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Development of New Generation Personal Monitors for Fine Particulate Matter (PM) and its Metal Content

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ABOUT THE NUATRC

The Mickey Leland National Urban Air Toxics Research Center (NUATRC or the Leland Center) was established in 1991 to develop and support research into potential human health effects of exposure to air toxics in urban communities. Authorized under the Clean Air Act Amendments (CAAA) of 1990, the Center released its first Request for Applications in 1993. The aim of the Leland Center since its inception has been to build a research program structured to investigate and assess the risks to public health that may be attributed to air toxics. Projects sponsored by the Leland Center are designed to provide sound scientific data useful for researchers and for those charged with formulating environmental regulations.

The Leland Center is a public-private partnership, in that it receives support from government sources and from the private sector. Thus, government funding is leveraged by funds contributed by organizations and businesses, enhancing the effectiveness of the funding from both of these stakeholder groups. The U.S. Environmental Protection Agency (EPA) has provided the major portion of the Center's government funding to date, and a number of corporate sponsors, primarily in the chemical and petrochemical fields, have also supported the program.

A nine-member Board of Directors oversees the management and activities of the Leland Center. The Board also appoints the thirteen members of a Scientific Advisory Panel (SAP) who are drawn from the fields of government, academia and industry. These members represent such scientific disciplines as epidemiology, biostatistics, toxicology and medicine. The SAP provides guidance in the formulation of the Center's research program and conducts peer review of research results of the Center's completed projects.

The Leland Center is named for the late United States Congressman George Thomas "Mickey" Leland from Texas who sponsored and supported legislation to reduce the problems of pollution, hunger, and poor housing that unduly affect residents of low-income urban communities.

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EXECUTIVE SUMMARY

BACKGROUND

This study was developed in response to the Mickey Leland National Urban Air Toxics Research Center's (NUATRC) request for applications (RFA) titled, "Development of New Generation Personal Monitors for Fine Particulate Matter (PM) and its Metal Content." Exposures to ambient PM have been associated with both acute and chronic human health effects. Furthermore, air toxics metals constitute an important component of fine PM. Epidemiological studies have often used central, fixed-site monitors to determine exposures, although these monitors do not adequately represent actual human exposures. The RFA identified the need to better validate risk estimates from epidemiological studies with personal monitoring studies. This would require the development of state-of-the-art personal monitors to allow simultaneous separation of particles by size and to characterize size-specific chemical constituents. The samplers should also be quiet, lightweight and userfriendly.

The purpose of this research study was to develop a personal PM sampler that would both allow separation of airborne particles by size and facilitate analyses of the particles for their air toxic metal content. The sampler would operate with the help of a high efficiency personal air pump, to be developed by SKC, Inc., through an independent contract with the NUATRC. Laboratory evaluation of the sampler/pump system for PM mass and metal constituents and pilot field evaluation were planned.

STUDY METHOD RESULTS

The study developed a Personal Cascade Impactor Sampler (PCIS), which is a miniaturized cascade impactor consisting of four impaction stages followed by an after-filter. Particles are separated in the following aerodynamic particle diameter ranges: <0.25; 0.25 to 0.5; 0.5 to 1.0; 1.0 to 2.5; and 2.5 to 10 μ m. The PCIS operates at a flow rate of 9.0 L/min using a very high efficiency, battery-operated light weight pump at a pressure drop of 11 inches of H₂O (2.7 kPa).

Pressure drop measurements were recorded for flow rates ranging from 5.0 to 15 L/min using a Magnehelic (Dwyer Instruments, Inc., Michigan City, IN). The targeted sampling flow rate was between 9.0 and 10 L/min, and the corresponding total pressure drop across the sampler was 11 to 13 inches of H_2O . Based on these results, the PCIS is expected to operate at this flow rate range for at least 24 hours.

The particle collection efficiency as a function of particle size for the 0.25, 0.50, 1.0 and 2.5 μ m cutpoint stages was determined using monodisperse latex particles and polydisperse ammonium sulfate and ammonium aerosols. The tests corroborated the cutpoints.

Each PCIS stage was evaluated using four impaction substrates: (a) 2.5 cm quartz filter, (b) 2.5 cm aluminum foil disk, (c) 2.5 cm PTFE filter and (d) 2.5 cm quartz filter coated with a thin layer of mineral oil (for the 0.25 μ m stage only).

For the 0.25 µm cutpoint stage, the coated quartz and the uncoated Polytetrafluoroethylene (PTFE) (Teflon) and quartz impaction substrates had the same 50% collection efficiency. This was observed at approximately 0.22 to 0.23 µm aerodynamic diameter, very close to the design cutpoint. The results suggested that the coating, used for minimizing particle bounce, would potentially interfere with chemical analysis of the collected particulate matter and is not necessary. For the 0.50 µm and 1.0 µm cutpoint stages, the 50% collection efficiency cutpoint of the Teflon and aluminum substrates was very close to the design cutpoint. However, the particle collection efficiency became higher for the quartz substrates. Particle collection efficiency decreased for particles larger than the cutpoint of this stage when the aluminum substrate was used, probably due to some particle bounce on that surface which would be more pronounced for particles larger than the cutpoint.

To identify a maximum particle mass loading range in a given PCIS stage, experiments were conducted for the 0.25 and 1.0 μ m stages using PTFE substrates. The findings demonstrated that particle bounce and reentrainment do not occur for loadings at least as high as 3.16 mg. Collection efficiencies of all three tested particle sizes remained well above 90% for mass loadings and as high as 700 mg.

Using a labile aerosol such as polydisperse ammonium nitrate, tests were performed to evaluate the ability of the PCIS to conserve labile constituents. The results indicated that the cutpoints of the stages were conserved. This is an important feature of the PCIS, particularly because in large metropolitan areas a significant fraction of fine particles is associated with such labile species as ammonium nitrate or semi-volatile organic compounds. Tests conducted with the PCIS in the wind tunnel facility of the School of Public Health, University of California, Los Angeles showed that the particle penetration characteristics of the PCIS 2.5 μ m stage were unaffected by the wind speeds. This demonstrates that the PCIS can be used for personal sampling in indoor or occupational environments under non-quiescent air.

A field study was conducted in which the PCIS was co-located with the Micro Orifice Uniform Deposit Impactor (MOUDI, Model 110, MSP Corp, Minneapolis, MN) and the Scanning Mobility Particle Sizer (SMPS, TSI Model 3936), Aerodynamic Particle Sizer (APS, TSI Model 3320) inside the Particle Instrumentation Unit. The findings indicated that losses from the PCIS were substantially lower than losses from the MOUDI. Very good overall agreement was obtained between the PCIS and SMPS-APS concentrations for the particle sizes 0.25 to 0.5; 0.5 to 1.0; and 1.0 to 2.5 μ m.

Size segregated $PM_{2.5}$ inorganic ion (nitrate and sulfate) concentrations, elemental carbon (EC) and organic carbon (OC) concentrations were compared using the PCIS and the MOUDI. PM_{2.5} sulfate and PM_{2.5} nitrate concentrations measured by the PCIS were in close agreement with concurrent MOUDI measurements. On comparison of the size-fractionated PM_{2.5} EC and OC concentrations, the PCIS and the MOUDI agreed well. However, the MOUDI underestimated the OC and EC contents for particles <0.25 µm. On comparing nitrate and total carbon measurements, the PCIS and the MOUDI agreed remarkably well (within \pm 15%) for these measurements for all size ranges, including the 0 to 0.25 um range. This indicates that the differences in labile nitrate and OC concentrations between the PCIS and the MOUDI are due to the sample collection process.

The compatibility of the PCIS and a prototype of the Leland Legacy Pump developed by SKC, Inc., was tested for a 24-hour period in a single run by operating the PCIS in conjunction with the pump. The test run was successful since the flow rate and pressure drop are conserved over the 24-hour period. Since the battery was able to power the pump for a maximum of 15 hours only, the pump was run for the full 24 hours with the help of a battery and AC adapter. The final configuration of the pump is expected to run continuously for 24 hours on a single battery.

While the demonstration of the utility of the PCIS for low-level metals analysis was one of the objectives of the study, time and budgetary constraints did not allow for a robust and meaningful study to be conducted. The sensitivity of the PCIS and limit of detection for various metals for a 24-hour sampling period needs to be demonstrated in the laboratory and in the field. This is a future goal for this project.

IMPLICATIONS

The PCIS developed in this study can provide sizedependent information on the mass and elemental concentrations of personal exposure to particulate matter. Because the particles are collected by impaction, losses of volatile compounds from the collected particulate matter are substantially reduced.

The combination of a relatively high flow rate and low pressure drop makes it possible for the sampler to operate in conjunction with a small, battery-operated personal pump. Furthermore, the low pressure-drop of 11 inches of H_2O (2.75 kPa) makes it possible to operate the sampler with a very low noise level. The combination of low noise level and weight of the sampler (approximately 150 g), allows it to be used easily by such subjects as children and the elderly. A field study involving human subjects would demonstrate the utility of the PCIS in exposure and health effects studies.

ABSTRACT

This report summarizes the development and findings from laboratory and field evaluations of a Personal Cascade Impactor Sampler (PCIS). The PCIS is a miniaturized cascade impactor, consisting of four impaction stages followed by an after-filter. Particles are separated in the following aerodynamic particle diameter ranges: <0.25; 0.25 to 0.5; 0.5 to 1.0; 1.0 to 2.5; and 2.5 to 10 µm. The PCIS operates at a flow rate of 9.0 L/min using a very high efficiency, battery-operated light pump with a pressure drop of 11 inches of H_2O (2.7 kPa). Laboratory tests conducted with monodisperse polystyrene latex particles as well as polydisperse ammonium sulfate and ammonium nitrate aerosols corroborated the cutpoints. Particle loading tests indicate that the PCIS stages can collect up to 3.16 and 0.7 mg of fine and coarse PM, respectively, without any loss in collection efficiency that might have resulted from particle bounce.

For field evaluation, the PCIS was co-located in Los Angeles with other samplers. These included the Micro Orifice Uniform Deposit Impactor (MOUDI), the Scanning Mobility Particle Sizer (SMPS) and Aerodynamic Particle Sizer (APS) in Los Angeles. The PCIS and MOUDI results agree very well for coarse PM ($PM_{2.5-10}$) mass. The fine PM ($PM_{2.5}$) mass measured by PCIS is also in excellent agreement with the SMPS-APS measurement (approximately 1.02 times) but is slightly higher (approximately 1.2 times) than the MOUDI measurement. Size-fractionated $PM_{2.5}$ elemental composition, inorganic ions (nitrate and sulfate), elemental carbon (EC), organic carbon (OC) and total carbon measurements by the PCIS and the MOUDI are also compared.

The performance of the PCIS was also evaluated in a wind tunnel. Findings indicate that the particle penetration characteristics of the PCIS 2.5 μ m stage are unaffected by the wind speeds, and that size-dependent particle penetration for the two wind speeds tested, namely 3.0 and 8.0 km/hour, show a very close agreement. This is particularly important because it demonstrates that the PCIS can be used throughout the various ambient conditions found in all normal environments.

INTRODUCTION

Personal monitors are needed for accurate assessment of individual exposures to ambient particles and/or particles in an occupational environment. Nevertheless, the development of reliable personal particle monitors has presented several technical challenges. These include the need for a smaller sampling pump, the requirement for a collection medium with a reduced volume or surface area, and the need for a self-contained energy source. All limit the amount that can be collected within a given time period (Clayton et al., 1993; Morandi et al., 1988; Spengler et al., 1985).

A variety of personal samplers has been developed to date. Among those now commercially available is the PM_{2.5} Personal Exposure Monitor (PEM Model 200 MSP Corp., Minneapolis, MN), which operates at a flow rate of 4.0 L/min with a 50% cutpoint at 2.5 µm in aerodynamic diameter (Buckley et al., 1991). A new cyclone-type, 4.0 L/min personal PM_{2.5} sampler (GK2.05 KTL Cyclone, BGI Inc., Waltham, MA) was developed for and used in the European six-city EXPOLIS study. This sampler was later used in other European and American studies for both personal and microenvironment sampling (Koistinen et al., 1999). Another commercially available personal sampler, the Institute of Occupational Medicine sampler (IOM Personal Inhalable Dust Sampler, SKC Inc., Eighty Four, PA), has a 50% cutpoint at 10 µm. The IOM sampler has a reusable filter cassette with a sampling head designed to measure a worker's exposure to total airborne particulates (Mark and Vincent, 1986). A High Flow Personal Sampler (HFPS) for PM_{2.5} was developed by Adams and coworkers (2001). The HFPS operates at a flow rate of 16 L/min, much higher than conventional personal samplers, and has been particularly useful in studies of particle exposure among urban transport users. These studies require personal sampling over short time periods that represent typical commuter journey times. A unique Multipollutant Personal Sampler developed by Demokritou and coworkers (2000) can be used for simultaneous measurements of particulates (for example, PM₁₀ and $PM_{2.5}$) and gaseous pollutants (for example, SO₂, NO₂, O_3). This sampler is particularly useful for studies examining associations between personal particulate and gaseous exposures.

All of the personal monitoring devices discussed so far provide very little information on the PM size distribution (at best they measure PM size distributions below 2.5μ m). The purpose of this program was to

develop a personal sampler for PM that will allow separation of airborne particles by size and analyses of the particles for their toxic metal and polyaromatic hydrocarbon (PAH) compound content. The sampler operates with the help of a high efficiency personal air pump (Leland Legacy sampler pump, SKC Cat no. 100-3000), which is developed by SKC, Inc., through an independent contract with the NUATRC. Both the development of the personal sampler and of the pump are closely coordinated.

The PCIS is a miniaturized cascade impactor, consisting of four impaction stages followed by an afterfilter. Particles are separated in the following aerodynamic particle diameter ranges: <0.25; 0.25 to 0.5; 0.5 to 1.0; 1.0 to 2.5; and 2.5 to 10 $\mu m.$ The PCIS operates at a flow rate of 9.0 L/min, a rate that is high by personal sampling standards. This makes chemical analyses of the size-fractionated particles possible within a period of 24 hours or less. The PCIS is designed to clip onto a subject's collar in the breathing zone, and the pump is designed to clip onto the subject's belt. The pump can also be put in a backpack worn by the subject. The only other personal cascade impactors reported in the aerosol literature were developed by Rubow et al. (1987) and Demokritou and coworkers (2002). In contrast to the PCIS, the Rubow impactor operates at a flow of 2.0 L/min, and its smallest cutpoint size is 0.5 µm. The personal cascade impactor developed by Demokritou and coworkers (2002) operates at a flow rate of 5.0 L/min, with 0.5 µm as the smallest cutpoint size. The development of the PCIS described in this report, because of its much higher flow rate and its ability to classify particles as small as 0.25 mm in aerodynamic diameter, constitutes a major improvement over prior state-of-the-art monitors in the field of personal devices.

The development of new state-of-the-art personal monitors such as the PCIS, taken together with advances in the area of source apportionment, will greatly add to the quality, depth and feasibility of studies designed to link personal exposures to specific outdoor or indoor sources and will help determine the degree to which these sources influence personal exposure. Moreover, size-fractionated PM personal levels can provide the much-needed data for determining particle dose as opposed to area exposure. Information obtained from more accurate personal monitoring devices that are userfriendly and inexpensive will allow large populations to be studied, thereby providing the much-needed data on the relationship between outdoor and indoor concentrations and personal exposures as a function of particle size and chemical composition. Additionally, the PCIS may also be used to expand present knowledge in the field of industrial exposure assessment.

SPECIFIC AIMS

The main objective of this project was to develop a personal sampler for PM that will allow separation of particles by size and to analyze the particles for their toxic metal content. The sampler will operate with the help of a high efficiency personal air pump to be developed by SKC, Inc., through an independent contract with the NUATRC. Both the development of the personal sampler and the pump will be closely coordinated.

The specific aims were:

- 1. To develop the PCIS
- 2. To evaluate the compatibility of the PCIS with the pump
- 3. To conduct a laboratory evaluation of the PCIS/Pump system for PM mass and metal constituents and pilot field evaluation

METHODS AND STUDY DESIGN

PCIS DESCRIPTION AND DESIGN

The PCIS is a miniaturized cascade impactor, consisting of four impaction stages followed by an afterfilter as shown in Figure 1 (a-d). The sampling flow rate is 9.0 L/min, and the measured total pressure drop across the sampler is 11 inches of H_2O (2.7 kPa). It operates together with a small battery-operated Leland Legacy pump developed by SKC Inc. Eighty Four, PA. The low pressure drop of 11 inches of H₂O makes it possible to operate the sampler with a very low noise level, which is a very desirable feature in personal monitoring. The entire sampler is enclosed in a cassette holder, 4 cm in diameter and 6.0 cm high, made of soft aluminum in order to avoid particle losses during electrostatic deposition. The total weight of the sampler is approximately 150 g; thus it is easily used by children or the elderly. The pump weighs about 450 g (including the battery) and is placed inside a small pouch with snap latch and foam inserts to protect the pump during transport.



Figure 1. Pictures of the PCIS.

Design of the impaction stages was based on the welldocumented impaction theory (Marple and Willeke, 1976). The principal parameter determining particle capture is the Stokes number of a particle having a 50% probability of impacting, St_{50} , defined as follows:

$$St_{50} = \frac{\rho_p \, d_{50}^{\ 2} U C_c}{9\mu D} \tag{1}$$

where d_{50} is the geometrical diameter of particles having a 50% probability of impacting, U is the average velocity of the jet (cm/s), D is the impactor's nozzle diameter (round nozzle) or width (rectangular nozzle, like in PCIS), ρ_{ρ} is the particle density (g/cm³), μ is the dynamic viscosity of the air (g/cm/sec), and C_c is the Cunningham slip correction factor. The slip correction factor is given by the equation (Hinds, 1982):

$$C_c = 1 + \frac{2}{Pd_p} \left[6.32 + 2.01 \exp\left(-0.1095P \, d_p\right) \right]$$
[2]

where P is the absolute pressure upstream of the impaction zone (in cm of mercury) and d_{p} is the particle diameter in $\mu m.$

The design and operating parameters of each impaction stage are shown in Table 1. We chose rectangular slit geometry as opposed to round nozzles in the PCIS design because rectangular nozzles are much easier to fabricate; small cutpoint diameters would need thousands of round nozzles to replace one narrow slit.

Table	1.	Design	and	operating	parameters	of	the	Personal	Cascade
Impac	tor	Sampler	(PCI	S). Sampli	ng Flow Rate	e: 9	.0 l/r	nin	

Impaction Stage Cutpoint (m)	Experimentally Determined Cutpoint (m)	W ^a (cm)	U ^b (cm/s)	ΔP^{c} (inches of H ₂ O)	L ^d (cm)	S/W ^e	Re ^f
2.5	2.60	0.09	877.2	0.3	1.9	2.10	473.7
1.0	0.95	0.05	1428.6	0.6	2.1	3.78	428.6
0.5	0.52	0.036	2631.6	1.5	1.9	5.25	473.7
0.25	0.23	0.014	4286	3.9	2.5	13.50	360.0
Teflon Filter	-	3.7	10.3	4.7	-	-	-

^a Acceleration slit nozzle width of each jet.

^b Average jet velocity.

^c Pressure drop (measured for all stages and after-filter)

^d Length of the acceleration jet. Also length of the deposit.

^e S: Jet-to-plate distance

^f Reynolds's number

The actual 50% cutpoint has been estimated from the collection efficiency curves of each impaction stage, which are described in subsequent sections of this report. Particles in the size range of 0.25 to 10 µm are accelerated in rectangular-shaped nozzles and collected on commercially available 2.5 cm filter substrates made of quartz (Pallflex Corp., Putnam, CT), PTFE (Teflon) (3.0 um pore, Gelman Science, Ann Arbor, MI) or aluminum foil disks that serve as the impaction substrate. In each stage, the impacted particles are collected on a small surface area of 1.0 cm^2 or less of the 2.5 cm filter substrates. This is a substantially smaller surface area than the 13.8 cm^2 area of a standard 4.7 cm filter. The reduced area of the particulate deposit makes it possible to use smaller liquid extraction volumes for such chemical analyses as ion chromatography or inductively coupled plasma-mass spectroscopy, thereby decreasing their limit of detection. Particles smaller than 0.25 µm are collected on a 3.7 cm filter (2.0 µm pore, Gelman Science, Ann Arbor, MI).

LABORATORY EVALUATION OF THE 0.25, 0.50, 1.0 AND 2.5 μm stages

The particle collection efficiency as a function of particle size for the 0.25, 0.50, 1.0 and 2.5 µm cutpoint stages was determined using polydisperse ammonium sulfate aerosols generated by means of a nebulizer (HOPE, B & B Medical Technologies, Inc., Orangevale, CA). The experimental set-up is shown schematically in Figure 2. Aqueous ammonium sulfate solutions (roughly 1.0 mg of ammonium sulfate in 1.0 mL of deionized water) were nebulized using room air at 20 psi. The



Figure 2. Schematic of the experimental set-up used for the laboratory characterization of the PCIS.

generated aerosol was passed through a 1.0 L chamber with ten Polonium 210 ionizing units (Staticmaster, NRD Inc., Grand Island, NY) to reduce particle charge close to the Boltzmann equilibrium. After passing through the neutralizer, the aerosol was first mixed with room air of relative humidity 20 to 30% in a 35 L chamber and then passed through the specific impaction stage. Penetration (or collection efficiency) of particles in the size range of 0.015 to 0.8 µm was determined by measuring their number concentration 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 0.3 L/min of the total flow rate of 9.0 L/min through the impactor. The PCIS flow rate was monitored continuously throughout the experiments using a calibrated in-line direct-reading flow meter (Model A-32457-46, Cole-Parmer Instrument Company, Vernon Hills, IL).

In addition to the SMPS, the DataRAM (RAM-1, MIE Inc., Billerica, MA) was used to evaluate the collection efficiency of the 0.50, 1.0 and 2.5 μ m stages for particles in the 0.2 to 10 μ m range, using laboratory-generated monodisperse polystyrene latex (PSL) particles (Bangs Laboratories Inc., Fisher, IN) in a process otherwise identical to that described above in the SMPS tests. The DataRAM could not be used to monitor particles smaller than 0.2 μ m, because the sensitivity of the instrument decreases sharply below this particle size. The SMPS could not be used for particles larger than approximately 0.8 μ m since they are removed by the impactor placed in the inlet of the instrument.

For selected experiments, monodisperse fluorescent aerosols were used as the test aerosol. Suspensions of 2.5% by weight fluorescent yellow-green latex microspheres of particle density 1.047 g/cm³ (Fluoresbrite, Polysciences, Warrington, PA) were atomized by a pocket nebulizer (Retec X-70/N) using room air at 20 psi. Monodisperse sizes having physical diameters of 0.20 to 0.5 µm were used. The suspensions were diluted with deionized water that removes surfactant and minimizes the generation of satellite droplets. After passing through the neutralizer, the aerosol was mixed with room air in a 35 L chamber and passed through the test system. The test system consists of the specific impactor stage to be calibrated, followed by parallel 3.7 and 4.7 cm Teflon filter samplings. At the end of each run, each Teflon filter was extracted with ethyl acetate to dissolve the latex particles and release their fluorescent dye into solution. The quantities of the fluorescent dye in the extraction solutions were measured by a fluorescence spectrophotometer (FD-500 Fluorescence Detector, GTI, Concord, MA) to determine particle concentration. This method can detect as little as 1.0 µg of PSL particles per mL of solution and is linear up to about 1.0 mg of particles per mL of ethyl acetate solution. Each PCIS stage was evaluated by using the following surfaces as impaction substrates: (1) a 2.5 cm quartz filter; (2) a 2.5 cm aluminum foil disk; (3) a 2.5 cm PTFE filter; and (4) a 2.5 cm quartz filter coated with a thin layer of mineral oil (for the 0.25 µm stage only). The first, second and third impaction surfaces were used without any coating or adhesive material for all PCIS stages. For the 0.25 µm cutpoint stage, the collection efficiency as a function of particle size for the coated quartz substrate was compared to those of the uncoated substrates. This comparison was done to determine the degree to which particle bounce occurs. These experiments were only conducted for the 0.25 µm PCIS stage because the higher impaction jet velocity of that stage would increase the likelihood of particle bounce.

Particle Loading Capacity Tests

One of the main problems with conventional impactors is the potential loss of the collection efficiency with particle loading. Previous investigations assessing the effect of loading on the performance characteristics of the impactor found that the collection efficiency of impactors using coated substrates degrades over time. This is because the grease becomes ineffective with particle accumulation (Reischl and John, 1978; Turner and Hering, 1987; Tsai and Cheng, 1995). Incoming particles bounce off previously deposited particles. This degradation poses a serious limitation to sampling high concentration aerosols or sampling over prolonged time periods, both of which would lead to high particle loading.

Experiments were conducted to identify a maximum particle mass loading range (in micrograms or milligrams of PM) in a given PCIS stage beyond which the efficiency of particle collection for any particle size would decrease because of particle bounce. These tests were conducted for the 0.25 and 1.0 µm stages using PTFE substrates. The $0.25 \ \mu m$ stage was chosen for particles in the 0.1 to 0.5 µm range because the highest jet velocities of that stage would accentuate particle bounce. Evaluation of the degree to which particle bounce occurs for coarse (that is, 2.5 to 10 µm) particles was conducted using the 1.0 µm stage. Although these particles are collected on the 2.5 µm stage, collection efficiency results with any of the substrates used, as discussed in the previous section, demonstrate that particle bounce is much less pronounced because this stage has a jet velocity substantially lower than the 1.0 µm cutpoint stage (Table 1). Hence, the 1.0 µm stage was chosen because it provides a "worst case" scenario for particle bounce of coarse PM collected by the PCIS.

The effect of particle loading on the performance of the 0.25 µm stage was investigated by generating polydisperse ammonium sulfate aerosol through a process similar to that used to characterize the impactor's collection efficiency. The volume median diameter of the generated aerosol, measured by means of the SMPS, was approximately 0.3 to 0.4 µm (Geometric Standard Deviation, $\sigma g = 1.8$). Measures by the SMPS upstream and downstream of the impactor in the beginning of this experiment further confirmed that over 95% of the aerosol by mass is collected by the impactor. The stability of the concentration of the generated aerosol throughout the loading test was confirmed by the DataRAM that was sampling in parallel to the PCIS and the SMPS samplers. Particle penetration through the PCIS 0.25 µm stage was determined every 15 min for selected particle sizes, namely 0.1, 0.3 and 0.5 µm, by manually adjusting the voltage of the SMPS and measuring the concentrations for each size upstream and immediately downstream of the PCIS. Particle loading for the 1.0 µm stage was investigated by generating monodisperse PSL particles in a process similar to that used for characterizing the collection efficiency of the impactor. Particle sizes of 2.6, 5.4 and 7.6 µm were used. The DataRAM was used to measure particle penetration through the PCIS 1.0 µm stage. Particle penetration was determined in a similar manner every 15 min by

measuring the concentration of particles upstream and downstream of the PCIS. Each experiment lasted approximately one to two hours. At the end of each experiment, the Teflon filter used as an impaction substrate was weighed using a Mettler 5 Microbalance (MT 5, Mettler-Toledo Inc., Hightstown, NJ), under controlled relative humidity (40% to 50%) and temperature (22° to 24° C).

Laboratory Tests for the Evaluation of Volatilization Losses

In addition to laboratory characterization using stable aerosols, tests were performed to evaluate the performance of the PCIS using polydisperse ammonium nitrate aerosols using the same process described above. A solution of approximately 1.0 mg of nitrate per 1.0 mL of deionized water was used in the nebulizer to generate a targeted aerosol mass median diameter (MMD) of 0.3 to 0.4 µm. These tests were conducted to provide an estimate of the degree to which labile constituents of ambient aerosols, such as ammonium nitrate, are preserved as they are drawn through the stages of the PCIS. First, the particle collection efficiency of the 0.25 and 0.50 µm stages for volatile ammonium nitrate aerosol were compared to those obtained using non-volatile ammonium sulfate aerosols. Subsequently, the nitrate PCIS concentrations (determined by adding the concentrations of all five stages) were compared with a co-located MOUDI Model 110, (MSP Corp, Minneapolis, MN) operating at a flow rate of 30 L/min. The PTFE substrates were used both in the PCIS as well as the MOUDI. In addition to the MOUDI, the size distribution of the generated ammonium nitrate aerosol was also measured using a SMPS, sampling in parallel to the PCIS and MOUDI. For the SMPS data, each particle size interval was converted from mobility equivalent diameter to aerodynamic diameter. This conversion was made using the following equation (Peters et al. 1993):

$$\sqrt{C_a} d_a = \sqrt{\frac{C_{me} \rho_p}{\chi \rho_o} d_{me}}$$
[3]

where C_a is the slip correction factor for the aerodynamic diameter; d_a is the aerodynamic diameter; d_{me} is the mobility equivalent diameter; C_{me} is the slip correction factor for the mobility equivalent diameter; χ is the dynamic shape factor; ρ_ρ is the density of the particle; and ρ_o is the unit density (1.0 g/cm³).

When performing this conversion, the mobility equivalent diameter was assumed to be equal to the equivalent volume diameter (Kasper, 1982), while particles were assumed to be perfect spheres (dynamic shape factor, $\chi = 1$). For each test, number concentrations for each size interval were then converted to mass concentrations using following equation:

$$C_m = \frac{\pi}{6} \rho_p N_c \left(\frac{d_a}{\sqrt{\rho_p}}\right)^3$$
[4]

where C_{m} is mass concentration and N_{c} is number concentration.

For each type of aerosol, particle mass concentrations obtained from the MOUDI were grouped in the following size ranges: <0.18; 0.18 to 0.32; 0.32 to 0.5; and 0.5 to 1.0 µm. For the PCIS, the ranges were grouped as follows: <0.25; 0.25 to 0.5; and 0.5 to 1.0 µm. The MOUDI does not have a 0.25 µm cutpoint stage. Therefore to make the MOUDI cutoff size ranges comparable to those of the PCIS, half of the nitrate mass determined in the 0.18 to 0.32 µm size range was added to that measured in the 0 to 0.18 µm range and half to that measured in the 0.32 to 0.50 µm range. This conversion was justified by assuming that the generated nitrate particles are lognormally distributed (an assumption corroborated by the concurrently obtained SMPS data); hence, about 50% of the particle mass in the 0.18 to 0.32 µm size range should be found above and 50% below 0.24 µm, which is the geometric mean of that range. The uncertainly introduced is minimal given the narrow width of this particle size range.

Wind Tunnel Tests

The performance of the PCIS was evaluated in the wind tunnel facility of the School of Public Health, UCLA. In its original design, the wind tunnel had a 1.6 m x 1.6 m cross-section and was operated at two wind speeds (3.0 and 8.0 km/hour). The blower downstream of the sampler test area was capable of driving air speed in the wind tunnel up to 8.0 km/hour. A plywood baffle was placed about 0.5 m upstream of the aerosol generation system to promote mixing. The vibrating aerosol orifice was itself mounted on a shaft moved both up and down and sideways to promote uniform injection. Three isokinetic samplers were placed around the PCIS. The PCIS was placed so that it was equidistant from the three isokinetic samplers. Two of these samplers

were lateral to the PCIS while the third one was above the PCIS. The positioning of the isokinetic samplers corresponded to uniformity in concentration around the PCIS.

The sampling characteristics of the PCIS 2.5 μ m cut stage were determined by comparing the mass concentration obtained by the PCIS 2.5 μ m stage to that measured by isokinetic samplers. Six different particle sizes (1.0, 2.0, 2.5, 5.0, 7.5 and 9.0 μ m) were selected to evaluate the performance of the PCIS.

Monodisperse particles were generated by a vibrating orifice aerosol generator (VOAG) (Model 3450, TSI Inc., St. Paul, MN). When a solution containing a non-volatile solute is sprayed through an orifice, the solvent eventually evaporates from the droplets, and nonvolatile particles of solute are obtained. The particle diameter for the non-volatile solute is given by:

$$D_p = C^{1/3} D_d$$
 [5]

where $\rm D_p$ is particle diameter, C is volumetric concentration of the non-volatile solute in the solution, and $\rm D_d$ is the initial droplet diameter.

A vibrating orifice produces one droplet per cycle, and \mathbf{D}_{d} is given by:

$$D_d = (6Q/\pi f)^{1/3}$$
 [6]

where Q is the liquid flow rate and f is the disturbance frequency.

The droplet size depends primarily on the orifice size for a given solution feed rate and the frequency of disturbance. For these experiments, a 20 μ m orifice was used for generating particles. Typical VOAG operating parameters were 0.150 mL/min of feed rate at 65 to 70 kHz.

Uranine-tagged oleic acid was used as a non-volatile solute for generating particles. Acetone was the solvent. To prepare the tracer solution, approximately 2.0 g of uranine dye was dissolved in 50 mL of methanol. The solution was left overnight to dissolve the dye while allowing for concomitant settling of any undissolved uranine dye. Twenty to forty percent of this solution was then added to the oleic acid-acetone solution to generate particles of a desired size using Equations 5 and 6. To confirm that the generated aerosols were monodisperse, the generated particles were observed under a microscope. This also served to corroborate the size of the particles.

The isokinetic samplers and the PCIS 2.5 µm stage were positioned at the same distance from the sample injection point specifically on the same axial plane. The isokinetic samplers were constructed from 2.5 cm in-line stainless steel filter holders (P/N 1209, Gelman Sciences Inc., Ann Arbor, MI) fitted with 8.5 mm ID brass probes that extended 32 mm from the face of the filter holder and sampled at a flow rate of 10 L/min for wind speeds of 3.0 and 8.0 km/hour. The three isokinetic samplers were sampled along with the PCIS for each particle size and wind speed. Millipore membrane filters (SMWP 02500, Millipore, Bedford, MA) were used to collect uranine- tagged oleic acid particles in the isokinetic samplers and the PCIS. Each of the experiments was characterized by particle size and wind speed and lasted for about 10 to 15 min, which was sufficient to obtain detectable mass on the filters.

Detection of the deposited uranine-tagged oleic acid particles on the Millipore filters was performed using a fluorescence detector (Model FD-500, Programmable Fluorescence Detector, GTI, Concord, MA). Prior to detection, the uranine was first extracted in a borate buffer solution. The buffer was prepared by mixing solutions of boric acid (12.5 g of boric acid in 100 mL of water) and sodium borate (19.05 g of sodium borate in 100 mL of water). Fifty mL of boric acid and 59 mL of sodium borate were then mixed and diluted to 200 mL using distilled water.

Material from Millipore filters was then extracted in glass vials using the buffer solution. Generally the extractions were done using 5.0 to 10 mL of buffer solution. Standard uranine dye solutions of 0.005, 0.01, 0.025, 0.05, 0.075 and 0.1 ppm were used to plot the calibration curve.

For each wind speed, the mass concentrations obtained by means of the three isokinetic samplers and the PCIS 2.5 μ m stage were compared. For each particle size, the averaged value of the mass concentration for the three isokinetic samplers was used. Finally, the particle penetration through the PCIS was plotted against the particle diameter for each wind speed.

FIELD EVALUATION OF THE PCIS IN CO-LOCATION WITH OTHER SAMPLERS IN CLAREMONT/LOS ANGELES

The performance of the PCIS was evaluated in a field study that began in March, 2002. Outdoor sampling was conducted at Claremont, CA, which was at that time the location of the Southern California Supersite mobile laboratory. This site is located approximately 45 miles east of downtown Los Angeles. Sampling was conducted during 24-hour periods once a week, on a Tuesday, Wednesday or Thursday, in order to coincide with one of the sampling days of the speciation network (which takes place every third day). However, during "intensive" particulate characterization studies and for our support of co-located health effects exposure studies, sampling schedules were followed that are consistent with these special studies. Typically, ambient data are averaged over 24 hours for exposure or source contribution measurements, although the time integrals may vary. The PCIS was co-located with the MOUDI and the SMPS-APS. All devices were placed inside the Particle Instrumentation Unit, a mobile laboratory trailer developed through funds provided by the U.S. Environmental Protection Agency. The development of a battery-operated pump by SKC Inc. was not accomplished by the time the field tests were conducted. Thus, in all field tests, the PCIS was used in conjunction with a battery-operated personal pump developed by BGI (BGI 400, BGI Inc., Waltham, MA). In July 2002, sampling was continued outdoors at the University of Southern California, the next sampling site of the Southern California Supersite. The school is located two miles south of downtown Los Angeles and represents a typical urban area in which the aerosol is primarily emitted by vehicles.

Mass Concentration Measurements

Substrates of PTFE were used both in the PCIS as well as the MOUDI for measuring size-segregated mass concentrations. The PCIS operates at a lower flow rate of 9.0 L/min as opposed to the MOUDI (which operates at 30 L/min). Unlike the PCIS, the MOUDI does not have a 0.25 µm cutpoint stage. Therefore, to make the MOUDI cutoff size ranges comparable to those of the PCIS, the procedure described earlier in this report was used where half the mass determined in the 0.18 to 0.32 μ m size range was added to that measured in the 0 to 0.18 µm range and half was added to that in the 0.32 to 0.50 µm range. The purpose of this conversion was to bring the MOUDI and PCIS aerosol size ranges closer to each other; it assumes that the aerosol size distribution in the range is log normal. Because this assumption may or may not be valid, it may introduce some errors in the comparison experiments between the MOUDI and PCIS.

The semi-continuous SMPS-APS particle mass was determined by integrating the cumulative number count for the respective size fractions and converting to mass by assuming particles to be perfect spheres with the average density of particles in the Los Angeles basin of 1.6 g/cm^3 . Prior to this conversion, for the SMPS data, each particle size interval was converted from mobility equivalent diameter to aerodynamic diameter using Equation 3.

Chemical Species Measurements

Size-segregated $PM_{2.5}$ inorganic ion (nitrate and sulfate) concentrations, elemental carbon (EC) and organic carbon (OC) concentrations were compared using the PCIS and the MOUDI. Concentrations of nitrate and sulfate ions in the particulates were determined by an ion chromatography technique similar to that employed by Harrison and coworkers (1996).

The Teflon filters from the personal samplers were extracted in small beakers using 10 mL of a 10% isopropanol solution prepared by dissolving 10% HPLC grade isopropanol (J.T. Baker, Phillipsburg, PA) in 10 mL of ultra-pure water. Extraction was accomplished by shaking in an ultrasonic vibrator for about one hour at room temperature. Under these conditions, inorganic species were completely dissolved. The solution was then filtered through a 0.45 µm PTFE filter, and transferred to a vial prior to analysis by ion chromatography. A Dionex DX 300 ion chromatograph (Dionex Corporation, Sunnyvale, CA), equipped with a 4 mm AS4A anion specific column and an AG4A guard column, was employed for the analysis of the nitrate and sulfate anions. The chromatographic conditions were as follows: eluant composition, 1.8 mM $Na_2CO_3/1.7$ mM NaHCO₃; regenerant, 25 mM H₂SO₄; and eluant flow rate, 2.0 mL/min. Blank samples from the field were also analyzed for nitrate and sulfate to determine background level concentrations. Each sample was analyzed in duplicate for quality assurance of the acquired data. Quality control was maintained by running appropriate additional standards of known concentrations and by the use of spiked samples in the sample run.

The EC and OC content in PCIS substrates (quartz) was determined by a thermo-analysis technique. Each substrate was placed in a platinum boat containing MnO_2 . The sample was acidified with an aliquot of HCl and heated to 115° C to dehydrate the sample and to form CO_2 , which served as an index of particle-associated carbon. The boat was then inserted into a dual zone furnace, where MnO_2 -oxidized OC at 550° C and EC at 850° 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).

In addition to the EC and OC analysis, size-segregated PM_{2.5} nitrate concentrations from the PCIS and the MOUDI were measured using a continuous sizesegregated nitrate monitor from Aerosol Dynamics Inc. (ADI) described in more detail by Stolzenburg et al. (2002). The ADI cascaded Integrated Collection and Vaporization Cell (ICVC) is an automated collection and analysis method for high-time resolution determination of the major chemical constituents of ambient particles. Near continuous (10 minute) data were obtained by connecting the ADI monitor to ports placed downstream of each MOUDI and PCIS stages. For these measurements, Zefluor (0.5 µm pore, Gelman Science, Ann Arbor, MI) substrates were used both in the MOUDI and the PCIS. The concentrations downstream of a MOUDI stage were measured immediately after its corresponding PCIS stage for 10 min. For any given particle range, this procedure was repeated two to three times to ensure that there are no significant changes in the concentrations of the ambient PM_{2.5} aerosol during the course of these experiments.

Size-segregated total carbon concentrations in $PM_{2.5}$ measured by the MOUDI and the PCIS were also measured using a continuous carbon monitor (Sunset Labs, Forest Grove, OR). The experimental procedure was identical to that described in the previous paragraph regarding nitrate measurements, with the exception that direct-reading measurements in each PCIS and MOUDI stage were alternately taken every 15 minutes (as opposed to 10 minutes in the case of nitrate tests). A total of seven field experiments were conducted for nitrate and eight for carbon measurements.

The PCIS samples at 9.0 L/min and collects on Zefluor (Teflon) substrates that are ideal for metals analysis using Inductively Coupled Plasma Mass Spectroscopy (ICP-MS). However, the sensitivity of the PCIS and limit of detection for various metals, assuming a standard extraction volume of 10 mL or less for a 24-hour sampling period, needs to be demonstrated in the laboratory and in the field. The ICP-MS is an extremely sensitive method, and a non-detect value may most likely suggest the absence of the metals of concern. While the demonstration of the utility of the PCIS for low-level metals analysis was one of the objectives of the study, time and budgetary constraints did not allow for a robust and meaningful study to be conducted. This is a future goal for this project.

STATISTICAL METHODS AND DATA ANALYSIS

All data are reported as means plus or minus the standard deviation of measurements. A paired Student'st test was performed to evaluate the significance of the differences in measurements by the PCIS and other samplers. A P value of <0.05 was considered significant. Where appropriate, least squares regression analysis was performed to evaluate correlation between grouped pairs of measurements (Microsoft Excel, Windows Millennium Edition).

RESULTS AND DISCUSSION

MEASUREMENT OF PRESSURE DROP VERSUS FLOW RATE

Figure 3 presents a plot of the pressure drop across the entire PCIS as a function of the sampling flow rate, including all four impaction stages and the 3.7 cm Teflon after-filters. Pressure drop measurements were recorded for flow rates ranging from 5.0 to 15 L/min using a Magnehelic (Dwyer Instruments, Inc., Michigan City, IN). The targeted sampling flow rate is between 9.0 to 10 L/min, and the corresponding total pressure drop across the sampler is found to be 11 to 13 inches of H_2O . Based on these results and the specifications of the personal pump, the PCIS can operate at this flow rate range for at least 24 hours without any need for re-charging the battery of the pump.



Figure 3. Pressure drop across PCIS as a function of flow rate.

LABORATORY EVALUATION OF THE 0.25, 0.50, 1.0, AND 2.5 μm STAGES

Results from the evaluation tests of the 0.25, 0.50, 1.0. and 2.5 µm stages are summarized in Figures 4 through 7. Particle collection efficiency data for each stage are plotted as a function of aerodynamic particle diameter. The collection efficiencies using three different surfaces as impaction substrates are shown in the same graph for direct comparison. The results plotted in Figure 4 for the 0.25 µm cutpoint stage indicate that there is no substantial difference among the coated quartz, the uncoated PTFE, and the quartz impaction substrates. All three substrates have the same 50% collection efficiency cutpoint, at approximately 0.22 to 0.23 µm in aerodynamic diameter, thus very close to the design cutpoint. Particle collection efficiency increases rapidly to more than 85% for particles $0.35\ \mu m$ and above. No tests were conducted for particles larger than 0.5 µm because they are presumably collected by the preceding impaction stage, which has a 50% cutpoint at that size.

The slope of the collection efficiency curve obtained for the PTFE is slightly steeper than those obtained for the coated and uncoated quartz substrates. This may be due to the capture of particles smaller than the cutpoint on the fibrous quartz surface because of partial entrainment of the impinging jet streamlines on that surface. In contrast, the PTFE is a relatively harder impaction surface, and this entrainment should be minimal. The good overall agreement between the collection efficiencies of the coated and uncoated quartz and PTFE substrates suggests that the use of coating, which would minimize particle bounce but substantially complicate chemical analysis of the collected particulate matter, is not necessary. An example of the effect of particle bounce on the collection efficiency is illustrated by the results obtained for the uncoated aluminum impaction substrate (Figure 4). The substantially lower collection efficiency values for any particle size obtained for that substrate can be attributed to particle bounce.

The results plotted in Figure 5 for the 0.50 μ m cutpoint stage indicate that the 50% collection efficiency cutpoint is approximately 0.48 to 0.50 μ m in aerodynamic diameter for the Teflon and aluminum substrates, very close to the design cutpoint. Particle collection efficiency becomes higher for the quartz substrates, particularly for particles smaller than the design (or theoretical) cutpoint of that stage, including the actual 50% cutpoint, estimated to be 0.43 μ m for this substrate. Particle collection efficiency increases rapidly to more than 85% for particles 0.60 μ m and above for the

quartz and Teflon substrates. Furthermore, the slope of the collection efficiency curve obtained for the PTFE and aluminum substrates is slightly steeper than those obtained for the quartz substrate. Similar to the results obtained for the 0.25 µm PCIS stage, the increased efficiency and decreased steepness of the quartz substrates are due to capture of particles smaller than the cutpoint on the fibrous quartz surface because of partial entrainment of the impinging jet streamlines on that surface. Particle collection efficiency also decreased for particles larger than the cutpoint of this stage when the aluminum substrate was used, probably due to some particle bounce on the surface. The particle bounce would be more pronounced for particles larger than the cutpoint. This phenomenon is more likely to occur in the aluminum substrates because of the higher surface hardness of aluminum as compared to Teflon or quartz.

Results from the evaluation of the 1.0 μ m stage are shown in Figure 6. The collection efficiency curves for quartz and Teflon substrates appear to be quite similar, while the efficiencies obtained for the aluminum substrates appear substantially lower, similar to the results observed for the 0.25 and 0.50 μ m stages. Again, the decreased collection efficiency of aluminum is attributed to particle bounce onto the harder surface of this substrate. No substantial difference was seen between the collection efficiency characteristics of the PTFE and quartz substrates, as both surfaces lead to the same cutpoint (1.0 μ m). In contrast, the 50% cutpoint obtained with the aluminum substrate was closer to 1.5 μ m.

Figure 7 presents the particle collection efficiency as a function of aerodynamic diameter for the 2.5 μ m stage of the PCIS for different impaction substrates. The plotted data indicate that collection efficiency trends versus



Figure 4. Collection efficiency of the 0.25 μ m stage as a function of particle aerodynamic diameter using ammonium sulfate as test aerosol.



Figure 5. Collection efficiency of the 0.50 μm stage as a function of particle aerodynamic diameter using ammonium sulfate as test aerosol.



Figure 6. Collection efficiency of the 1.0 μm stage as a function of particle aerodynamic diameter using polystyrene latex particles as test aerosol.



Figure 7. Collection efficiency of the $2.5 \ \mu m$ stage as a function of particle aerodynamic diameter using polystyrene latex particles as test aerosol.

particle size are quite similar for the three substrates. Contrary to the observations made for the 0.25, 0.50 and 1.0 μ m PCIS stages, particle bounce seems to be less pronounced for the aluminum substrates in this stage, given the high collection efficiency observed for particles larger than 2.5 μ m. This is likely a result of the substantially lower impactor jet velocity of this stage (Table 1).

The sharpness of the collection efficiency curve of an impactor can be defined in terms of σ_g , the geometric standard deviation. The geometric standard deviation is defined as the square root of the ratio of the aerodynamic particle diameter for the 84% collection efficiency to the aerodynamic particle diameter for the 16% collection efficiency (Marple and Willeke, 1976). The values of σ_g for each stage and substrates were estimated and are listed in Table 2.

 Table 2. Sharpness of the collection efficiency for each stage of the PCIS as a function of substrate.

Substrate Type	Collection Efficiency Curve Sharpness
0.25 μm	
PTFE Teflon	1.28
Quartz	1.53
Aluminum	1.31
0.50 μm	
PTFE Teflon	1.25
Quartz	1.58
Aluminum	1.39
1.0 μm	
PTFE Teflon	1.25
Quartz	1.32
Aluminum	1.34
2.5 μm	
PTFE Teflon	1.19
Quartz	1.39
Aluminum	1.22

Generally, lower σ_g values indicate the higher precision in particle separation characteristics of a given impaction stage, which is a highly desirable feature of an impactor as it leads to a finer resolution in the size distribution of an aerosol. Based on this definition, the value of σ_g is approximately 1.2 to 1.3 for the PTFE and aluminum substrates, indicating reasonably sharp aerodynamic particle separation characteristics when these two substrates were used. The values of σ_g increase to about 1.5 for the quartz substrates, especially for the lower cutpoint stages of the PCIS, because of the increased collection efficiency observed for this type of

substrate for particles smaller than the theoretical cutpoint of the impactor.

Finally, collection efficiencies for different particle sizes and different PCIS stages obtained by means of either the DataRAM or analysis of fluorescence are shown in Table 3. The results shown in this table clearly indicate that the collection efficiencies for any particle size, and for any of the two PCIS stages, obtained by means of the DataRAM or the fluorescent monodisperse aerosols are in excellent agreement with those obtained by means of the SMPS.

Table 3. Comparison of collection efficiencies obtained by means of the DataRAM and analysis of fluorescence and those obtained using the SMPS. The PCIS stages were used with PTFE teflon substrates. Mean and Standard Deviation are based on three experiments per configuration.

Particle Size (µm)	SMPS Collection Efficiency	DataRAM Collection Efficiency	Analysis of Fluorescence	
0.25 µm Stage				
0.15	0.08 (± 0.02)	0.13 (± 0.04)	0.11 (± 0.04)	
0.25	0.33 ±0.05)	0.39 (± 0.08)	0.35 (± 0.07)	
0.50	0.92 (± 0.02)	0.95 (± 0.04)	0.97 (± 0.01)	
0.50 µm Stage				
0.15	0.05 (± 0.03)	0.08 (± 0.03)	0.08 (± 0.04)	
0.50	0.48 (± 0.06)	0.53 (± 0.05)	0.55 (± 0.08)	
0.75	0.93 (± 0.03)	0.95 (± 0.05)	0.92 (± 0.05)	

Particle losses in each PCIS stage were evaluated by first removing the impaction substrate block of that stage and by measuring the concentrations of the polydisperse ammonium sulfate (0.05 to 0.5 μ m) or monodisperse (0.2 to 10 μ m) PSL particles upstream and downstream of the stages using the SMPS and DataRAM, respectively. Results are shown in Figure 8. As evident from the



Figure 8. Particles losses in each of the PCIS stages.

figure, losses in each PCIS stage are quite low, within 10% or less, and they increase with particle size. This observation suggests that particle losses are caused primarily by inertial deposition on the walls of the acceleration nozzles of the impactor as the flow turns, exiting a stage. For larger particles, inertial effects are the most severe and, hence, higher losses are expected, in agreement with the experimental results.

Particle Loading Capacity Tests

Results from the loading tests of the 0.25 µm stage are shown in Figure 9. The collection efficiency of the PCIS 0.25 µm stage for three different particle sizes, 0.1, 0.3 and 0.5 µm, is plotted as a function of particle loading, expressed in mg of accumulated particles. The data demonstrate unequivocally that particle bounce and reentrainment, which would have been manifested by a detectable decrease in the collection efficiency measured for any size, do not occur for loadings at least as high as 3.16 mg. It should be noted that this is an unrealistically high particulate loading for any stage of the PCIS, considering typical PM levels in ambient indoor or occupational environments. For example, for a 24-hour sampling period, and at a PCIS nominal flow rate of 9.0 L/min, this loading would correspond to an aerosol concentration of about 240 μ g/m³ in each PCIS stage.



Figure 9. Collection efficiency of the 0.25 μm stage as a function of particle mass loading.

The collection efficiency of the PCIS 1.0 μ m stage as a function of particle loading expressed in μ g of accumulated particles is plotted in Figure 10. Three different particle sizes, 2.6, 5.4 and 7.6 μ m, were tested. The plotted data reveal a slightly decreasing trend in the



Figure 10. Collection efficiency of the 1.0 μm stage as a function of particle mass loading.

collection efficiency as particle loading increases, probably the result of particle bounce and reentrainment. This hypothesis is further corroborated by the faster decline in collection efficiency observed with increasing particle size. However, collection efficiencies of the three particle sizes tested remain well above 90% for mass loadings as high as 700 µg. This loading would correspond to an average aerosol concentration of about 54 µg/m³ for a 24-hour sampling period and at a sampling PCIS flow rate of 9.0 L/min. These results suggest that the PCIS retains its high collection efficiency for coarse PM even at the unusually high concentrations.

Laboratory Tests for the Evaluation of Volatilization Losses

The evaluation of 0.25 and 0.5 μ m stages using polydisperse ammonium nitrate aerosols is presented in Figure 11. As is evident, the cutpoints of the stages are conserved even when a labile aerosol is passed through



Figure 11. Collection efficiency curves for the 0.25 and 0.50 μm PCIS stages using ammonium nitrate as the test aerosol.

the PCIS. No notable differences in the particle collection efficiency were seen in data obtained for stable sulfate and volatile nitrate aerosols. Previous studies have shown that losses of volatile compounds from the collected particulate matter are substantially reduced (if not eliminated) when these particles are collected by impaction as compared to filtration (Wang and John, 1988; Sioutas et al., 1994). These studies also showed that ambient ammonium nitrate concentrations obtained with impactors were three to four times higher than those obtained with standard Teflon filter samplers. This is an exceedingly important feature of impactors, particularly as a significant fraction of fine particles is associated with labile species such as ammonium nitrate or with semi-volatile organic compounds. Potential nitrate losses (or lack thereof) from the PCIS after-filter are discussed in the following section.

The results from the laboratory evaluation of the PCIS, the MOUDI and the SMPS using polydisperse ammonium nitrate aerosols are shown in Table 4. As evident from the data, a very good agreement persists between the concentrations of the PCIS and the SMPS for all the three size ranges: <0.25; 0.25 to 0.5; 0.5 to 1.0 μ m. The total ammonium nitrate concentrations measured by the PCIS and the SMPS agree to within 10% or less. The concentrations for the MOUDI, however, appear to be quite low for size ranges <0.25 μ m in almost all the tests. This could well be attributed to the high flow rate of the MOUDI (30 L/min) and consequently a

 Table 4. Comparison between concentrations of ammonium nitrate for PCIS, MOUDI, and SMPS.

Particle size	Concentration	Concentration (µg/m ³)				
range	PCIS	MOUDI	SMPS			
(µm)						
Test I						
0.5-1.0	18.3	26.7	17.8			
0.25-0.5	63.3	82.2	73.0			
< 0.25	123.3	25.6	98.0			
Total	204.0	144.5	188.8			
Test II						
0.5-1.0	22.1	38.9	15.8			
0.25-0.5	40.0	58.9	52.6			
< 0.25	83.3	13.3	74.4			
Total	145.4	111.1	142.8			
Test III						
0.5-1.0	29.7	31.1	18.4			
0.25-0.5	63.3	86.7	66.0			
< 0.25	91.0	24.4	94.0			
Total	184.0	142.2	178.4			
Test IV						
0.5-1.0	18.3	20.6	15.3			
0.25-0.5	73.3	88.9	72.8			
< 0.25	94.0	43.3	98.8			
Total	185.6	152.8	186.9			

relatively high pressure drop across the 0.18 µm stage as well as the after-filter stage, which would enhance volatilization of ammonium nitrate. The pressure drops across the 0.18 µm and after-filter MOUDI stages are 24.4 and 50 inches of H₂O, respectively. By comparison, in the last two PCIS stages the pressure drops were 3.9 and 4.7 inches of H_2O . This difference in pressure drop apparently causes substantial volatilization of ammonium nitrate from the MOUDI filter. The ability of the PCIS to fractionate by size and collect particles under a low pressure drop, which in turn minimizes volatilization losses, is an exceedingly important feature. This is especially so since, in large metropolitan areas like Los Angeles, a significant fraction of fine particulate matter is associated with such labile species as ammonium nitrate and semi-volatile organic compounds. Furthermore, personal activities including smoking and cooking are also known to generate particles with a large fraction of volatile constituents. The excellent agreement between the SMPS and the PCIS ammonium nitrate concentrations clearly supports the conclusion that the volatile species are preserved during particle collection by the PCIS.

Wind Tunnel Tests

The results of the wind tunnel tests are summarized in Figure 12. As is evident from the figure, the particle penetration characteristics of the PCIS 2.5 μ m stage are unaffected by the wind speeds. The penetration curves for both wind speeds tested, 3.0 and 8.0 km/hour, show a very close agreement with those determined in control laboratory experiments for that PCIS stage. This finding is particularly important because it demonstrates that the



Figure 12. Penetration efficiency of the 0.25 μ m PCIS stage vs. particle aerodynamic diameter at two different wind speeds.

PCIS can be used for personal sampling in indoor or occupational environments under non-quiescent air conditions. The wind tunnel tests show the expected 50% cut at 2.5 µm.

FIELD EVALUATION OF THE PCIS IN CO-LOCATION WITH OTHER SAMPLERS IN CLAREMONT/LOS ANGELES

Mass Concentration Measurements

Coarse Particle (PM_{10-2.5}) Concentrations

Coarse particle concentrations measured by the PCIS were compared to concurrent measurements by the MOUDI. Inter-comparisons between the PCIS and the MOUDI indicate an overall excellent overall agreement with an average $PM_{10-2.5}$ PCIS to MOUDI ratio of 1.007 (± 0.07). In Figure 13, the coarse particle mass concentrations obtained with the PCIS and those obtained with the MOUDI are plotted together with the linear regression lines and the regression coefficients. As evident from the figure, the PCIS concentrations (R²= 0.95).



Figure 13. Coarse PM mass concentrations obtained from PCIS and MOUDI.

Fine Particle (PM_{2.5}) Concentrations

Fine particle ($PM_{2.5}$) mass concentrations measured by the PCIS were compared with those obtained with MOUDI and the semi-continuous SMPS-APS samplers. The $PM_{2.5}$ concentration ranges and ratios (mean with standard deviation) of the PCIS, the MOUDI and the SMPS-APS are summarized in Table 5. Intercomparisons between the PCIS and the SMPS-APS indicate an overall excellent agreement with an average

Table 5. $\ensuremath{\mathsf{PM}_{2.5}}$ mass concentrations and ratios for PCIS, MOUDI, and SMPS-APS.

Instruments Compared		Concentration Range (gm ⁻³)	Concentration Ratio (mean \pm S.D.)
PCIS to N	IOUDI		1.16 (± 0.22)
	PCIS	5.50-46.60	
	MOUDI	3.37-42.75	
PCIS to S	MPS-APS		1.02 (± 0.34)
	PCIS	3.40-63.80	
	SMPS-APS	4.83-57.34	

 $PM_{2.5}$ ratio of 1.02 (± 0.34). However, the MOUDI $PM_{2.5}$ concentrations are slightly lower than those measured by the PCIS; the average PCIS to MOUDI $PM_{2.5}$ concentration ratio was 1.16 (± 0.22). This difference could be attributed to a possible underestimation of particles <0.25 µm in size by the MOUDI (explained in more detail in the following paragraph). The fine particle mass concentrations obtained with the PCIS and those obtained with the MOUDI and the SMPS-APS are plotted in Figures 14 and 15 together with the linear regression



Figure 14. PM_{2.5} mass concentrations obtained with PCIS and MOUDI.



Figure 15. PM_{2.5} mass concentrations obtained with PCIS and SMPS-APS.

lines and the regression coefficients. As evident from these figures, the PCIS concentrations are highly correlated with both the MOUDI and the SMPS-APS $PM_{2.5}$ data. The value of R^2 for the PCIS versus the MOUDI is 0.90; for the PCIS versus the SMPS-APS it is 0.70.

Size-fractionated Mass Concentrations

Figure 16 depicts size-fractionated average mass concentrations obtained with the PCIS and the MOUDI along with standard errors in the respective size fractions using data from a total of 13 experiments.



Figure 16. Comparison of size fractionated mass concentrations obtained with PCIS and MOUDI.

A summary of the PCIS and the MOUDI sizefractionated mass concentrations is shown in Table 6. Good overall agreement between the PCIS and the MOUDI was obtained for particles in the ranges of 0.25 to 0.5; 0.5 to 1.0; and 1.0 to 2.5 µm, with the average PCIS to MOUDI concentration ratios being 0.98 (\pm 0.21), 0.88 (\pm 0.13) and 1.21 (\pm 0.35), respectively. Paired *t*-tests between the PCIS and MOUDI concentrations for these three size ranges indicate that these concentrations are not significantly different. Statistically significant differences (p < 0.001), however, were observed for the

 $\label{eq:constraint} \begin{array}{l} \textbf{Table 6. Size-fractionated mass concentrations obtained with PCIS and MOUDI. \end{array}$

Aerodynamic Size Range (m)	Concentration Range (gm ⁻³)		Concentration Ratio (mean ± S.D.)	p- value
	PCIS	MOUDI		
10-2.5	7.18-21.99	7.71-21.32	1.01(±0.70)	0.94
2.5-1.0	1.60-12.81	1.27-16.13	1.21(±0.35)	0.82
1.0-0.5	0.70-13.73	0.51-18.52	0.88(± 0.13)	0.67
0.5-0.25	0.77-11.65	2.17-6.13	0.98 (± 0.21)	0.93
< 0.25	3.5-13.27	1.69-8.5	2.02 (± 0.59)	<0.001*

<0.25 µm range, with the MOUDI underestimating these concentrations. The average PCIS to MOUDI ratio was 2.02 (\pm 0.59). This difference could well be attributed to the high flow rate of the MOUDI (30 L/min) and consequently a relatively high pressure drop across its 0.18 µm stage and after-filter. These drops could enhance the volatilization of labile species such as ammonium nitrate and lead to an underestimation of overall mass of particles <0.25 µm in size. The pressure drop across the 0.18 µm and after-filter MOUDI stages are 24.4 and 50 inches of H₂O, respectively, compared to 3.9 and 4.7 inches of H₂O of the last two PCIS stages. This difference in pressure drop apparently causes substantial volatilization of labile species from the MOUDI filter. Similar discrepancies between the last stage of the PCIS and the MOUDI were also observed during the laboratory tests using ammonium nitrate as a surrogate of labile species. These tests clearly indicated that losses (if any) from the PCIS are substantially lower than those in the MOUDI.

Figure 17 shows a comparison of the size-fractionated mass concentrations obtained with the PCIS and the SMPS-APS samplers together with standard errors in the respective size fractions. A total of fourteen 24-hour



Figure 17. Comparison of size-fractionated mass concentrations obtained with PCIS and SMPS-APS.

sampling experiments were conducted. The sizefractionated mass concentrations and their ratios together with respective p-values for paired *t*-tests between the PCIS and the SMPS-APS are summarized in Table 7. Very good overall agreement is obtained between the PCIS and the SMPS-APS concentrations for the particle sizes 0.25 to 0.5; 0.5 to 1.0; and 1.0 to 2.5 µm with average PCIS to SMPS-APS mass ratios of 1.16 (\pm 0.26), 1.03 (\pm 0.15) and 0.96 (\pm 0.14). For particles <0.25 µm in size, the PCIS slightly overestimates the mass concentrations as compared to the SMPS by a factor of $\ensuremath{\text{Table 7. Size-fractionated}}\xspace$ mass concentrations and ratios for PCIS and SMPS-APS.

Aerodynamic Size Range (m)	Concentration Range (gm ⁻³)		Concentration Ratio (mean ± S.D.)	p- value
	PCIS	SMPS-APS		
2.5-1.0	0.70-12.81	0.69-21.73	0.96 (± 0.14)	0.56
1.0-0.5	0.47-13.73	1.09-7.58	1.03 (± 0.15)	0.98
0.5-0.25	0.70-16.70	0.58-15.69	1.16 (± 0.26)	0.83
< 0.25	2.00-13.27	2.30-14.23	1.31 (± 0.24)	0.44

1.31 (\pm 0.24). This difference may be due to the different size classification principles employed by the PCIS and the SMPS, with the former measuring aerodynamic and the latter measuring mobility particle diameters. As indicated by previous studies (McMurry et al., 2002), particles in urban areas originating from vehicular emissions contain a high fraction of fractal agglomerates. These particles are relatively hollow and would be classified in the ultrafine PM mode aerodynamically. However, due to their large surface area, the SMPS would place a substantial portion of these particles in the accumulation mode because of their increased mobility diameter. Paired *t*-tests between the PCIS and the SMPS-APS concentrations for these four size ranges indicate that these concentrations are not significantly different at the p=0.05 level.

Chemical Species Measurements

Figure 18 shows size-segregated $PM_{2.5}$ sulfate concentrations measured by the MOUDI and the PCIS together with standard deviation in respective size fractions. The $PM_{2.5}$ sulfate concentrations measured by



Figure 18. MOUDI and PCIS sulfate concentrations.

the PCIS are in close agreement with concurrent MOUDI measurements (Table 8). Paired *t*-tests between the PCIS and MOUDI concentrations for the <0.25; 0.25 to 0.5; 0.5 to 1.0; and 1.0 to 2.5 μ m size ranges indicate that these concentrations are not significantly different (p=0.50, p=0.17, p=0.96 and p= 0.12, respectively).

Table 8. Summary of comparison between the PCIS and MOUDI for $\rm PM_{2.5}$ chemical species concentrations.

Chemical Specie	Mean of the ratios \pm S.D.	Mean difference \pm S.D.	p-value
Nitrate	1.04 ± 0.36	-0.82 ± 2.49 (-10.04%)	0.70
Sulfate	0.98 ± 0.10	0.038 ± 0.26 (0.97%)	0.97
EC	1.30 ± 0.37	0.18 ± 0.22 (20.28%)	0.19
OC	1.94 ± 0.52	5.2 ± 2.35 (46.94%)	<0.01*

* Statistically significant difference in values.

Overall $PM_{2.5}$ nitrate concentrations as measured by the PCIS also agree closely with the MOUDI concentrations (Table 8). Figure 19 depicts sizefractionated PM_{2.5} nitrate measurements by the MOUDI and the PCIS together with standard deviations in respective size fractions. Paired *t*-tests between the PCIS and MOUDI concentrations for size ranges <0.25; 0.25 to 0.5; 0.5 to 1.0; and 1.0 to 2.5 µm indicate that these concentrations are not significantly different (p=0.52, p=0.33, p=0.44 and p=0.09, respectively). The difference between the PCIS and MOUDI concentrations appears to approach significance in the smaller particle size range, with the PCIS measuring about 1.69 times higher concentrations than the MOUDI. Lower MOUDI concentrations in the <0.25 µm range may be due to evaporative losses from the MOUDI after-filter, as discussed previously. The low nitrate content of the urban aerosol in that size range does not affect the overall good PM_{2.5} agreement between the concentrations of the PCIS and the MOUDI.



Figure 19. MOUDI and PCIS nitrate concentrations.

Figures 20 and 21 show a comparison of the sizefractionated PM2.5 EC and OC concentrations obtained with the PCIS and the MOUDI together with standard deviations in the respective size fractions. For these measurements, the PCIS and the MOUDI agree well, within 15% for particle size ranges from 2.5 to $0.25 \ \mu m$. However, the MOUDI underestimates the OC and EC contents for particles <0.25 µm. The average PCIS-to-MOUDI ratios are 2.43 (\pm 0.78) and 1.32 (\pm 0.37) for this size range of OC and EC, respectively. While EC concentrations of the PCIS and the MOUDI for the <0.25 μ m range are not significantly different (p=0.12), those based on OC are significantly different (p=0.01). The lower EC concentrations may be due to the slight difference in the cutpoints of the impaction stages of the PCIS and the MOUDI (0.25 versus 0.18 µm, respectively). Since a substantial mass fraction of EC may be found in the $<0.25 \mu m$ range, small differences in the size ranges in which PM are classified by the two samplers may



Figure 20. Comparison of elemental carbon concentrations obtained with MOUDI and PCIS.



Figure 21. Comparison of organic carbon concentrations obtained with MOUDI and PCIS.

result in appreciable differences in their concentrations. This is further supported by the slightly higher EC concentrations for the MOUDI in the 0.25 to 0.50 μ m range and by the overall good agreement in the total PM_{2.5} EC concentrations obtained between these samplers, as shown in Table 8.

The significantly lower MOUDI concentrations observed in the last stage and in the after-filter could be attributed to volatilization of carbon particles collected under low pressure in these stages. This was discussed in previous sections. An additional explanation for the discrepancy between the PCIS and the MOUDI in the <0.25 μ m range may be that more gaseous organic compounds are absorbed on the quartz after-filter of the PCIS. While organic vapor phase absorption on the quartz filter of the MOUDI may also be possible, the much lower phase velocity and pressure drop of the PCIS after-filter would favor this process in the PCIS. This hypothesis is further corroborated by OC measurements using continuous monitors, described in subsequent paragraphs.

Figures 22 and 23 show a comparison of nitrate and total carbon measurements respectively by the PCIS and the MOUDI using continuous ADI and Sunset Labs monitors with Zefluor substrates. As can be seen in the figures, the PCIS and the MOUDI agree remarkably well (within \pm 15%) for these measurements for all size ranges, including the <0.25 µm range. This finding is of particular note because it indicates that, for this size range, differences in labile nitrate and OC concentrations observed in the time-integrated experiments of the PCIS and the MOUDI are due to the sample collection process but not to differences in the cutpoints between the two



Figure 22. Comparison of elemental nitrate concentrations obtained with MOUDI and PCIS using ADI continuous monitor (Zefluor substrate).



Figure 23. Comparison of total carbon concentrations obtained with MOUDI and PCIS using ADI continuous monitor (Zefluor substrate).

samplers. Sampling artifacts such as adsorption to and desorption from impaction and filter substrates are phenomena associated with prolonged sampling. Such phenomena include changes in temperature, humidity and vapor phase concentrations that typically occur over periods of several hours. The near-continuous OC and nitrate data indicate that the particle concentrations of the PCIS and the MOUDI penetrating the 0.25 μ m stage are virtually identical when measured by a continuous monitor, whose readings would not be affected by sampling artifacts. However, when sampling these labile species over longer time periods, the increased pressure drop across the MOUDI after filter on which these particle-bound species are collected will enhance evaporation of volatile compounds, as evident from the lower nitrate and organic carbon concentrations measured by that sampler compared to the PCIS. In addition, the possibility of increased adsorption of organic vapors on the PCIS after filter cannot be ruled out.

COMPATIBILITY OF THE PCIS AND THE LELAND LEGACY PUMP

The compatibility of the PCIS and a prototype Leland Legacy Pump developed by SKC, Inc. was tested in a single test over a 24-hour period. The measured flow rate remained the same (9 L/min) throughout the test period. The test run was successful, and the pump and sampler ran smoothly. Adjustments were necessary to ensure continuous sampling. The maximum time for the battery run was 15 hours, requiring the pump to be operated with both the battery and an AC adapter for the 24-hour run time. Furthermore, the maximum programmable sampling time for the pump was 999 minutes. To have 24 hours or 1440 minutes of uninterrupted sampling, it was necessary that the pump be started and stopped manually. Thus, the 24-hour run was not completely automated, and it did not have an external power source. These issues are expected to be resolved in the final configuration of the Leland Legacy pump. Further tests are required to establish the ability of the battery to power the pump for the full 24 hours and compatibility of the pump and sampler system.

IMPLICATION OF FINDINGS

Personal monitors should be inexpensive and easy to use and should provide size-dependent information on the mass and chemical composition of personal exposures to particulate matter. Existing personal monitoring devices can only provide limited resolution (for example, resolution of PM_{10} or $PM_{2.5}$) of particle mass and chemical composition. Such devices may also be bulky, not user-friendly and expensive for large-scale personal monitoring studies. The PCIS overcomes all of these limitations as it can provide size-dependent information on the mass and elemental concentrations of personal exposure to particulate matter. The combination of a relatively high flow rate (by personal sampling criteria) and low pressure drop makes it possible for the sampler to operate in conjunction with a small, battery-operated personal pump. Furthermore, the low pressure drop of 11 inches of H₂O (2.75 kPa) make it possible to operate a sampler with a very low noise level, a very desirable feature in personal monitoring. The total weight of the sampler is approximately 150 g, making it easy to use by such subjects as children and the elderly. This is a very important feature of personal samplers, since recent epidemiological studies have shown that such sensitive subpopulations as children or the elderly may be more severely affected by PM pollution.

These features, combined with a relatively high flow rate for a personal sampler, make it possible to perform 24-hour sampling with sufficient amounts collected in each stage to determine the size-dependent elemental composition of PM personal exposures. An additional advantage of particle collection by impaction is that losses of volatile compounds from the collected particulate matter are substantially reduced (if not eliminated). This is a highly desirable feature of personal samplers, especially since a significant fraction of fine particles is associated with such labile species as ammonium nitrate or semi-volatile organic compounds, particularly in large metropolitan areas like Los Angeles. Furthermore, personal activities including smoking and cooking are also known to generate particles with a large fraction of volatile constituents. Thus sampling artifacts will be minimized and personal exposure levels will not be underestimated, a problem frequently encountered when using filtration as the means for particle collection.

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ABBREVIATIONS

APS	Aerodynamic particle sizer
EC	Elemental carbon
FIDF	Flame ionization detector
HFPS	High flow personal sampler
IOM	Institute of Medicine
MOUDI	Micro orifice uniform deposit impactor
OC	Organic carbon
PAH	Polyaromatic hydrocarbon
PCIS	Personal cascade impactor sampler
PM	Particulate matter
PTFE	Polytetrafluoroethylene
SMPS	Scanning mobility particle sizer
VOAG	Vibrating orifice aerosol generator

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