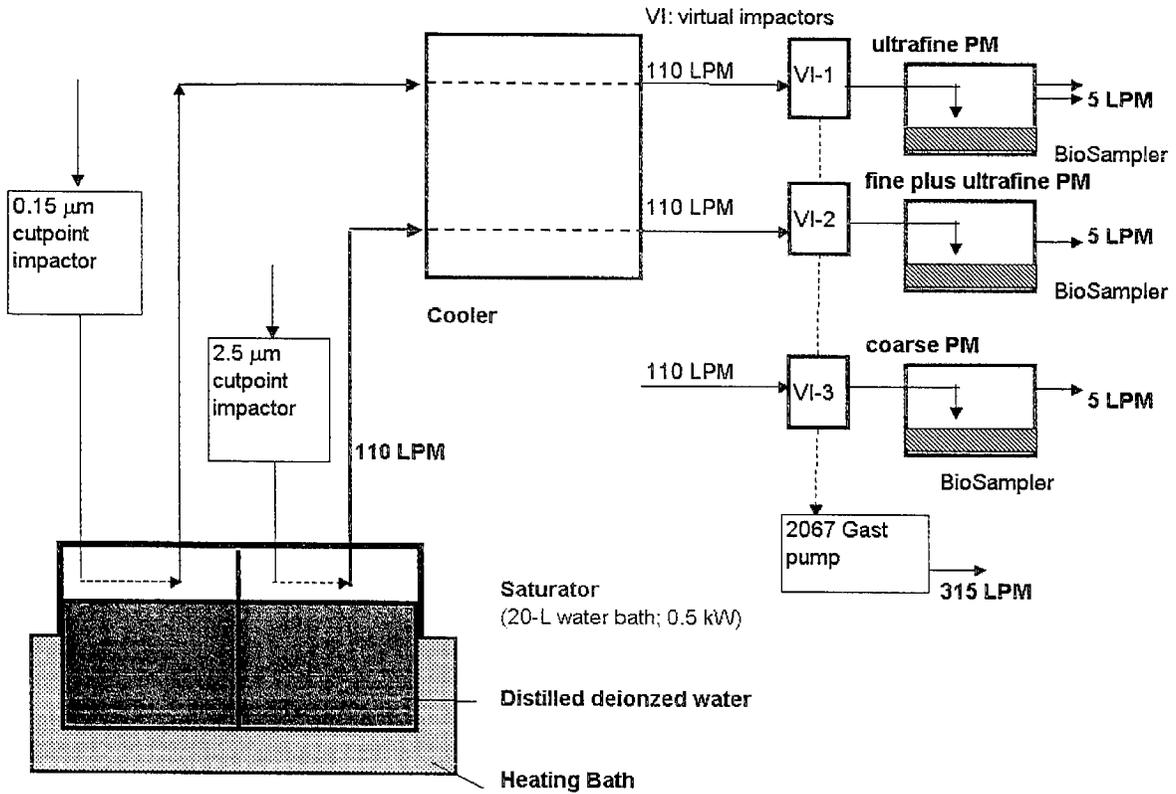


Figure 1b. Versatile Aerosol Concentration Enrichment System (VACES) for in vitro studies

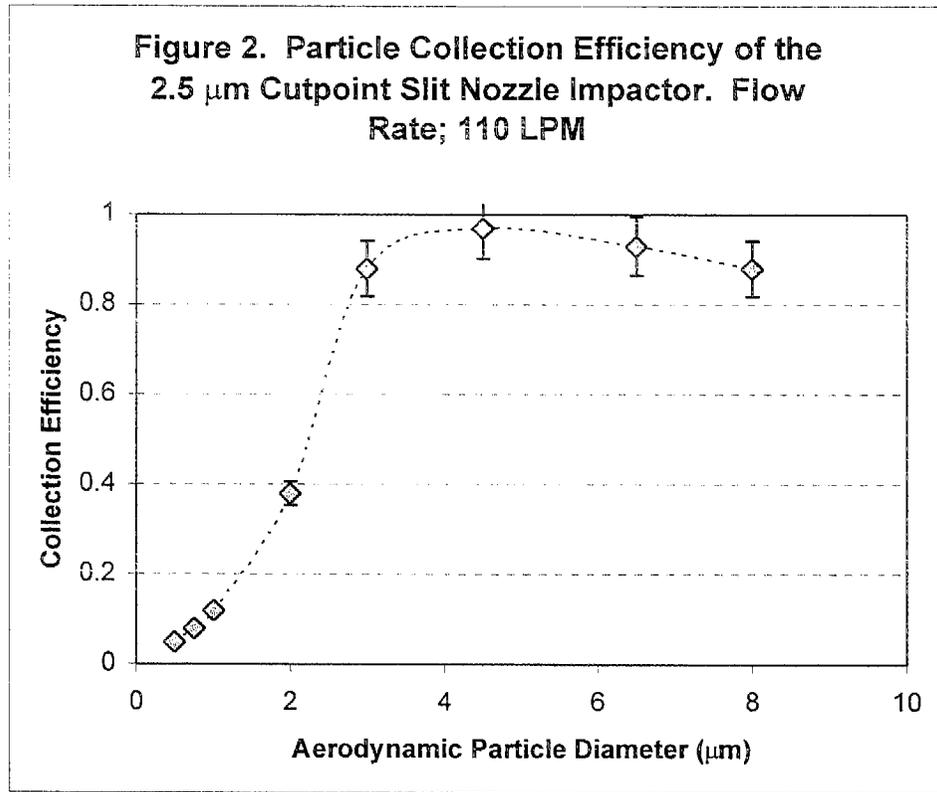


Figure 3. Pressure drop across the 0.18 μm cutpoint, multi-slit impactor as a function of flow rate

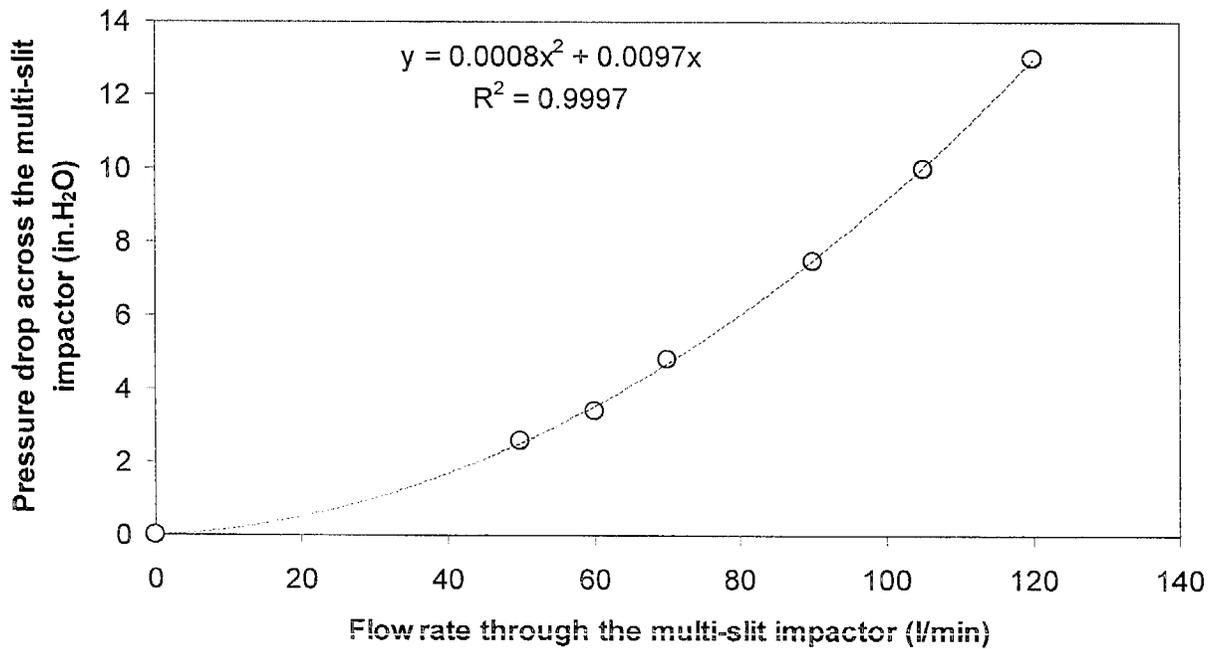


Figure 4. Removal efficiency of multi-slit low pressure drop impactor as function of particle diameter.

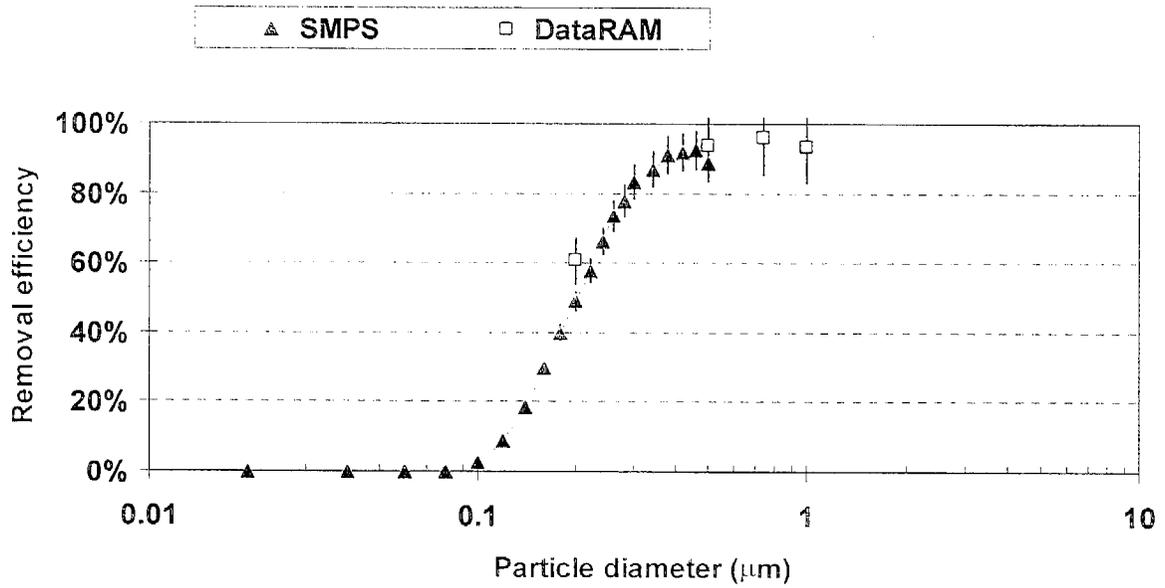


Figure 5. Pressure drop across the BioSampler nozzle as a function of flow rate

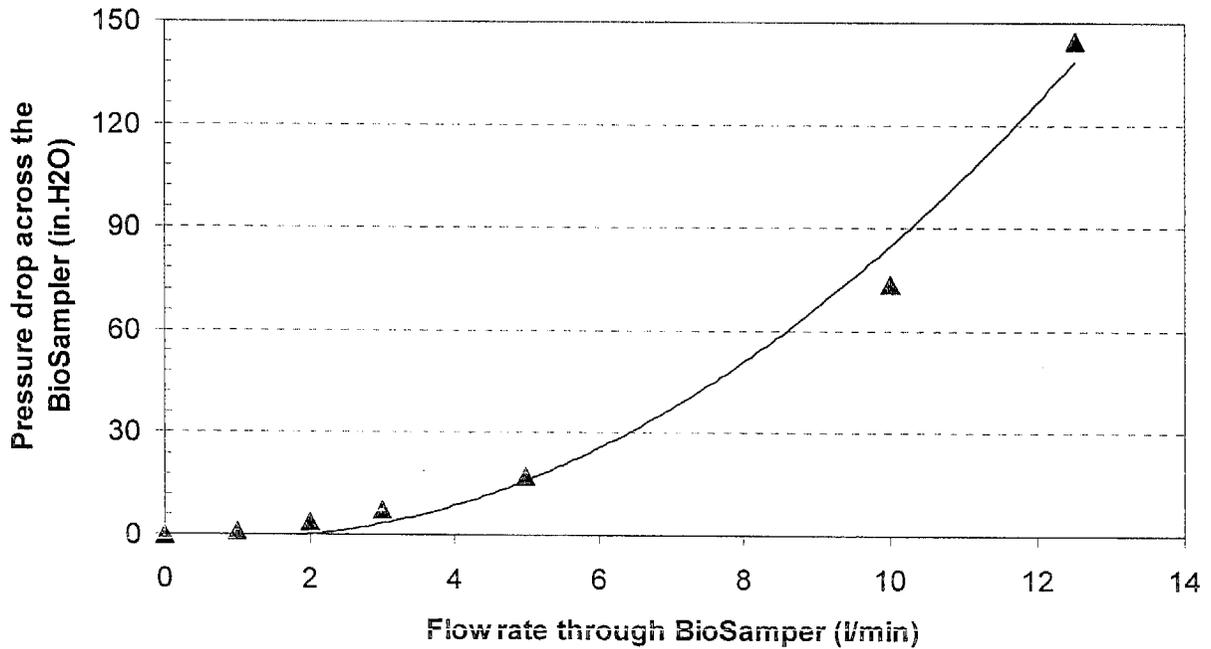


Figure 6. Particle collection efficiency of BioSampler as a function of particle aerodynamic diameter. Sampling flow rate: 5 LPM

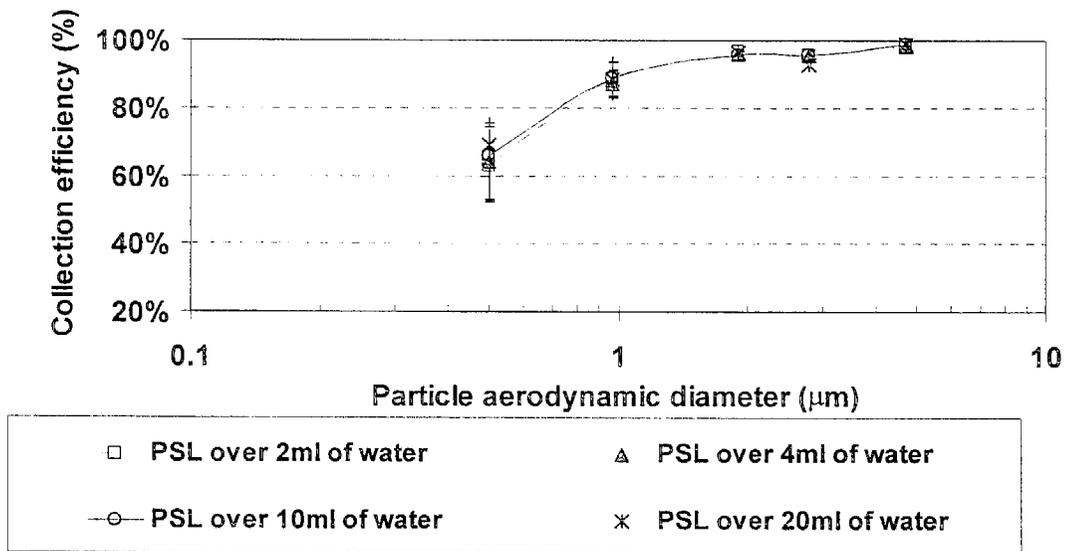


Figure 7. Characterization of the Versatile Aerosol Concentration Enrichment System for three minor flows. Total intake flow: 220 lmin⁻¹

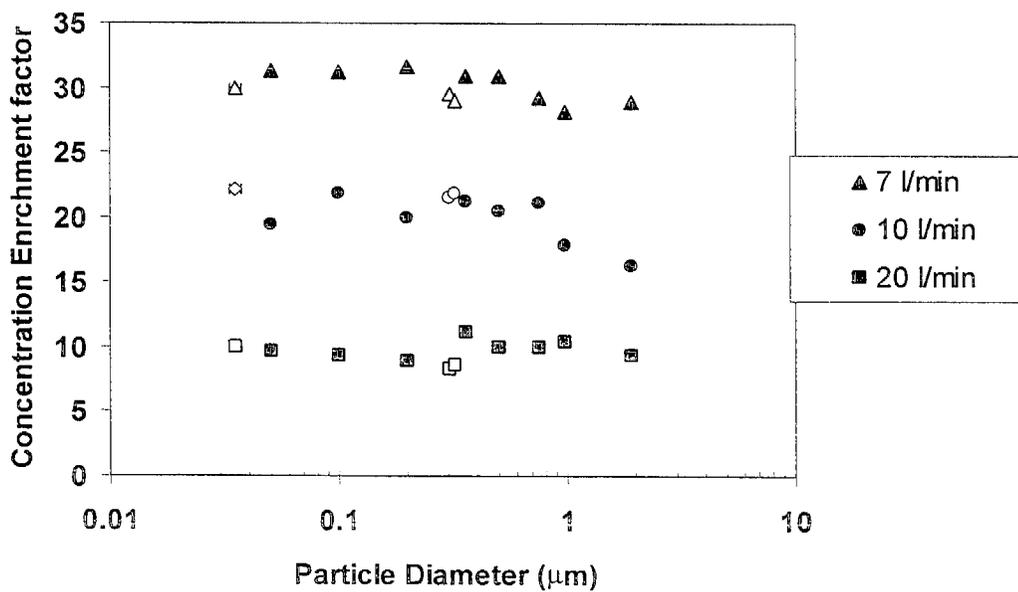


Figure 8. Structure and fractal dimension of agglomerates produced by two computer simulation algorithms (after Schaefer, 1988). Diffusion-limited aggregation was simulated for two subcases, (a) particle-cluster aggregation and (b) cluster-cluster aggregation. Particle-cluster aggregation refers to the release of single particles which attach to a growing cluster by Brownian diffusion. In cluster-cluster aggregation, agglomerates of primary particles are released and collide by Brownian motion.

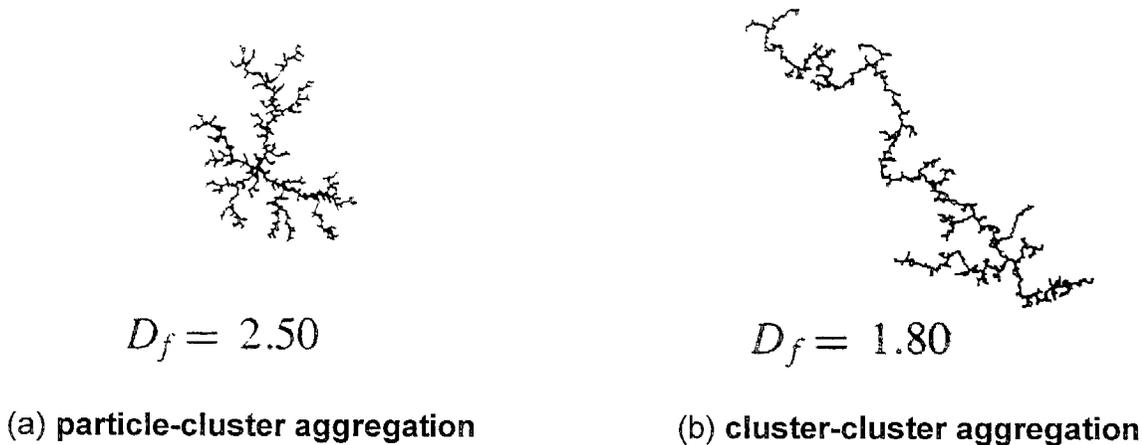


Figure 9. Ambient (MOUDI) and VACES Coarse Particle Concentrations

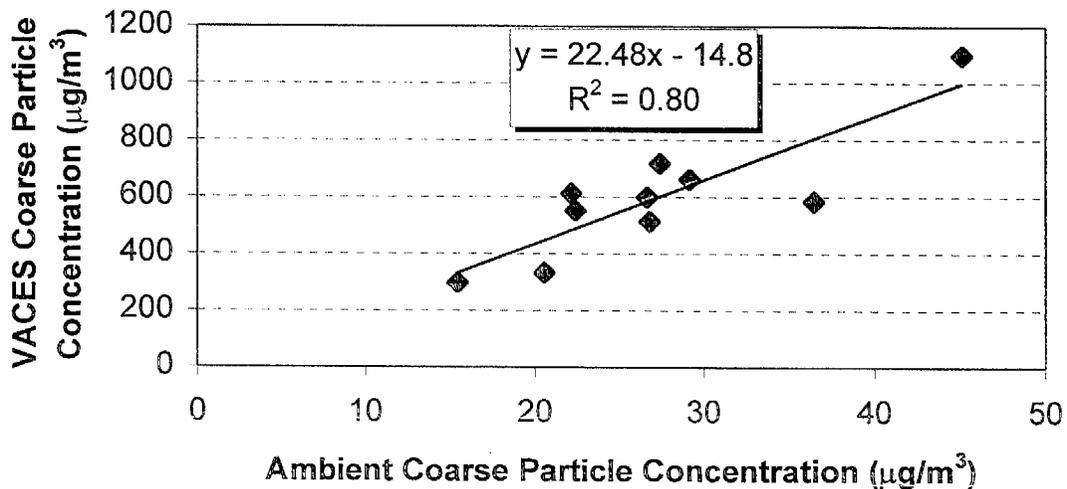


Figure 10. Ambient (MOUDI) and VACES PM_{2.5} Mass Concentrations

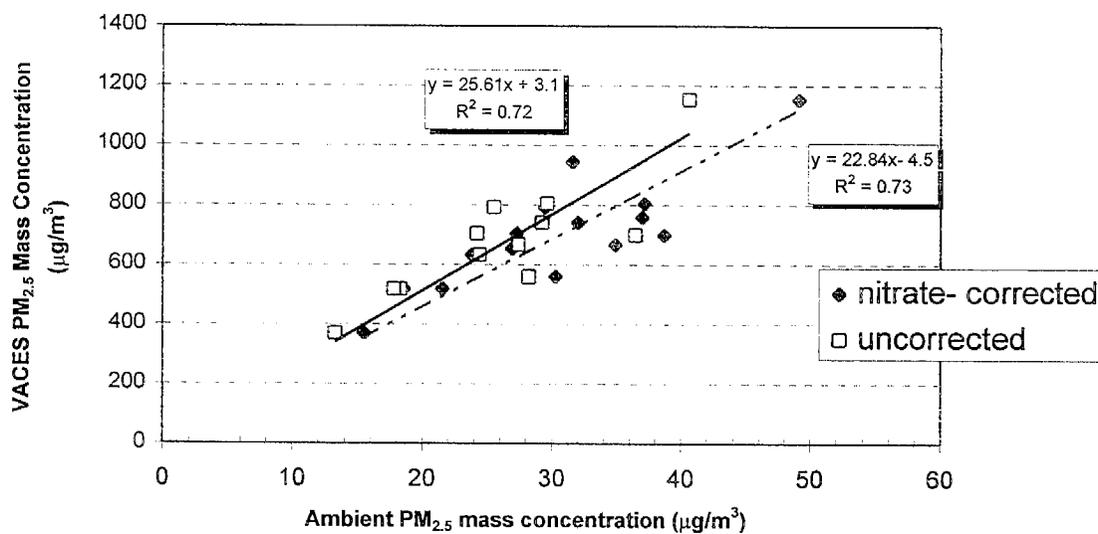


Figure 11. Ambient (MOUDI) and VACES PM_{2.5} Sulfate Concentrations

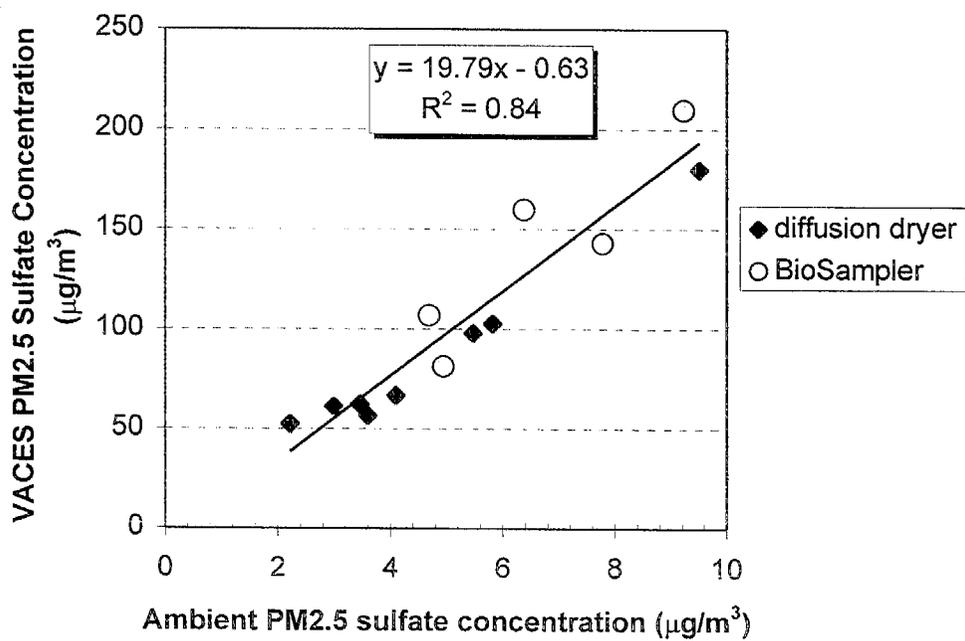


Figure 12. Ambient (MOUDI) and VACES PM2.5 Nitrate Concentrations

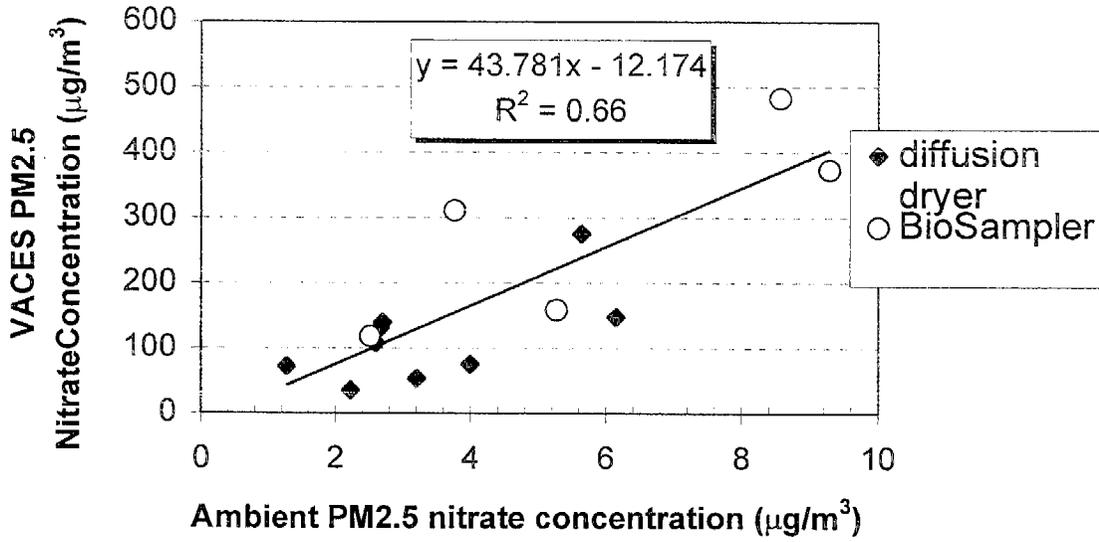


Figure 13

Concentration Enrichment of Selected Trace Elements and Metals by VACES

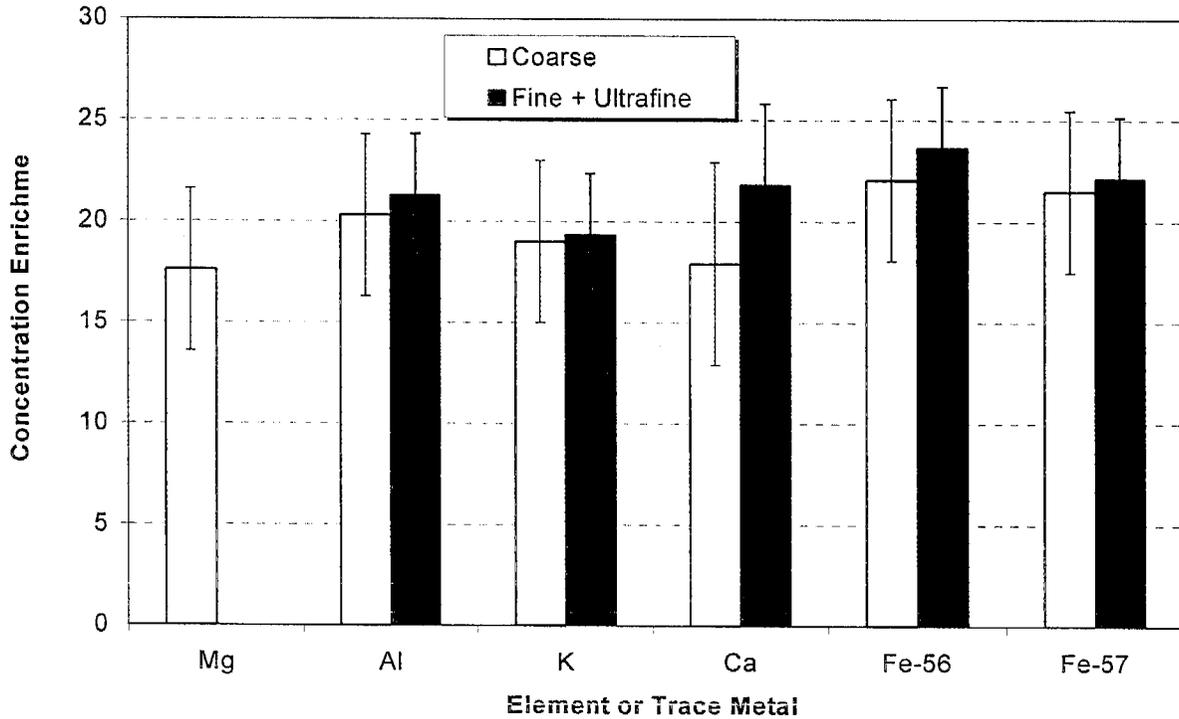


Figure 14. Concentration enrichment of ambient ultrafine particle Elemental Carbon (EC) by VACES

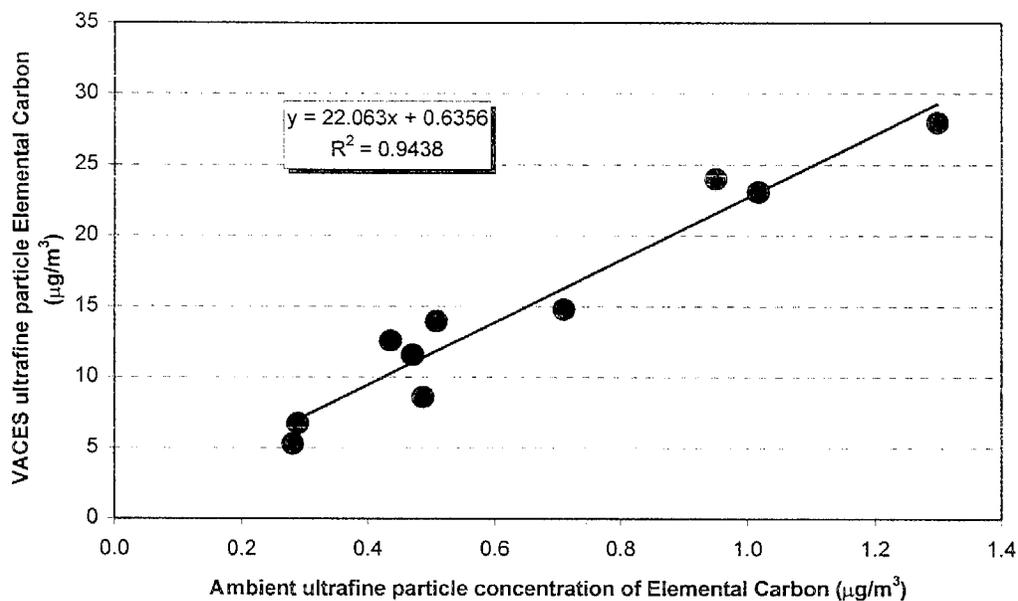


Figure 15. Fractal dimension distribution for agglomerates from the VACES. The count mean D_f value was found to be between 1.6 and 1.8. Samples were taken at the Center for Health Sciences at UCLA using a LPI.

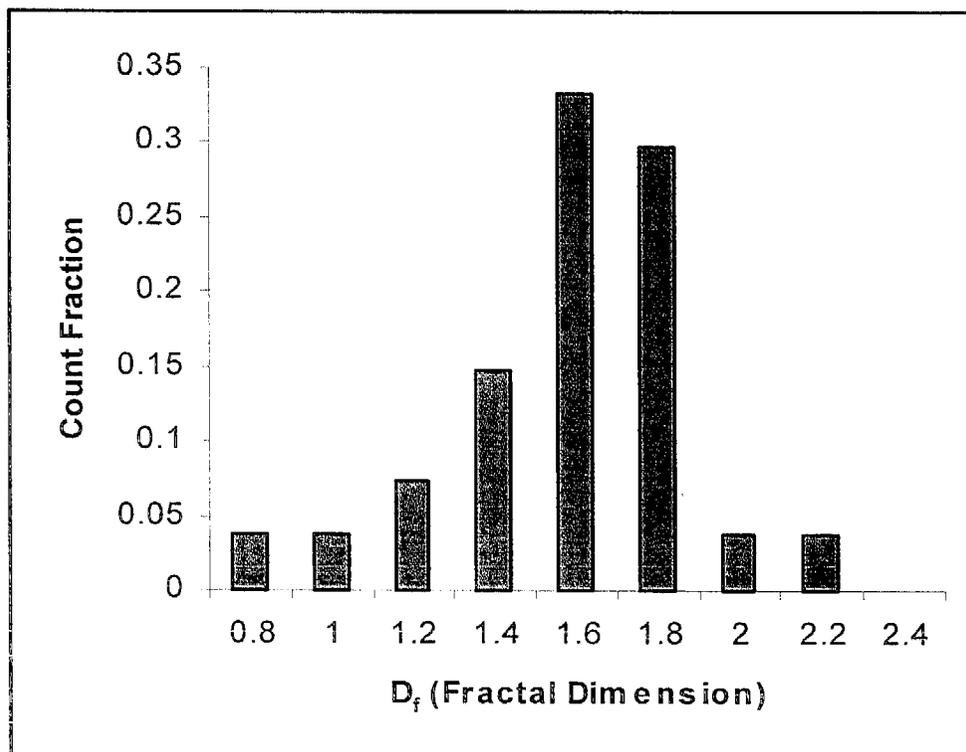


Figure 16. Fractal dimension distribution for agglomerates sampled from the ambient aerosol. The count mean D_f value was found to be between 1.6 and 1.8. Samples were taken at the Center for Health Sciences at UCLA on 8/22/00 using a LPI.

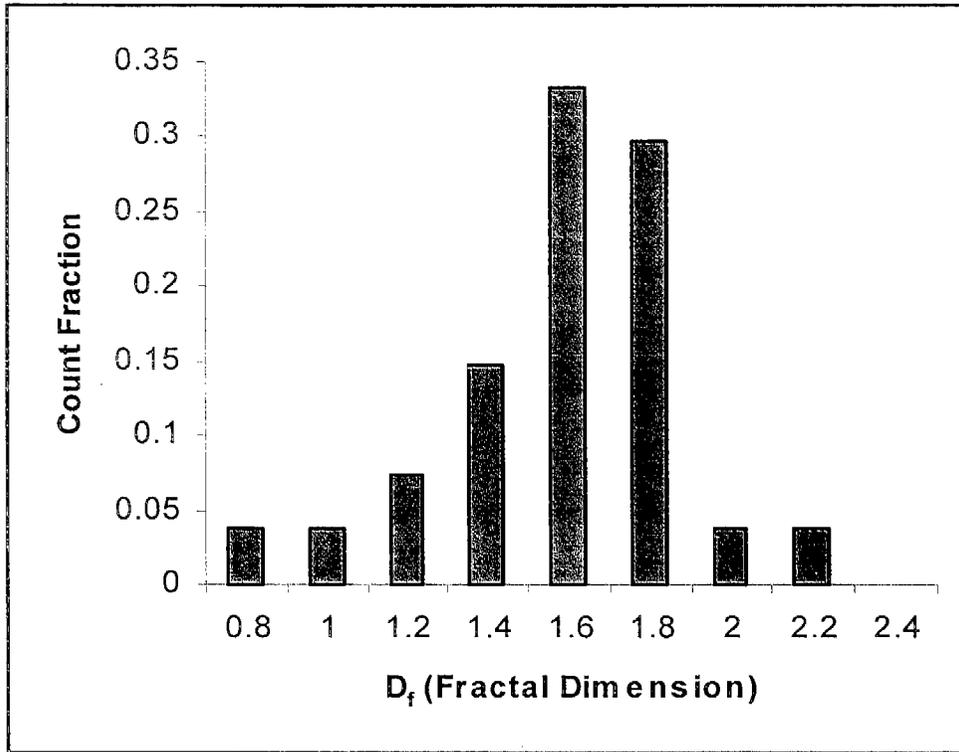


Figure 17. Size distribution of ambient aerosols before and after the VACES measured by SMPS

