Inhalation Toxicology, 16:851–862, 2004
Copyright © Taylor & Francis Inc.
ISSN: 0895-8378 print / 1091-7691 online
DOI: 10.1080/08958370490506664

Taylor & Francis healthsciences

Development and Performance Evaluation of a High-Volume Ultrafine Particle Concentrator for Inhalation Toxicological Studies

Tarun Gupta, Philip Demokritou, Petros Koutrakis
Exposure Epidemiology and Risk Program, School of Public Health, Harvard University, Boston, Massachusetts, USA

This article presents the development and performance evaluation of a high-volume ultrafine particle concentrator. The ultrafine particle concentrator consists of several units, including a size-selective inlet; a condensational growth unit; a series of two virtual impactors (concentrators); a thermal size restoration device; an air cooler; and a size-selective outlet. Ambient ultrafine particles are condensationally grown to supermicrometer sizes and then are concentrated by a factor of 40 to 50 using a two-stage virtual impactor. Subsequently, ultrafine particle size distribution is restored, using a thermal method. The Harvard ultrafine concentrated ambient particle system (HUCAPS) delivers 58 lpm of concentrated aerosol that can be used for in vivo or in vitro inhalation toxicological studies. Overall, pressure drop through the system is only 2.2 kPa, which is adequately low for inhalation toxicological exposure tests. The performance of this system was evaluated using single-component artificial aerosols with a variety of physicochemical properties as well as ambient air. These experiments showed that for an optimum supersaturation ratio of 3.0, all ultrafine particles grow and get concentrated by about the same enrichment factor, regardless of their composition and surface properties.

Numerous epidemiological studies have shown that particulate matter (PM) and especially its $PM_{2.5}$ (d_p [aerodynamic diameter] $< 2.5\ \mu$m) mass is associated with respiratory and cardiovascular adverse health effects, including premature mortality, asthma exacerbation, decreased lung function, and increased risk of myocardial infraction among others (Schwartz & Dockery, 1992; Dockery et al., 1993; Dockery & Pope, 1994; Peters et al., 2000; Gamble, 1998; Pope et al., 1999).

Ultrafine particles ($d_p < 0.1\ \mu$m) are formed through gas-to-particle conversion mechanisms and are associated with both anthropogenic and natural sources (Whitby & Svendrup, 1980). Inhaled ultrafine particles deposit onto the respiratory tract almost exclusively by diffusional mechanisms (ICRP, 1994). Inhalation of fumes, consisting mainly of ultrafine particles, leads to the well-known effects of metal or polymer fume fever (Drinker et al., 1927; Gordon et al., 1992). The pulmonary toxicity of ultrafine particles has been also demonstrated in several controlled laboratory exposure studies using simple particle models (Oberdorster et al., 1994; Johnston et al., 2000). The particles used in these studies were artificially generated, single-chemical-component particles, or particles collected from a single source such as diesel exhaust. Therefore, these exposures may not adequately represent the heterogeneous mixture of actual ambient particles. The various components of ambient particles may also synergistically interact to produce toxic health effects, not seen with artificially generated atmospheres. It was also shown in studies that the oxidative stress of real particles differs significantly from that of the artificially generated particles (Lippmann, 1989).

Despite the tremendous progress that has been made over the last few years in understanding the biological mechanisms of PM, there is a need to further examine the toxicological properties of ambient ultrafine particles in controlled inhalation chamber studies. Ambient particle concentrators based on virtual (inertial) impaction technology have been used successfully during recent years to conduct in vivo animal and human inhalation exposures to concentrated accumulation mode ($PM_{0.1–2.5}$) and coarse mode particles ($PM_{2.5–10}$) (Sioutas et al., 1995; Godleski et al., 1996; Clarke et al., 2000; Demokritou

Received 10 March 2004; accepted 7 July 2004.
This experimental study was supported by the U.S. EPA/Harvard Particle Health Effects Center (grant R827 353-01-0) and the NIEHS Harvard Center (grant ES 0002). Also, we thank Dr. Mike Wolfson and Stephen Ferguson for their helpful comments and technical support.
Address correspondence to Philip Demokritou, PhD, Exposure Epidemiology and Risk Program, Department of Environmental Health, School of Public Health, Harvard University, 401 Park Drive, Box 15677, Landmark Center-West, Boston, MA 02215, USA. E-mail: pdemokri@hsph.harvard.edu

et al., 2003). However, virtual impaction technology cannot be applied to separate and concentrate ultrafine particles because adequate inertia cannot be induced without supersonic flows and excessive pressure drops. Subjecting the ultrafine particles to such a substantial vacuum will potentially cause an unacceptable amount of volatilization of labile species. In addition, such a substantial vacuum at the chamber side is not practical for inhalation studies. Previous studies have used condensational growth to enlarge ultrafine particles above 1 μm using water vapor as a condensing medium in conjunction with virtual impaction technology as a method to concentrate ultrafine particles (Sioutas & Koutrakis, 1996; Sioutas et al., 1999; Kim et al., 2000). Grown droplets are separated from the majority of the surrounding air without being subjected to excessively low pressures. However, these systems are restricted by a low volumetric output flow (only a few liters per minute, lpm), uncontrolled supersaturation, and insufficient concentration enrichment of ultrafine particles. In order to conduct any meaningful dosimetric analysis, output aerosol flows of 30–50 lpm and concentrated by about 30–50 times are required for in vivo inhalation toxicological studies.

In this article, we present the development and performance evaluation of a high-volume ultrafine particle concentrator, which can deliver about 58 lpm of highly concentrated (~40–50 times) aerosol for inhalation toxicological exposure studies.

METHODS

Description of the System

The Harvard ultrafine concentrated ambient particle system (HUCAPS) is shown in Figure 1. The system operates at an input airflow of 5000 lpm and delivers 58 lpm of highly concentrated ambient ultrafine aerosol. Ambient aerosol is drawn through a size-selective inlet where particles larger than 2.5 μm are removed. Subsequently, it is drawn sequentially through the saturator and condenser (supersaturator), where saturation and supersaturation conditions are obtained, respectively. After ultrafine particles grow to supermicrometer sizes, the aerosol is drawn through a series of two virtual impactors to increase their concentration levels. After concentration enrichment, the original size distribution of ambient ultrafine particles is restored in the thermal size restoration device. The concentrated aerosol is cooled down to room conditions as it passes through a conditioner, and then particles larger than 0.2 μm are removed by inertial impaction in a size selective outlet. Finally, the concentrated aerosol is introduced into the inhalation exposure chamber. The main components of HUCAPS are described in detail next.

Size-selective inlet. This high-volume (5000 lpm) size-selective inlet is a two-stage conventional inertial cascade impactor, with the first stage removing particles larger than 10 μm and the second stage removing particles larger than 2.5 μm from the aerosol stream (Figure 1). A PM_{10} stage upstream of $PM_{2.5}$ stage is used to take care of the high particle mass loading possible at different ambient locations. Both stages 1 and 2 are rectangular slit impactors and use polyurethane foam (PUF) as an impaction substrate. The use of a PUF impaction substrate instead of a rigid flat-surface impaction substrate makes it possible to collect, without using any adhesives, relatively large amounts of particles (milligram to gram levels) with minimum particle bounce and reentrainment (Kavouras & Koutrakis, 2001; Demokritou et al., 2002b, 2002c). The physical and theoretical characteristics of the size-selective inlet stages are presented in Table 1.

The experimental setup used for the performance evaluation of the size-selective inlet is shown in Figure 2a. The two impactor stages were tested separately. A fluidized-bed aerosol generator (3400A, TSI, Inc., St. Paul, MN) was used to generate artificial polydisperse aerosol using Al_2O_3 spheres (nominal size 2–20 μm, density = 3.97 g/cm^3). The generated aerosol was introduced in a duct and mixed with room air. Particle number concentration and size distribution were measured upstream and downstream of the impactor (alternating sequentially) using an aerodynamic particle sizer (APS, model 3321, TSI, Inc., St. Paul, MN). The sampling probes were isokinetically designed. Each APS measurement lasted for 2 min, and the reported results are based on the average of at least 5 consecutive (alternating) tests. The relative standard deviation of the average for each particle size was less than 5%. Flow was monitored using a calibrated orifice and was drawn using a vacuum pump (MFG Corp., Benton Harbor, MI).

Condensational growth unit. The condensational growth unit consists of two main components: saturator and condenser (supersaturator) (Figure 1). The saturator is a 1.5 m long, 25 cm × 35 cm rectangular duct. A steam injection tube is located inside the saturator, near the entrance. As the air enters the saturator, steam is injected and mixed with the air. The flow inside the saturator is turbulent to maximize steam mixing. Steam is generated, at atmospheric pressure, using a feedback-controlled electric humidifier (electric steam generator, steam capacity = 14.2 kg/h, model ESDDR-11, PURE). Only a few grams of steam (~70 g/min, ~100°C) are mixed with huge amount of inlet air (~6000 g/min, ~25°C). Also, the mixing time is quite short (a few milliseconds). These conditions were chosen to limit the potential impact of the high temperature to a very small fraction of the particles. The sample air exits the saturator at close to saturation conditions and with a temperature increase of less than 2°C. Subsequently, the saturated sample air enters the condenser (supersaturator) where it cools down to achieve the target supersaturation condition. While the HUCAPS can produce supersaturation ratios (Sr) of up to 5, supersaturation ratio in the range of 2.9–3.2 is sufficient (Demokritou et al., 2002a; Gupta et al., 2003).

The condenser is a coolant-to-air heat exchanger (HX-100 Doyle and Roth single pass shell and tube air heat exchanger), with the air flowing through the tubes and the coolant through the shell side. Airflow through each tube is laminar to minimize particle losses. There is an external refrigeration unit (chiller),

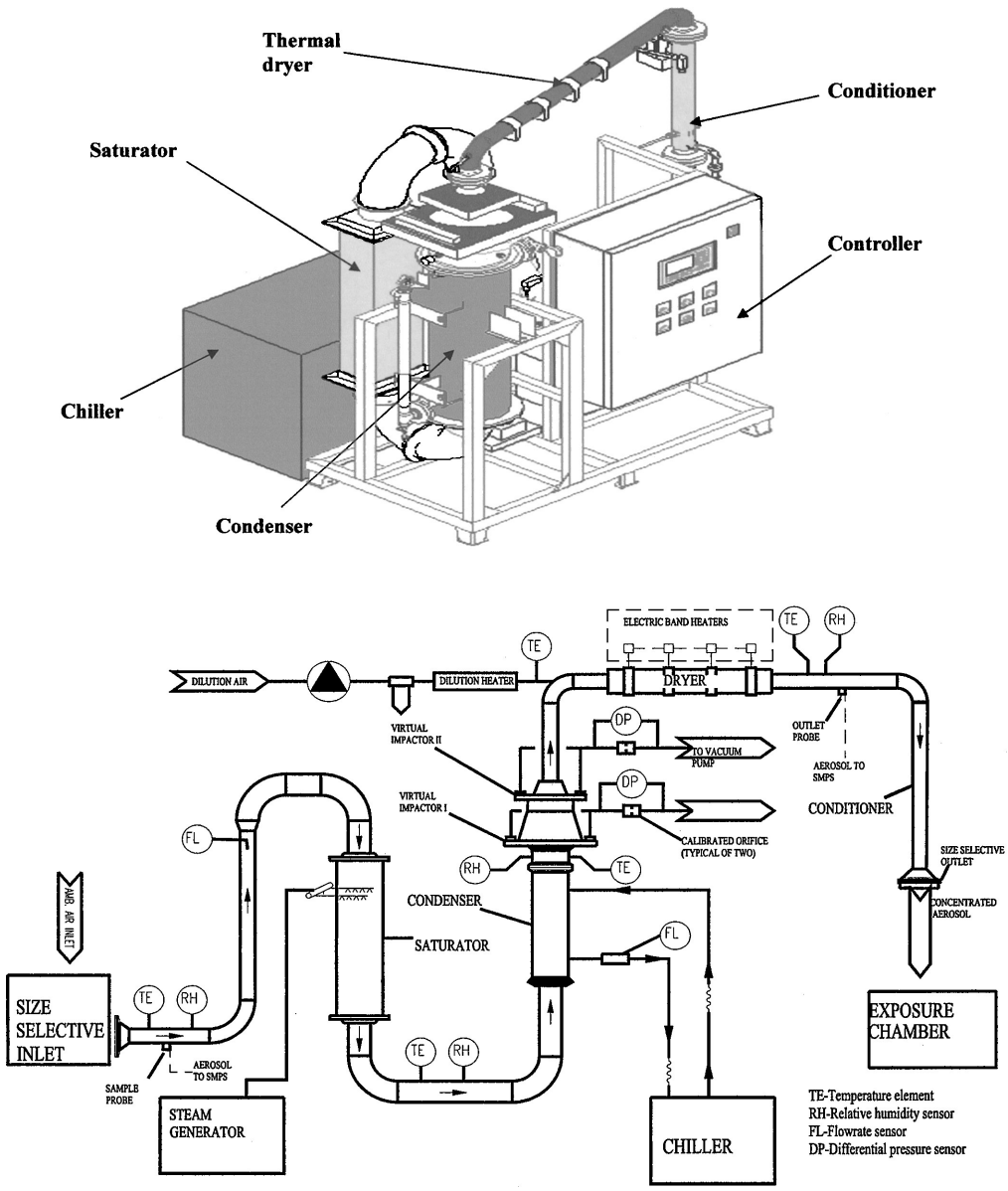


FIG. 1. The Harvard ultrafine concentrated ambient particle system (HUCAPS).

which is used to cool down the coolant (glycol–water solution) required in the condenser (Figure 1). The coolant temperature is adjustable and can be used to adjust the aerosol temperature (hence supersaturation ratio) inside the condenser. The system is fully automated. A computerized system is used to monitor and control all the operational parameters of the HUCAPS, including relative humidity, air temperature, refrigerant temperature, supersaturation ratio, and airflow in every section of the system.

Virtual impactors. Particles grown by condensation are subsequently drawn through a series of two virtual impactors (stages I and II) (Figure 1). Virtual inertial impactors are often used to separate and concentrate particles of a desired particle size range. In a virtual impactor, the inlet flow, Q_0, is typically divided into two flow streams, with the major flow, Q_M, carrying most of the particles smaller than a distinct cutpoint, and the minor flow, Q_m, carrying most of the particles above the cutpoint, together with a small fraction of the smaller particles. Particles larger than the impactor cutpoint follow the relatively straight minor flow, while particles smaller than the impactor cutpoint follow the deflected air streamlines of the major flow. As a result, particles with sizes above the impactor cutpoint are collected in the minor flow and are concentrated by a nominal factor of Q_0/Q_m, while in the minor flow, the concentrations of particles with sizes below the impactor cutpoint remain at their original values.

TABLE 1
Physical characteristics and experimental results for size-selective inlet and outlet for HUCAPS

	Size-selective inlet		Size-selective outlet	
Stage	PM_{10}	$PM_{2.5}$	A	B
Physical characteristics				
L^a (cm)	106.7	167.6	2.3	3.1
W^b(cm)	1.27	0.25	0.06	0.06
S^c (cm)	1.27	0.25	0.06	0.06
S/W	1.0	1.0	1.0	1.0
Theoretical characteristics				
Q^d (lpm)	5000	5000	58	58
Re	10400	6618	5596	4220
Experimental results				
$d_{50}^e(\mu m)$	10.0	2.6	0.2	0.3
s^f	1.47	1.65	1.68	1.52
$\sqrt{Stk}^g$	0.55	0.56	0.24	0.29
ΔP^h (Pa)	24.9	189.3	3612	1993

[a]Nozzle length.
[b]Nozzle width.
[c]Substrate to nozzle distance.
[d]Volumetric flow rate.
[e]Particle size corresponding to a 50% collection efficiency.
[f]Collection efficiency curve sharpness.
[g]Square root of Stokes number.
[h]Pressure drop.

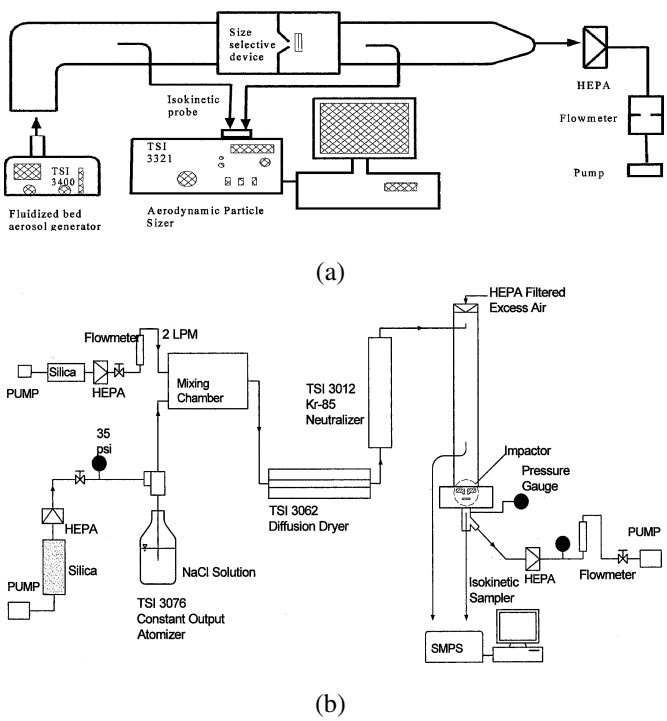


FIG. 2. Schematics for experimental characterization of size-selective inlet and outlet. (a) Experimental setup for characterization of size-selective inlet. (b) Experimental setup for characterization of size-selective outlet.

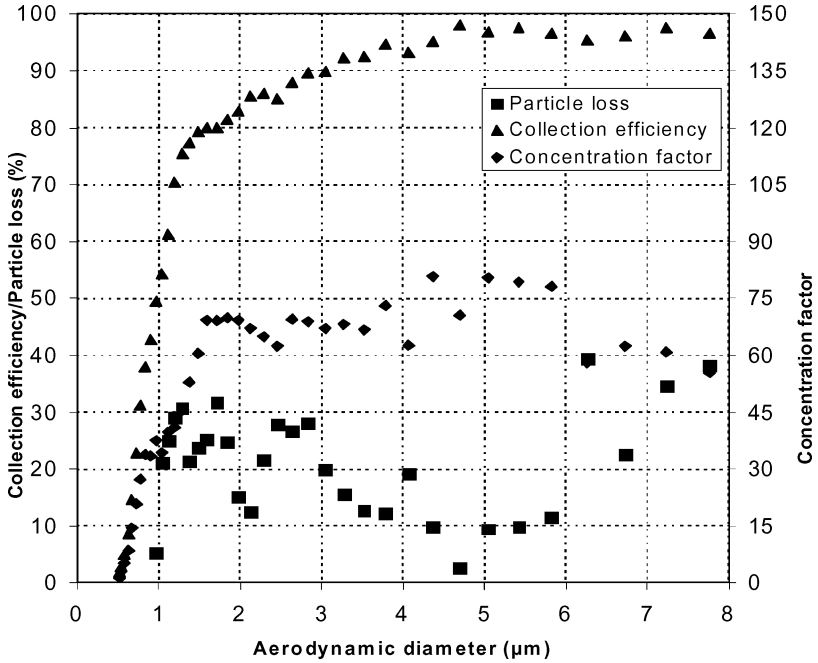
The high-volume multislit nozzle virtual impactors used in the HUCAPS consist of two main components, the acceleration nozzles and collection nozzles. The experimental setup and results from the characterization of stages I and II have been reported previously (Demokritou et al., 2003). Table 2 summarizes the geometrical characteristics and operational parameters for both virtual impactors (stages I and II). The overall cutpoint of the combined two stages is approximately 1.0 μm. Pressure drop through the combined two stages is only 224 Pa.

Thermal dilution–dryer section. Restoring the size distribution of the condensationally grown ultrafine particles while maintaining their chemical composition is of great importance for the performance of the system in inhalation toxicological studies. In the past, diffusion dryers have been used to dry grown ultrafine particles (Kim et al., 2000). Major problems associated with the use of diffusion dryers include saturation of the desiccant in a relatively short period of time, and extremely low aerosol flow capacity (<3 lpm). Recently, a thermal method was employed to restore particle size distribution for a prototype ultrafine particle concentrator (Demokritou et al., 2002a). Heating up the aerosol stream provides the necessary latent heat needed to evaporate the water from the particles. By raising the air temperature, an increase in the water vapor holding capacity of the air is obtained. It was shown that the performance of this thermal method depends on particle concentration, aerosol temperature, and residence time (aerosol flow) (Demokritou et al., 2002a).

TABLE 2

Operational parameters for virtual impactors of HUCAPS

Length of acceleration slit (cm)	Reynolds number	Pressure drop: Minor flow (Pa)	Pressure drop: Major flow (Pa)	r^c (%)	d_{50}^f (μm)
Stage I (inlet flow = 5000 lpm)					
111.8	9928	149.4	4483.8	10	0.9
111.8	9928	87.2	4608.4	5	1.1
111.8	9928	49.8	4732.9	2.5	1.4
Stage II (inlet flow = 500 lpm)					
32.8	3386	24.9	523.1	10	1.7
32.8	3386	5.0	548.0	5	2.0
32.8	3386	5.0	597.8	2.5	2.3



Combined Stage I and II (System inlet flow = 5000 lpm)

Virtual impactor stage I: Q_M^a (lpm)	Virtual impactor stage I: Q_m^b (lpm)	Virtual impactor stage I: Re	Virtual impactor stage I: r^c (%)	Virtual impactor stage II: Q_M^a (lpm)	Virtual impactor stage II: Q_m^b (lpm)	Virtual impactor stage II: Re	Virtual impactor stage II: r^c (%)	System pressure drop^d (Pa)	CF_{avg}^e	d_{50}^f (μm)
4500	500	9928	10	450	50	3386	10	224.2	57	1.0

[a] Virtual impactor major flow.
[b] Virtual impactor minor flow.
[c] Ratio of minor to total flow.
[d] Pressure drop measured at the exit (minor flow) of virtual impactor stage II.
[e] Average concentration factor for 1–10 μm aerodynamic particle size range.
[f] Particle size corresponding to a 50% collection efficiency.

In order to minimize the semivolatile component losses, the aerosol temperature inside the thermal dryer should be as close to ambient conditions as possible. For this system it was kept below 35°C. There is an equilibrium partition (dependent on temperature, composition of particles, and total particle surface area, as well as other variables) of semivolatile organic species between gas and particle phases in ambient air. For simplicity, and thus ignoring effects of humidity and temperature changes during the concentration process, increasing the concentration of particles leads to a larger fraction of the total amount (gas plus particle phase) of these semivolatiles in the particle phase. The particles must be heated from about 25°C to about 35°C for a relatively short period of time, with the consequence that a certain amount of the semivolatiles are shifted from the particle to the gas phase. Assume that a new equilibrium is achieved when the concentrated particles are heated to 35°C (it is not easy to calculate whether the rates of volatilization for the typical semivolatile organic species are high enough to come close to equilibrium during the transition through the heated dryer). If we also assume that the partition factor (gas phase vs. particle phase) increases by a factor of two with a 10°C increase in temperature, only about 2–3% of the particle-phase semivolatiles need to go to the gas phase, since the absolute ratio of gas-phase to particle-phase semivolatiles has decreased by a factor of 40–50. Hence with only a few percent of the particle-phase semivolatiles of the concentrated particles likely to volatilize, there is little likelihood that the relative losses from the smaller particles relative to the larger sizes will have any significant impact on the biological effects.

Considering these factors it was decided, in addition to heating up the concentrated aerosol, to also dilute it with a small air flow (8 lpm) of preheated, particle-free (filtered) air for achieving rapid and comprehensive drying of grown droplets. Figure 1 also shows the basic setup of this thermal dilution–dryer system that was used in the HUCAPS to restore the particle size distribution. The preheated dilution air at a temperature ~120°C was introduced via an isokinetic, concentric tube into the duct (ID 7.6 cm, length 1.5 m) (Figure 1) and rapidly mixed with the incoming concentrated aerosol from the concentrators. After mixing, the aerosol is heated up further to ~35°C in the dryer, using electric band heaters, to evaporate the water from the particles.

The final temperature of the outlet aerosol depends not only on the temperature and amount of dilution air but also on the dryer temperature. Various tests were performed to study the effects of changes in dilution airflow, dilution air temperature, and dryer temperature on the restoration of ambient ultrafine particle size distributions. A major technical challenge was to restore the size distribution as close as possible to the original ambient ultrafine particle size distribution with minimum particle loss, and using a minimum amount of dilution air. Aerosol temperatures at both the entrance and exit of the dryer were monitored and controlled. The size distribution of particles at the exit of the thermal dryer was measured using a scanning mobility particle sizer (SMPS, TSI, Inc., St. Paul, MN). Each SMPS measurement was 3 min long. Aerosol flow through the dryer was also measured. In addition, the thermal dilution–dryer assembly was separately tested to make sure that the dilution air is particle free.

Conditioner (an air cooler). The concentrated aerosol is then cooled down to room conditions (~20°C and 50% relative humidity, RH) as it passes through the conditioner section. The conditioner is composed of two concentric tubes. The outer tube carries chilled water and the inner tube supplies the concentrated aerosol from the thermal dilution–dryer section to the size-selective outlet. Adjusting the flow of chilled water in the outer tube controls the aerosol temperature.

Size-selective outlet. The cooled-down concentrated stream of aerosol is then passed through a size-selective outlet before it is supplied to the inhalation exposure chamber. The size-selective outlet is designed to remove accumulation mode particles (2.5 μm $\geq d_p \geq$ 0.2 μm). The size-selective outlet has a pressure drop ~2 kPa. In order to minimize the time for which the aerosol remains under the low-pressure conditions, the size-selective outlet was placed downstream of the conditioner rather than upstream along with the two size-selective inlet stages. It is based on a slit-nozzle inertial impactor technology (Demokritou et al., 2002b, 2002c). Again PUF is used as an impaction substrate. The physical characteristics of the two available configurations (A and B) of size-selective outlet are presented in Table 1.

The experimental setup used for characterization of size-selective outlet is shown in Figure 2b. Two different acceleration nozzle configurations (A and B) were developed and each one of them was characterized individually. Polydisperse aerosol (nominal size 10–400 nm, density = 2.17 g/cm^3) was generated from 1% aqueous solution of sodium chloride using a constant-output atomizer (nanoparticle generator, model 3076, TSI, Inc., St. Paul, MN). After passing through the charge neutralizer, the generated aerosol was then introduced at the top of a duct. Particle number concentration and size distribution were measured upstream and downstream of the impactor (alternating sequentially) using an SMPS. Each SMPS measurement lasted for 3 min, and the reported results are based on the average of at least 5 consecutive tests. The relative standard deviation of the average for each particle size was less than 5%. The pressure drop was measured using a differential pressure gauge.

Experimental Setup

It is important to note that this article gives an overall presentation of the HUCAPS, whereas our other studies test part of the system and present specific findings in detail. For instance, Gupta et al. (2003) present a parametric investigation of the effect of the supersaturation ratio on the performance of HUCAPS and critically assess the relationship between particle hygroscopicity, growth and enrichment.

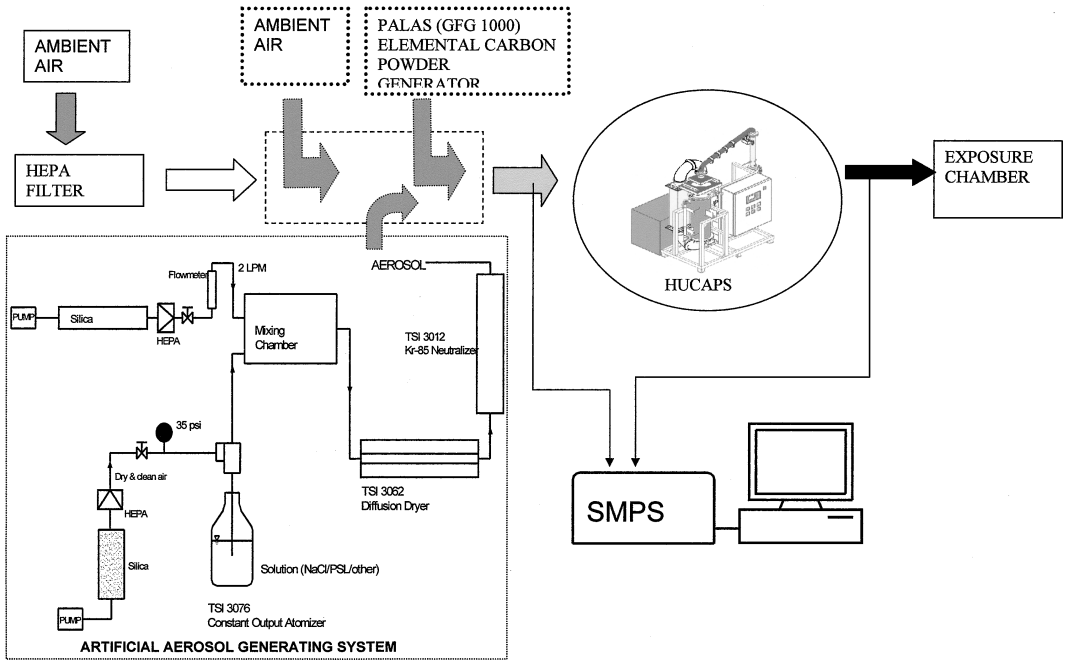


FIG. 3. Experimental setup for laboratory performance evaluation of HUCAPS.

Characterization of HUCAPS Using Aerosols of Different Physicochemical Properties

Figure 3 shows the basic experimental setup used to conduct a series of performance evaluation tests on the HUCAPS. For all these experiments, HUCAPS was operated at its optimum conditions with a supersaturation ratio of about 3.0. Aerosols with different chemical composition and hygroscopicity (sodium chloride, potassium sulfate, polystyrene latex (PSL), and elemental carbon) were generated. The HUCAPS performance was tested with each type of artificial aerosol separately.

Sodium chloride, potassium sulfate, and PSL polydisperse aerosols (count median diameter ~45–60 nm, GSD ~1.7) were generated from aqueous solutions/suspensions using a constant output atomizer (nanoparticle generator, model 3076, TSI, Inc., St. Paul, MN). The aerosol was passed through a Kr^{85} charge neutralizer to bring charges to the Boltzmann equilibrium distribution. Prior to its entry into the HUCAPS, the aerosol was introduced into a duct where it was mixed with HEPA-filtered room air.

Figure 3 also shows an elemental carbon powder generator (PALAS GFG 1000, Karlsruhe, Germany). This aerosol generator was used to generate elemental carbon particles with a count median diameter of 40–45 nm and GSD ~1.5, using electric spark discharge of carbon graphite electrodes in argon gas.

To evaluate the system's ability to increase the concentration and restore the size distribution of the ultrafine particles, particle number concentration and size distribution were measured upstream and downstream of the HUCAPS (alternating sequentially) using an SMPS. Each SMPS measurement was 3 min long. About five sets of upstream and downstream measurements were recorded for each type of aerosol during the entire duration of the experiments (~3–4 h). The concentration enrichment factor (CF), defined as the ratio of the outlet (downstream) to sample (upstream) aerosol number concentration, was also determined for these experiments.

Characterization of HUCAPS Using Ambient Air

Finally, ambient air was used as the test aerosol for the overall performance evaluation of the HUCAPS. The particle number concentration and size distribution were measured, at the entrance and exit of the HUCAPS, for 3-min intervals each using the SMPS. The performance of the HUCAPS was monitored (and optimized) during several months of operation. The effects of changes in the composition, size distribution, and number concentration of ambient air on the HUCAPS performance were examined. Also, the ability of HUCAPS to restore the original ambient ultrafine particle size distribution was carefully monitored. For this, the percentage differences of the mean, modal, and median mobility diameters and geometric standard deviation (GSD) between the ambient and concentrated aerosol size distributions, averaged over the entire duration of the experiment, were calculated for each measurement day. These four parameters and the average CF were calculated for the size range of 20–300 nm particle mobility diameter using the following

formulas:

$$\text{Average mean}_{\text{diff}} = \left(\frac{\text{Mean}_{\text{avg}}^{\text{conc}} - \text{Mean}_{\text{avg}}^{\text{amb}}}{\text{Mean}_{\text{avg}}^{\text{amb}}}\right) \times 100 \quad [1]$$

$$\text{Average mode}_{\text{diff}} = \left(\frac{\text{Mode}_{\text{avg}}^{\text{conc}} - \text{Mode}_{\text{avg}}^{\text{amb}}}{\text{Mode}_{\text{avg}}^{\text{amb}}}\right) \times 100 \quad [2]$$

$$\text{Average median}_{\text{diff}} = \left(\frac{\text{Median}_{\text{avg}}^{\text{conc}} - \text{Median}_{\text{avg}}^{\text{amb}}}{\text{Median}_{\text{avg}}^{\text{amb}}}\right) \times 100 \quad [3]$$

$$\text{Average GSD}_{\text{diff}} = \left(\frac{\text{GSD}_{\text{avg}}^{\text{conc}} - \text{GSD}_{\text{avg}}^{\text{amb}}}{\text{GSD}_{\text{avg}}^{\text{amb}}}\right) \times 100 \quad [4]$$

where $\text{Mean}_{\text{avg}}^{\text{amb}}$ and $\text{Mean}_{\text{avg}}^{\text{conc}}$ are the average of the mean mobility diameters of the ambient and concentrated aerosol size distributions, respectively, measured for the entire duration of the experiment. The terms in Eqs. (2), (3), and (4) are defined in a similar fashion.

RESULTS AND DISCUSSION

Characterization of Size-Selective Inlet and Outlet

Figure 4 shows the results from the characterization of the PM_{10} and $PM_{2.5}$ stages of the size-selective inlet. The experimentally calculated cutpoint (d_{50}, μm), the collection efficiency sharpness (s), and the pressure drop (ΔP, kPa) are presented in Table 1. The PM_{10} inlet is designed to remove large particles ($d_p > 10$ μm). The experimentally determined cutpoint is 10.0 μm, which corresponds to $\sqrt{\text{Stk}} = 0.55$ (where Stk is the Stokes number). The experimentally determined cutpoint of the $PM_{2.5}$ inlet is 2.6 μm ($\sqrt{\text{Stk}} = 0.56$) (Table 1). It is worth mentioning that the experimentally determined cutpoints of these stages are smaller than that theoretically calculated for a flat rigid surface impaction substrate. The cutpoints are similar to those obtained for impactors using polyurethane foam as an impaction substrate (Demokritou et al., 2002b, 2002c). By using PUF, lower cutpoints are obtained even though particles are accelerated to similar velocity as when using a flat rigid surface. Hence, this results in comparatively lower pressure drops for these stages, which is another advantage of using PUF substrates. This cutpoint decrease has been explained by penetration of some air streamlines into the porous polyurethane foam surface (Kavouras & Koutrakis, 2001).

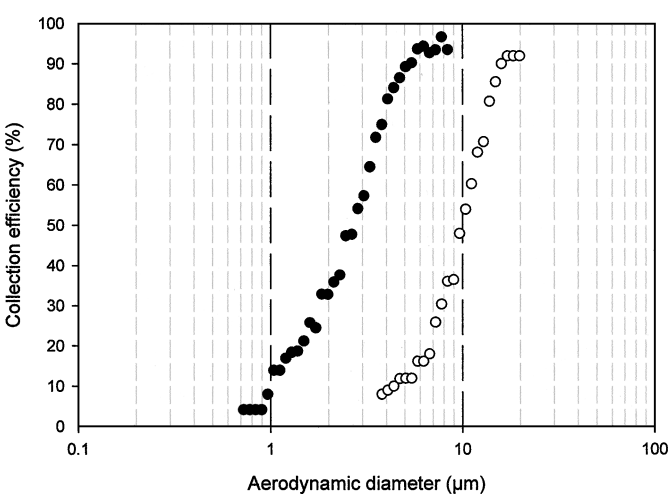


FIG. 4. Collection efficiency curves of PM_{10} and $PM_{2.5}$ size-selective inlets.

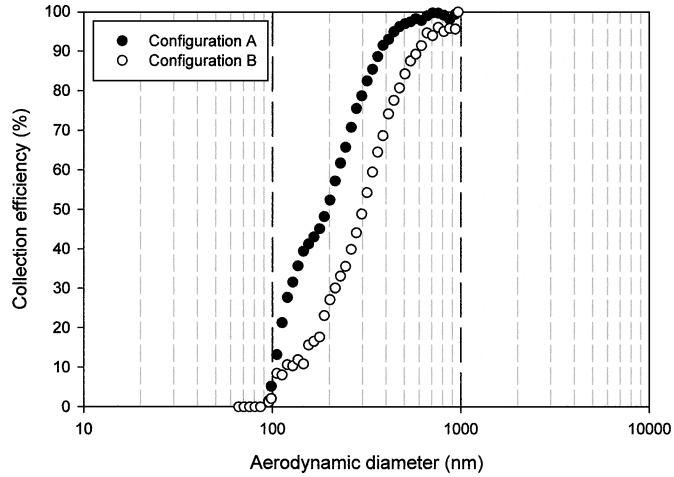


FIG. 5. Collection efficiency curves for the two configurations of size-selective outlet.

Figure 5 shows the relationship between collection efficiency and the aerodynamic diameter for the two different configurations of the size-selective outlet. The experimentally determined cutpoint for configuration A ($L = 2.29$ cm) is 0.2 μm, for which the pressure drop is 3.6 kPa (Table 1). The cutpoint for configuration B ($L = 3.05$ cm) is 0.3 μm with a net pressure drop of 2.0 kPa. The advantage of configuration B is the lower pressure drop with correspondingly lower losses for semivolatile aerosol components. No matter which configuration is used, PUF has shown excellent retention of particle collection properties even under very high particle mass loading conditions (Demokritou et al., 2004).

Particle Losses in the HUCAPS in Absence of Growth

Figure 6 depicts the measured ultrafine particle losses, as a function of the particle size, in the absence of condensational growth process. This was tested by passing the air sample through the entire HUCAPS without introducing steam into the system. The background ultrafine particle losses throughout the HUCAPS were on average <10% and were about the same for the entire ultrafine particle size range. Hence, changes in particle size distribution due to internal losses are expected to be minimal.

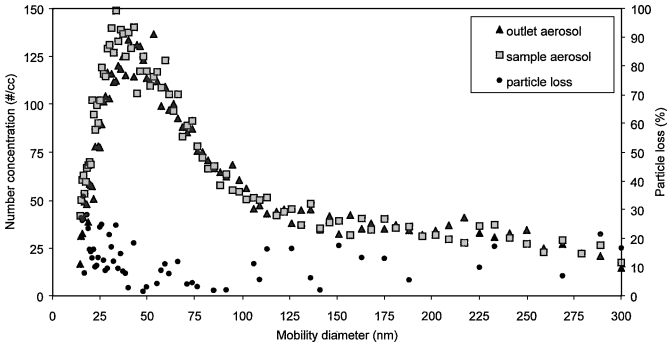


FIG. 6. Particle losses as a function of mobility diameter for the HUCAPS in the absence of condensational growth.

Best Conditions for Size Distribution Restoration

In order to obtain the best possible conditions for size distribution restoration, various tests were performed with varying dilution airflow rates, dilution air temperatures and dryer temperatures. Table 3 presents selected results from a series of experiments carried out using ambient air to fully understand the size distribution restoration process. For all these experiments a constant flow of 50 lpm of concentrated aerosol entered the thermal dilution–dryer section. Table 3(a) shows the results when a fixed amount (25 lpm) of dilution air was used, and the temperature of the dilution air was varied using the dilution–heater. However, no heating in the dryer section was employed. Increase in temperature of dilution air led to an increase in the final outlet aerosol temperature and in partial restoration of the size distribution. There was a steady increase in the overall concentration enrichment factor with increase in dilution air temperature which can be attributed to the fact that partially restored ultrafine particles are less prone to sampling losses as compared to grown ultrafine particles.

Table 3(b) shows the results when a fixed amount of dilution air (25 lpm), heated to a fixed temperature (138°C) was used. The temperature of the dryer section was varied to ascertain its effect on the restoration of outlet aerosol size distribution. A significant shift in the outlet aerosol size distribution was noticed when dryer temperature increased. The size distribution restoration process is more sensitive to increasing dryer temperature as compared to increasing dilution air temperature. One explanation for this may be that not only the dilution air but also the total volume of the outlet aerosol was heated in the dryer section, which led to complete particle size restoration. Also, a substantial amount of heat is lost to the walls of the thermal dryer section.

The set of experiments just described resulted in an outlet flow of 75 lpm with an increase of ultrafine aerosol concentration by a factor of up to 25. However, for toxicological exposure studies at least a CF of about 40 is required, and thus we were limited by the amount of dilution air that could be used for the size restoration process. Further experiments showed that the best possible thermal conditions for the size distribution restoration were: dilution airflow = 8 lpm, dilution air temperature ($T_{\text{dil-flow}}$) ~120°C, total outlet flow = 58 lpm, and outlet air temperature (T_{outlet}) ~35°C. Minor adjustments of these parameters were required depending on day-to-day variations in the number concentration and size distribution (composition) of ambient air in Boston.

TABLE 3
Selected results showing the effect of dilution and heating on the size-distribution restoration

Dilution airflow (lpm)	Total outlet airflow (lpm)	Dilution air temperature (°C)	Outlet air temperature (°C)	Number concentration (#/cm^3)	Aerosol	CF	Mobility dilameter Mean (nm)	Mode (nm)	Median (nm)
(a) Effect of variation on dilution air temperature									
25	75	22.8	22.8	70,900	Outlet	26	118	122	110
25	75	43.9	24.4	64,500	Outlet	24	115	113	104
25	75	54.4	25.6	73,800	Outlet	27	113	113	103
25	75	82.2	27.8	92,000	Outlet	34	107	106	101
25	75	105.6	28.9	100,000	Outlet	37	104	82	87
				2710	Sample		93	27	69
(b) Effect of variation on outlet (dryer) air temperature									
25	75	137.8	32.8	126,000	Outlet	31	91	74	80
25	75	137.8	33.3	116,000	Outlet	28	88	71	77
25	75	137.8	33.9	98,800	Outlet	24	85	64	74
25	75	137.8	35.0	110,000	Outlet	27	78	64	68
25	75	137.8	35.6	105,000	Outlet	26	78	62	68
				4100	Sample		71	61	65

Note: Sr = 3.0, always a constant flow of 50 lpm of concentrated aerosol entering the thermal dilution–dryer section.

TABLE 4
Concentration enrichment factors for different aerosol types studied at Sr = 3.0

Aerosol composition	Sample aerosol number concentration (#/cm^3)	Concentrated aerosol number concentration (#/cm^3)	Concentration enrichment factor
Sodium chloride	3050 ± 50	$125{,}000 \pm 7500$	42 ± 3 ($n = 5$)
Potassium sulfate	1980 ± 30	$76{,}200 \pm 6000$	39 ± 3 ($n = 5$)
Polystyrene latex (PSL)	1120 ± 10	$41{,}000 \pm 2400$	36 ± 3 ($n = 5$)
Elemental carbon	$30{,}300 \pm 2000$	$1{,}015{,}000 \pm 48{,}000$	34 ± 4 ($n = 5$)
Outdoor air	1710 ± 50	$72{,}517 \pm 3000$	42 ± 4 ($n = 5$)
Indoor air	4350 ± 100	$169{,}000 \pm 7000$	37 ± 3 ($n = 5$)

Note: Dilution flow = 8 lpm, $T_{\text{dil-flow}}$ ~121°C, total outlet flow = 58 lpm, T_{outlet} ~35°C, n = pairs of upstream and downstream measurements.

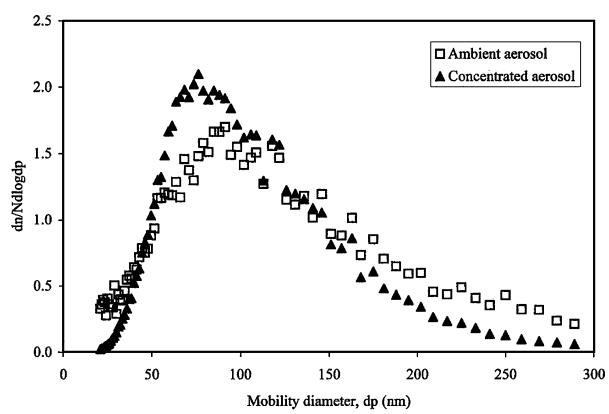
Performance of HUCAPS with Aerosols of Different Physicochemical Properties

Table 4 summarizes the comparison of concentration enrichment factors for each type of artificially generated test aerosol and ambient air for the optimum supersaturation ratio of 3.0. HUCAPS performance results with elemental carbon and outdoor air are new findings of this study, and these are compared to the results obtained using other artificially generated aerosols reported previously in Gupta et al. (2003). It is evident from the results that all particles grow and get concentrated by about the same enrichment factor, regardless of particle hygroscopicity and chemical composition.

Performance of HUCAPS with Ambient Air

Figure 7 shows the results of an ambient air test conducted on 6 February 2004. The test conditions were: dilution flow = 8 lpm, dilution air temperature = 121°C, total outlet flow = 58 lpm, outlet aerosol temperature = 35°C, and supersaturation ration (Sr) = 3.0. The data shown in the Figure 7 represent measured aerosol size distributions that have been normalized relative to the total aerosol number concentrations. A concentration enrichment factor of about 53 was obtained for this particular set of measurements from 6 February 2004. It is important to maintain the same size distribution between ambient and concentrated ultrafine aerosol for inhalation toxicological studies. The mean, median, and modal mobility diameters and geometric standard deviation (GSD) for ambient and concentrated aerosols agreed reasonably well (Figure 7). This is an indication that the thermal restoration method satisfactorily restored the size distribution of the concentrated aerosol.

The performance of HUCAPS with ambient air at Sr = 3.0 was monitored for several months in Boston. Table 5 presents the results from 10 selected measurement days. The last measurement day (6 February 2004) represents result from HUCAPS characterization at Research Triangle Park–Durham, NC. Overall, an enrichment factor of about 40 was observed for different measurement days, indicating that the day-to-day variations in the number concentration and chemical composition of particles as well as meteorological conditions have little effect on the performance of HUCAPS. For these measurement days, the percentage differences for mean, mode, median, and GSD were about 15–18% and varied from day-to-day measurements. These four parameters in addition to the average CF were calculated for the size range of 20–300 nm particle mobility diameters. The minor distortion of the size distribution suggests that physicochemical properties of ultrafine particles do not have major influence on the HUCAPS performance.



Mobility diameter (nm)	Ambient aerosol	Concentrated aerosol	Difference (%)
Mean	98.4	92.8	5.7
Median	85.1	82.0	3.6
Modal	91.4	76.4	16.4
GSD	1.72	1.61	6.4
Total concentration (#/cc)	8665	460642	
Overall concentration enrichment factor = 53			
Dilution flow = 8 LPM, $T_{\text{dil-flow}}$ = 121°C, Total outlet flow = 58 LPM, T_{outlet} = 35°C, Sr = 3.0			

FIG. 7. A representative plot showing HUCAPS performance with ambient air.

TABLE 5
HUCAPS performance with ambient air for different measurement days

Aerosol	Date	Duration of experiment (h)	Average CF	Average $\text{mean}_{\text{diff}}$ (%)	Average $\text{mode}_{\text{diff}}$ (%)	Average $\text{median}_{\text{diff}}$ (%)	Average GSd_{diff} (%)
Indoor air	5 March 2003	1.0	38 ± 2	9.7 (84)	19.6 (65)	16.9 (62)	16.1 (1.9)
Indoor air	6 March 2003	3.5	39 ± 5	18.7 (80)	11.7 (71)	19.1 (67)	17.0 (2.0)
Indoor air	26 March 2003	2.5	35 ± 3	10.5 (103)	5.1 (95)	16.4 (89)	11.9 (2.1)
Indoor air	27 March 2003	2.0	39 ± 2	18.4 (78)	14.4 (72)	18.8 (73)	16.5 (1.9)
Indoor air	2 April 2003	2.5	37 ± 5	19.5 (75)	17.1 (72)	18.2 (63)	17.5 (1.9)
Outdoor air	26 June 2003	1.0	33 ± 5	8.5 (99)	19.3 (44)	13.2 (61)	10.5 (2.1)
Outdoor air	3 July 2003	3.0	43 ± 6	13.0 (87)	14.5 (54)	5.2 (62)	8.3 (1.8)
Outdoor air	8 August 2003	1.0	42 ± 4	6.8 (78)	15.3 (53)	18.5 (58)	15.1 (2.0)
Outdoor air	9 August 2003	3.0	44 ± 4	14.6 (88)	16.0 (64)	19.3 (62)	14.5 (1.9)
Outdoor air	6 February 2004	3.0	50 ± 5	0.3 (91)	6.2 (71)	12.7 (71)	18.8 (1.8)

Note: Average ambient aerosol size distribution parameters: $\text{Mean}_{\text{avg}}^{\text{amb}}$, $\text{Mode}_{\text{avg}}^{\text{amb}}$, $\text{Median}_{\text{avg}}^{\text{amb}}$, and $\text{GSD}_{\text{avg}}^{\text{amb}}$ are inside parentheses. Dilution flow = 8 lpm, $T_{\text{dil-flow}} \sim 121°\text{C}$, total outlet flow = 58 lpm, $T_{\text{outlet}} \sim 35°\text{C}$, Sr = 3.0. Average $\text{Mean}_{\text{diff}} = \{[\text{Mean}_{\text{avg}}^{\text{conc}} - \text{Mean}_{\text{avg}}^{\text{amb}}]/[\text{Mean}_{\text{avg}}^{\text{amb}}]\} \times 100$. Average $\text{Mode}_{\text{diff}} = \{[\text{Mode}_{\text{avg}}^{\text{conc}} - \text{Mode}_{\text{avg}}^{\text{amb}}]/[\text{Mode}_{\text{avg}}^{\text{amb}}]\} \times 100$. Average $\text{Median}_{\text{diff}} = \{[\text{Median}_{\text{avg}}^{\text{conc}} - \text{Median}_{\text{avg}}^{\text{amb}}]/[\text{Median}_{\text{avg}}^{\text{amb}}]\} \times 100$. Average $\text{GSD}_{\text{diff}} = \{[\text{GSD}_{\text{avg}}^{\text{conc}} - \text{GSD}_{\text{avg}}^{\text{amb}}]/[\text{GSD}_{\text{avg}}^{\text{amb}}]\} \times 100$.

Since the size distribution of concentrated ultrafine particles is very similar to that of the unconcentrated ambient aerosol, we expect that the basic physicochemical properties are not likely to be changed during the process of concentration. Nonetheless, there remains the possibility that subtle changes may occur that could result in different biological effects. However, we are unaware of any feasible quantitative analytical techniques that could be used to detect such subtle changes. Whenever suitable measurement techniques become available, it would be highly appropriate to pursue such investigations.

CONCLUSIONS

The HUCAPS system delivers 58 lpm of concentrated aerosol, with little distortion of the ultrafine aerosol size distribution, which can be used to perform in vivo inhalation and in vitro exposure studies. The performance of HUCAPS was tested using single-component artificial aerosols of different physicochemical properties as well as ambient air. Optimum thermal conditions for size distribution restoration of ultrafine particles were determined and the performance of the HUCAPS was evaluated using ultrafine particles of ambient origin.

The maximum concentration factor of 40–50 times is obtained at approximately Sr = 3.0. For this study, the Sr was calculated simply based on the ratio of the water vapor pressure at the saturator and condenser temperatures to the saturation vapor pressure (Hinds, 1999). Also, for the theoretical calculations, homogeneous temperature and RH conditions were assumed at the entrance/exit of the condenser. It is true that the supersaturation ratio calculated by this method does not take into account the dynamics of heat and mass transfer within the condensing tubes, so the actual "effective" Sr (which, conceivably, would be some sort of average over both the cross-section of each tube and over the length of each tube) is certainly less than the calculated value. However, the optimum operational conditions for the HUCAPS was determined empirically, and the controls allow for reproducibly achieving the calculated optimal value of Sr = 3. Thus, while it might be interesting to perform complicated calculations to estimate the effective Sr, it is not necessary to do this to be able to use the HUCAPS to produce suitable atmospheres for toxicological tests.

In addition to the performance evaluation tests, operating HUCAPS for long durations of time tested the stability of the overall system. The ultrafine particle concentration enrichment factor remained relatively constant throughout this time period (<10% variation). Overall, pressure drop through the system is only 2.2 kPa, which is adequately low for inhalation toxicological chamber tests.

REFERENCES

Clarke, R. W., Coull, B., Reinisch, U., Catalano, P., Killingsworth, C., Koutrakis, P., Kavouras, I., Murthy, G. G. K., Lawrence, J., Lovett, E., Wolfson, M., Verrier, R., and Godleski, J. 2000. Inhaled concentrated ambient particles induce pulmonary inflammation and hematological changes in canines. *Environ. Health Perspect*. 108:1179–1187.

Demokritou, P., Gupta, T., and Koutrakis, P. 2002a. A high volume apparatus for the condensational growth of ultrafine particles for inhalation toxicological studies. *Aerosol Sci. Technol*. 36:1061–1072.

Demokritou, P., Kavouras, I. G., Ferguson, S. T., and Koutrakis, P. 2002b. Development of a high volume cascade impactor for toxicological and chemical characterization studies. *Aerosol Sci. Technol*. 36:925–933.

Demokritou, P., Gupta, T., Ferguson, S. T., and Koutrakis, P. 2002c. Development and laboratory performance evaluation of a personal cascade impactor. *J. Air Waste Manage. Assoc*. 52:1230–1237.

Demokritou, P., Gupta, T., Ferguson, S., and Koutrakis, P. 2003. Development of a high-volume concentrated ambient particles system (CAPS) for human and animal inhalation toxicological studies. *Inhal. Toxicol*. 15:111–129.

Demokritou, P., Lee, S. J., Ferguson, S., and Koutrakis, P. 2004. A compact multistage impactor for the characterization of atmospheric aerosols. *J. Aerosol Sci*. 35:281–299.

Dockery, D. W., and Pope, C. A. 1994. Acute effects of particulate air pollution. *Annu. Rev. Public Health* 15:107–132.

Dockery, D. W., Pope, C. A., Xu. X., Spengler, J. D., Ware, J. H., Fay, M. E., Ferris, B. G., and Speizer, F. E. 1993. An association between air pollution and mortality in six U.S. cities. *N. Engl. J. Med.* 329:1753–1808.

Drinker, P., Thomson, R. M., and Finn, J. L. 1927. Metal fume fever II: Resistance acquired by inhalation of ZnO_2 on two successive days. *J. Ind. Hyg. Toxicol*. 9:98–105.

Gamble, J. 1998. $PM_{2.5}$ and mortality in long term prospective cohort studies: Cause effects or statistical associations? *Environ. Health Perspect*. 106(9):535–549.

Godleski, J., Sioutas, C., Katler, M., and Koutrakis, P. 1996. Death from inhalation of concentrated ambient air particles in animal models of pulmonary disease. *Respir. Crit. Care Med.* 155(4):A246.

Gordon, T., Chen, L. C., Fine, J. M., Schleisinger, R. B., Su, W. Y., Kimmel, T. A., and Amdur, M. O. 1992. Pulmonary effects of zinc oxide in human subjects, guinea pigs, rats, and rabbits. *Am. Ind. Hyg. Assoc. J*. 53:503–509.

Gupta, T., Demokritou, P., and Koutrakis, P. 2003. Effects of physicochemical properties of ultrafine particles on the performance of an ultrafine particle concentrator. *Aerosol Sci. Technol*. 38(S2): 1–9.

International Commission on Radiological Protection. 1994. *Human respiratory tract model for radiological protection: A report of committee 2 of the ICRP*. Oxford: Pergamon Press.

Johnston, C. J., Finkelstein, J. N., Mercer, P., Corson, N., Gelein, R., and Oberdorster, G. 2000. Pulmonary effects induced by ultrafine PTFE particles. *Toxicol. Appl. Pharmacol.* 168:208–215.

Kavouras, I. G., and Koutrakis, P. 2001. Use of polyurethane foam as the impaction substrate/collection medium in conventional inertial impactors. *Aerosol Sci. Technol*. 34:46–56.

Kim, S., Chang, M. C., Kim, D., and Sioutas, C. 2000. A new generation of portable coarse, fine and ultrafine particle concentrators for use in inhalation toxicology. *Inhal. Toxicol*. 12(suppl.):121–137.

Lippmann, M. 1989. Progress, prospects, and research needs on the health effects of acid aerosols. *Environ. Health Perspect*. 79:203–205.

Oberdorster, G., Ferin, J., and Lehnert, B. E. 1994. Correlation between particle size, in vivo particle persistence and lung injury. *Environ. Health Perspect*. 102:173–179.

Peters, A., Verrier, R. L., Schwartz, J., Gold, D. R., Mittleman, M., Baliff, J., Oh, J. A., Allen, G., Monahan, K., and Dockery, D. W. 2000. Air pollution and incidence of cardiac arrythmia. *Epidemiol*. 11:11–17.

Pope, C. A., Verrier, R. L., Lovett, E. G., Larson, A. C., Raizenne, M. E., Kanner, R. E., Schwartz, J., Villegas, M., Gold, D. R., and Dockery, D. W. 1999. Heart rate variability associated with particulate air pollution. *Am. Heart J*. 138:890–899.

Schwartz, J., and Dockery, D. W. 1992. Increased mortality in Philadelphia associated with daily air pollution concentrations. *Am. J. Epidemiol*. 135:12–19.

Sioutas, C., and Koutrakis, P. 1996. Inertial separation of ultrafine particles using a condensational growth/virtual impaction system. *Aerosol Sci. Technol*. 25:424–436.

Sioutas, C., Koutrakis, P., and Burton, R. M. 1995. A technique to expose animals to concentrated fine ambient aerosols. *Environ. Health Perspect*. 103:172–177.

Sioutas, C., Kim, S., and Chang, M. 1999. Development and evaluation of a prototype ultrafine particle concentrator. *J. Aerosol Sci*. 30:1001–1017.

Whitby, K. T., and Svendrup, G. M. 1980. California aerosols: Their physical and chemical characteristics. *Environ. Sci. Technol*. 10:477.

Copyright of Inhalation Toxicology is the property of Taylor & Francis Ltd and its content may not be copied or emailed to multiple sites or posted to a listserv without the copyright holder's express written permission. However, users may print, download, or email articles for individual use.