

Field evaluation of a personal cascade impactor sampler (PCIS)

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Abstract

This paper presents the field evaluation of a personal cascade impactor sampler (PCIS). 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 – 0.5 , 0.5 – 1.0 , 1.0 – 2.5 and 2.5 – 10 μm . The PCIS operates at a flow rate of 9 liters per minute (l/min) using a very high efficiency, battery-operated light pump at a pressure drop of 11 in H_2O (2.7 kPa). For field evaluation, the PCIS was collocated with other samplers including the micro-orifice uniform deposit impactor (MOUDI), scanning mobility particle sizer (SMPS) and aerodynamic particle sizer (APS) in Los Angeles and Claremont, CA. PCIS and MOUDI agree very well for coarse particulate matter (PM) ($\text{PM}_{2.5-10}$) mass concentrations. The fine PM ($\text{PM}_{2.5}$) mass as measured by PCIS is in excellent agreement with SMPS–APS measurement (~ 1.02 times) and slightly higher (~ 1.1 times) than the MOUDI measurements. Time-integrated (size fractionated) $\text{PM}_{2.5}$ mass, inorganic ions (nitrate and sulfate), elemental carbon (EC) and organic carbon (OC) concentrations obtained with PCIS and MOUDI were found to be in very good agreement with few differences in the <0.25 μm size fraction, especially for OC and nitrate measurements. Near-continuous and size fractionated $\text{PM}_{2.5}$ nitrate and total carbon measurements by PCIS and MOUDI using the ADI and Sunset labs monitors are in close agreement for all size fractions, indicating that any differences between MOUDI and PCIS measurements for time-integrated data might be due to artifacts associated with long-term sampling and not to differences in individual cut-points. The performance of the PCIS was also evaluated in wind tunnel tests at wind speeds up to 8 km/h. These tests showed that particle sampling efficiency and separation characteristics of the PCIS are unaffected by the wind speeds for particles up to 10 μm in aerodynamic diameter.

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1. Introduction

Ambient particulate matter (PM) is an air pollutant that has become a major public health concern. A great number of clinical and epidemiological studies have indicated cause and effect associations between respira-

tory effects and exposures to ambient PM (Dockery et al., 1993a; Gordian et al., 1996; Schwartz and Dockery, 1992). Most of the health effects have been associated with the ambient mass concentration of particles smaller than 10 μm (PM_{10}) or, more recently, 2.5 μm in aerodynamic diameter ($\text{PM}_{2.5}$). Results from other studies suggest that particle components, such as sulfate (SO_4^{2-}) and aerosol strong acidity (H^+), also may be associated with increased mortality and other adverse health impacts (Ayres et al., 1989; Bates and Sizto, 1989;

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Bates et al., 1990; Dockery et al., 1993b; Raizenne et al., 1989, 1993; Thurston et al., 1992, 1993). More recent studies in Southern California (Li et al., 2003) demonstrated increased biological potency of the spatially inhomogeneous urban ultrafine particles, which was related to their high content of redox cycling organic chemicals and their ability to damage mitochondria.

In light of these findings, the sufficiency of the National Ambient Air Quality Standard (NAAQS) for PM, in terms of both its regulated level and species has become a matter of concern. In order to address these concerns, knowledge of relationship among the various particle measures and their personal exposure profiles becomes essential. Both indoor and outdoor concentrations have been found to be poor estimators of personal exposures to PM₁₀ and its components, as neither indoor nor outdoor concentrations suffice to account for the observed interpersonal variability in their exposures. Daytime personal PM₁₀ exposures were found to be approximately 50% higher than corresponding indoor and outdoor levels (Thomas et al., 1993), while personal SO₄²⁻ and H⁺ exposures were found to be higher than indoor, but lower than outdoor concentrations (Suh et al., 1994).

Understanding of individual exposures to PM can be significantly improved by the use of personal monitors, as these samplers incorporate the effects of factors like indoor pollutant sources and human time-activity patterns. Coupled with recent advances in the area of molecular source tracers (Schauer et al., 2002), these samplers can enhance the quality of studies linking personal exposures to specific outdoor or indoor sources and help determine degrees to which they influence personal exposure.

A personal sampler for PM that allows separation of airborne particles in several size ranges has been developed and evaluated in the laboratory (Misra et al., 2002). The personal cascade impactor sampler (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, 0.25–0.5, 0.5–1.0, 1.0–2.5 and 2.5–10 µm. It operates at a high flow rate (91/min) by personal sampling standards that make chemical analyses of the size-fractionated particles possible within a period of 24-h or less. The only other personal cascade impactors reported in the aerosol literature were developed by Rubow et al. (1987) and Demokritou et al. (2002). The Rubow et al. impactor operates at a flow of 21/min and its smallest cutpoint size is 0.5 µm. Demokritou et al. (2002) have developed a personal cascade impactor that also operates at a flow rate of 51/min with smallest cutpoint size of 0.5 µm. The development of the PCIS therefore constitutes a major improvement over the prior state-of-the-art in the field of personal monitoring because of its much higher flow

rate and its ability to classify particles as small as 0.25 µm in aerodynamic diameter.

In this paper, we present results from a field evaluation of the PCIS conducted in two locations of Southern California, at Claremont and Los Angeles, respectively. The field evaluation was performed via a comparison to collocated samplers including micro-orifice uniform deposit impactor (MOUDI, Model 110, MSP Corp, Minneapolis, MN), scanning mobility particle sizer (SMPS, TSI Model 3936) and aerodynamic particle sizer (APS, TSI Model 3320). Correlations between the instruments will serve to validate the sampler's field performance. The performance of PCIS is also evaluated in the wind tunnel.

2. Methods

2.1. Description of the PCIS

Design and a comprehensive laboratory evaluation of the PCIS are described in detail by Misra et al. (2002). Thus, only a brief description is presented in this paper. The PCIS is a miniaturized cascade impactor, consisting of four impaction stages and an after-filter that allows the separation and collection of airborne particles in five size ranges (Figs. 1a–d). The sampling flow rate is 91/min and the measured total pressure drop across the sampler is 11 in of H₂O (2.7 kPa). The PCIS is used in combination with the Leland Legacy Sample Pump (SKC Inc., Cat. No. 100–3000). The total weight of the sampler is approximately 150 g, thus easy to be used by subjects such as children or 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. Particles in the size range of 0.25–10 µm are accelerated in rectangular-shaped nozzles and collected on commercially available 25 mm substrates made of quartz (Pallflex Corp., Putnam, CT), aluminum or Zefluor (0.5 µm pore, Gelman Science, Ann Arbor, MI). Particles smaller than 0.25 µm are collected on a 37 mm after-filter made of either quartz or Teflon (PTEF, 2 µm pore, Gelman Science, Ann Arbor, MI). For real-life usage, the PCIS is designed to clip onto a subject's collar in the breathing zone and the pump clips on the subject's belt.

2.2. Field experiments

The performance of the PCIS was evaluated in a field study, which was conducted from March to November 2002. Outdoor sampling was initiated at Claremont, CA, the location at the time of the Southern California Supersite mobile laboratory facility. This site is located approximately 45 km east of downtown Los Angeles. Sampling was conducted during 24-h periods once a

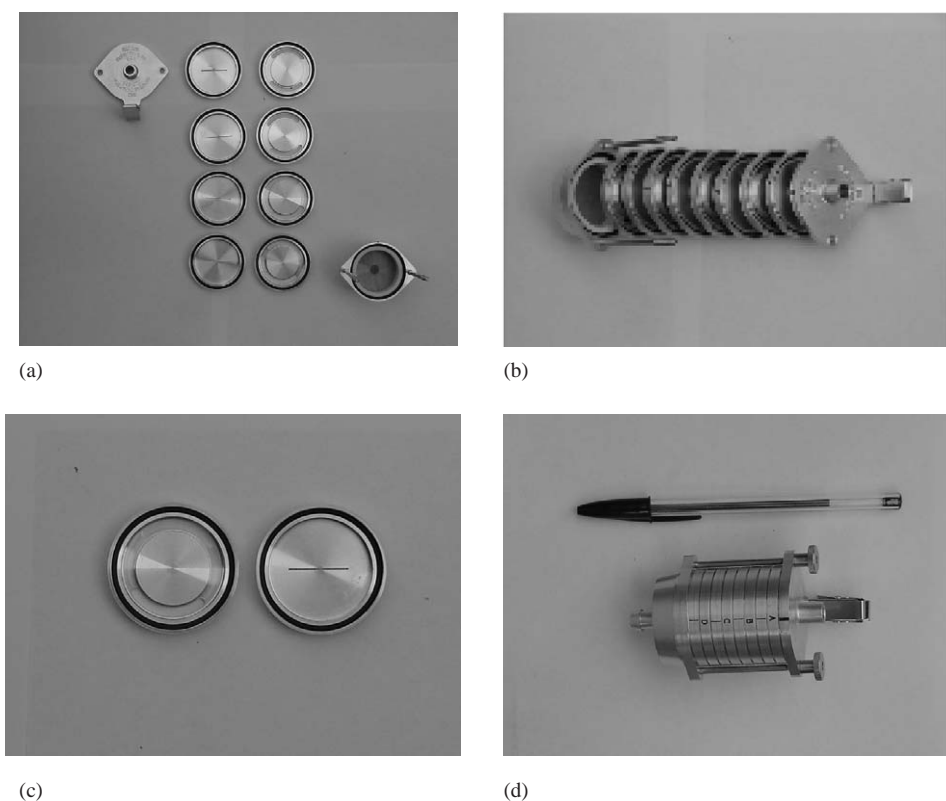


Fig. 1. Pictures of the PCIS.

week. The PCIS was collocated with the micro-orifice uniform deposit impactor (MOUDI™, MSP Corp. Minneapolis, MN) and the scanning mobility particle sizer (SMPS™, TSI Model 3936)—aerodynamic particle sizer (APS™, TSI Model 3320) tandem inside the Particle Instrumentation Unit, a mobile laboratory trailer that was developed through funds provided by the US Environmental Protection Agency and is being currently used in large-scale field studies that are part of the Southern California Particle Center and Supersite activities. As of July 2002, sampling was continued outdoors in the environment of University of Southern California, the next sampling site of the Southern California Supersite. The school is located about 3 km south of downtown Los Angeles and represents a typical urban area in which the aerosol is primarily emitted by vehicles.

Size-segregated $PM_{2.5}$ mass, inorganic ions (nitrate and sulfate), elemental carbon (EC) and organic carbon (OC) concentrations measured by PCIS and collocated MOUDI were compared. MOUDI operates at a higher flow rate of 301/min compared to PCIS. Concentrations obtained from the MOUDI were grouped in the following size ranges: <0.18, 0.18–0.32, 0.32–0.5, 0.5–1.0, 1.0–2.5 and 2.5–10 μm , whilst for PCIS, the ranges were: <0.25, 0.25–0.5, 0.5–1.0, 1.0–2.5 and 2.5–10 μm .

The MOUDI does not have a 0.25 μm cutpoint stage. In order to therefore make the MOUDI cutoff size ranges comparable to those of the PCIS, 50% of the mass measured in the 0.18–0.32 μm size range was added to that in the 0–0.18 μm range and 50% to that in the 0.32–0.50 μm . It should be noted that this conversion was only an effort to bring the MOUDI and PCIS aerosol size ranges closer to each other; it assumes that the aerosol size distribution in that range is log-normal, an assumption that may or may not be valid, and is thus likely to introduce some errors in the comparison experiments between MOUDI and PCIS. For mass and inorganic ions concentration measurements, the MOUDI substrates were 47 mm Teflon filters, followed by a 37-mm Teflon after-filter, whereas the PCIS substrates were 25 mm Zeffluor substrates followed by a 37 mm Teflon after-filter as the last stage. For EC and OC analysis, 25 and 35 mm prebaked aluminum foil substrates were used for the PCIS and MOUDI stages, respectively, followed by 37 mm prebaked quartz filters.

For mass concentration measurements, the substrates were first weighed before and after each field tests using a Mettler 5 Micro-balance (MT 5, Mettler-Toledo Inc., Highstown, NJ), under controlled relative humidity (e.g. 40–45%) and temperature (e.g., 22–24°C) conditions. At the end of each experiment, filters were stored in the

control humidity and temperature room for 24 h prior to weighing in order to ensure removal of particle-bound water. Particle-bound sulfate and nitrate concentrations were determined by means of ion chromatography (IC) similar to that employed by Harrison and Peak (1996). To determine the EC and OC content of PM, the substrates were analyzed by means of a thermo-analysis technique (Fung, 1990).

The semi-continuous SMPS–APS particle mass concentrations were determined by integrating the cumulative number count for the respective size fractions, and converting to mass by assuming particles to be perfect spheres and an average particle density 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 the following equation (Peters et al., 1993):

$$\sqrt{C_a}d_a = \sqrt{\frac{C_{me}\rho_p}{\chi\rho_0}}d_{me}, \quad (1)$$

where d_a is aerodynamic diameter, d_{me} is the mobility equivalent diameter, C_a is the slip correction factor for the aerodynamic diameter, C_{me} is the slip correction factor for the mobility equivalent diameter, χ is the dynamic shape factor, ρ_p is the density of the particle (1.6 g/cm^3), and ρ_0 is the unit density (1 g/cm^3). When performing this conversion, the mobility equivalent diameter was assumed to be equal to the equivalent volume diameter, while particles were assumed to be perfect spheres (dynamic shape factor, $\chi = 1$). The hourly SMPS-APS PM concentrations were integrated into 24 h intervals and compared to those measured concurrently by the PCIS.

In addition to the IC analysis, size segregated $\text{PM}_{2.5}$ nitrate concentrations from PCIS and MOUDI were measured using a continuous nitrate monitor from aerosol dynamics Inc. (ADI) described in more detail by Stolzenburg et al. (2002). Near continuous (10-min) data were obtained by connecting the ADI monitor (sampling at a flow of 1 l/min) to ports placed downstream of each MOUDI and PCIS stages. 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 2–3 times in order to ensure that there are no significant changes in the concentrations of the ambient $\text{PM}_{2.5}$ aerosol during the course of these experiments.

Size segregated total carbon concentrations in $\text{PM}_{2.5}$ by MOUDI and PCIS were also measured using a continuous carbon monitor (Sunset Laboratory Semi-Continuous OC-EC Carbon Aerosol Analyzer, Sunset Labs, Forest Grove, OR) operating at a flow of 9 l/min. Given that the carbon monitor flow rate is equal to that of the PCIS, the entire flow after a given PCIS stage was

drawn into the carbon analyzer, whereas MOUDI concentrations in each stage were measured by diverting 9 of the 30 l/min downstream of each stage, as described above for nitrate measurements. 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 30 min (as opposed to 10 min in the case of nitrate tests). In both series of experiments, the PCIS was used with Zefluor impaction substrates and the MOUDI with Teflon substrates. A total of 8 field experiments were conducted for nitrate and 10 for carbon measurements.

2.3. Wind tunnel tests

The ability of the PCIS to sample isokinetically particles at moderate wind speeds was evaluated in the wind tunnel facility of School of Public Health, UCLA. The wind tunnel is described in detail in Hinds and Kuo (1995) and by Kennedy et al. (2001). The tunnel has a $1.6 \times 1.6\text{ m}^2$ cross-section and was operated at two wind speeds (3 and 8 km/h). A plywood baffle was placed about 0.5 m upstream of the aerosol generation system to promote mixing. A vibrating orifice aerosol generator (VOAG) (Model 3450, TSI Inc., St. Paul, MN) was used to generate monodisperse particles. The droplet size primarily depends on the orifice size for a given solution feed rate and the frequency. For these experiments, a $20\text{ }\mu\text{m}$ orifice was used for generating Typical VOAG operating parameters were 0.150 ml/min of feed rate at 65–70 kHz. Uranine tagged oleic acid was used as a non-volatile solute for generating particles with acetone as the solvent. Approximately, 2 g of uranine dye was dissolved in 50 ml of methanol to prepare the tracer solution and was left overnight to dissolve the dye in the solution with concomitant settling of the undissolved uranine dye. A 20–40% of this solution was then added to the oleic acid–acetone solution to generate monodisperse aerosols in the size of 2–10 μm . The monodispersity of the generated aerosols was confirmed by observing the generated particles under a microscope, which also corroborated the size of the particles.

The vibrating aerosol orifice was itself mounted on a shaft, which moved both up and down and sideways to promote uniform injection. Three isokinetic samplers were placed around the PCIS. The PCIS was placed such 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. An earlier work by Hinds and Kuo (1995) describes the positioning of isokinetic samplers in detail.

Considering that isokinetic sampling is an issue of concern mainly for super-micrometer particles, tests

were only conducted for the particle range of 2–10 μm . The PCIS was thus used in a modified configuration, whereby the all impaction stages were moved except of the 2.5 μm cut off stage, which was followed by the PCIS after-filter. The sampling characteristics of the 2.5 μm cut PCIS stage were determined by comparing the coarse (2.5–10 μm) particle mass concentration obtained by the PCIS 2.5 μm stage to that measured by the isokinetic samplers. The isokinetic samplers and the PCIS 2.5 μm stage were positioned at the same distance from the sample injection point (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 and 8 km/h. Millipore membrane filters (SMWP 02500, Millipore, Bedford, MA) were used to collect uranine tagged oleic acid particles in the isokinetic samplers, whereas a 25 mm Zefflur filter was used as an impaction substrate for the 2.5 μm stage of the PCIS. Each of the experiments was characterized by particle size and wind speed and lasted for about 10–15 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 their fluorescence detection, the extraction of the uranine was done using a buffer solution. The buffer solution was prepared by dissolving 12.4 g of boric acid in 1000 ml of water (solution A) and 19.05 g of sodium

borate in 1000 ml of water (solution B). Diluting 50 ml of solution A and 59 ml of solution B to 200 ml using distilled water yielded the buffer solution.

The Millipore filters were then extracted in glass vials using the buffer solution. Most of the extractions were done using 5–10 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, comparison between the mass concentrations obtained by means of the three isokinetic samplers and the PCIS 2.5 μm stage was performed. 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.

3. Results and discussion

3.1. Coarse particle ($PM_{10-2.5}$) mass concentrations

Coarse particle concentrations measured by PCIS were compared to concurrent measurements by MOUDI. Inter-comparisons between PCIS and MOUDI indicate an overall excellent agreement with an average $PM_{10-2.5}$ PCIS to MOUDI ratio of 1.007 (± 0.07). The coarse particle mass concentrations obtained with PCIS and those obtained with MOUDI are plotted in Fig. 2 along with the linear regression lines and the regression coefficients. As evident from the figure, the PCIS concentrations are also highly correlated with MOUDI ($R^2 = 0.95$).

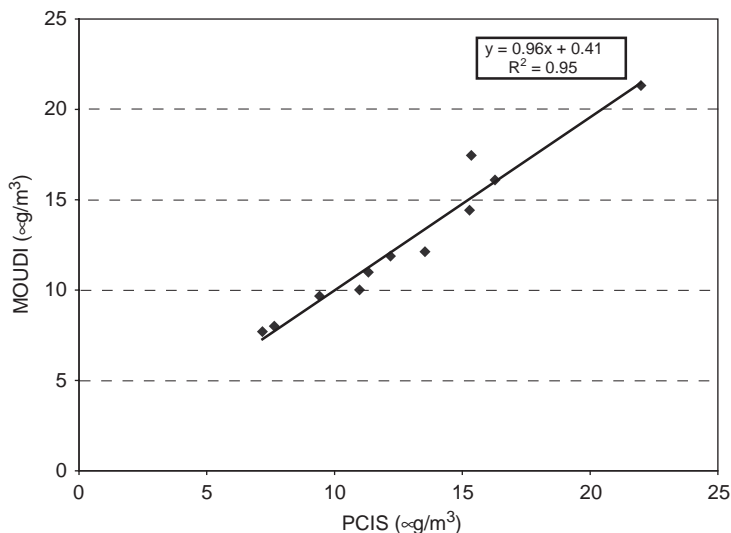


Fig. 2. Coarse PM mass concentrations obtained with PCIS and MOUDI.

3.2. Fine particle ($PM_{2.5}$) mass concentrations

The 24-h averaged 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. A summary of these comparisons is given in Table 1. Inter-comparisons between PCIS and SMPS-APS indicate an overall excellent agreement

between these monitors, with an average PCIS to SMPS-APS $PM_{2.5}$ ratio of 1.02 (± 0.34). However, MOUDI $PM_{2.5}$ concentrations are slightly lower than those measured by the PCIS, with the average PCIS to MOUDI $PM_{2.5}$ concentration ratio being 1.11 (± 0.05). The fine particle mass concentrations obtained with PCIS and those obtained with MOUDI and SMPS-APS are plotted in Figs. 3a and b, respectively, along with the

Table 1

Summary of comparison between the PCIS, SMPS-APS and MOUDI for $PM_{2.5}$ species concentrations obtained from time-integrated measurements

Instruments Compared	Species	Mean of the ratios \pm S.D.	Mean difference ($\mu\text{g}/\text{m}^3$) \pm S.D. (mean relative difference)	<i>p</i> -value
PCIS: SMPS-APS	Mass	1.02 \pm 0.34	0.30 \pm 8.43 (1.19%)	0.96
PCIS: MOUDI	Mass	1.11 \pm 0.05	2.15 \pm 0.93 (9.28%)	0.82
PCIS: MOUDI	Sulfate	1.11 \pm 0.10	0.19 \pm 0.22 (8.04%)	0.86
PCIS: MOUDI	Nitrate	1.22 \pm 0.21	0.64 \pm 0.63 (6.53%)	0.93
PCIS: MOUDI	EC	1.19 \pm 0.37	0.18 \pm 0.22 (20.28%)	0.39
PCIS: MOUDI	OC	1.94 ^a \pm 0.52	5.2 \pm 2.35 (46.94%)	<0.01

^a Statistically significant difference in values at $p = 0.05$ level.

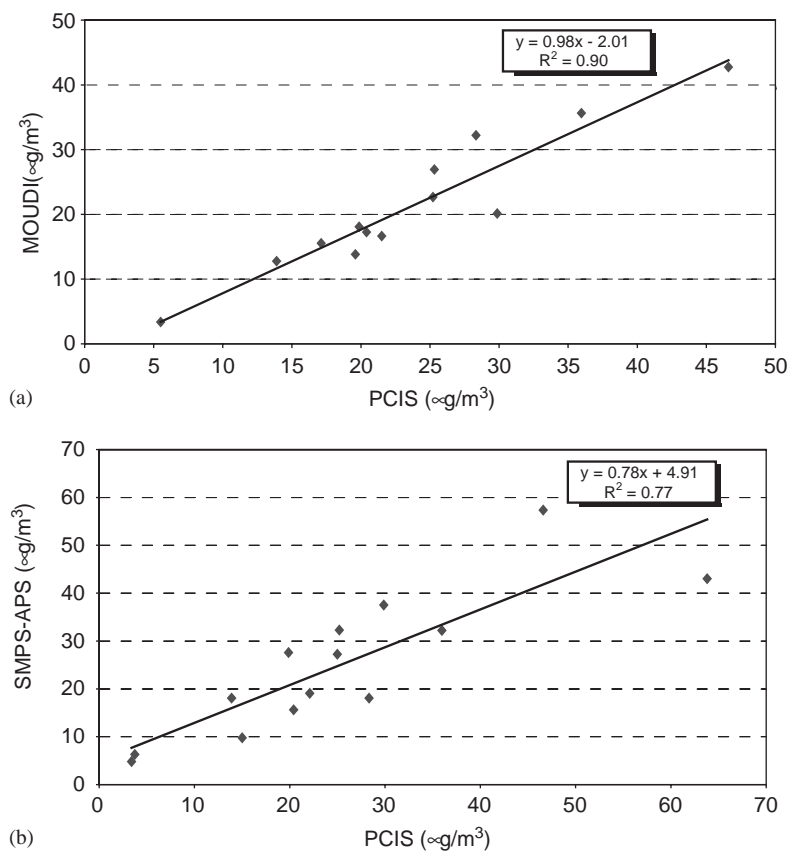


Fig. 3. (a) $PM_{2.5}$ mass concentrations obtained with PCIS and MOUDI and (b) $PM_{2.5}$ mass concentrations obtained with PCIS and SMPS-APS.

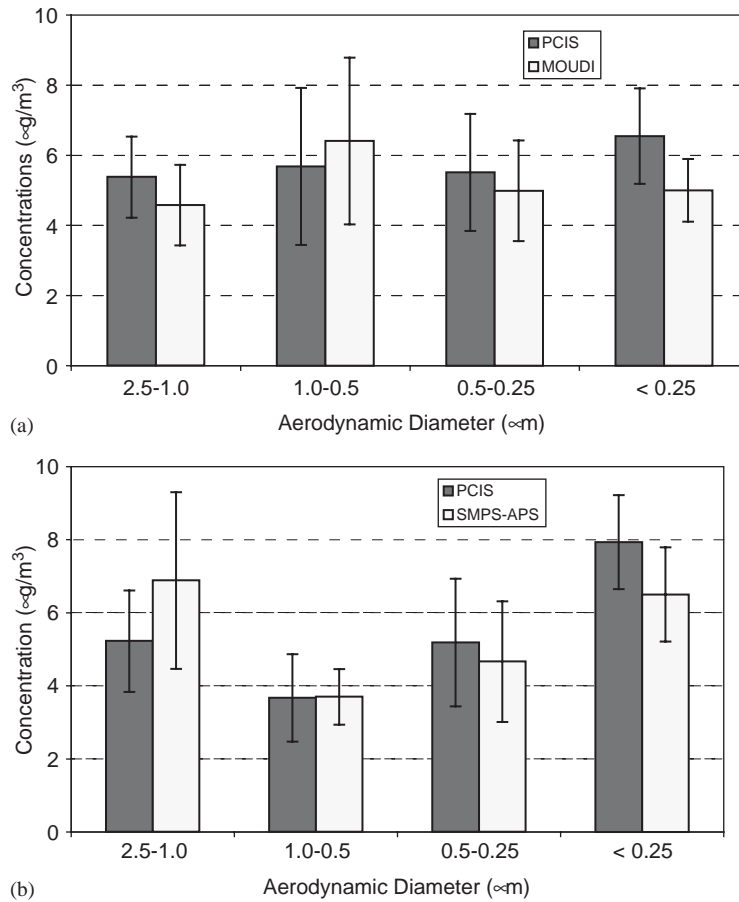


Fig. 4. (a) Comparison of size-segregated mass concentrations obtained with PCIS and MOUDI and (b) comparison of size-segregated mass concentrations obtained with PCIS and SMPS-APS.

linear regression lines and the regression coefficients. As evident from these figures, the PCIS concentrations are highly correlated with both MOUDI and SMPS-APS $PM_{2.5}$ data, with the R^2 for PCIS vs. MOUDI and PCIS vs. SMPS-APS being 0.90 and 0.77, respectively.

3.3. Size-fractionated $PM_{2.5}$ mass concentrations

Fig. 4a depicts size-fractionated average $PM_{2.5}$ mass concentrations obtained with PCIS and MOUDI (error bars represent the standard deviation of field measurements) in the respective size fractions, based on a total of 13 experiments. Good overall agreement between the PCIS and MOUDI was obtained for particles in the aerodynamic diameter ranges of 1–2.5, 0.5–1.0 and 0.25–0.5 μm , with the average PCIS to MOUDI concentration ratios being 1.21 (± 0.15), 0.84 (± 0.12) and 1.09 (± 0.18), respectively. However, for the <0.25 μm range, MOUDI slightly underestimates the mass concentrations; the average PCIS to MOUDI ratio is 1.28

(± 0.12). This difference may be attributed to the high flow rate of MOUDI (301/min) and consequently a relatively high pressure drop across its 0.18 μm stage as well as the after-filter, which would enhance the volatilization of labile species, resulting in the underestimation of overall mass of particles <0.25 μm in size. Paired t -tests between PCIS and MOUDI concentrations for these four size ranges indicates that these concentrations are not statistically significantly different at the $p = 0.05$ level (including the last stage).

Fig. 4b shows a comparison of the size fractionated average $PM_{2.5}$ mass concentrations obtained with the PCIS and SMPS-APS samplers, along with standard deviations, in the respective size fractions. A total of 14 24-h sampling experiments were conducted for these comparisons. Very good overall agreement was obtained between the PCIS and SMPS-APS concentrations for the particle sizes 2.5–1.0, 0.5–1.0 and 0.25–0.5 μm with average PCIS to SMPS-APS mass ratios of 0.96 (± 0.14), 1.03 (± 0.15) and 1.16 (± 0.21). For particles

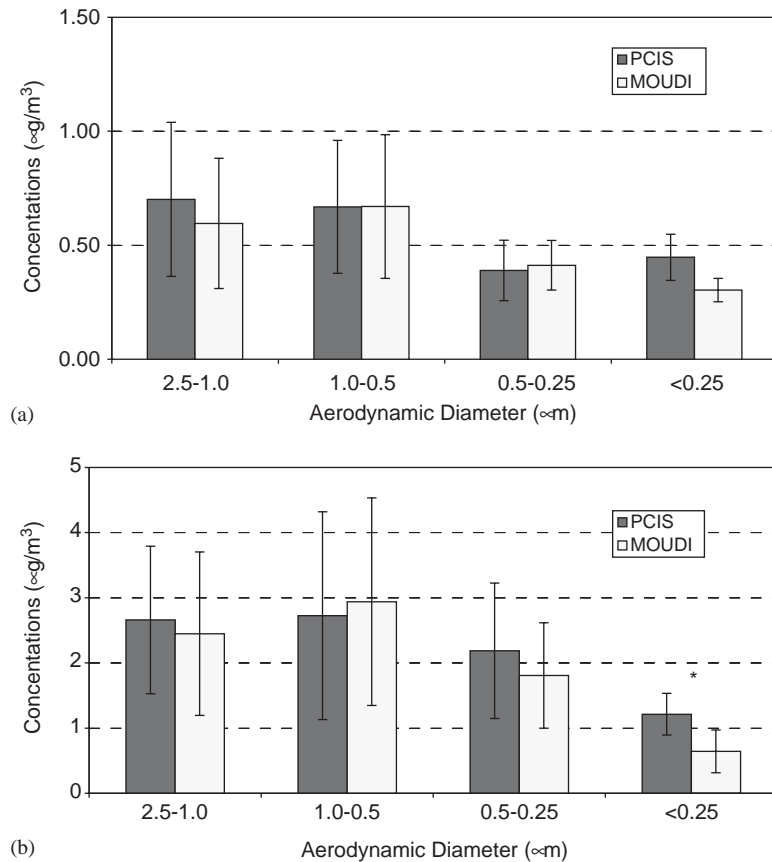


Fig. 5. (a) Comparison of size-segregated sulfate concentrations obtained with PCIS and MOUDI and (b) comparison of size-segregated nitrate concentrations obtained with PCIS and MOUDI. The asterisk denotes statistically different concentrations at the $p = 0.05$ level.

<0.25 µm in size, the PCIS overestimates the mass concentrations as compared to SMPS by a factor of 1.31 (± 0.24). Although not statistically significant at the $p = 0.05$ level, this difference may be due to the different size classification principles employed by the PCIS and 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-like 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 due to their increased mobility diameter. Similar observations between the mass concentrations measured by impactors and those measured by the SMPS–APS tandem were made in a recent study by Shen et al. (2002). As in the case of the PCIS–MOUDI comparisons, paired t -tests between PCIS and SMPS–APS concentrations for these four size ranges indicates that these concentrations are

not statistically significantly different at the $p = 0.05$ level.

3.4. $PM_{2.5}$ chemical species concentrations

Fig. 5a shows size-segregated $PM_{2.5}$ average sulfate concentrations measured by MOUDI and PCIS in respective size fractions. $PM_{2.5}$ sulfate concentrations measured by PCIS are in close agreement with concurrent MOUDI measurements (within 10% or less, Table 1). Paired t -tests between the PCIS and MOUDI concentrations for the 1–2.5, 0.5–1.0, 0.25–0.50 and < 0.25 µm size ranges indicate that these concentrations are not statistically significantly different ($p = 0.82$, $p = 0.99$, $p = 0.90$ and $p = 0.27$, respectively).

Overall $PM_{2.5}$ nitrate concentrations measured by PCIS are also in close agreement with MOUDI (Table 1). Fig. 5b depicts the size fractionated $PM_{2.5}$ nitrate measurements by MOUDI and PCIS along with standard deviations in respective size fractions. Paired t -tests between the PCIS and MOUDI concentrations for size ranges 1.0–2.5, 0.5–1.0, 0.25–0.5 µm indicate that

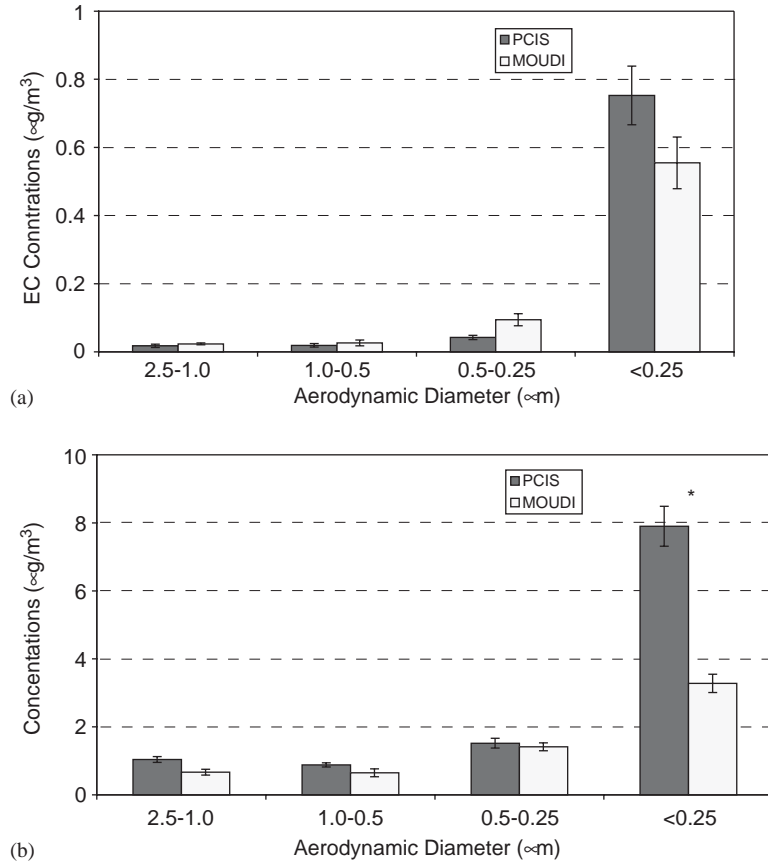


Fig. 6. (a). Comparison of size-segregated EC concentrations obtained with PCIS and MOUDI and (b) comparison of OC concentrations obtained with PCIS and MOUDI. The asterisk denotes statistically different concentrations at the $p = 0.05$ level.

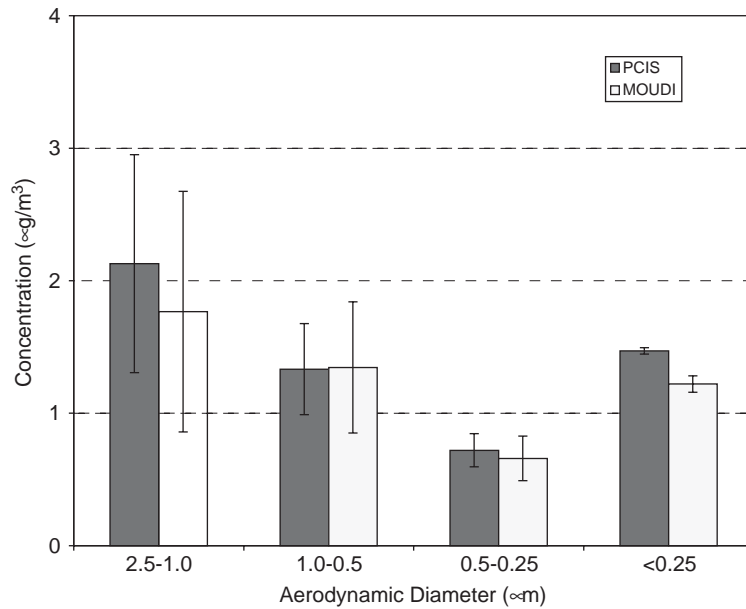


Fig. 7. Comparison of nitrate concentrations obtained with MOUDI and PCIS using continuous monitor (the MOUDI cutpoint of the last stage is 0.18 µm).

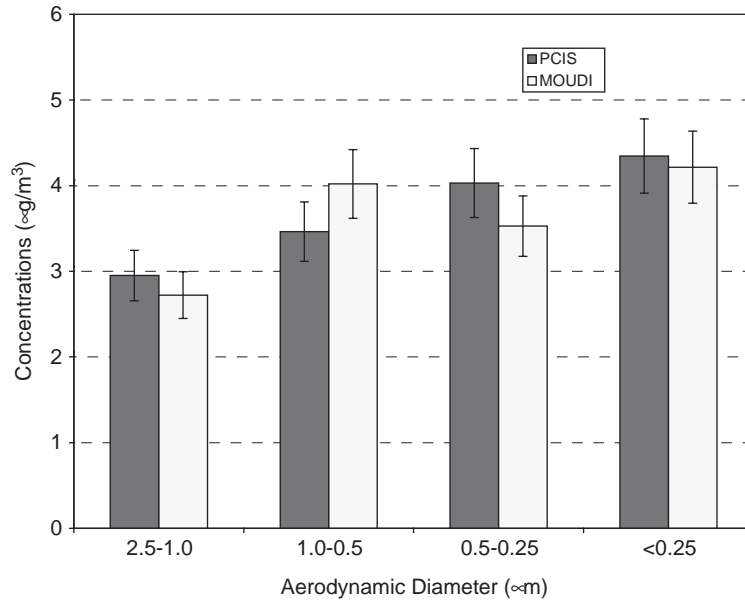


Fig. 8. Comparison of total carbon concentrations obtained with PCIS and MOUDI using continuous monitor (the MOUDI cutpoint of the last stage is 0.18 µm).

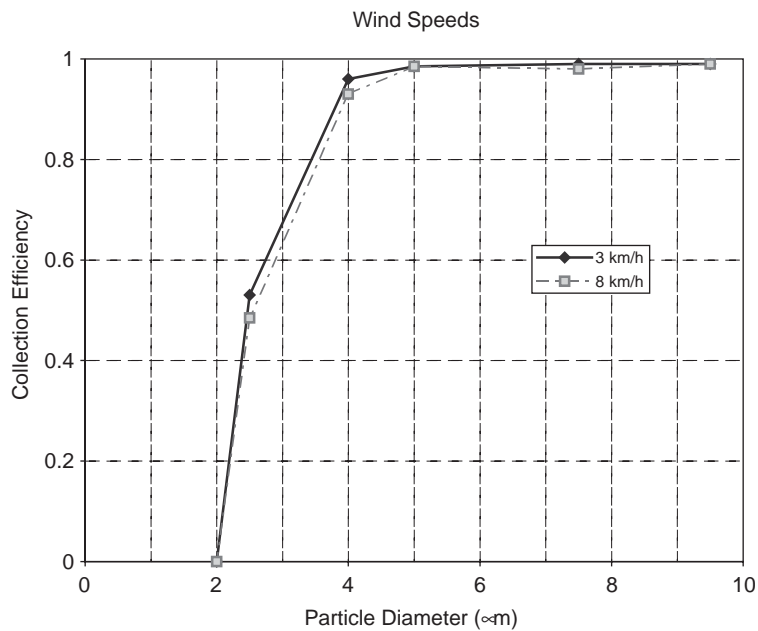


Fig. 9. Collection efficiency of the 2.5 µm PCIS stage vs. particle diameter at two different wind speeds.

these concentrations are not statistically significantly different ($p = 0.90$, $p = 0.93$, $p = 0.78$, respectively). The difference in the PCIS–MOUDI concentrations appears to be significant ($p = 0.04$) in the smaller particle size range of $<0.25 \mu\text{m}$, with the PCIS measuring about $2.7(\pm 0.13)$ times higher concentrations than the

MOUDI. Lower MOUDI concentrations in the $<0.25 \mu\text{m}$ range may be due to evaporative losses from its after-filter stage, as discussed previously. The low nitrate content of the urban aerosol in that size range does not affect the overall good $\text{PM}_{2.5}$ agreement between the concentrations of PCIS and MOUDI.

Figs. 6a and b show a comparison of the average size fractionated EC and OC concentrations, respectively, obtained with the PCIS and MOUDI along with standard errors in the respective size fractions. For these measurements, PCIS and MOUDI agree well (i.e., within 15%) for particle size ranges from 2.5 to 0.25 μm . However the MOUDI underestimates the OC and EC contents for particles $<0.25\ \mu\text{m}$, with the average PCIS to MOUDI ratio being $2.43(\pm 0.78)$ and $1.32(\pm 0.37)$, respectively, for this size range. While EC concentrations of the PCIS and MOUDI for the $<0.25\ \mu\text{m}$ range are not statistically significantly different ($p = 0.12$), those based on OC are ($p < 0.01$). The lower EC concentrations may be due to the slight difference in the classification of the aerosol in the 0–0.25 μm range by PCIS and MOUDI. As previously discussed, the algorithm used to bring the cutpoints 0.0.18 and 0.25 μm of MOUDI and PCIS closer assumes a log-normal distribution of aerosol in these size ranges which may not be always true. Considering that a substantial mass fraction of EC may be found in the 0–0.25 μm range (Hughes et al., 1998), small differences in the size ranges in which PM are classified by the two samplers may result in appreciable differences in their concentrations. This is further supported by the slightly higher MOUDI EC concentrations in the 0.25–0.50 μm range and by the overall good agreement in the total $\text{PM}_{2.5}$ EC concentrations obtained between these samplers, as shown in Table 1.

The significantly lower MOUDI OC concentrations observed in the last stage and after-filter could be attributed to volatilization of organic particles collected under low pressure in that stage. Evaporative losses of semi-volatile compounds from impactor stages are a function of pressure drop across these stages (Zhang and McMurry, 1987). The pressure drop across all MOUDI stages is considerably higher than the corresponding PCIS stages (the pressure drop across the 0.18 μm and after-filter MOUDI stages are 0.06 and 0.125 atmospheres, respectively, compared to 0.009 and 0.01 atmospheres for the last two PCIS stages). This difference in pressure drop may cause some volatilization of labile organic species from particles collected on the MOUDI filter. An additional explanation for the discrepancy between PCIS and MOUDI for the $<0.25\ \mu\text{m}$ range may be increased adsorption of gaseous organic compounds on the quartz after filter of the PCIS. Organic vapor adsorption in the preceding PCIS and MOUDI stages can be ruled out, given that particles are collected on aluminum substrates, but it is quite likely to occur on the quartz after-filters of either sampler. The much lower phase velocity and pressure drop of the PCIS after-filter compared to MOUDI would favor this process in the PCIS. This hypothesis is further corroborated by total carbon measurements using continuous monitors, which are described in subsequent paragraphs.

Figs. 7 and 8 show the comparisons for nitrate and total carbon measurements, respectively, using the semi-continuous ADI and Sunset Labs monitors. As evident from the figures, PCIS and MOUDI agree remarkably well (within $\pm 15\%$) for these measurements for all size ranges, including the 0–0.25 μm range. This finding is of particular note, because it suggests that the differences in labile nitrate and OC concentrations between PCIS and MOUDI for that size range observed in the time-integrated experiments are due to the sample collection process, and not as much to differences in the cutpoints between the two samplers. Sampling artifacts such as adsorption to- and desorption from impaction and filter substrates are phenomena that are associated with prolonged sampling due to changes in parameter such as temperature, humidity and vapor phase concentrations that occur typically in time scales of several hours. The near-continuous carbon and nitrate data indicate that the particle concentrations of PCIS and MOUDI penetrating the 0.25 μm stage are virtually identical when measured by a continuous monitor, the readings of which would not be affected as much by these sampling artifacts. However, when sampling these labile species in 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 OC concentrations measured by that sampler compared to the PCIS. The possibility of increased adsorption of organic vapors on the PCIS after-filter also cannot be ruled out, which would explain the higher OC values obtained in the time integrated experiments.

3.5. Wind tunnel tests

The results of the wind tunnel test are summarized in Fig. 9, which reveals that particle penetration characteristics of the PCIS 2.5 μm stage are unaffected by the wind speeds. Particle collection efficiency increases sharply as particle diameter becomes larger than about 2 μm to about 50% at 2.5 μm (the design cutpoint of that stage) and exceed the 90% value at about 3.5 μm . The collection efficiency curves for all the wind speeds tested, e.g., 3 and 8 km/h, show a very close agreement to that determined in control laboratory experiments for that PCIS stage, described by Misra et al. (2002). This finding is particularly important because it demonstrates that the PCIS can be used for personal sampling in indoor or occupational environments under non-quiescent air conditions.

4. Summary and conclusions

The performance of the PCIS, a miniaturized cascade impactor consisting of four impaction stages and an

after-filter, was evaluated in a field study. PM_{2.5} mass concentrations measured by PCIS and SMPS-APS were compared and found to be in good agreement for all size fractions. The results also show excellent agreement between PCIS and MOUDI for coarse PM (PM_{2.5–10}) mass. The fine PM (PM_{2.5}) mass as measured by PCIS is slightly higher than MOUDI measurement, mostly due to an underestimation of particles <0.25 μm by the MOUDI. PM_{2.5} sulfate measurements by MOUDI and PCIS were found to be in good agreement with some small differences in individual size bins. A comparison of PCIS–MOUDI nitrate concentrations shows significant difference in the smaller particle size range <0.25 μm, with the PCIS measuring about 2.7 (±0.13) times higher concentrations than the MOUDI. Lower MOUDI concentrations in the <0.25 μm range are attributed to possible evaporative losses from the MOUDI after-filter. EC and OC measurements follow a trend similar to mass concentration measurements. MOUDI and PCIS agree well for particles in the range 2.5–0.25 μm, however MOUDI underestimates the carbonaceous content for particles <0.25 μm. The relatively small discrepancy between PCIS and MOUDI for EC measurements is attributed to a difference in classification of particles in 0–0.25 μm range by PCIS and MOUDI. However, the significant difference in OC concentrations could be attributed to either volatilization of OC particles collected under low pressure in the last stages and/or to increased adsorption of gaseous organic compounds on the quartz after-filter of the PCIS.

In an attempt to eliminate sampling artifacts associated with time-integrated measurements, nitrate and total carbon measurements by PCIS and MOUDI using continuous monitors were compared. PCIS and MOUDI agreed well (within ±15%) for these measurements for all size ranges, including the <0.25 μm range. We thus conclude that differences for nitrate and OC concentrations between PCIS and MOUDI observed in the time-integrated experiments are due to the sample collection process and not to differences in the cutpoints between the samplers.

The sampling efficiency of PCIS was also evaluated in wind tunnel tests, which confirmed that sampling and collection of super-micrometer particles by the PCIS is unaffected at wind speeds as high as 8 km/h.

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