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Development and evaluation of a high-volume dichotomous sampler for chemical speciation of coarse and fine particles

Satya B. Sardar^a, Michael D. Geller^a, Constantinos Sioutas^{a,*}, Paul A. Solomon^b

^a University of Southern California, Department of Civil and Environmental Engineering, 3620 South Vermont Avenue, Los Angeles, CA 90089, USA

^bUnited States Environmental Protection Agency, Office of Research and Development, Las Vegas, NV 89193-3478, USA

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Abstract

This study presents the development and field evaluation of a compact high-volume dichotomous sampler (HVDS) that collects coarse ($PM_{2.5-10}$) and fine ($PM_{2.5}$) particulate matter. The key features of this device is the utilization of a round nozzle virtual impactor with a 50% cutpoint at 2.5 µm aerodynamic diameter, operating at a high inlet flow rate (10001 min^{-1}) with a calculated Reynolds number of approximately 1.1×10^5 . In its primary configuration as tested, the sampler size-fractionates PM_{10} into coarse and fine fractions with a minor flow ratio of 10% (minor/total flow rate), with major and minor flow rates of 900 and 1001 min^{-1} , respectively. Performance evaluation for concentration enrichment was conducted with a 4% minor flow ratio (401 min^{-1} minor flow) as well. Tests demonstrated near ideal results at both 10% and 4% minor flow ratios, indicating enrichment to be independent of minor flow rates within the range evaluated. Reasonable agreement was found between the new sampler and collocated Partisol and MOUDI for ambient measurements. A tandem virtual impactor super-concentration system was also tested with the 10001 min⁻¹ virtual impactor followed by a 1001 min^{-1} round nozzle virtual impactor and proved to be an efficient system to achieve higher concentration enrichment of ambient particles up to 180 times ambient levels. Investigation of the effect of different ambient parameters like RH and wind speed on coarse PM concentration corroborates results from earlier studies. The HVDS is an effective system to collect coarse and fine PM simultaneously, allowing for comprehensive standard chemical analyses over short sampling intervals.

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1. Introduction

Abundant epidemiological and toxicological research has focused attention on the role of fine particles ($PM_{2.5}$) on observed health effects (Anderson, 2000; Brown, Stone, Findlay, Macnee, & Donaldson, 2000; Ferin et al., 1990). In addition to fine particulate matter, coarse PM (aerodynamic diameter between 2.5–10 µm), which has a different pattern and degree of deposition in the respiratory system, may also consist of several potentially toxic components. Recent records from a number of epidemiological studies have established significant statistical associations between

* Corresponding author. Tel.: +1 213 740 0999; fax: +1 213 744 1428.

E-mail addresses: sardar@usc.edu (S.B. Sardar), sioutas@usc.edu (C. Sioutas).

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coarse PM and observed health outcomes (Brunedkreef and Forsberg, 2005; Castillejos, Borja-Aburto, Dockery, Gold, & Loomis, 2000; Chen et al., 2005). In addition to epidemiological evidence, atmospheric coarse particle toxicity has been demonstrated in laboratory in vitro studies (Becker et al., 2005; Kleinman, Bhalla, Mautz, & Phalen, 2000; Ostro, Hurley, & Lipsett, 1999). The aforementioned studies corroborate the potential toxicity of coarse PM and its public health implications, and establish the need for identification of their chemical constituents in the atmosphere.

Coarse particles are generated mechanically by crushing, grinding and attrition processes and differ from the fine fraction not only in size, but also due to different formation mechanisms, chemical composition, sources and physicochemical behavior. Sources of coarse PM include resuspended dust, sea salts, crustal materials from roads and construction, brake linings, mining and farming activities, as well as bioaerosols, such as fungal spores and pollens. Because of the different physicochemical and biological characteristics and deposition pattern, coarse PM is likely to have different health impacts as compared to fine PM.

Coarse PM can be measured either as the difference between collocated PM_{10} and $PM_{2.5}$ samplers or more directly using dichotomous samplers (virtual impactor). The coarse PM estimation using the difference method may be affected by increased analytical noise and biases inherent in using two separate instruments. Furthermore, the relatively low ambient coarse concentrations sometimes make accurate estimation of coarse PM by difference difficult. Coarse PM measurement with a dichotomous sampler (virtual impactor) is an effective way to increase coarse PM concentration by enriching ambient concentrations and reducing the previously discussed errors associated with the difference method. Most importantly a nearly clean coarse particle fraction is obtained with less than 10% intrusion by fine PM.

The objective of this work was to develop and evaluate the next generation, high volume, single nozzle dichotomous virtual impactor sampler-coarse aerosol concentrator to enrich ambient coarse particles without altering their physicochemical characteristics, including trace component analysis. This sampler operates at twice the flow rate of its predecessor (Solomon, Moyers, & Fletcher, 1983) and has improved separation characteristics. This device is designed to simultaneously collect coarse and fine PM over short time intervals, thereby making it possible to identify and measure potentially toxic components and individual organic tracer compounds for improved source apportionment results.

2. Methods

The design of the coarse particle chemical speciation sampler, illustrated in Fig. 1, consists of two main components: (1) a commercially available PM_{10} inlet manufactured by Tisch Environmental, Inc. (PM_{10} Hi-Volume Sampler, Tisch Environmental, Cleaves, OH), and (2) a 2.5 μ m cut point round nozzle virtual impactor (coarse particle concentrator) developed by the Aerosol Laboratory of the University of Southern California.

Particles smaller than 10 μ m in aerodynamic diameter are drawn through the virtual impactor, which was designed to have a theoretical 50% cutpoint of 2.5 μ m for a total intake flow rate of 10001min⁻¹. The 50% cutpoint can be estimated from the Stokes number, *St*, defined as (Hinds, 1999)

$$St = \frac{\rho_{\rm p} U_i d_{\rm p}^2 C_{\rm c}}{9\mu d_0},\tag{1}$$

where d_p , ρ_p , and C_c are the particle diameter, density, and slip correction, μ is the air viscosity (1.81×10^{-4} g/cm s), U_i is the velocity through the acceleration nozzle, and d_0 is the inside diameter of the acceleration nozzle. The single jet, round nozzle virtual impactor has an acceleration nozzle diameter of 1.4 cm and a collection nozzle diameter of 2.0 cm ID. The two nozzles were separated by a gap of 2.0 cm. These design parameters were selected to yield a Stokes number of 0.3 for particles of 2.5 µm in aerodynamic diameter at a flow of 10001 min⁻¹. The entire assembly of the virtual impactor is contained in an aluminum cylinder, 15 cm in diameter and 17.5 cm in length. The flow regime in the virtual impactor is determined by the Reynolds number (Re), defined as

$$Re = \frac{d_0 U_i \rho}{\mu},\tag{2}$$

where U_i is the average jet velocity through the acceleration nozzle of the impactor and μ and ρ are the dynamic viscosity and density of air, respectively. The calculated Reynolds number corresponding to the operating configuration of the virtual impactor is 1.1×10^5 , theoretically considered a highly turbulent flow regime, similar to its predecessor, which operated efficiently at a Re_i of approximately 8.0×10^4 (Solomon et al., 1983).



Fig. 1. Schematic of the HVDS.

As the particle-laden air exits the acceleration nozzle, the flow (10001 min^{-1}) is divided into two streams. The major flow (9001 min^{-1}) draws particles smaller than the 2.5 µm cutpoint of the virtual impactor through a 20.3 cm × 25.4 cm (8 in. × 10 in.) filter, while all particles above the cutpoint, including a small fraction of small particles, are carried by the minor flow through a 90 mm diameter filter. Any typical filter material can be used. In its nominal configuration, the high volume dichotomous sampler (HVDS) operates with a minor flow of 1001 min⁻¹, which results in only a 10% intrusion of fine particles in the coarse fraction. As it will be discussed in subsequent sections of this paper, the minor flow can be reduced to as low as 401 min^{-1} (resulting in only a 4% intrusion of fine particles in the coarse fraction) without any repercussions on the ideal concentration enrichment achieved by the instrument. The corresponding pressure drop across the major flow of the virtual impactor is 40 in. of water.

2.1. Characterization of the 2.5 µm cut point round nozzle virtual impactor

The evaluation of the HVDS and its components were conducted using a variety of different technologies and ambient aerosols at the University of Southern California near downtown Los Angeles. The sampling location was approximately 150 m downwind of a major freeway, and represents an urban mix of industrial, vehicular and construction sources. Several continuous and semi-continuous particle measurement instruments were used to check the performance of the system. The sampling varied from 3 to 12 h depending on the observed PM levels.

The first series of experiments were done to investigate the relationship between the concentration enrichment achieved by the 2.5 μ m cut point round nozzle virtual impactor as a function of particle size. Enrichment was determined by measuring the concentrations upstream and downstream of the virtual impactor with an Aerodynamic Particle Sizer (APS Model 3320, TSI, Inc., Shoreview, MN). These experiments were conducted at two different minor to total flow ratios of 4% and 10%, with corresponding minor flows set at 40 and 1001min⁻¹, respectively. The total flow of the virtual impactor was kept at 10001min⁻¹ by adjusting accordingly the ratio between the major and minor flow rates. For each configuration, the concentrations of ambient and concentration-enriched aerosols were

measured alternately in 5-min intervals. This experimental procedure was repeated 10 times for each configuration by the APS.

In addition to ambient aerosols, the performance of the HVDS was evaluated in few laboratory experiments. Monodisperse polystyrene latex (PSL) aerosols were generated by atomizing aqueous suspensions of these particles in ultra pure deionized water using a commercially available nebulizer in a process described by Misra, Geller, Shah, Sioutas, and Solomon (2001). For these experiments, we used four different PSL particle sizes (2.6, 3.1, 5.4 and 7.7 μ m in aerodynamic diameter). The aerodynamic diameters were calculated based on the physical batch diameter of the suspensions and a PSL density of 1.05 g cm⁻³.

Following the enrichment evaluation, the HVDS was operated in collocation with a modified Micro Orifice Uniform Deposit Impactor (MOUDI, MSP Corporation, Minneapolis, MN) and a Dichotomous Partisol Sampler (Model 2025, Rupprecht & Patashnik Co., Inc., East Greenbush, NY). In the HVDS, the minor flow was drawn through 90 mm Teflon filters (PTFE 2 μ m pore size, Gelman, Ann Arbor, MI) at 1001 min⁻¹ flow rate in all experiments, while the major flow, at 9001 min⁻¹ was drawn through a 20.3 cm by 25.4 cm rectangular Teflon filter. The MOUDI sampled at a flow rate of 301 min⁻¹ and was modified to collect the coarse (2.5–10 μ m) and fine (0–2.5 μ m) size fraction by removing all of its stages and placing its after filter following the 2.5 μ m stage. The Partisol operated at a flow rate of 16.71 min⁻¹, with major and minor flows of 15 and 1.671 min⁻¹, respectively. The MOUDI and Partisol samples were collected on 47 mm PTFE filters (except of the MOUDI after stage, which used 37 mm filters) and all filters were weighed with a microbalance (MT 5, Mettler Toledo Inc., Highstown, NJ) after equilibration in a controlled temperature (20 ± 5 °C) and relative humidity (RH) controlled room (40 ± 5%). Filters were weighed twice in order to increase precision. In case of a difference of more than 3 μ g between consecutive weightings, the filters were weighed a third time or reweighed until two consecutive weights differed by less than 3 μ g. After gravimetric analysis the coarse particles, the Teflon filters from the coarse concentrator and the Partisol sampler were subsequently analyzed for elemental species using X-ray fluorescence (XRF).

Quartz-fiber filters also were collected by the collocated HVDS and Partisol sampler, albeit at a different time then when the Teflon filters were collected. The quartz-fiber filters were pre-baked at $550 \,^{\circ}$ C for 8 h and stored in baked aluminum foil lined containers prior to deployment. Approximately 1 cm² was removed from the center of each quartz filter for measurement of elemental and organic carbon by thermal optical analysis (EC and OC are not reported in this paper). The remainder of each quartz filter was used to determine inorganic ion concentrations by ion chromatography. Samples that were lower than three times the lower limit of detection (LOD) of either XRF or IC were excluded from statistical analysis.

As part of an intensive study, we evaluated the concentration enrichment of the HVDS (10001 min^{-1}) in tandem with a 1001 min^{-1} round nozzle virtual impactor (2.5 µm cutpoint), which was connected inline with the high-volume virtual impactor's minor flow. The performance of the 1001 min^{-1} virtual impactor-coarse particle concentrator is described in greater detail by Kim, Jaques, Chang, Froines, and Sioutas (2001). The concentration enrichment for the tandem system of virtual impactors (1000 and 1001 min^{-1}) was recorded by an APS. A weather data recorder (Wizard III, Weather Systems Company, San Jose, CA) also complimented the HVDS by providing information on the diurnal variability of the temperature, RH, wind speed and wind direction for each experiment.

3. Results and discussion

The concentration enrichment of the 2.5 μ m cutpoint round nozzle virtual impactor is presented in Figs. 2a and b as a function of particle aerodynamic diameter at two different minor flows rates, 100 and 401 min⁻¹, respectively. The plotted data correspond to 5-min averaged APS measurements, and the final concentration enrichment results in Figs. 2a and b are the average of at least 10 consecutive tests. The enrichment factor for a minor flow of 1001 min⁻¹ (Fig. 2a) increased from near 0 to its ideal value of 10 (based on the intake to minor flow ratio) as the aerodynamic particle size increased from 1 to 10 µm. A sharp increase of concentration enrichment is observed for particles in the range of 2–3 µm aerodynamic diameter, and all particles between 4 and 10 µm were enriched by approximately 10 times their ambient levels. Similarly, the concentration enrichment achieved with a minor flow of 401 min⁻¹ (Fig. 2b) was close to the ideal value of 25. The curve for the 401 min⁻¹ minor flow also showed a sharp rise between 2 and 3 µm, with a constant enrichment of 25 times for larger particles (aerodynamic diameter > 4 µm). The data from Figs. 2a and b show that the 50% cutpoint of the virtual impactor, which is defined as the aerodynamic diameter at which the enrichment factor is half of its ideal value, is approximately 2.5 µm and is independent of minor flow rate.



Fig. 2. Concentration enrichment factor as a function of particle aerodynamic diameter at a minor flow rate of (a) 100 and (b) 401 min⁻¹.

Table 1 Coarse PM concentrations $(\mu g\,m^{-3})$ measured by the HVDS, MOUDI and Partisol

Sample number	MOUDI	Partisol	High-volume dichotomous sampler	High-volume dichotomous sampler/MOUDI	High-volume dichotomous sampler/Partisol	High-volume dichotomous sampler/average of MOUDI-Partisol
1	19.6	18.4	19.7	1.01	1.07	1.04
2	14.6	14.3	13.5	0.93	0.94	0.93
3	27.2	24.6	23.1	0.85	0.94	0.89
4	12.6	12.5	10.4	0.83	0.83	0.83
5	14.8	12.6	12.2	0.82	0.96	0.89
6	12.1	11.1	10.4	0.87	0.94	0.90
7	24.9	29.1	22.8	0.92	0.79	0.85
8	19.4	20.1	18.1	0.93	0.90	0.92
9	27.0	21.8	22.2	0.82	1.02	0.91
10	16.1	NA	13.6	0.84		
11	18.1	19.4	17.8	0.98	0.92	0.95
12	17.3	18.1	16.8	0.97	0.93	0.95
13	22.6	19.8	18.9	0.84	0.96	0.89
Average	18.9	18.5	16.9	0.89	0.93	0.91
SD	5.1	5.3	4.5	0.07	0.07	0.05

The sharpness of the collection efficiency curve of any conventional or virtual impactor can be defined in terms of the geometric standard deviation (σ_g), which is the square root of ratio of the aerodynamic particle diameter corresponding to 84% collection efficiency to that corresponding to 16% efficiency (Marple & Willeke, 1976). Based on this definition, the value of σ_g is approximately 1.2 for both the 4% and 10% minor flow ratio configurations, thereby indicating sharp aerodynamic particle separation characteristics. An important conclusion from this set of experiments is that the size distribution of the enriched coarse particulate matter is not altered from that of the ambient air because concentration enrichment is mostly independent of particle size for coarse particles.



Fig. 3. HVDS vs. MOUDI coarse PM concentrations.



Fig. 4. HVDS vs. Partisol coarse PM concentrations.

A comparison of the coarse PM mass concentrations measured by the three collocated samplers is provided in Table 1. The dichotomous sampler operated at a minor flow rate of $100 \,\mathrm{l\,min^{-1}}$, which yields an ideal enrichment of 10. The coarse concentrations measured by the HVDS and Partisol were corrected for the 10% penetration of fine PM mass, which corresponds to the ratio of the minor to total flow. Similarly, the MOUDI and Partisol concentrations that are reported here and in the rest of this manuscript have been normalized to the intake flows of these instruments.

The average coarse PM mass concentrations measured by the three samplers were comparable, and the ratio of the coarse concentrations measured by the HVDS to the MOUDI and Partisol were 0.89 ± 0.07 and 0.93 ± 0.07 , respectively. Figs. 3 and 4 show the comparison of the coarse concentrations measured by the HVDS vs. the MOUDI and Partisol results, respectively. As seen in Figs. 3 and 4, the coarse PM concentrations measured by the three samplers are highly correlated with high coefficients of determination (R^2) and slopes approaching unity. Therefore, data presented in Table 1 and Figs. 3 and 4 establish the excellent overall agreement between the three instruments. The slightly lower (roughly 10%) concentration, measured by the high-volume dichotomous sampler compared to the MOUDI or Partisol might be an effect of small cutpoint differences between the high-volume virtual impactor and those of the MOUDI and Partisol as well as potential losses in the HVDS due to the high air velocities.

A summary of coarse, fine, and PM_{10} (sum of fine and coarse) particulate nitrate, sulfate and trace element concentrations measured by the HVDS and Partisol is presented in Table 2. Fig. 5 represents the relationship between the two samplers based on PM_{10} particulate nitrate and sulfate. The PM_{10} concentrations are moderately well correlated

Table 2 Comparison between HVDS and Partisol based on nitrate, sulfate and metal concentrations ($\mu g m^{-3}$)

Species	Coarse		Fine		PM ₁₀		Ratio (high-volume dichotomous sampler/Partisol)		
	Partisol	High-volume dichotomous sampler	Partisol	High-volume dichotomous sampler	Partisol	High-volume dichotomous sampler	Coarse	Fine	PM ₁₀
Nitrate	5.7 ± 0.98	3.6 ± 0.74	4.46 ± 1.84	4.8 ± 2.08	10.17 ± 1.68	8.57 ± 2.15	0.65 ± 0.12	1.16 ± 0.38	0.84 ± 0.15
Sulfate	2.47 ± 0.92	1.97 ± 0.59	4.76 ± 1.69	4.26 ± 1.66	7.23 ± 2.34	6.24 ± 1.82	0.83 ± 0.21	0.90 ± 0.30	0.88 ± 0.21
Na	0.61 ± 0.25	0.52 ± 0.18					0.85 ± 0.17		
Si	1.06 ± 0.66	0.97 ± 0.59					0.91 ± 0.37		
Fe	0.65 ± 0.40	0.64 ± 0.30					0.98 ± 0.13		
Κ	0.18 ± 0.09	0.16 ± 0.07					0.89 ± 0.23		
Al	0.41 ± 0.27	0.41 ± 0.30					1.00 ± 0.28		
Ca	0.62 ± 0.42	0.61 ± 0.33					0.98 ± 0.17		
Ti	0.072 ± 0.02	0.073 ± 0.025					1.00 ± 0.18		



Fig. 5. Comparisons of PM₁₀ nitrate and sulfate concentrations between HVDS and Partisol.

 $(R^2 = 0.69)$. Reasonable agreement is obtained also for PM₁₀ even though there are likely differences in the inlet cutpoints of the PM₁₀ inlets of the various samplers. Good overall agreement was obtained for sulfate, as shown for the data listed in Table 2. The slightly lower coarse PM sulfate concentrations reported by the HVDS can be explained by a probable cutpoint difference between the two instruments. Investigations by Geller, Fine, and Sioutas (2004) in Los Angeles have shown that hygroscopic inorganic species like sulfate and nitrate constitute approximately 30–50% of total mass in the size fraction of 1–3 µm range, hence a small cutpoint variation between the two instruments is likely to result in a measurable difference in their concentrations.

The HVDS measured on average about 65 (\pm 12)% of nitrate in the coarse mode compared to the Partisol, while for the fine PM the average concentration ratio was 1.16 (\pm 0.38). Two probable causes, or a combination of both, may explain the low nitrate coarse particle value. The first one relates to small cut point differences between the two samplers and was discussed in the preceding text. The argument of cutpoint variation is further supported by the higher fine PM nitrate concentrations measured by the HVDS and the overall good agreement for the PM₁₀ data. Apart from the cutpoint shift, a second possibility may be volatilization losses associated with labile nitrate species from the coarse PM filter of the HVDS. Previous studies in Southern California indicate that coarse mode nitrate is associated predominantly with sodium and ammonium (Kleeman, Hughes, Allen, & Cass, 1999; Sardar, Fine, & Sioutas, 2005) ions. The vast majority of fine PM nitrate however is in the form of ammonium nitrate (Hughes et al., 1999, 2002). A significant amount of ammonium nitrate can be lost during sampling (Kleeman et al., 1999) by volatilization and the



Fig. 6. Comparison of multimetal and elemental concentrations between HVDS and Partisol based on coarse PM concentrations.

lower coarse PM nitrate concentrations of the HVDS can be rationalized because of its higher filter face velocity (by 16-fold) compared to that of the Partisol, which would result in higher pressure drop across its filter, hence higher nitrate losses (Cheng & Tsai, 1997). Interestingly, the filter face velocities in the fine PM collector of the two instruments are comparable (the HVDS face velocity is 1.6 times higher than that of the Partisol) and therefore no major discrepancy in nitrate measurements was observed for that size range. It should be noted that the lower coarse PM nitrate concentration of the high-volume sampler are not a result of the design of the sampler's key components (i.e., PM_{10} inlet and virtual impactor), but intrinsic to the very high filtration velocity associated with passing a flow of 1001 min^{-1} through a 90 mm filter. Decreasing the minor flow to perhaps 401 min^{-1} may reduce some of these volatilization losses, although this needs to be verified in future investigations.

The comparison between HVDS and Partisol based on concentrations of trace elements and metals is summarized in Table 2. The following metals and elements were measured in sufficiently high concentrations in the coarse particle fraction by both samplers to allow comparisons: Al, Si, Ca, K, Fe, Ti and Na. Other trace elements, including Ni, Va, Cr, Pb, Sb, Sn, Cd, Ba and Cu, were detected in all samples of the HVDS, but in almost none of the Partisol samples because of its much lower sampling flow rate. Comparisons could therefore not be made for these elements because of insufficient data. Results from the field tests indicate that the coarse PM concentrations of the two samplers are in very good overall agreement for these trace elements. As shown in Table 2, the average concentration ratios between the HVDS and Partisol range between 0.85 and 1.00 for these species.

Fig. 6 represents the relationship between the collocated Partisol and HVDS for coarse PM trace elements and metals by integrating all the data points. The linear regression of this integrated comparison indicated an excellent overall agreement. The individual slopes, intercepts and R^2 for each trace elements measured are shown in Table 3. An important conclusion that can be derived from Fig. 6 is that the correlation is independent of the amount of the chemical constituent in the aerosol.

In addition to the HVDS and the Partisol measurements, the particle concentration enrichment of a tandem system of two virtual impactors—the coarse 10001 min^{-1} virtual impactor followed by a 1001 min^{-1} round nozzle virtual impactor (2.5 µm cut point) operating at a 5% minor flow ratio—was measured. The purpose of this analysis was to investigate the capability of enriching ambient coarse particles to higher degrees, and thus substantially improve the response time and detection limit characteristics of any on-line detector that may be used downstream of the sampler. This enhanced system may thus provide additional benefits for measuring minute traces of explosives and biohazards at ultra trace levels in ambient air, thereby contributing to national security and anti-terrorist strategic planning.

For these experiments, the minor flow of the second virtual impactor was connected directly to the APS, the sampling flow rate of which matches that of the virtual impactor. Ambient and concentration-enriched aerosol concentrations were measured alternately at 5-min intervals with the APS. Fig. 7 shows the average concentration enrichment factor

0.93

0.83

0.70

0.13-1.0

0.21-1.44

0.027-0.11

Trace element	Slope	Intercept ($\mu g m^{-3}$)	R^2	Concentration range $(\mu g m^{-3})$			
Na	0.74	0.09	0.92	0.19–1.0			
Si	0.90	0.01	0.90	0.24–2.3			
Fe	0.70	0.23	0.76	0.18-1.46			
Κ	0.69	0.05	0.77	0.05-0.34			

Table 3 Summary results of linear regression analysis of trace elements and metals measured by the HVDS and Partisol

-0.03

0.15

0.02

1.12

0.89

0.71

Al

Ca

Ti



Fig. 7. Concentration enrichment factor as a function of particle aerodynamic diameter for the tandem system of virtual impactors at a minor flow rate of 51 min^{-1} .

as a function of aerodynamic particle diameter for 10 field experiments. Data plotted in Fig. 7 reflect the collective concentration enrichment achieved by the virtual impactor tandem system and are thus the product of the size—dependent enriched curves of each system. The resulting 50% cutpoint of the tandem system (about 2.9 µm) is larger than the cut points of the individual virtual impactors (2.5μ m) because it corresponds roughly to the 75% cutpoint of each system. It is also evident in this figure that the concentration enrichment at 2.5 µm is about 25%, thus the product of the 50% cutpoint of the two virtual impactors. Considering that the majority of the mass-based concentration of ambient coarse PM typically is associated with particles larger than 3 µm in aerodynamic diameter (Lestari, Ali, & Kenneth, 2003), we do not expect this somewhat increased cutpoint of the tandem system to affect its overall performance in measuring total coarse PM concentrations. This argument is further corroborated by the overall enrichment factor based on the total mass concentrations in the size range of 2.5–10 µm PM measured by the APS. The average ambient and enriched coarse PM concentrations were determined to be 10.1 ± 3.6 and 1,723.2 ±223.6 µg m⁻³, respectively, thus indicating an overall concentration enrichment of 170 (of an ideal 200) accomplished by the dichotomous sampler.

During evaluation of the tandem impactor system, coarse PM mass concentrations were recorded during a 5-day period by a Tapered Element Oscillating Microbalance (TEOM, Series 1400a, Thermo Electron Corporation) that was connected directly to the outlet of the minor flow of the second virtual impactor. The mass concentration measured by the tandem system is compared to that determined by a Continuous Coarse PM Monitor (CCM), which is described in greater detail by Misra et al. (2001). Fig. 8a shows the time series of coarse PM concentrations measured by these two technologies during summer 2005. The graph has concentrated coarse PM concentrations by the tandem virtual impactor system on the primary *y*-axis (left side) and ambient coarse PM concentrations on the secondary *y*-axis (right side) plotted as a function of time and day. The primary axis scale is larger than the secondary axis scale by a factor equal to the ideal enrichment of 200. Thus, overlap of the two time series indicates close to ideal aerosol concentration enrichment.



Fig. 8. (a) Time series for virtual impactor tandem system and CCM measured coarse PM concentrations ($\mu g m^{-3}$) and (b) ambient parameters (RH and wind speed) during tandem system experiments.

The data plotted in Fig. 8a clearly shows excellent overall agreement obtained for the two samplers, with the average ratio of tandem virtual impactor system to CCM of 178 (\pm 14) over the 5-day period. Furthermore, as part of this analysis, we have attempted to identify the factors that are responsible for the diurnal variation of coarse PM mass concentrations. Meteorological parameters such as wind speed and direction and the ambient RH can considerably influence coarse particle mass concentrations. Fig. 8b represents the RH and the wind speed at the sampling site for the entire 5-day period of this analysis. Figs. 8a and b show that coarse PM concentrations increase during the daytime as wind speed increases. This was interpreted as the result of more effective wind-driven resuspension of coarse particles in the latter part of the day leading to higher concentrations. Harrison et al. (2001) have studied the influence of wind on coarse PM and found two primary contributors to coarse PM. The first is a non-wind component that corresponds to particles emitted from industrial and construction activities, traffic-induced resuspension and biological particles, and the second is a wind-derived fraction that arises mainly from natural sources such as sea spray and surface soils or dusts on paved areas. A recent study by Charron and Harrison (2005) also corroborated the association of coarse PM and wind speed. Finally, Watson et al. (1994) reported in the Southern California Air Quality Study (SCAQS) in 1987 that the major component of PM_{10} in summer was suspended surface dusts corresponding to higher wind speed. Figs. 8a and b also show an inverse correlation of coarse PM concentration and ambient RH, with higher coarse PM concentrations during daytime corresponding to lower RH. The period between April and July (late spring through early fall) in the Los Angeles Basin is characterized by regular occurrence of marine layers and associated low cloud and fog formations with high RH in the overnight and early morning hours. The early morning stable atmospheric conditions during marine layer episodes and to a lesser extent, the higher soil moisture associated with high RH, results in less resuspension of road and soil dust, the main contributors to coarse mode aerosol.

4. Conclusions and summary

This study details the development and characterization of the next generation, single, round nozzle, HVDS with a flow capacity of $1000 \, l \, min^{-1}$. The field evaluation of the HVDS yielded good agreement with other impactor-based measurements. Characterization of the coarse virtual impactor displayed near ideal enrichment at two different minor flow ratios (10% and 4%) relative to the total flow rate of $1000 \, l \, min^{-1}$, yielding enrichments of coarse particles relative to that observed in ambient air of 10 and 25, respectively. The $PM_{2.5-10}$ mass concentrations measured by the HVDS correlated well with both MOUDI and dichotomous Partisol results. Sulfate and nitrate species demonstrated slightly lower agreement between the HVDS and the Partisol due to a combined effect of cutpoint differences and possible volatilization of labile ammonium nitrate from the HVDS as result of the high face velocity on the filter collecting coarse PM. Nevertheless, excellent agreement was observed between the two samplers for observed concentrations of several trace elements. The benefit of the higher flow rate and resulting lower limits of detection were observed in the trace elements analysis, since a number of species were observed in the coarse particles collected by the HVDS that were not observed on filters collected by the dichotomous Partisol. The tandem virtual impactor super-concentration system was tested and proved to be capable of achieving enrichment of ambient particles up to 180 times. This study substantiates results of previous studies concerning the effect of wind speed and RH on coarse PM concentrations in ambient air.

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