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Versatile aerosol concentration enrichment system (VACES) for simultaneous in vivo and in vitro evaluation of toxic effects of ultrafine, fine and coarse ambient particles Part I: Development and laboratory characterization

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Abstract

This study presents the development and bench-testing of a versatile aerosol concentration enrichment system (VACES) capable of simultaneously concentrating ambient particles of the coarse, fine and ultrafine size fractions for conducting in vivo and in vitro studies. The VACES consists of three parallel sampling lines (concentrators), each operating at an intake flow rate of $110\,l\,min^{-1}$. Coarse particles are concentrated using a single round nozzle virtual impactor. Concentration enrichment of $PM_{2.5}$ and ultrafine particles is accomplished by first drawing air samples through two parallel lines, having 2.5 and 0.18 μm cutpoint pre-impactors, respectively, to remove particles larger than these sizes from the air sample. Both of the smaller PM fractions are drawn through a saturation–condensation system that grows particles to 2–3 μm droplets, which are subsequently concentrated by virtual impaction. A diffusion dryer is used in the fine and ultrafine concentrators to remove excess vapor and return the concentrated particles to their original size, prior to supplying them for in vivo exposures. The VACES can also provide highly concentrated liquid suspensions of particles of these three modes for in vitro toxicity studies. This is accomplished by connecting the concentrated output (minor) flows of each of the VACES parallel concentrators to a liquid impinger (BioSampler), used in a modified configuration, to collect particles under near-ambient pressure.

Detailed laboratory characterization of the individual components of the VACES are presented in this paper, including evaluation of its ability to preserve particle mass, number, and chemical species during the concentration enrichment process. Our experimental results showed that concentration enrichment is

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accomplished with very high efficiency, minimal particle losses and without any significant dependence on particle size or chemical composition.

Keywords: Ultrafine; fine and coarse particle concentrators; PM toxicity

1. Introduction

Despite evidence from epidemiological studies associating ambient particulate pollution with adverse health effects in humans (Bates & Sizto, 1989; Thurston, Hayes, Bates, & Lippmann, 1994; Pope, Dockery, & Schwartz, 1995), there are still uncertainties regarding physiochemical properties of particles that affect health risk and underlying pathophysiological mechanisms (Vedal, 1997). In many cases, the apparent lack of agreement between epidemiological and toxicological studies may be attributed to the inability of controlled laboratory investigations to deliver a sufficient dose of ambient aerosols, which might support the epidemiological findings. Ambient aerosol concentrations are, in general, too low to induce measurable acute toxic effects, and particles generated from laboratory solutions do not represent the full matrix of potentially toxic components present in ambient aerosols.

Ambient particulate matter smaller than 10 μm in aerodynamic diameter (PM_{10}), is commonly identified by the following three size modes: ultrafine (less than about 0.1 μm); accumulation (between 0.1 and 2.5 μm); and coarse (2.5–10 μm). The accumulation and ultrafine modes are often combined into the so-called fine mode (or $PM_{2.5}$). Results from epidemiological studies suggest that fine particles may contribute more to toxic outcomes than do coarse particles. In contrast, recent investigations conducted in Mexico City (Loomis, Castillejos, Borja-Aburto, & Dockery, 1999) and the Netherlands (Kleinman, Sioutas, Chang, & Cassee, 2000) have demonstrated higher toxicity associated with coarse than fine particles. These studies suggest that the coarse mode may be more important as a predictor of daily mortality in certain populations, or regions. Additionally, results from recent epidemiological studies (e.g., Heyder et al., 1996; and Peters, Dockery, Heinrich, & Wichman, 1997) suggest that the ultrafine fraction of PM may be of toxicological importance. Reported effects may be associated with the greater number of particles and/or greater surface area that predominates in this size range.

The recent development of ambient particle concentrators (e.g., Sioutas, Koutrakis, Ferguson, & Burton, 1995; Sioutas et al., 1997; Gordon, Gerber, Fang, & Chen, 1999; Kim, Chang, & Sioutas, 2000a) has made it possible to investigate exposures to "real-life" ambient aerosols at increased, but still realistic concentrations. Their results suggest that concentrated ambient aerosol exposure systems may provide a useful method for assessing health effects associated with ambient particles (e.g., Godleski, Sioutas, Katler, & Koutrakis, 1996; Gong et al., 2000). Nevertheless, currently available aerosol concentrators focus on concentrating only one and/or two mode(s) of ambient particulate matter. For example, the concentration enrichment of the Harvard Fine Particle Concentrator (Sioutas et al., 1995) depends on particle size, with the 0.4–1 μm particles of the accumulation mode being concentrated far more effectively than particles smaller than 0.4 μm (Sioutas et al., 1997; Kim, Sioutas, Chang, Gong, & Linn, 2000b). Consequently, this system cannot be used to increase the concentration of particles below 0.15 μm (i.e., the ultrafine particles). Another type of concentrator, using centrifugal forces,

achieves restricted particle concentration enrichment (mostly in the 0.5–1.0 μm aerodynamic size range) due to coarse particle loss by impaction and losses of ultrafine particles by diffusion (Gordon et al., 1999).

A recently developed portable concentrator, designed to concentrate fine and ultrafine PM (Kim et al., 2000a), uses a super-saturation/condensation system to rapidly enlarge particles to super-micrometer droplets, followed by size-separated concentration by means of a virtual impactor. This earlier system is useful as a portable unit, because of its relatively small size, and of its ability to concentrate particles of either the coarse, or fine (including the ultrafine) fractions of PM. However, this system is limited to concentrating one size fraction at a time and one type of exposure for a given experiment. Furthermore, it does not have the ability to separate the ultrafine from the accumulation modes of the concentrated $PM_{2.5}$.

The purpose of this study, herein, is to develop and bench-test a versatile aerosol concentration enrichment system (VACES) capable of simultaneously concentrating particles of the coarse, fine and ultrafine size fractions for use in in vivo and in vitro studies. This paper focuses on the development and characterization of the individual units of the VACES in the laboratory, including evaluation of its ability to preserve particle mass, number, and chemical species throughout the concentration enrichment process. The performance of the entire VACES was further evaluated in a field study, conducted outdoors at Rancho Los Amigos National Rehabilitation Center in Downey, CA, and presented in the second part (Kim, Jaques, Chang, & Sioutas, 2001) of this series.

2. Methods

2.1. Design of the VACES

The versatile aerosol concentration enrichment system incorporates the following features:

1. The ability to isolate and enrich the concentration of ultrafine particles, and supply them to an exposure chamber at virtually atmospheric pressure (i.e., 0.99 atm).
2. The ability to allow concurrent animal exposures to different particle size fractions: coarse, fine, and ultrafine.
3. The ability to operate at flow rates as high as 330 l min^{-1} and concentrate one of the three PM modes to nearly a factor of 33 (e.g., to a minor flow of 10 l min^{-1}). This feature makes it possible to use more animals, or humans in inhalation studies, potentially increasing the confidence level in the observed outcomes.
4. Simultaneous collection of high particulate mass quantities (i.e., on the order of mg) of all size fractions into relatively small liquid volumes (e.g., 4–10 ml) within a liquid impinger (BioSampler, SKC Inc, Eighty-Four, PA). The resulting highly concentrated liquid suspensions can be readily used for ambient particle size-dependent in vitro toxicity studies.

Two, of several, possible configurations of the VACES system are presented in Figs. 1a and b. Fig. 1a shows the configuration for simultaneous inhalation (in vivo) exposures to coarse, fine and ultrafine particles, and Fig. 1b shows the version of the same system, but used for in vitro toxicity testing. In general, both versions use the same configuration and components, except

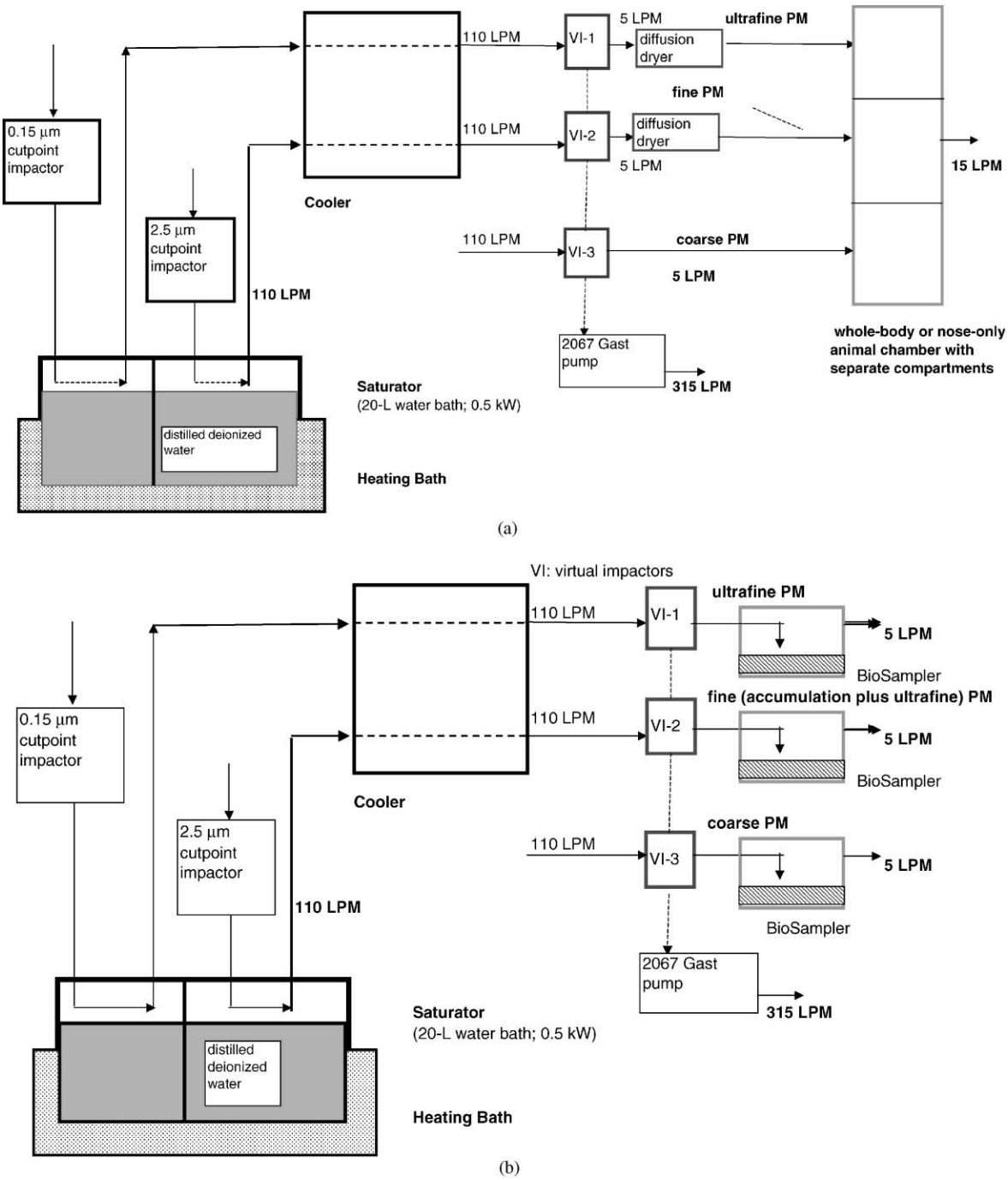

Fig. 1. (a) Versatile aerosol concentration enrichment system (VACES) for concurrent in vivo studies to coarse, fine and ultrafine PM. (b) Versatile aerosol concentration enrichment system (VACES) for in vitro studies.

for how the particles are treated and collected downstream of the concentrator. First, the general system is discussed, and then two of the optional configurations (i.e., in vivo, and in vitro) are described, below.

2.1.1. In vivo VACES

The VACES consists of three parallel sampling lines. In each line, ambient coarse, fine and ultrafine aerosols are separately drawn at 110 l min^{-1}. For enrichment of the coarse PM fraction, ambient particles are drawn through a round nozzle, single-stage virtual impactor that has a 50% cutpoint at 1.5 μm. Coarse particles in this sampling line can be concentrated by, as much as a factor of 33, and supplied to the exposure chamber at a flow rate of 3.3 l min^{-1}. In much greater detail, Kim et al. (2000a) detail the design and performance characteristics of the virtual impactor, which is used, herein, for enrichment of coarse ambient aerosols.

As with the coarse concentrator, the $PM_{2.5}$ and ultrafine PM sampling lines of the VACES use a virtual impactor for concentrating ambient aerosols within an identical or similar configuration to that of the coarse concentrator. Differences between the coarse and the fine and ultrafine concentrators are: (1) the $PM_{2.5}$ and ultrafine PM units use 2.5 and 0.18 μm pre-impactors, respectively, to remove particles larger than these sizes from the air samples; (2) both of the smaller fractions are drawn through a saturation–condensation, particle-to-droplet growth system prior to concentration enrichment; and (3) both systems require a diffusion dryer to remove the excess vapor from the concentrated and saturated aerosol and return the particles to their original size, prior to supplying them for in vivo exposures.

In the sampling line concentrating fine (i.e., accumulation plus ultrafine) PM, air samples are first drawn through a single-slit nozzle impactor, having a 50% cutpoint at 2.5 μm at a flow rate of 110 l min^{-1}. The impactor's acceleration nozzle is 0.2 cm wide and 5 cm long. At a sampling flow rate of 110 l min^{-1}, particles are accelerated to a velocity of 1834 cm s^{-1}, and the corresponding pressure drop across the impactor is 1.5 in of H_2O (i.e., 0.37 kPa).

In order to remove all but the ultrafine PM, particles in the third sampling line of the VACES are drawn through a multi-nozzle, high volume conventional impactor having a target 50% cutpoint of 0.18 μm in aerodynamic diameter. Separation of these particles is accomplished under a very low-pressure drop (i.e., 7 in of H_2O), which is essential for inhalation studies that cannot be conducted under a substantial vacuum. The impactor, specially designed for this study, consists of five slit-shaped parallel nozzles, each being 5 cm long and 0.015 cm wide. The resulting velocity through each rectangular jet is approximately 4200 cm s^{-1}. A 5.0×0.2 cm quartz fiber strip is placed at a distance of 0.04 cm underneath each acceleration nozzle. The quartz strips are coated with mineral oil and serve as bounce-free impaction substrates for collecting particles above 0.18 μm in aerodynamic diameter.

After size-selective separation, the $PM_{2.5}$ and ultrafine fractions are separately drawn through a 10 l stainless steel container that is partially filled with warm, ultrapure distilled-deionized water. To saturate the aerosol, the ambient particles pass over the liquid surface, mixing with the vapor to result in a particle–vapor mixture at about 30 (± 2) degrees Celsius ($^{\circ}C$). To obtain insulated and stable conditions, the stainless steel container is placed inside a heating bath (VWR Scientific, Model 1024) that has a maximum heating power of 0.5 kW. Downstream, the saturated aerosol fractions are simultaneous cooled through two aluminum tubes (inner

diameter of 2.2 cm × 80 cm in length) that are embedded in a saline–ice mixture, maintained at about −6 to −8°C. Through the tubes, the resultant warmed and saturated aerosol is cooled by, about, 9–10°C, i.e., to 20°C or 21°C. The resulting supersaturation causes all particles to grow to about 2.5–3 μm droplets. Subsequently, each sampled fraction (i.e., the $PM_{2.5}$ and Ultrafine) of enlarged droplets are drawn through their respective virtual impactors. The virtual impactors (made of anodized aluminum) are designed to separate particles into two different size ranges, approximately above (within the minor flow) and below (within the major flow) 1.5 μm, respectively. The concentration of the larger droplet-associated particles in the minor flow (i.e., those of interest) is enriched by a factor ideally equal to the ratio of total sample flow rate to minor flow rate.

The separate fractions of concentrated droplets are drawn through two separate diffusion dryers (TSI Model 3062, TSI Inc., St. Paul, MN), which are placed downstream of the minor flow collection nozzle of each virtual impactor. A single, oil-less rotary vane pump (Gast model 2067, Gast Manufacturing, Cerritos, CA) is used to independently draw all three major flows of the parallel virtual impactors. The pump can draw at up to 360 l min^{-1} under a vacuum of 150 in of water, while consuming only 0.5 kW at 110 V. Moreover, it is relatively light (20 lb), occupies very little space, and does not require any special power installation. Operating at a maximum flow rate of 10 l min^{-1}, each diffusion dryer reduces the relative humidity of the incoming aerosol from 100% to less than 50% to dry the droplets to their original particle size distribution.

2.1.2. In vitro VACES

Fig. 1b shows the alternative configuration of the VACES, used for simultaneous coarse, fine and ultrafine PM collection for in vitro toxicology experiments. For this in vitro configuration, instead of passing through a diffusion dryer, the concentrated coarse, fine and ultrafine particles in each parallel sampling line are drawn through a liquid impinger (BioSampler, SKC West Inc., Fullerton, CA). The performance of this device is described in greater detail by Willeke, Lin, and Grinshpun (1998). Unlike conventional impingers, in which the aerosol is impacted into a reservoir filled with liquid, particles in the BioSampler are injected into a swirling flow so that they can be collected by a combination of inertial and centrifugal forces onto the surface over which the air flow swirls.

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

Particle collection using liquid impingers has been shown to be advantageous over the traditional filtration or impaction methods for collection of airborne particles, because impingers are not easily overloaded (Willeke et al., 1998), and because impingement eliminates the need for elaborate extraction procedures (Zucker, Draz, & Muller, 2000). Under normal operating

conditions (at its nominal flow rate of 12.5 l min^{-1}), the BioSampler has collection efficiency close to 100% for particles larger than about 1.5 μm. For particles smaller than 1.0 μm in aerodynamic diameter, the collection efficiency decreases sharply to less than 50% (Willeke et al., 1998). Operating in conjunction with the VACES, however, the BioSampler can collect any of the PM size ranges with 100% efficiency and at sampling flow rate that is at least a 10-fold higher than its nominal operating flow rate. Thus, the supersaturational growth of even ultrafine PM to super-micrometer particles enables effective trapping of these particles by the impinger and allows us to "concentrate" large volumes of ambient PM into a very small solution (i.e., order of 5–10 ml). Furthermore, the ability to collect large volumes of particles directly into a small volume of any solution is a particularly attractive feature when intratracheal instillation is used as the method to conduct particle toxicity tests.

Overall, the VACES is very compact in size, and is easily transported to field sites used for investigating in vitro cytotoxicity, and/or animal, or human inhalation exposures (i.e., in vivo) to concentrated particulate matter. The modular design of this concentrator makes it readily adaptable to accommodate higher output flow rates that are required for human exposures, but which is easily achieved by placing several single-nozzle virtual impactors in parallel.

2.2. Experimental characterization of the VACES individual components

2.2.1. 2.5 *and* 0.18 μm *low pressure drop slit impactors*

The collection efficiency of the 2.5 μm cutpoint slit impactor was determined using monodisperse aerosols generated by atomizing suspensions of PSL particles (size range: 0.5–10 μm; Bangs Laboratories Inc., Fisher, IN) generated with a constant output Nebulizer (HEART, VORTRAN Medical Technology, Inc., Sacramento, CA). Prior to passing through the slit impactor, the generated aerosols were diluted with filtered air, and passed through ^{210}Po neutralizers. The mass concentrations of the monodisperse aerosols upstream and downstream of the impactor were measured with a nephelometer (DataRAM, MIE, Inc., Billerica, MA). For each test, repeated measurements of the concentrations upstream and downstream of the impactor were taken. The concentrations of the generated aerosols were in the range of 100–400 μg m^{-3}, thus several orders of magnitude higher than the limit of detection of the DataRAM (about 1–5 μg m^{-3}), and below the upper limit (about 40 mg m^{-3}). The response of the DataRAM is dependent on particle size (Sioutas, Kim, Chang, Terrell, & Gong, 2000). Since penetration through an impactor is defined by the ratio of the downstream to the upstream concentrations, the value of this parameter should not be dependent on particle size when determined using monodisperse aerosols.

The collection efficiency of the multi-slit 0.18 μm cutpoint (for the ultrafine fraction) impactor was estimated using ambient laboratory air as the test aerosol. For particles in the size range of 0.015–0.5 μm, penetration was determined by measuring their number concentration upstream and downstream of the impactor by means of the Scanning Mobility Particle Sizer (SMPS, Model 3096, TSI Inc., St. Paul, MN). The SMPS sampled 0.2 l min^{-1} of the total flow rate of 110 l min^{-1} through the impactor. To account for possible diffusion losses of ultrafine particles through the SMPS sampling lines, the number concentration of ambient aerosols was measured, with and without the block that holds the acceleration slit nozzles of the impactor. In addition to the SMPS, the DataRAM was used to evaluate the collection efficiency of the multi-slit

impactor for particles in the 0.2–1.0 μm range, using artificially generated monodisperse PSL particles as described above. The DataRAM could not be used to monitor particles less than 0.2 μm, because the sensitivity of the instrument decreases sharply below this particle size.

Additional paired, but limited, field measurements were conducted by comparing the ultrafine impactor to the Microorifice Uniform Deposition Impactor (MOUDI, MSP Corp., Minneapolis, MN), which was used as the reference sampler. A 47 mm Teflon filter (2 μm pore size, Gelman Science, Ann Arbor, MI) was placed immediately downstream of the multi-slit impactor, sampling at a flow rate of $110\,l\,min^{-1}$. The MOUDI was placed at a distance of 1 m from the impactor and sampled at $30\,l\,min^{-1}$. Ambient particles smaller than 0.18 μm in aerodynamic diameter were collected on a 37 mm Teflon filter following the last impaction stage of the MOUDI. Particle mass on the filters was measured under the controlled relative humidity (40–45%) and temperature (22–24°C) conditions for, both, the MOUDI and multi-slit impactor by a Mettler Microbalance (MT5, Mettler-Toledo, Inc., Highstown, NJ).

2.2.2. Characterization of the BioSampler

At the standard operation flow rate of $12.5\,l\,min^{-1}$, the pressure drop across the BioSampler is close to 0.5 atm, which has been shown to cause excessive evaporation of liquid collection media such as water (Willeke et al., 1998). It is also expected that under these sampling conditions, excessive loses of semi-volatile components of ambient particles can occur. In order to reduce the pressure drop across the BioSampler used in conjunction with the VACES virtual impactors, a flow rate of $5\,l\,min^{-1}$ was used. The decrease in flow rate was expected to increase the cutpoint of the BioSampler. However, as most of fine and ultrafine particles are grown to 2.5–3 μm via super-saturation in the VACES, our primary concern was to ensure that particles above this size are efficiently collected by the modified BioSampler, and under a relatively low pressure drop.

Another modification of the BioSamplers used in conjunction with the VACES was the amount of water used in its reservoir for collecting impinged particles. In its nominal configuration, 20 ml of liquid are required in the BioSampler reservoir. However, from the standpoint of toxicological studies, it is highly desirable to maximize the concentration of the collected ambient particles in the liquid medium of the BioSampler. We, thus, investigated the effect of different volumes of water on the collection efficiency of the BioSampler at the reduced flow rate of $5\,l\,min^{-1}$. We specifically tested the BioSampler using water volumes of 2, 4, 10 and 20 ml, respectively. For each liquid volume, the collection efficiency of the BioSampler for sampling monodisperse aerosols (0.5–5 μm) was measured with the DataRAM nephelometer, up- and downstream the impinger. At $5\,l\,min^{-1}$, the pressure drop across the BioSampler was approximately 17 in of H_2O. The exhaust of the DataRAM pump was returned downstream of the BioSampler in order to avoid sampling biases, which might occur when this instruments samples under a vacuum (the manufacturer recommends this sampling strategy).

2.2.3. Characterization of $PM_{2.5}$ and ultrafine aerosol concentration enrichment units

As the coarse particle concentrator component of the VACES was previously developed (Kim et al., 2000), the laboratory tests, herein, focus on the experimental characterization of the fine and ultrafine concentrators of the double sample-line configuration of the VACES. It

should be noted that the use of the 0.18 μm impactor to remove all but ultrafine particles is optional. The VACES can also be used to concentrate fine PM including the ultrafine fraction from 220 l min^{-1} to a flow as small as 7 l min^{-1}. Thus experiments were conducted at a sampling flow of 220 l min^{-1} as a worse case scenario, since this flow rate represents the most challenging configuration for the saturator and the cooler of the VACES. To achieve this, individual monodisperse aerosols were generated, and simultaneously sampled through the paired 110 l min^{-1} VACES sample lines (see Figs. 1a and b).

The experimental characterization of the VACES was conducted using laboratory monodisperse particles as well as "real-life" ambient particles as test aerosols. Monodisperse aerosols were generated by atomizing suspensions of ultrafine and fine particles using the same setup described above for characterizing the slit impactors. Different types of suspensions were used, including monodisperse PSL fluorescent latex particles (size range 0.05–2 μm; Polysciences, Inc., Warrington, PA), and as monodisperse silica beads (0.36 μm; Bangs Laboratories, Inc., Carmel, IN). In addition, aqueous solutions of ammonium sulfate and ammonium nitrate were atomized. Finally, indoor aerosol was also used as a test aerosol. For particles less than 0.7 μm, the size distributions of the polydisperse aerosols were determined using the SMPS. The generated aerosols were dried, neutralized, and drawn through the saturator at 220 l min^{-1}. Three different minor flow rates were tested, 7, 10, and 21 l min^{-1}, respectively (corresponding to theoretical enrichment factors of 30, 22, and 10.5, respectively). The TSI Condensation Particle Counter (CPC 3022, TSI, Inc., St. Paul, MN) was connected immediately upstream of the saturator and downstream of the diffusion drier to measure the number concentrations of the original and concentrated aerosols. For each particle size, concentration enrichment was defined as the ratio of the concentration measured downstream of the diffusion dryer to that measures upstream of the saturator.

3. Results and discussion

3.1. 2.5 *and* 0.18 μm *low pressure drop slit impactors*

Figs. 2 and 4 present the collection efficiency of the 2.5 and 0.18 μm cutpoint slit impactors as a function of particle aerodynamic diameter, respectively. The data in Fig. 2 confirm that the 50% cutpoint of the single-slit impactor used to remove coarse particles from the air sample is at about 2.5 μm in aerodynamic diameter. The sharpness of the collection efficiency curve of an impactor can be defined in terms of the geometric standard deviation (σ_g), which is the ratio of the aerodynamic particle diameter corresponding to 84% collection efficiency to the 50% cutpoint (Marple & Willeke, 1976). Based on this definition, the value of σ_g is approximately 1.2 for this impactor, thereby indicating reasonably sharp aerodynamic particle separation characteristics.

Since the 0.18 μm cutpoint multi-slit impactor has been designed specifically for toxicology studies (which cannot be conducted under a substantial vacuum), its pressure drop as a function of flow rate was measured (Fig. 3). At the inlet flow of 110 l min^{-1}, which is used for enrichment of the concentrated ultrafine fraction of ambient aerosols in this study, a pressure drop of about 7 in of H_2O (1740 Pa) resulted. The ability of this impactor to remove all but ultrafine particles with a very low pressure drop is a very important feature of the VACES, as

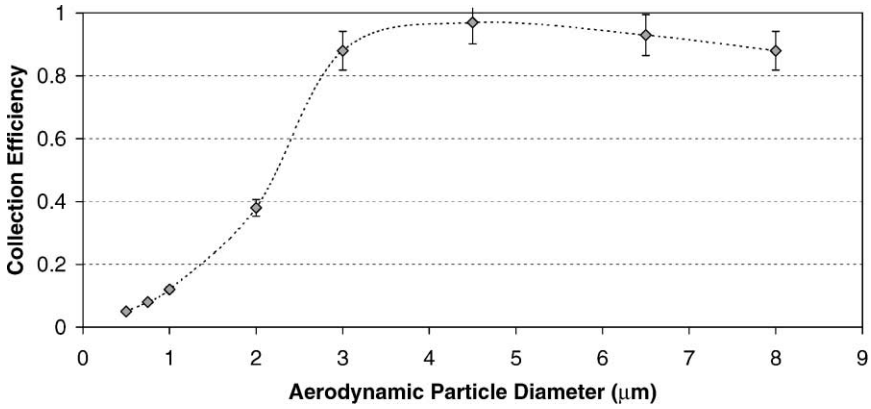

Fig. 2. Particle collection efficiency of the 2.5 μm cutpoint slit nozzle impactor. Flow rate: 110 l min^{-1}.

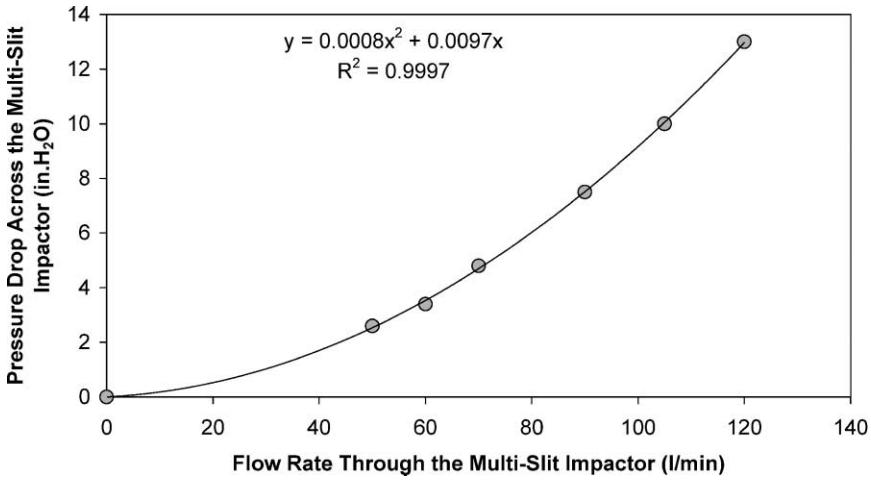

Fig. 3. Pressure drop across the 0.18 μm cutpoint, multi-slit impactor as a function of flow rate.

it makes it possible to supply the concentrated ultrafine aerosol to exposure chambers under a pressure of 0.98 atm.

The collection efficiency of multi-slit impactor, determined from the decrease of both number (SMPS) and mass (DataRAM) concentrations, measured downstream of the impactor, is also plotted as a function of particle aerodynamic diameter (Fig. 4). The data demonstrate that the two aerosol monitors are in agreement, where they overlap (i.e., between 0.2 and 0.5 μm), and suggest continuity where the measurement of the SMPS ends and the DataRAM begins (i.e., about 0.5 μm). Particle collection efficiency, as measured by the SMPS, increases sharply starting at 0.1 μm, and reaches the value of about 90% at particles larger than 0.3 μm in aerodynamic diameter. The data plotted in Fig. 4 show that the 50% cutpoint of the multi-slit nozzle impactor has an electrical mobility diameter of 0.18 μm.

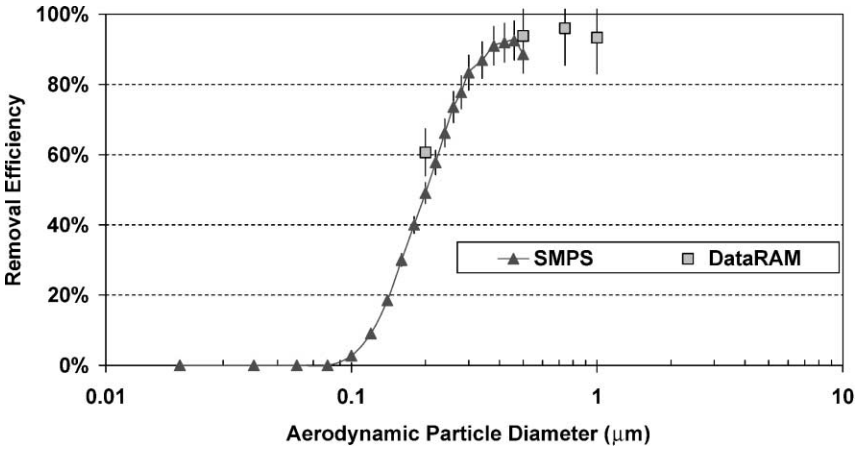

Fig. 4. Removal efficiency of multi-slit low pressure drop impactor as function of aerodynamic particle diameter.

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

Ambient ultrafine mass concentration ($\mu g\ m^{-3}$)	Multi-slit impactor ultrafine mass concentration ($\mu g\ m^{-3}$)	Ratio of mass concentrations between multi-slit impactor and MOUDI[b]
1.89	2.48	1.31
2.78	2.47	0.89
3.28	3.16	0.96
3.81	4.13	1.08
4.23	5.05	1.19
	Average	1.07
	Standard deviation	0.15

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

Comparisons between the mass concentrations, as measured by the multi-slit impactor and the reference MOUDI, are presented in Table 1. Despite the small number of data points, the mass concentrations of ultrafine particles, measured by the two impactors, are in very good agreement. The average multi-slit impactor-to-MOUDI ratio for ultrafine particle mass concentration, is 1.07 ($\pm$0.15). The agreement between the two samplers is remarkable, given that, a small difference in the cutpoint (e.g., 0.15 versus 0.2 μm) could result in substantial differences in particle mass collected by the two impactors. This would be because typically the mass-based concentration of ambient $PM_{2.5}$ decreases sharply at particle sizes smaller than 0.2 μm (Whitby & Svendrup, 1980), and a significantly higher mass concentration would be measured by the sampler having the largest cutpoint impactor.

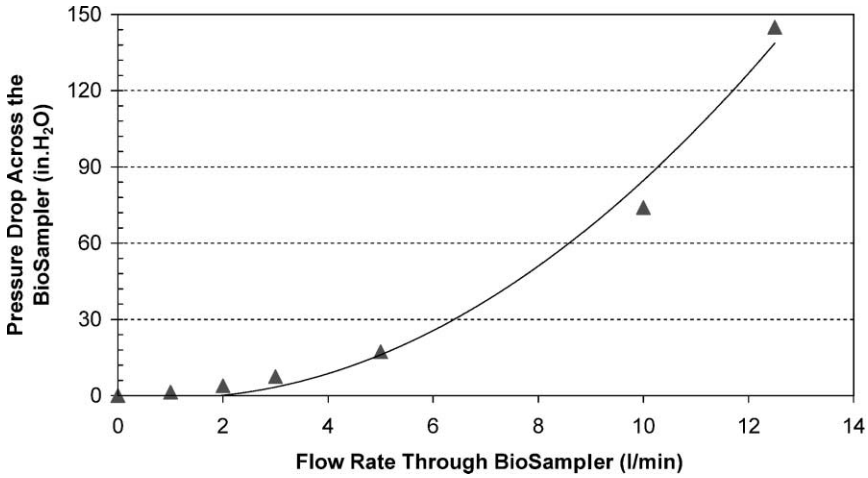

Fig. 5. Pressure drop across the BioSampler nozzle as a function of flow rate.

3.2. Characterization of the modified BioSampler

Fig. 5 shows the pressure drop across the BioSampler nozzle as a function of flow rate. The pressure drop at 5 l min^{-1} is 17 in of H_2O (0.035 atm), which is a substantially lower value than the 145 in of H_2O, at the standard flow rate of 12.5 l min^{-1}. As a result of this small pressure drop, less than 0.5 ml of water volatilized after 6 h of sampling ambient concentrated air, at relative humidities ranging from 30% to 65%. By comparison, 80% or more of 20 ml of water normally evaporates within 2 h under reduced pressure at 12.5 l min^{-1} (Willeke et al., 1998). Furthermore, the small pressure drop is essential in preserving labile semi-volatile species such as ammonium nitrate and a host of organic compounds that would be more pronounced under the high pressure drop across the sampler (Zhang & McMurry, 1987).

The collection efficiency of the BioSampler at 5 l min^{-1} is presented as a function of particle size for various amounts of water (2–20 ml) in the BioSampler reservoir (see Fig. 6). Error bars represent the standard deviation of repeated tests. The results show that, for any particle size, the collection efficiency of the BioSampler does not vary with the volume of water in its reservoir (between 2 and 20 ml).

The collection efficiency of the BioSampler is close to 100% for particles larger than 2 μm at a flow rate of 5 l min^{-1}, suggesting this sampler is adequate for collecting the enlarged and concentration-enriched ultrafine and fine particles. For particles less than 1 μm in aerodynamic diameter, the collection efficiency decreases sharply to about 50% at 0.5 μm. Any significant decrease in the collection efficiency due to particle bounce was not observed up to about 5 μm in aerodynamic diameter.

Additionally, a major advantage of the impinger's high particle collection efficiency is the maximization of concentration-enriched ambient fine and ultrafine PM suspended in aqueous solution, which is greatly useful for in vitro toxicity testing. To ensure complete wetting of the bottom of the BioSampler reservoir 5 ml is also sufficient, a feature that ensures effective particle captures by the glass impinger.

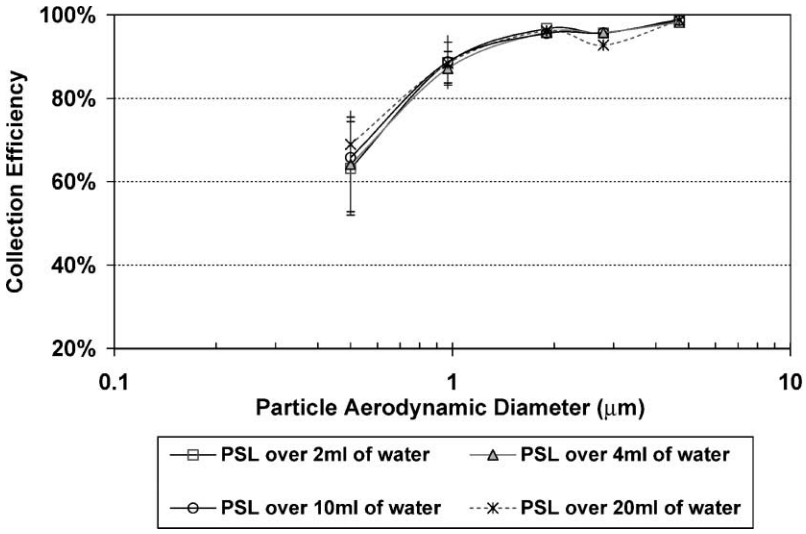

Fig. 6. Particle collection efficiency of BioSampler as a function of particle aerodynamic diameter. Sampling flow rate: $5\,\mathrm{l\,min^{-1}}$.

Operating in conjunction with our prototype ultrafine, fine or coarse particle concentrator, the BioSampler can collect any of the PM size ranges with virtually 100% efficiency and at sampling flow rates as much as 10–30 times higher than its nominal operating flow rate. Thus, the condensation growth of even ultrafine PM to super-micrometer particles enables effective trapping of these particles by the impinger, thereby allowing concentration of large quantities of ambient PM into a small liquid volume (i.e., 5–10 ml).

3.3. *Characterization of* $PM_{2.5}$ *and ultrafine aerosol concentration enrichment units*

Results from the laboratory evaluation of the VACES at three different minor flow rates are summarized in Fig. 7. In all three minor flow configurations, the major flow rate is adjusted to yield a total intake flow of $220\,\mathrm{l\,min^{-1}}$. Hence, the maximum obtainable concentration enrichment factors for each configuration are 31, 22, and 10.5, respectively. The concentration enrichment factors, as a function of aerodynamic particle diameter (see Fig. 7), have been obtained using monodisperse PSL aerosols in the size range of 0.05–1.9 μm (indicated with the solid data labels), except for the data corresponding to 0.028, 0.16, and 0.36 μm particles (indicated with transparent data labels). The number median diameters (NMD) of polydisperse indoor air, ammonium sulfate and ammonium nitrate aerosols are 0.028, 0.16 and 0.36 μm, respectively. The size distributions of these aerosols were obtained using the SMPS.

The resulting enrichment factors at minor flow rates of $7, 10$, and $20\,\mathrm{l\,min^{-1}}$ are 30.1, 20.4, and 9.6, respectively, which are very close to the ideal values (indicated by the dotted lines in Fig. 7). In addition, hygroscopic ammonium sulfate and ammonium nitrate aerosols did not show any observable difference in the enrichment factors compared to the hydrophobic PSL particles.

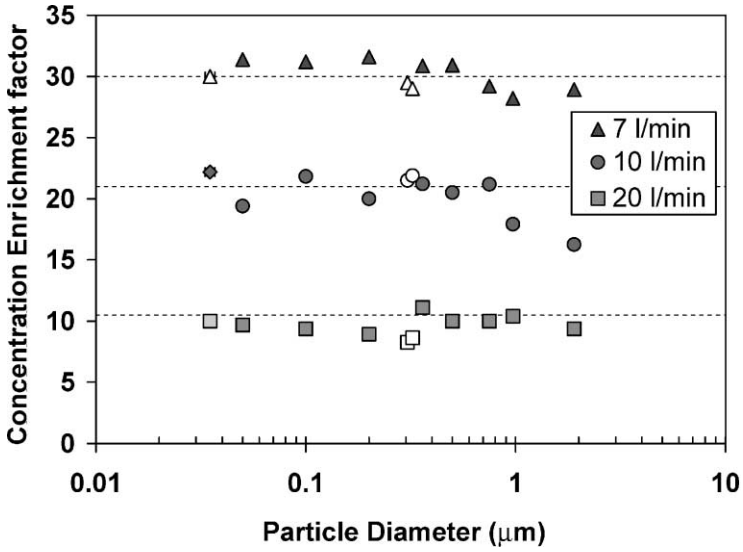

Fig. 7. Characterization of the versatile aerosol concentration enrichment system for three minor flows. Total intake flow: 220 l min^{-1}. Transparent data labels correspond to indoor air (NMD = 0.028 μm) ammonium sulfate (NMD = 0.16 μm) and ammonium nitrate (NMD = 0.36 μm) particles. Solid data labels correspond to PSL particles.

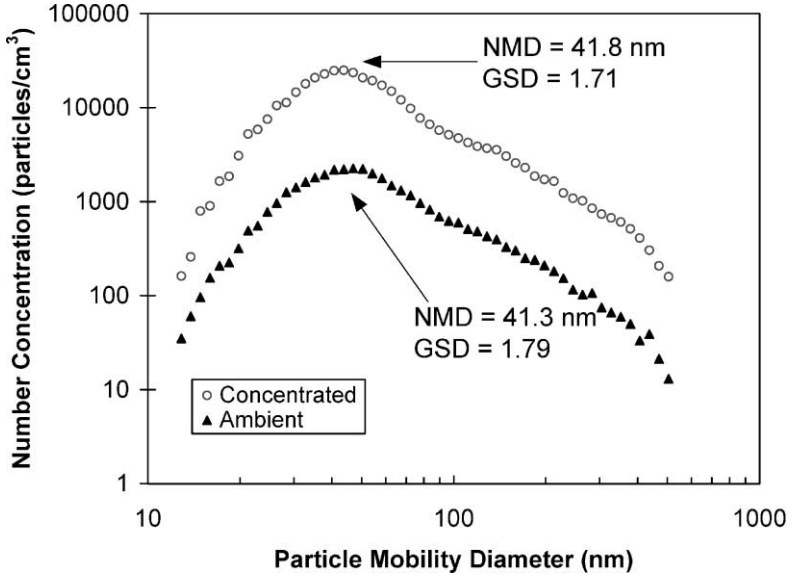

Fig. 8. Size distribution of ambient aerosols before and after the VACES measured by SMPS.

Finally, Fig. 8 shows the concentration enrichment as a function of particle mobility diameter obtained by measuring the size distributions of ambient aerosols upstream of the VACES and immediately downstream of the diffusion dryer of the VACES line sampling fine PM by means of the SMPS. These experiments were conducted at a minor flow rate of 20 l min^{-1}

(thus the ideal concentration enrichment is by a factor of 11). Each experiment started by first measuring the ambient particle number concentration by means of the TSI 3022 condensation particle counter for 5 min. Subsequently, the concentration immediately downstream of the 0.18 μm impactor was measured for an additional 5 min, followed by a concentration measurement downstream of the ultrafine VACES concentrator for 5 min. The above cycle was repeated three times in each experiment.

The results of Fig. 8 show categorically that size distribution was fairly well preserved during the concentration enrichment process, as the number median diameters (41 nm) and geometric standard deviation (1.7) of the concentrated and ambient aerosols are virtually identical. These results confirm that drying by diffusion returns the concentrated droplets to their original size with minimal distortion.

4. Summary and conclusions

This study focused on the development and laboratory evaluation of a versatile particle concentration enrichment system (VACES), capable of enriching concurrently the concentration of ambient coarse, fine and ultrafine particles by a factor up to 30, either suspended in air or collected into any liquid suitable for in vitro tests. The latter is accomplished by connecting the concentrated output (minor) flows of each of the VACES parallel concentrators to a liquid impinger (BioSampler).

A high flow rate, low pressure drop multi-slit nozzle impactor was developed as size-selective inlet to remove all but ultrafine particles. The 50% cutpoint was about 0.18 μm and the collection efficiencies for particles larger than 0.3 μm was more than 95%. The comparison in ultrafine mass concentrations obtained using the multi-slit impactor and a reference MOUDI using indoor aerosols showed excellent agreement between the two samplers.

The BioSampler was tested at a reduced flow rate and with as small volume of water as possible. The pressure drop across the BioSampler decreased from 145 in of H_2O (36,100 Pa) to less than 20 in of H_2O (4980 Pa) as the flow rate was reduced from 12.5 to 5 l min^{-1}. This low pressure drop across the sampler becomes important especially in collecting semi-volatile species such as ammonium nitrate or semi-volatile organic compounds, both significant constituents of Los Angeles $PM_{2.5}$. The collection efficiency of the BioSampler at the reduced flow rate was still high enough to collect particles larger than 2 μm. In addition, the collection efficiency of BioSampler was not influenced by the amount of water in the sampler's reservoir, thereby making it possible to achieve highly concentrated suspensions for use in in vitro studies.

The ability of the VACES to concentrate particles was first tested in laboratory experiments using different type of particles in the size range of 0.05–1.9 μm and at three minor flow rates of two 7, 10, and 20 l min^{-1} with the total intake flow rate of 220 l min^{-1}. The enrichment factors based on number concentrations were close to the ideal values. Hygroscopic aerosols, such as ammonium sulfate and ammonium nitrate were concentrated as effectively as hydrophobic PSL particles.

The experimental characterization of the VACES demonstrated that the concentration enrichment does not depend on particle size or chemical composition. Volatile species such

as ammonium nitrate are preserved through the concentration enrichment process under the laboratory conditions used in this study.

In summary, the VACES concentrated ambient aerosols with a very high efficiency and without any substantial distortion in their physico-chemical characteristics. One of the most important features of the VACES is the ability to concurrently concentrate all three major PM size modes and to provide them either suspended in air or collected into water. This enables toxicologists to simultaneously conduct in vivo and in vitro evaluations of toxic effects of ambient particles.

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