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Field Validation of the New Miniature Versatile Aerosol Concentration Enrichment System (mVACES)

Zhi Ning, Katharine F. Moore, Andrea Polidori, and Constantinos Sioutas
University of Southern California, Department of Civil and Environmental Engineering, Los Angeles, California, USA

Recently a new compact aerosol concentration enrichment system was developed at the University of Southern California, specifically intended to provide particle-laden air at flow rates and pressures suitable for interfacing with on-line continuous aerosol instrumentation for chemical analysis such as mass spectrometers. The re-design and engineering of the miniature Versatile Aerosol Concentration Enrichment System (mVACES) and primarily laboratory-based validation of the individual components and overall system has been previously reported (Geller et al. 2005). From September to December 2005, a field performance validation study of the mVACES was conducted in Los Angeles, California at a mixed urban site influenced by both freeway traffic and construction. A variety of continuous and semi-continuous physical and chemical composition measurements were performed to assess the performance of the mVACES compared to accepted methods for validation. Near-ideal performance for aerosol concentration enhancement by the mVACES was observed for mass and number distribution with minimal evidence for distortion of the size distribution. Similarly, near-ideal concentration enhancement factors were observed for both inorganic and organic species suggesting that the mVACES works equally well across the range of externally mixed urban aerosol. The data suggest that aerosol concentration enhancements up to an ideal factor of 20 in a delivered flow on the order of 1.5 liters min^{-1} are readily achievable in an urban environment for the ambient conditions studied.

INTRODUCTION

The concentrations and characteristics of ambient atmospheric aerosols affect many atmospheric and environmental processes. Continuing advancements in techniques to improve the chemical and physical characterization of aerosol particles provide insights into aerosol formation, evolution and subsequent effects. Of particular interest here is the widespread recognition, based upon increasing epidemiological and toxicological evidence, of the association of increased ambient particulate matter (PM) levels with adverse human health effects (Dockery et al. 1989; Schwartz & Dockery 1992; Thurston 1996; Pope et al. 2002; Li et al. 2003). Despite significant progress, substantial uncertainties remain regarding the relevant particles' physiochemical properties and the underlying pathophysiological mechanisms (Delfino et al. 2005). Therefore, further improvements in the accuracy and speed of aerosol measurement techniques are essential not only to understand the sources and atmospheric processes contributing to the ambient PM but also to better characterize exposure in health effects studies.

Particle concentration technologies have proven to be effective in both studies of health effects (Sioutas, Koutrakis, & Burton 1995; Sioutas et al. 1995; Demokritou et al. 2002a; Li et al. 2003) and ambient sampling efforts (Zhao et al. 2005; Geller et al. 2002; Khlystov et al. 2004) while introducing minimal artifacts. Early particle concentrators used slit virtual impactors operated at high flow rates with correspondingly large pressure drops to concentrate the aerosol (Sioutas, Koutrakis, & Burton 1995; Sioutas et al. 1995, 1997). These systems are large, immobile, power-consuming and not effective at concentrating particles less than 150 nm in diameter—a size range associated with major health risks observed in numerous recent toxicological *in vivo* and *in vitro* studies (e.g., Li et al. 2003; Xia et al. 2004; Kleinman et al. 2005). An improved particle concentrator—the Versatile Aerosol Concentration Enrichment System (VACES)—was subsequently developed to effectively concentrate both fine (<1000 nm) and ultrafine (<150 nm) particles at flow rates from 5–60 liters min^{-1} (lpm) without altering the particle physical and chemical properties of the aerosol (Kim et al. 2001a, 2001b; Misra et al. 2004). Operation of the VACES, however, requires constant attendance by trained personnel. Further, while using the VACES to concentrate aerosol upstream of a particle mass spectrometer was demonstrably successful (Zhao et al. 2005), it is not ideally suited to the task due to the mismatch

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Address correspondence to Constantinos Sioutas, University of Southern California, Department of Civil and Environmental Engineering, 3620 South Vermont Ave., Los Angeles, CA 90089, USA. E-mail: sioutas@usc.edu

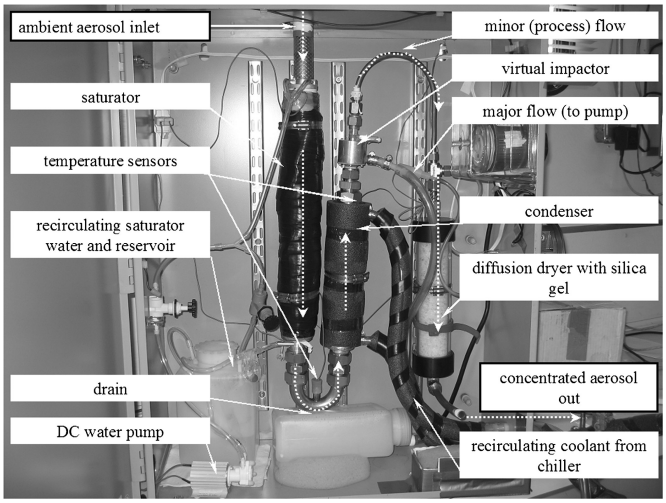


FIG. 1. The mVACES system with components marked (a schematic without the inlet can be found in Geller et al. [2005], Figure 1). The aerosol flow through the mVACES is shown with the white dashed-line arrows.

between the flow rate the VACES provides (minimum 5 lpm) and the input flow rates typically required (on the order of 1 lpm or less). Additionally the size of the VACES restricts its rapid and mobile deployment, particularly where available instrument space is at a premium. Another new particle concentrator taking advantage of the strengths of the VACES while mitigating its limitations was therefore developed.

The "miniature-VACES" (mVACES) is the compact particle concentrator designed to meet this challenge (Figure 1). The system is smaller, lighter (20 kg), more readily controlled than the conventional VACES, and the principal components (exclusive of the pump and chiller) are fixed inside an easily-deployable cabinet (0.6 m length × 0.3 m width × 0.8 m height). The intake flow rate of 30–40 lpm and concentrated aerosol output flows of 1–2 lpm (Geller et al. 2005) are well-suited for use with downstream aerosol mass spectrometry instrumentation. Briefly, ambient aerosols are drawn through a $PM_{2.5}$ inlet and are exposed to heated, moisture-laden air in the saturator section (2.54 cm ID, 45 cm long). The warm, moist process stream is rapidly cooled in the condenser (2.54 cm ID, 27 cm long) and drops formed via water supersaturation. Drops larger than approximately 1.5 μm in diameter are concentrated in the minor flow of the round virtual impactor immediately downstream. The concentrated drops are dried, the original size distribution is recovered and concentrated aerosols are produced. The saturator and condenser are shorter than the length to develop parabolic flow velocity profiles. The presence of the U-tube will also induce vortices into the flow through the condenser. For typical operating conditions, the observed temperature and relative humidity of the air downstream of the saturator are $\approx 30^\circ$C and $>95\%$, respectively, and the air temperature downstream of the condenser is 21–22°C. While complete cooling of the process stream does not occur (the condenser's wall temperature is maintained at $\approx 0^\circ$C), sufficient conditions for drop activation, discussed further below, are produced. Geller et al. (2005) provide a more detailed description of the mVACES and its operation. Additionally, the mVACES can be easily modified to permit aqueous particle collection (described in the next section) which can be useful for both toxicity assays and chemical analyses.

The mVACES' performance depends critically upon providing conditions within the concentrator sufficient for the rapid growth of sub-micron aerosol to super-micron sizes. The water supersaturation achieved internally by rapid cooling in the condenser of the near-saturated air at elevated temperature exiting the saturator must exceed that required for drop activation and growth. The diameter of the smallest particle, d^*, activated for a known supersaturation ratio, S, is given by the Kelvin equation (Hinds 1999):

$$d^* = [4\sigma MW/\rho_p R\ T\ In(S)] \quad [1]$$

where, σ, MW, and ρ_p are the surface tension, molecular weight, and density of water, R is the ideal gas constant, and T the temperature in °K. For the typical mVACES' operating conditions described above and a minimum particle diameter of 20 nm, S must be ≥ 1.15 by Equation (1). The Kelvin equation only considers the effect of particle curvature on drop activation. If particle composition is considered (e.g., the Raoult effect incorporated and the Köhler equation derived), the minimum value of S required to activate a completely soluble 20 nm ammonium sulfate aerosol particle is reduced to 1.02. For externally mixed ambient aerosol (e.g., urban background) complete solubility is a poor assumption and the Kelvin equation S value (1.15) is recovered as the insoluble fraction of the input aerosol increases to 100%. Therefore, successful mVACES operation requires a minimum S ratio of at least 1.02–1.15. The mVACES' ideal theoretical centerline supersaturation ratio, defined here as the ratio of the water vapor partial pressure of the air at the entrance and exit of the condenser for the typical conditions cited above, is 1.55–1.70. This simple calculation represents an approximate maximum value for S because dynamic factors, such as water vapor diffusion, which would tend to reduce S are neglected. This result suggests, however, that it is likely 20 nm aerosol will be activated and subsequently concentrated by the mVACES. Clearly these calculations are highly idealized and are intended to give approximate bounds for S only. Many factors affecting drop activation and S are not accounted for and may compromise the validity of these calculations. These factors are extensively discussed by Demokritou et al. (2002b) and particularly include gradients in the aerosol distribution, temperature and water supersaturation profiles within the condenser. Further, while the flow field inside the mVACES should be laminar at 30 lpm (Re $\approx$ 1700–1800), conditions are in the transition regime for higher flow rates (Re $\approx$ 2200–2400 at 40 lpm). It is possible that localized inhomogeneities in the flow field may also affect the supersaturation ratios achieved. Additionally, some particles, such as soot, may require much higher supersaturations to initiate condensational growth (Kotzick et al. 1997). Therefore,

while these calculations are a useful starting point for instrument design and in developing operating guidelines for the mVACES, there is no substitute for empirically determining its performance using a variety of laboratory-generated and ambient aerosols.

Previous work used almost exclusively laboratory-generated aerosol to evaluate the performance of the mVACES and its individual components (Geller et al. 2005). Several continuous and semi-continuous measurements of the aerosol characteristics were made before and after concentration enrichment in the mVACES, including particle size distributions from the sub- to super-micron mode, particle number counts and cumulative mass distribution. Geller et al. (2005) reported the mVACES is capable of concentrating particles without significant alteration of their physical properties. Also varying the composition of the single-component test aerosols from soluble inorganic to organic and hydrophobic species (e.g., ammonium nitrate, ammonium sulfate, adipic acid, glutaric acid, polystyrene latex spheres) produced little difference in the results, indicating performance is largely independent of particle solubility. Minimal data were collected using ambient indoor and atmospheric aerosol, although what was observed produced results consistent with those observed in the laboratory. Here we expand upon the work reported by Geller et al. (2005) using extensive ambient aerosol collection to validate the mVACES in an urban environment. Given the complexity of the urban aerosol, achieving consistent concentration enrichment factors and performance by the mVACES provides a more comprehensive validation than can be achieved in the laboratory. Of particular interest in the present study was the ability of the mVACES to concentrate effectively species such as trace elements and metals as well as organic compounds, such as organic carbon (OC) and polycyclic aromatic hydrocarbons (PAH), all of which are associated with health effects attributable to PM (Pope et al. 2002; Li et al. 2003, 2004).

EXPERIMENTAL METHODS

Field validation measurements were conducted over a four-month period, from September to December 2005 at an urban sampling site—the Particle Instrumentation Unit of the Southern California Particle Center and Supersite (Sardar et al. 2005)—near the University Park campus of the University of Southern California in Los Angeles. The sampling site is within 100–150 m of a major freeway and adjacent to a multi-story parking structure, industrial and construction sites. An instrumented trailer is maintained at the site and includes a weather station. The sampling inlets used were located at an elevation of ca. 4 m through the roof of the trailer and within ca. 3 m of each other. All instruments were equipped with $PM_{2.5}$ size selective inlets. Ambient conditions at the site varied during the sampling periods. Most samples were collected at 20–22°C, 30–60% RH, and $PM_{2.5}$ concentrations of 10–50 μg m^{-3} (the overall study mean was approximately 33 μg m^{-3}), although the actual range was 13–26°C, 13–80% RH, 6–180 μg m^{-3}. Table 1 lists the co-located instrumentation in the trailer used for this study. Many different instruments are operated in parallel with the mVACES in order to establish the reference physical and chemical properties that the mVACES' performance was validated against.

The mVACES was generally operated as described by Geller et al. (2005). The total inlet flow was set to 30 lpm and the minor flow to 1.5 lpm (except as noted below) for an ideal enrichment factor of 20. The condenser wall temperature ranged from −1 –1°C in response to the applied conditions and the saturator was set to 4–7°C above ambient. If ambient conditions were relatively warm, dry and/or clean (e.g., "offshore" flow conditions, $PM_{2.5} < 20$ μg m^{-3}), the saturator temperature was increased. The silica in the mVACES' dryer was changed every 1–2 days depending upon usage and ambient conditions. Occasionally, if the measurement method was particularly sensitive to humidity, an additional matched pair of diffusion dryers (Model 3062, TSI Inc., Shoreview, MN) was added downstream of the mVACES and to the separate ambient inlet. Instruments making continuous measurements, e.g., the Condensation Particle Counter (CPC TSI 3022, TSI Inc., Shoreview, MN) were directly attached to the mVACES' outlet. For the Microorifice Uniform Deposit Impactor (MOUDI, MSP Corp., Shoreview, MN) and filter-based comparisons (described below) a 47 mm filter holder was installed downstream of the mVACES' diffusion dryer ("dry" collection, Table 1), or the mVACES' diffusion dryer was removed and the aerosol was collected as drops in a centrifugal, all-glass wet collector (SKC Biosampler™, "wet" collection, Table 1). In its nominal configuration, the SKC Biosampler™ operates at a flow of 5 lpm. During "wet" collection, two of the three tangential nozzles of the SKC Biosampler™ were plugged in order to match its sampling flow to the minor flow of the mVACES. Drop-laden air was drawn through the central inlet of the SKC Biosampler™ and accelerated through the remaining open jet. Grown aerosol droplets were collected by impaction onto the wall of the impinger's base.

The MOUDI, a separate $PM_{2.5}$ filter sampler and the mVACES were concurrently operated over several days. The MOUDI was modified to collect a single $PM_{2.5}$ sample on the after-filter stage and filters were used as substrates. The MOUDI and filter sampler were operated at 30 lpm. Collection periods extended over several hours to ensure that sufficient aerosol mass was obtained for subsequent analysis. One to two sample sets were usually collected per day. Throughout each individual sampling period, collection would be temporarily suspended and the mVACES' performance checked using the DataRAM 2000 ($PM_{2.5}$ mass) and the CPC ($PM_{2.5}$ number concentration). Both Teflon and quartz filters/substrates were used. At the end of sampling the filters were removed from all three instruments and placed in clean plastic (Teflon) or clean plastic lined with baked aluminum foil (quartz) filter holders. After re-equilibration to laboratory conditions, the Teflon filters were re-weighed in the lab to determine the collected mass (Mettler MT 5 microbalance, Mettler-Toledo, Inc., Highstown, NJ). All filters were frozen immediately after collection until analysis. Impinger samples were

TABLE 1
Instrumentation and parameters investigated

Parameter	Instrument	mVACES aerosol output (collection method)	Individual sampling period	Dates (non-continuous)
Physical				
•mass	MOUDI[a,b]	Dry, (filter[b])	3–5 hours	9/13–10/2/2005
	filter[b]	Dry, (filter[b])	3–5 hours	9/13–10/2/2005
	DataRAM 2000[c]	Dry	1–2 min	9/13–12/30/2005
•size distribution	SMPS[d]	Dry	2–4 min	9/13–12/30/2005
•number	CPC[e]	Dry	1–2 min	9/13–12/30/2005
Chemical				
•Inorganic ions	MOUDI[a,b]	Wet, (Biosampler[i])	3–5 hours	11/23–12/9/2005
•Metals	filter[b]	Dry, (filter[b])	3–5 hours	9/13–10/2/2005
	MOUDI[a,b]	Wet, (Biosampler[i])	3–5 hours	11/23–12/9/2005
•speciated organic compounds	MOUDI[a,f]	Dry, (filter[f])	3–5 hours	9/27–10/1/2005 (PAHs)
	filter[f]	Wet, (Biosampler[i])	3–5 hours	12/18–12/27/2005
• Elemental and Organic Carbon (EC/OC)	MOUDI[a,f]	Dry, (filter[f])	3–5 hours	12/18–12/27/2005
	Aethalometer	Dry	20–40 min	12/8–12/30/2005
	semi-continuous OC-EC[h]	Dry	20–60 min	12/22–12/30/2005

[a]Micro-Orifice Uniform Deposit Impactor (Marple et al., 1991) (Model 100, MSP Corp., Shoreview, MN).
[b]37 mm (MOUDI) and 47 mm diameter, 2 μm pore size PTFE filters (Teflo, Pall Corp., East Hills, NY).
[c]DataRAM 2000, (Mie, Inc. [now ThermoElectron Corp., Franklin, MA]).
[d]Scanning Mobility Particle Spectrometer (Model 3936, TSI, Inc., Shoreview, MN).
[e]Condensation Particle Counter (Model 3022A, TSI, Inc., Shoreview, MN).
[f]37 mm (MOUDI) and 47 mm diameter, quartz fiber filters (Whatman Grade QM-A 1851037, Whatman Inc., Maidstone, England) baked at 550°C for 12 hours and stored in baked aluminum foil prior to deployment (see Fine et al., 2004).
[g]Two-channel (BC+UV) Model AE-21 (Thermo Andersen, Smyrna, GA).
[h]Carbon Aerosol Analysis Field Instrument (thermal/optical transmittance) after Birch and Cary (1996) (Sunset Laboratory, Inc., Tigard, OR).
[i]Used as an impactor, SKC BioSampler (SKC West Inc., Fullerton, CA).

also immediately returned to the lab after collection and the original sample pipetted directly into a pre-weighed Teflon vial. A small amount (3–5 ml) of deionized water was then added to clean/rinse the impinger base and also pipetted into the vial. The vials were re-weighed to determine the collected mass and refrigerated prior to analysis. The impinger was cleaned and rinsed with deionized water and allowed to dry prior to re-use. Laboratory and field blanks of the filters, impinger and deionized water were taken.

The CPC, DataRAM 2000, SMPS, and Aethalometer data were obtained by switching back-and-forth between an ambient inlet and downstream of the mVACES. The measured PM property was allowed to clearly stabilize at each position before switching and measurements were typically repeated to ensure their validity. As noted earlier, most of our experiments were conducted at RH $< 60\%$ values, quite typical of the semi-arid climate of Los Angeles, especially during daytime. In very few occasions where ambient RH was relatively high, data were obtained by connecting an additional diffusion dryer—identical to that used in the mVACES—to each instrument's inlet. This modification was deemed necessary as both particle size and light scattering by PM are affected by relatively high RH which these instruments depend upon (Lowenthal, Watson, & Saxena 2000). This method was also used throughout the study to repeat routine individual measurements by these RH-sensitive instruments to confirm their performance. Additionally, due to the relatively and continuously high ambient number concentrations ($> 10^4$ # cm^{-3}), concentration enhancement measured using the CPC alone is affected by the switch from the individual particle counting to the photometric mode at 1×10^5 # cm^{-3} which is very sensitive to the assumptions the algorithm is based upon. This reduces the ability to use the CPC to quantify accurately the concentration enhancement but it does not affect the SMPS results.

The Aethalometer and DataRAM were operated at 1.7 lpm, resulting in an ideal concentration enhancement factor of 17.6 (30/1.7). The SMPS (aerosol size distribution data over the range 6–225 nm) was operated with 15/1.5 lpm balanced

sheath/aerosol flow rates, the 0.0710 cm impactor and each scan took 2 minutes (90 seconds voltage ramp "up," 30 seconds "down"). While the Aethalometer was set to obtain 3- and 5-minute cumulative samples to avoid detection limit problems, we found that it took several sampling intervals for the reading to stabilize due to the large difference in concentration between ambient and the mVACES' output. The dual-channel Aethalometer provides information on two composition parameters simultaneously—$PM_{2.5}$ Black Carbon (BC) concentrations and an estimate of the PAH content based upon PM absorption of ultraviolet light.

In contrast to the instruments in the preceding paragraph, the semi-continuous EC/OC (Elemental Carbon/Organic Carbon, Sunset Labs Model 3F, Sunset Laboratory, Inc., Portland, OR) measurements were performed concurrently as two instruments were available. Their use at USC and typical operation has been previously reported (Arhami et al. 2006). These instruments use the thermal-optical transmittance method (Birch & Carey 1996) and the results—as well as their limitations—have been widely reported (Turpin, Cary, & Huntzicker 1990; Lim & Turpin 2002; Subramanian et al. 2004). The instrument downstream of the mVACES was operated with the parallel-plate denuder (Subramanian et al. 2004) and with the addition of particle-free make-up air to maintain the 6 lpm total instrument flow rate. Due to the total flow rate requirements of the Sunset Laboratory instruments, the ideal enrichment for the mVACES was reduced from a factor of 20 to a mean factor of 5 based upon flow modification/dilution for the data reported here. Individual sampling time periods varied depending upon estimates of ambient concentrations and were less than 100 minutes in length followed by approximately 15–20 minutes for analysis. The programmed temperature ramp during the analytical cycle provides 4 OC fractions from most (OC^1) to least volatile (OC^4). Chemical data provided are EC, OC, total carbon (TC) as well as the four different OC fractions.

Off-line chemical analyses on the filters/substrates and "wet" collections include ion chromatography (IC) ($SO_4^=$, NO_3^-, Na^+, K^+, NH_4^+), selected metals via inductively coupled plasma-mass spectroscopy (ICP-MS) (Be, S, Cr, Mn, Fe, Co, Cu, Zn, Mo, Sn, Ba, Al, K, Ti, Mg, and Ni), and speciated organic compounds. Lough et al. (2005) describe in detail the procedures followed for sample processing (e.g., filter/substrate extraction methods, digestion) for the IC and ICP-MS analyses. The extracts were analyzed by IC using a modified version of the NIOSH (National Institute for Occupational Safety and Health) Method 7903 and OSHA (Occupational Safety and Health Administration) Method 188. Two different methods were used to determine speciated organic compound concentrations for selected time periods. The first method was used to measure selected polycyclic aromatic hydrocarbon (PAH) compounds only while the second provided more extensive non-polar organic compound quantification (including PAHs). The first method—performed on extracts from quartz filter/substrates only—used the High Performance Liquid Chromatography (HPLC)-fluorescence technique developed by Eiguren-Fernandez and Miguel (2003). The second method used Gas Chromatography—Mass Spectrometry (GC-MS) techniques for the analysis. It is a slight modification to/evolution of the detailed Sheesley et al. (2004) method and was used for concentrated extracts of both "wet" collections and "dry" filter/substrates. The cited references provide additional information on the sample processing and analytical techniques. Organic speciation analysis was conducted on pooled mVACES and MOUDI samples in order to ensure collection of adequate PM amounts for the aforementioned analyses.

RESULTS AND DISCUSSION

We will present the physical characterization results first followed by the chemical composition and characterization results. It is important to recognize that equipment limitations and the necessity to follow sampling protocols consistent with the planned chemical analyses preclude performing every possible chemical analysis for each sample. Further, as performance validation of the mVACES is the principal focus of this work, discussion of the physical and chemical characteristics of the observed aerosol is subsequently limited. Our observations, however, are consistent with extensive measurements previously reported at the same site (Sardar et al. 2005; Miguel et al. 2004).

Physical Characterization of Ambient and Concentrated Aerosol

Over 200 data points were obtained of concentrated (mVACES) versus ambient total $PM_{2.5}$ mass measurements using the DataRAM during the course of the entire study due to the frequent use of this method to monitor the mVACES' performance. The results are shown in Figure 2. The slope of the linear regression line through the data is 16.56 which corresponds to 94% of the expected value of 17.6, thus well within the likely uncertainty in the measurement (recall the slightly

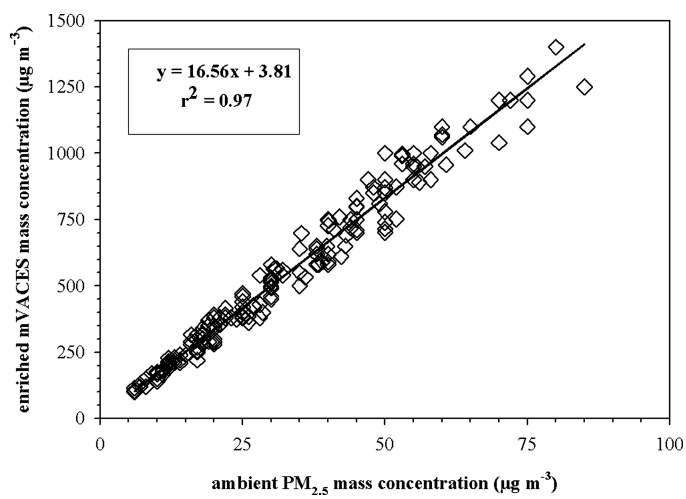


FIG. 2. Total concentrated versus ambient $PM_{2.5}$ mass concentrations observed during the entire study using the DataRAM 2000.

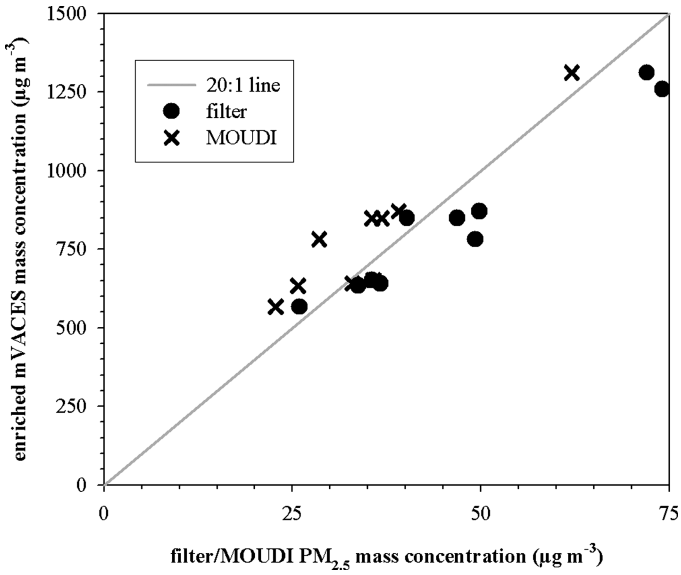
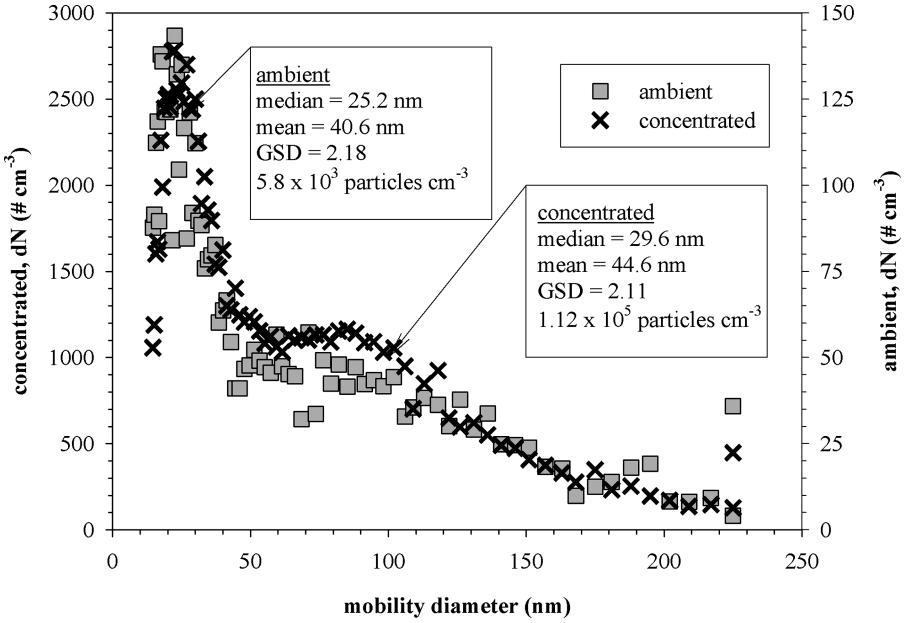


FIG. 3. Ambient filter and MOUDI $PM_{2.5}$ mass concentrations compared to simultaneous collection in the mVACES (Teflon filters only, 10 data points for ambient filter, 9 for MOUDI [pump failure]). A 20:1 line is shown to represent ideal concentration enhancement.

larger operating flow rate (1.7 lpm) of the DataRAM produces a somewhat smaller ideal concentration enrichment factor). The x-intercept is near-zero which is logically consistent and the correlation parameter is near unity. This is excellent agreement particularly over the varied aerosol mass loadings and ambient conditions experienced over the four-month study period.

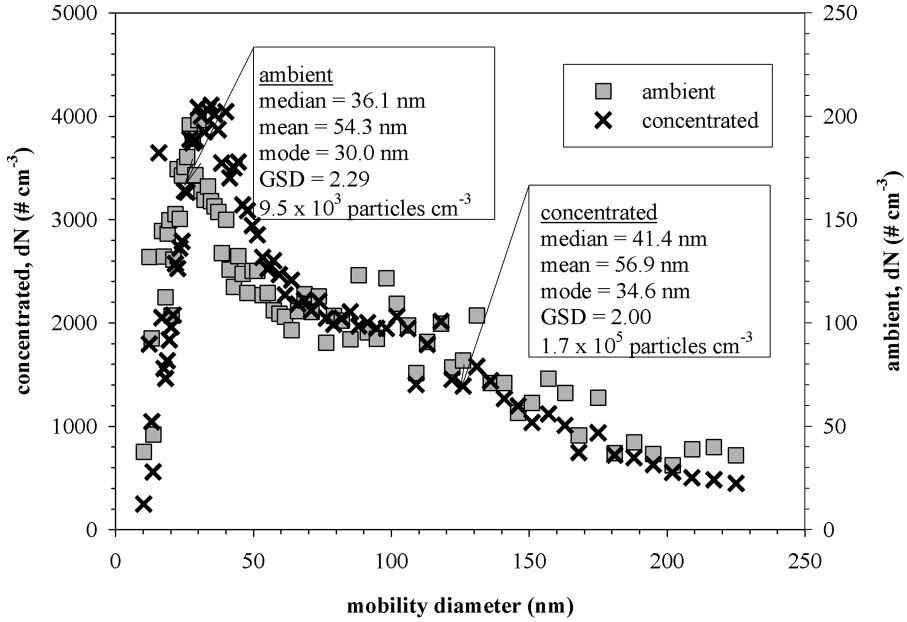
Similar results were found for mass concentration results derived from the mVACES, MOUDI and filter samples concurrently operated during a sub-set of the study days where Teflon filters were used for aerosol collection (Figure 3). Concentrations calculated for the mVACES (567–1311 μg m^{-3}, mean 841 μg m^{-3}) were enhanced on average 18.4 $\pm$ 1.8 (one standard deviation) times those on the filter and 22.8 $\pm$ 2.9 times those on the MOUDI, respectively. These are within 8% and 14%, respectively, of the ideal concentration enhancement of 20 and, again, well within the uncertainty of the measurement (largely due to uncertainty associated with filter weighing). While there is slightly more scatter in the MOUDI data, these results also suggest that comparisons between the mVACES and the MOUDI or filter can be used interchangeably.

The final physical comparison more sensitive to small particle size are particle size distribution data obtained using the SMPS system. Two examples are shown in Figure 4a and b. The number median diameters calculated for the ambient aerosol distributions shown in these figures—roughly 25 and 35 nm, respectively—are typical of the ambient values observed throughout this study. The degree of overlap of the two plots indicates the closeness of the mVACES to ideal enrichment performance. There is a 10–15% shift in the median, mode and mean diameters between the ambient and concentrated aerosols but that is well within the typical variation observed within 15 minutes at our location. The size distributions are well-preserved, and there is a factor of 19.5 (Figure 4a) and 20.3 (Figure 4b) between the total concentrated and ambient concentrations for particles $\geq$20 nm. Figure 4c shows the average

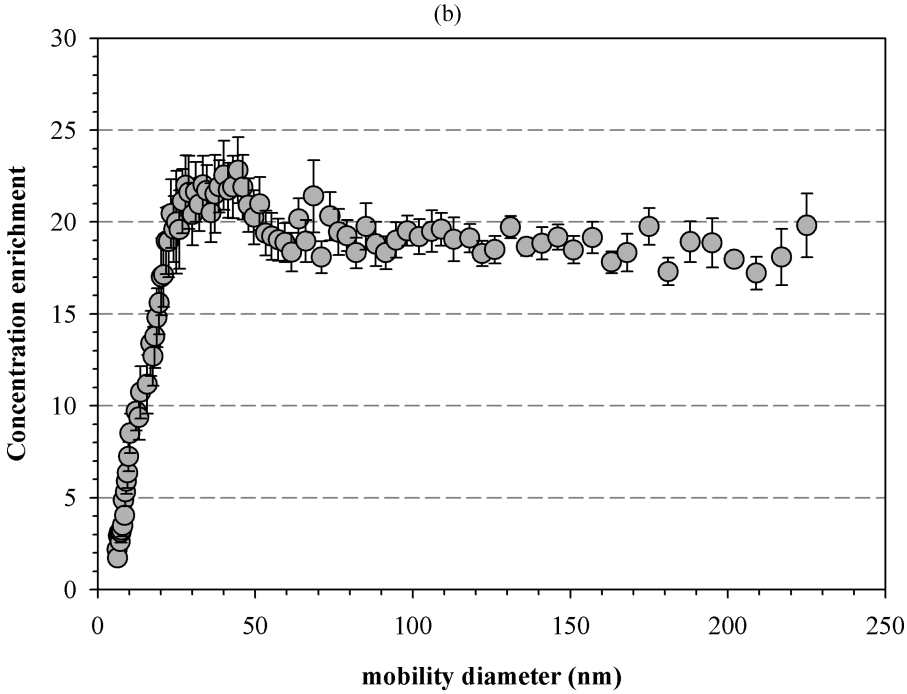


(a)

FIG. 4. (a) Ambient and concentrated particle size distributions, December 28, 2005, 4:40–4:57 p.m. PST (b) ambient and concentrated particle size distributions, October 19, 2005. The concentrated and ambient ordinate axes are offset by a factor of 20 for ideal concentration enhancement by the mVACES. (GSD = geometric standard deviation). (c) average ($\pm$1 standard deviation) concentration enrichment as a function of particle mobility diameter. *(Continued)*



(b)



(c)

FIG. 4. (Continued)

(± one standard deviation) concentration enrichment plotted as function of particle mobility diameter for the range of 6–225 nm, corresponding to the specific SMPS configuration used in our field tests. The plotted averages correspond to 30 field experiments performed when ambient number concentrations varied from 10,000–60,000 cm^{-3}. The concentration enrichment increases monotonically from about 2 to 20 (the ideal value) as particle size increases from 6 to 20 nm. The "50% cutpoint" or half of the ideal enrichment value (e.g., 10 of 20) is at about 10 nm. For particles in the range of 20–225 nm, the enrichment remains very close to its ideal value. These particles have been concentrated regardless of their composition and mobility diameter from ambient concentrations typical of an urban environment. Geller et al. (2005) obtained similar results using

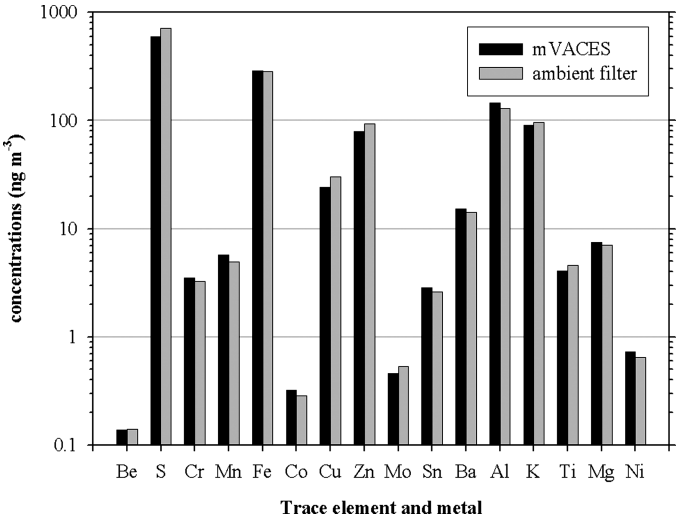


FIG. 5. Average observed concentration for 16 trace metals and elements for the mVACES and concurrent ambient filter measurements. Note use of the log scale on the ordinate axis.

laboratory-generated aerosols up to concentrations of 5×10^5 cm^{-3} (see Figure 5 and Table 1 in Geller et al. 2005). The preservation of the size distribution suggests that passage through the mVACES does not lead to substantial coagulation.

Overall, the physical comparisons between ambient and the concentrated aerosol stream produced by the mVACES suggest that the particles are being concentrated near ideally. Within our ability to measure it this appears to be true for mass (sensitive to larger particles' effective concentration) as well as for number (sensitive to the smaller particles).

Chemical Composition of Concentrated and Ambient Aerosol

Sixteen individual metals and elements were measured by ICP-MS in the concentrated and ambient aerosol. Table 2 has the consolidated results and Figures 5–7 are relevant examples of the results. For these data, the mVACES concentrations were based on the intake (30 lpm) as opposed to the minor (1.5 lpm) flow. The equivalent concentrations between the mVACES and the filter measurements indicate ideal (a factor of 20) concentration enrichment. For average concentrations that range over four orders of magnitude (sub-ng m^{-3} to 100s ng m^{-3}) very good overall agreement between the mVACES and the filter measurements (Table 2, Figure 5) is achieved for the sixteen species. There is not a discernible difference in results for species found at relatively high concentrations (e.g., Fe, Figure 6) or low concentrations (e.g., Sn, Figure 7). The mean linear regression slope for all data is 0.88 ± 0.22 which is well within the uncertainty of unity. This corresponds to a concentration enhancement in the mVACES of 17.6 ± 4.4 (one standard deviation). Correlation coefficients (Table 2) are also near unity. The sole exception is Mg, for which there is appreciable scatter in the data, although the mean and range of concentrations are consistent between

TABLE 2
Summary of trace metals and elements results

Species	Ambient filter concentrations [ng m^{-3}] Mean (range)	mVACES concentrations [ng m^{-3}] Mean (range)	Regression r^2	Regression Slope	No. of data points
Be	0.14 (0.53–1.2)	0.14 (0.45–1.1)	0.8	0.91	6
S	711 (515–1190)	594 (356–1022)	0.93	0.77	6
Cr	3.3 (0.8–10.1)	3.5 (0.81–10.9)	1	1.07	5
Mn	4.9 (1.8–11.9)	5.8 (1.7–14.8)	0.97	1.27	5
Fe	285 (107–691)	288 (77–704)	0.96	1.04	5
Co	0.29 (0.05–0.91)	0.32 (0.08–1.2)	0.98	1.36	5
Cu	30 (6.7–84.9)	24 (3.8–67.2)	1	0.79	4
Zn	93 (11–386)	79 (9.3–322)	1	0.83	5
Mo	0.53 (0.37–0.8)	0.46 (0.34–0.65)	0.96	0.68	4
Sn	2.6 (1.2–5.0)	2.9 (1.9–5.1)	0.97	0.88	5
Ba	14 (7.1–32)	15 (4.2–31)	0.83	0.85	6
Al	130 (26–367)	145 (44–322)	0.93	0.79	6
K	96.2 (32–266)	91 (34–236)	0.88	0.85	6
Ti	4.6 (1.2–13.8)	4.1 (1.2–11.9)	0.91	0.83	5
Mg	7.0 (1.6–14.7)	7.5 (3.5–13.5)	0.29	0.43	5
Ni	0.65 (0.32–1.51)	0.73 (0.30–1.6)	0.84	0.93	5

Mean slope 0.89 ± 0.22.

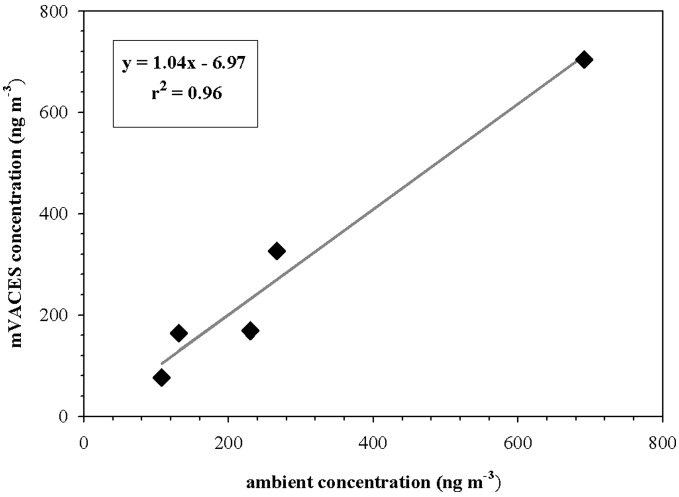


FIG. 6. mVACES versus concurrent ambient filter measurements with linear regression fit for Iron (Fe).

the two instruments (Table 2) and the slope (0.43) and correlation coefficient (0.29) are poor. While concentrations are low, they are well above the method detection limit (Lough et al. 2005). It is not known what the source of this discrepancy is—if contamination were responsible in the field it is difficult to imagine that only Mg would be affected. In general, the trace metal and elements results suggest the mVACES is concentrating these inorganic species effectively and with minimal artifact.

While trace metals, elements and other inorganic constituents may well have similar distribution patterns in the ambient aerosol, carbonaceous species are likely to have a different size distribution, as their sources—both primary and atmospheric formation mechanisms—will likely differ. Therefore, it is important to also characterize the mVACES' concentration enhancement for carbonaceous species as well, particularly as they are important contributors to aerosol mass at this location (Sardar et al. 2005). Elevated black carbon concentrations are associated with proximity to the freeway. Black carbon and surrogate "UV" PAH carbon measurements from December 2005 are shown in Figure 8 from the two-channel Aethalometer. The results for both species indicate again near-ideal enrichment (a factor of 17.7 ± 2.3 (15.0–22.5 range) for BC and 15.7 ± 1.0 (13.8–17.3 range) for UV PAH carbon). Regression lines for both yielded slopes of approximately 16.1 with $r^2 \geq 0.95$. These results are very consistent and are from a completely different measurement technique than those shown previously. The mix of organic carbon species measured likely varied over the course of the experiment but the concentration factors do not appear to be appreciably affected.

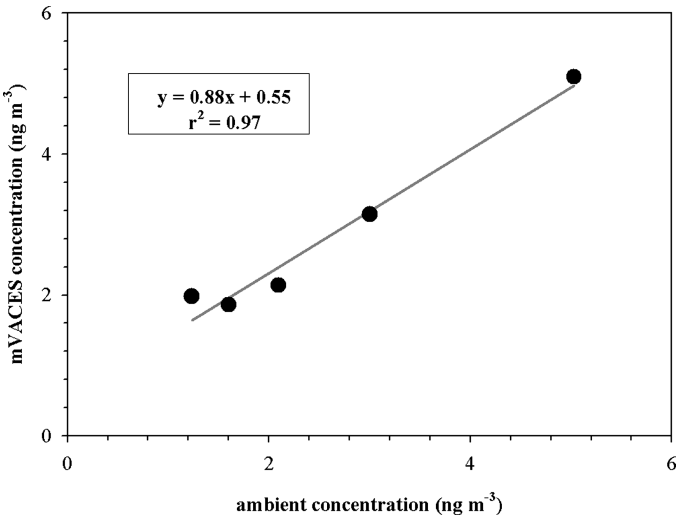


FIG. 7. mVACES versus concurrent ambient filter measurements with linear regression fit for Tin (Sn).

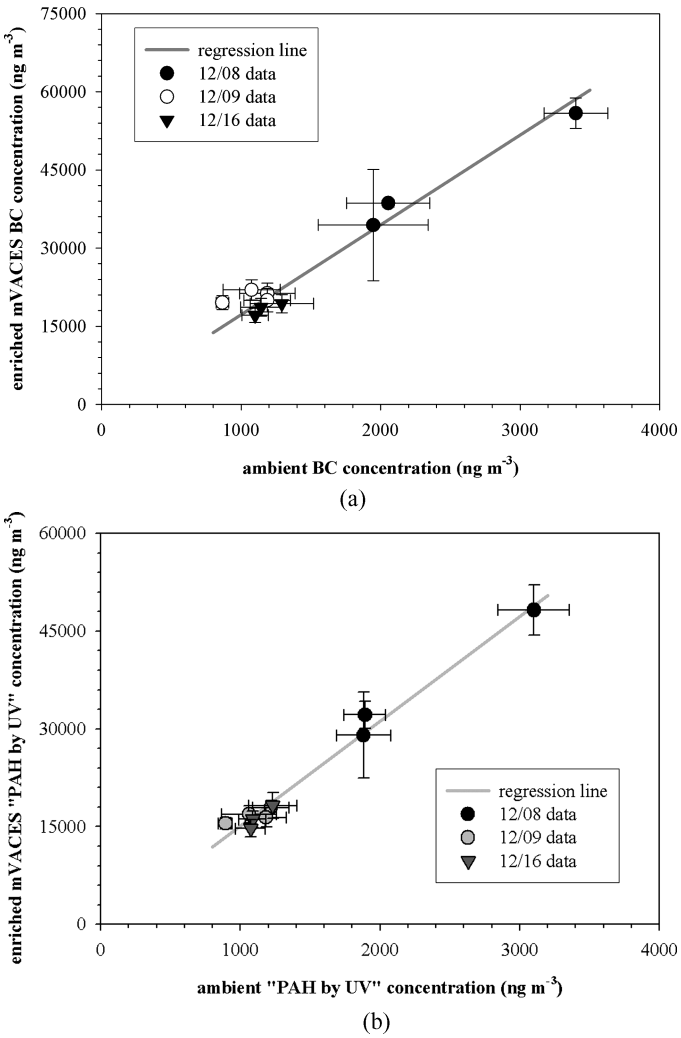


FIG. 8. mVACES versus concurrent ambient observations, two-channel Aethalometer, December 2005 (a) Black Carbon (BC) concentrations, and (b) Ultraviolet (UV) organic-carbon equivalent concentrations.

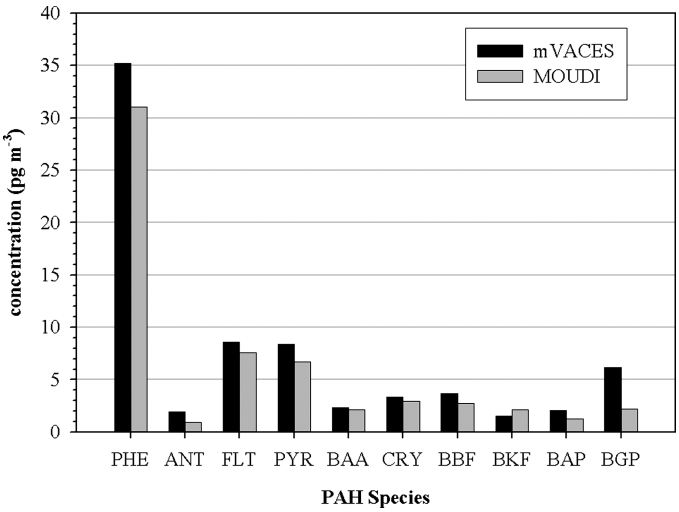


FIG. 9. mVACES and MOUDI concentrations for 11 selected polycyclic aromatic hydrocarbons (PHE—phenanthrene, ANT—anthracene, FLT—fluoranthene, PYR—pyrene, BAA—benz[a]anthracene, CRY—chrysene, BBF—benzo[b]fluoranthene, BKF—benzo[k]fluoranthene, BAP—benzo[a]pyrene, BGP—benzo[ghi]perylene), geometric mean concentrations, September–October 2005.

Polycyclic aromatic hydrocarbon concentrations were also measured for the following species: phenanthrene, anthracene, pyrene, fluoranthene, benzo(a)anthracene, chrysene, benzo(ghi) perylene, benzo(a)pyrene, and benzo(k)fluoranthene. PAHs are of interest due to their toxicity, ubiquity and association with urban air pollution (Finlayson-Pitts & Pitts, 1999). In contrast to the Aethalometer data (Figure 8b), specific identification of individual PAHs is possible. The PAH results are shown in Figure 9 comparing concentrations measured in a filter downstream of the mVACES and a simultaneously-operated MOUDI. In this plot, the mVACES data were also normalized to the inlet (30 lpm) flow to make the graph more legible. As in Figures 5–7, ideal enrichment in the mVACES results in equal concentrations for both instruments. Again, across the range of volatilities of the species measured, comparison between the two is quite good with the mVACES/MOUDI concentration ratio of 1.30 ± 0.33 (average ± one standard deviation) and a range of 0.73–1.96 for the nine species measured above the detection limit of the analysis. For most species, the difference in reported concentrations was within 1 pg m^{-3} which is good given the concentrations observed (generally less than 10 pg m^{-3}). The exceptions are phenanthrene and benzo(ghi)perylene. Phenanthrene was present in appreciably higher (ca. 30 pg m^{-3}) concentrations than the others (Figure 9), although here again, agreement is nevertheless very good (within 10%). While not perfect, these data are highly consistent with the other mVACES' performance results given the uncertainty of the analysis.

The final dry comparison is between simultaneous observations made of elemental carbon and four organic carbon fractions downstream of the mVACES and simultaneously collected from ambient $PM_{2.5}$ aerosol by the semi-continuous carbon aerosol analyzer instruments. The results are compiled in Table 3 by fraction and shown in Figures 10 and 11. For these analyses, the ideal concentration factor is reduced to ca. 5 primarily due to the dilution of the mVACES' output with the make-up air required to operate the aerosol analyzer. As we have seen previously with the Aethalometer Black Carbon data (Figure 8a), Thermal EC is concentrated efficiently by the mVACES compared to ambient (Figure 10). Ambient Thermal EC concentrations were consistent with ambient Aethalometer BC during these sampling periods (not shown). Thermal OC concentrations were about three

TABLE 3
Summary of semi-continuous OC/EC observations December 29–30, 2005

Measurement[a]	mVACES concentration [μgC m^{-3}] mean (range)	Ambient concentration [μgC m^{-3}] mean (range)	Measured enrichment factor mean ± std. dev.	Measured/calculated enrichment factor ratio mean ± std. dev. (range)
OC peak #1	9.3 (6.1–13.7)	2.3 (1.3–3.1)	4.06 ± 0.45	90 ± 10% (73–105%)
OC peak #2	5.6 (3.2–8.0)	1.5 (0.7–2.1)	4.0 ± 0.50	89 ± 12% (68–102%)
OC peak #3	3.1 (1.4–4.8)	0.6 (DL–1.0)[b]	5.67 ± 1.30	126 ± 29% (96–179%)
OC peak #4	5.2 (1.4–9.3)	0.7 (DL–1.1)[c]	8.3 ± 1.3	188 ± 30% (144–209%)
Thermal OC	23.5 (12.4–35.8)	4.8 (2.0–7.0)	4.91 ± 0.71	109 ± 16% (88–133%)
Thermal EC	7.7 (2.9–12.4)	1.6 (0.5–2.8)	4.83 ± 0.80	107 ± 15% (92–141%)
Total Carbon (TC)	31.2 (15.3–48.2)	6.5 (2.7–9.8)	4.86 ± 0.49	108 ± 11% (95–123%)

[a]8 data points, except where below detection limit (DL).
[b]7 ambient data points.
[c]4 ambient data points.

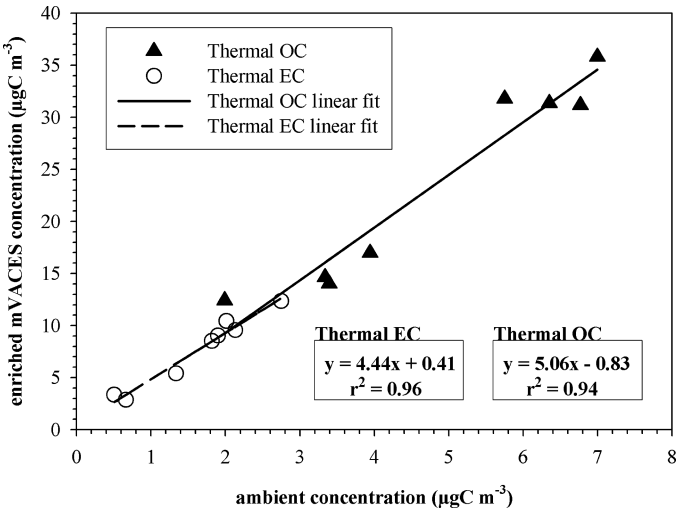


FIG. 10. mVACES versus ambient concentrations of Thermal EC and Thermal OC measured side-by-side, December 29–30, 2005.

times higher on average and were also concentrated efficiently by the mVACES (Figure 10). There is little scatter in the results over a reasonably broad range of concentrations. The more (OC^1) and less (OC^{2-4}) volatile OC fraction results (Figure 11) are also very consistent with the mVACES' effectively concentrating the peaks by near-ideal factors. For the least volatile, and smallest in concentration OC fraction (OC^4), the mVACES appears to substantially over-concentrate the aerosol or have a substantial positive artifact. The detection limit is shown in Table 3 and suggests that the ambient data suffer from the additional uncertainty

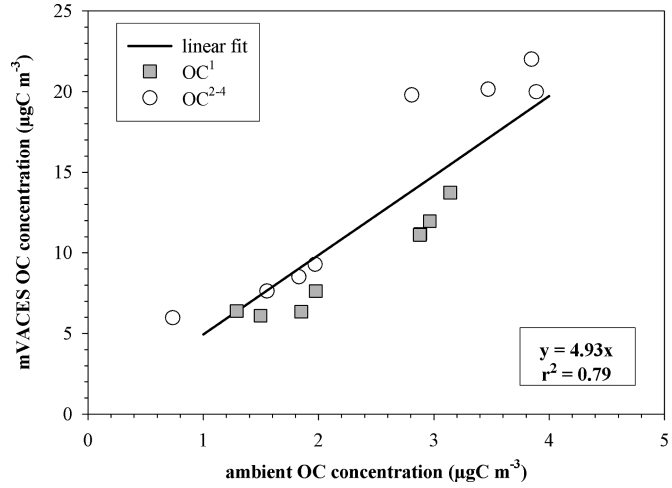


FIG. 11. mVACES versus ambient concentrations of the peak OC^1 and the sum of the remaining, less volatile OC peaks (OC^2–OC^4) measured side-by-side, December 29–30, 2005. The regression line is shown.

associated with near-detection limit concentrations. Overall the data listed in Table 3 and plotted in Figure 11 corroborate that concentration for all fractions is reasonably within the expected range. We have no reason to believe that the higher enrichment (as a %) observed for OC^4 is due to anything else but the very low ambient levels. Since this fraction is less volatile, its vapor phase (which would presumably be the cause of an adsorption artifact) is expected to be almost negligible, and its overall thermodynamic properties should be approaching those of elemental carbon, for which the mVACES yielded near ideal enrichment. We will, however, continue to pursue improved quantification of the OC^4 peak with additional ambient observations using longer collection periods.

The limited "wet" collection composition data currently available complement those reported for dry collection. Concentrations of 52 individual non-polar organic compounds—including selected PAHs, hopanes, steranes, and alkanes—measured in the slurry samples collected by the mVACES are compared against simultaneous dry ambient filter samples in Figure 12. Concentrations varied from approximately 0.15 ng m^{-3} (fluoranthene) to 10 ng m^{-3} (docosane). Overall agreement improved compared to that obtained for the dry collection PAH data previously discussed (Figure 9) obtained using a different analytical method. The mean "wet" collection/filter concentration ratio for the PAHs, hopanes-steranes and alkanes were 0.96 ($\pm$0.1), 1.12 ($\pm$0.26), and 0.94 ($\pm$0.11), respectively, thus very close to the ideal value of 1.0. These findings indicate that a wide variety of ambient hydrophobic aerosol species are thus effectively concentrated.

Similarly to the physical measurements, the chemical composition results shown here suggest that over the range of inorganic and organic species observed, the mVACES appears to concentrate the aerosol effectively at ideal/near-ideal factors regardless of composition. There is no evidence from the data available of the mVACES introducing significant positive or negative artifact for the species measured.

There is remarkable consistency in the reported results despite the variety of aerosol collection/measurement methods used. One limitation that all the techniques except for the filter-based methods and the semi-continuous carbon aerosol analyzers had is that measurements were made sequentially, not simultaneously. While some of this can be mitigated by rapid switching between mVACES and ambient source aerosol as quickly as possible (e.g., 1–2 minutes for the DataRAM 2000), there is still some unknown uncertainty added to the results. The SMPS, in particular, would be sensitive to this as it makes discrete measurements across the size range studied, not just a lump sum "total." Despite these uncertainties, which are quite common in field studies, the results are remarkably consistent in indicating the robustness of the mVACES' ability to concentrate ambient aerosols.

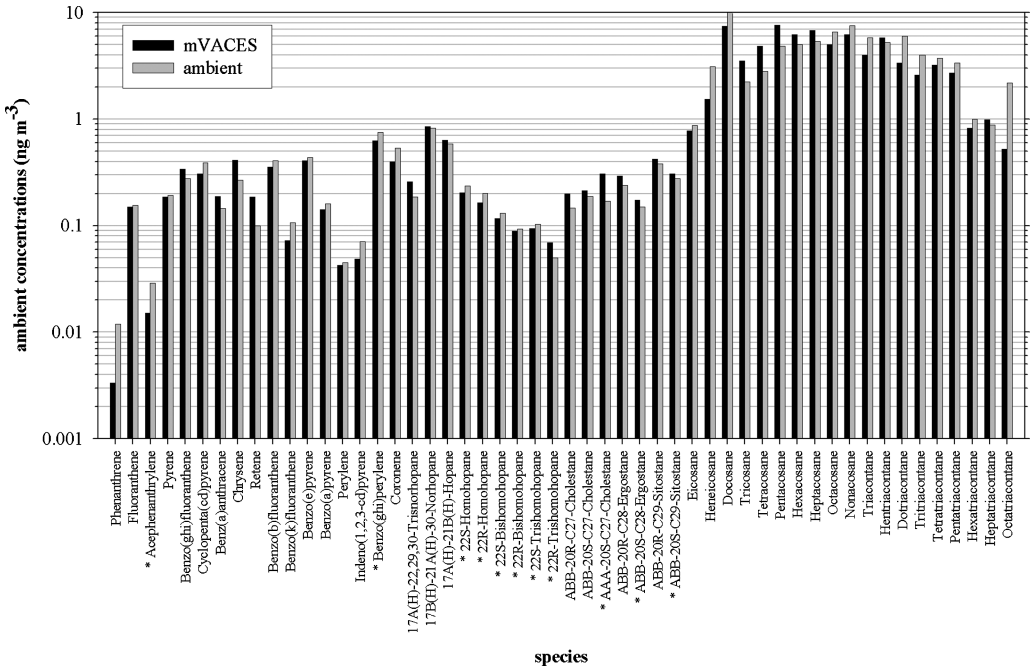


FIG. 12. mVACES versus concurrent ambient filter measurements for selected non-polar organic species (asterisks indicate quantification based upon the calibration for a similar compound). Note use of the log scale on the ordinate axis.

CONCLUSIONS

A new, compact aerosol concentration enrichment system has been designed and built at USC. It is suitable for use to concentrate aerosol particles upstream of some of the new on-line, low flow rate aerosol mass spectrometers. To complement previous validation work primarily with laboratory-generated aerosol particles, an extensive field validation campaign was undertaken in an urban environment. Both physical and chemical measurements support that the new mVACES concentrates aerosol as designed at rates predictable based upon the difference in major and minor flow rates through its virtual impactor. It appears to be both stable and robust.

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