Aerosol Science and Technology

ISSN: 0278-6826 (Print) 1521-7388 (Online) Journal homepage: http://www.tandfonline.com/loi/uast20

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To cite this article: P. Demokritou , I. G. Kavouras , S. T. Ferguson & P. Koutrakis (2002) Development of a High Volume Cascade Impactor for Toxicological and Chemical Characterization Studies, Aerosol Science and Technology, 36:9, 925-933, DOI: 10.1080/02786820290092113

To link to this article: http://dx.doi.org/10.1080/02786820290092113

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Aerosol Science and Technology 36: 925–933 (2002)
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Published by Taylor and Francis
0278-6826/02/$12.00 + .00
DOI: 10.1080/0278682029009211 3

Development of a High Volume Cascade Impactor for Toxicological and Chemical Characterization Studies

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This paper presents the design and development of a compact high volume cascade impactor (HVCI). The HVCI operates at a flow rate of 900 l/min and consists of 4 impaction stages equipped with circular slit-shaped acceleration nozzles and a backup filter. The backup filter is placed downstream of the fourth stage and is used to collect the ultrafine particles ($d_p < 0.1$ μm). The major feature of this novel sampler is its ability to collect relatively large amounts of particles (mg–g levels) onto relatively small polyurethane foam substrates without using adhesives. As previously reported, the capacity of the impaction substrate is 2.15 g of collected particles per cm^2 of foam. Although the impaction substrates are not coated with adhesives such as grease or mineral oil, particle bounce and re-entrainment losses were found not to be significant. Particles can be easily recovered from the foam substrates using aqueous extraction. The impactor was calibrated using polydisperse particles. The 50% cutpoints of the 4 stages were 9.90, 2.46, 1.0, and 0.1 μm, respectively. Interstage losses of ultrafine and fine particles were <10% and for coarse particles were <20%. The pressure drop across the 4 stages and the backup filter were 0.25, 0.75, 1.25, 19.9, and 3.3 kPa, respectively.

INTRODUCTION

A large number of studies have demonstrated the adverse effects of particle exposures on respiratory and cardiac health (Gold et al. 1999; Pope et al. 1999; Simpson et al. 1999; Klemm et al. 2000; Peters et al. 2000; Murphy et al. 1999; Soukup et al. 2000). The effect of the physicochemical characteristics of coarse, fine, and ultrafine particles on mortality and other health outcomes need to be examined separately (Burnett et al. 1997; Neas et al. 1999; Castillejos et al. 2000; Cohen et al. 1990; Venkatamaran and Kao 2000). A concentrator for ambient fine particles has been used to conduct in vivo human and animal inhalation studies (Sioutas et al. 1995a, b). The outcomes of these studies are very important in our efforts to investigate the acute, chronic, and subchronic particle health effects of fine particles (Cheng et al. 1990; Clarke et al. 1999, 2000; Kodavanti et al. 2000). In addition, it is necessary to investigate biological mechanisms through in vitro cellular studies to examine the toxicity of atmospheric pollutants (Imrich et al. 1999; Soukup et al. 2000). To adequately conduct these in vitro studies, it is necessary to collect relatively large amounts of particles (from about 0.2 g to several g).

Particles can be classified into different size categories using different impaction technologies (Hinds 1999). Several cascade impactors (sequential arrangements of inertial impactors in order of decreasing particle size) have been designed to classify particles from 5 nm to 50 μm (Berner et al. 1979; Vanderpool et al. 1987; Marple et al. 1991). However, these samplers are not adequate for collecting large quantities of particles for toxicological and particle characterization studies due to their limited collection capacity and low flow rate. Furthermore, in order to minimize particle bounce-off and re-entrainment, impaction substrates are usually coated with adhesives such as mineral oil and grease (Sehmel 1980; Wall et al. 1990; John et al. 1991; John and Sethi 1993; Pak et al. 1992).

In this paper we present the design and development of a high volume cascade impactor (HVCI) that utilizes foam as the collection medium. This impactor includes 4 impaction stages (10.0, 2.5, 1.0, and 0.1 μm) and a filter that collects particles below 0.1 μm. The major feature of the HVCI is its ability to relatively rapidly collect large amounts of particles (mg–g levels) onto small and inert pieces of impaction substrates without the use of adhesives.

Received 8 December 2000; accepted 20 March 2002.
The laboratory performance evaluation of the sampler was supported by U.S. EPA STAR grant R825 270-01-0 and the EPA/Harvard Particle Health Effects Center (EPA grant R827 353-01-0).
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DESIGN CONSIDERATIONS

The cutpoint of each impaction stage was calculated using the Stokes equation, as follows:

$$\mathrm{Stk} = \frac{\rho_p \cdot d_\alpha^2 \cdot U \cdot C_c}{9 \cdot \eta \cdot W}, \quad [1]$$

where ρ_p is the particle density (g/m^3), d_a is the particle diameter (μm), U is the jet velocity (m/s), η is the dynamic viscosity of the air (g/(m min)), W is the nozzle width (m), and C_c is the Cunningham slip correction factor. The slip correction factor is given by the following equation (Hinds 1999):

$$C_c = 1 + \frac{2}{P \cdot d_p} \cdot \left[6.32 + 2.01e^{(-01095 \cdot P \cdot d_p)}\right], \quad [2]$$

where P is the absolute atmospheric pressure (atm) upstream of the nozzle. The Stokes equation was used to determine the dimensions of the acceleration nozzles. For the first stage, a layer of vacuum grease (0.32 cm thick) is used as an impaction substrate. A razor blade is used to create a smooth impaction surface. This is necessary to minimize bounce-off and breakup losses of large particles ($da > 10$ μm) and to obtain a sharp cut-off curve (Demokritou et al. 2001a, b). For this stage, a $\sqrt{Stk_{50}}$ value of 0.7 was used (Hinds 1999). For the other 3 stages, which use polyurethane foam as an impaction substrate, lower $\sqrt{Stk_{50}}$ values were used (0.4–0.5). The Reynolds number (Re) was calculated using the following equation:

$$\mathrm{Re} = \frac{U \cdot \rho_{\mathrm{air}} \cdot A}{\eta} = \frac{(Q/L \cdot W) \cdot \rho_{\mathrm{air}} \cdot 2 \cdot W}{\eta} = \frac{2 \cdot Q \cdot \rho_{\mathrm{air}}}{\eta \cdot L}, \quad [3]$$

where ρ_{air} is the air density (g/m^3), A is the hydraulic diameter of the nozzle (which is equal to $2W$ for rectangular nozzles), L is the length of the nozzle (m), and Q is the flow rate (m^3/min).

Polyurethane foam was used as an impaction substrate. This is a polymeric material with stable physical characteristics, low chemical background (when cleaned properly), and high collection efficiency characteristics (Kavouras et al. 2000; Salonen et al. 2000). As we have previously reported, the use of polyurethane foam substrates improves the performance of inertial impactors by minimizing bounce-off and re-entrainment losses, as compared to coated and uncoated flat plate substrates (Kavouras et al. 2000; Kavouras and Koutrakis 2001). Since for the same nozzle geometry and flow rate lower cutpoints can be achieved as compared to those for flat plate substrates, the pressure drop is substantially reduced for the same size cutpoint (Kavouras et al. 2000; Kavouras and Koutrakis 2001). Another advantage of foam substrates is their ability to collect large amounts of particles per surface area (2.15 g/cm^2) (Kavouras et al. 2000). This increases the sensitivity of toxicological and chemical characterization studies (Salonen et al. 2000; Chang et al. 2000).

DESCRIPTION OF THE HVCI

A fully assembled HVCI is shown in Figure 1. The sampling device consists of 4 impactor stages and a backup filter. The HVCI is compact (40.00 [H] × 20.32 [O.D.] cm) and is relatively lightweight (~10 kg). The physical characteristics and the theoretical cutpoint of each impactor stage are presented in Table 1. A schematic diagram of the cascade impactor, including the backup filter, is depicted in Figure 2. Each stage consists of 3 slit-shaped acceleration nozzles laid out on the perimeter of a 12.7 cm diameter circle (Figures 1 and 2). The length of each nozzle corresponds to 99.2° (actinic perimeter angle, Figure 1). The distance between the nozzles is 2.30 cm (equal to 20.8°, actinic perimeter angle). The nozzle widths are 0.03, 0.13, 0.25, and 0.76 cm for the fourth, third, second, and first stages, respectively (Table 1). As a result of the nozzle construction process, the ends of the nozzles were rounded. These radially arranged slit-shaped nozzles were used to avoid jet-to-jet interactions, while reducing the overall size of the HVCI.

Polyurethane foam (density = 0.020 g/cm^3, Merryweather Foam, OH) is used as an impaction substrate for the second, third, and fourth stages. For the first stage, which is designed to remove particles with diameters above 10 μm, a greased flat surface is used as an impaction substrate. The optimum substrate-to-nozzle distance (S) and the size of impaction substrate (D) were determined experimentally. S is 0.76, 0.25, 0.12, and 0.06 cm for

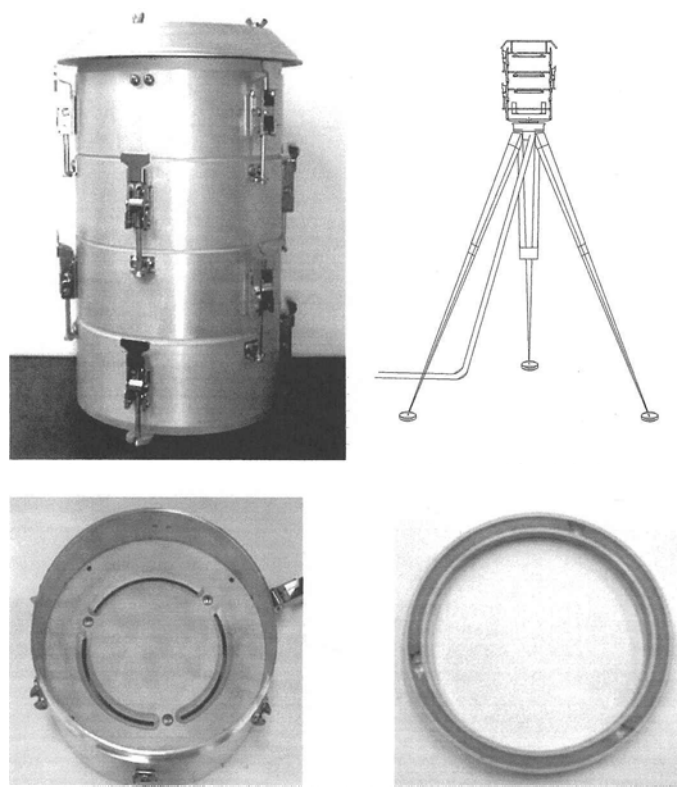


Figure 1. The HVCI.

Table 1
Physical characteristics and theoretically and experimentally calculated characteristics of cascade impactor (L: nozzle length; W: nozzle width; T: nozzle throat length; S: substrate-to-nozzle distance; D: substrate width; U: nozzle air velocity; Re: Reynolds number; d_{50}: 50% cutpoint; $\sqrt{Stk}$: square root of Stokes number; s: collection efficiency curve sharpness; ΔP: pressure drop)

Stage	1*	2	3	4
	Physical characteristics			
Nozzles	3	3	3	3
L (cm)	10.99	10.99	8.0	9.60
W (cm)	0.759	0.254	0.132	0.028
S (cm)	0.759	0.254	0.125	0.056
S/W	1.0	1.0	0.95	2.0
D (cm)	2.3	1.28	0.6	0.6
	Theoretical calculations			
U (cm/s)	599	1791	4735	18601
Re	6058	6058	8322	6935
	Experimental results			
d_{50}	9.9	2.46	1.0	0.09
s	1.17	1.43	1.44	1.33
$\sqrt{Stk}$	0.70	0.53	0.51	0.34
ΔP (kPa)	0.25	0.75	1.25	19.93

*Grease is used as an impaction substrate.

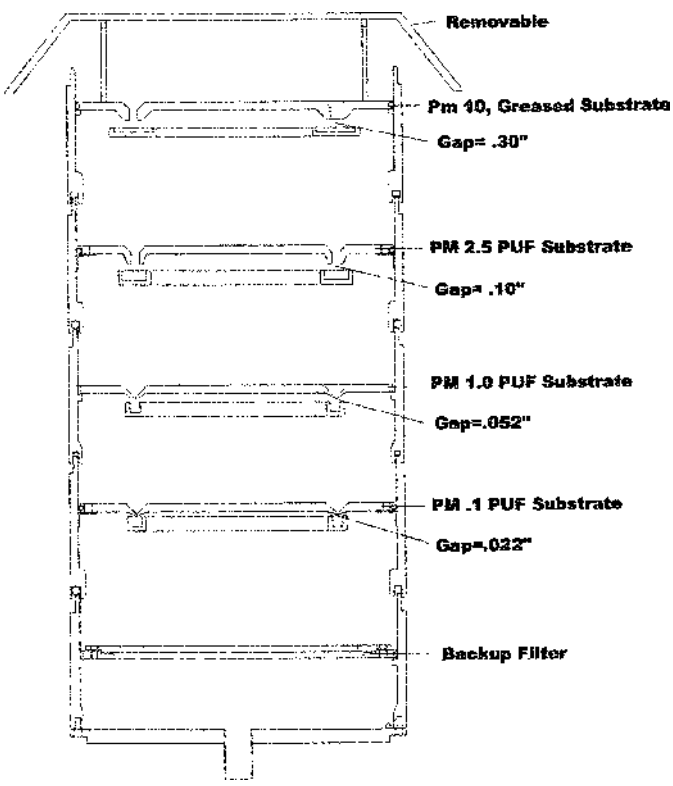


Figure 2. Schematic section of the HVCI.

the first, second, third, and fourth stages, respectively (Table 1). The widths of the impaction substrate are as follows: 1st stage: 2.3 cm (3.0W); 2nd stage: 1.3 cm (5.1W); 3rd stage: 0.64 cm (4.8W); 4th stage: 0.64 cm (22.8W) (Table 1). Furthermore, the thickness of the polyurethane foam for all stages is 0.64 cm to assure that particles do not penetrate through the Polyurethane Foam (PUF) and reach the substrate holder.

Finally, ultrafine particles are collected on a backup filter. The criteria for the selection of the backup filter material were as follows: (a) high capacity; (b) low blank levels; (c) high collection efficiency; and (d) low pressure drop. Three filter materials were evaluated, including a polypropylene filter (Monadnock, Grade 5300), a glass fiber filter (Gelman, type A/C), and a teflon coated quartz filter (Pallflex, Emfab-TX40).

METHODS

The experimental setup is shown schematically in Figure 3. The apparatus consisted of 3 components: the particle generation system, the dilution tube, and the particle monitor instrument. The particle generation system used an aqueous suspension of either hollow glass spheres (nominal size 2–20 μm, density = 1.10 g/cm^3 Polysciences, PA) or solid glass spheres (nominal size 3–10 μm, density = 2.48 g/cm^3 Polysciences, PA). To achieve continuous and stable generation of aerosolized particles, the aqueous suspension flowed into a nebulizer (Retec Model X-70/N, with a pressure of 7 psi), with excess flow out of the nebulizer reservoir into a waste container. The output of the nebulizer passed into the top of the anodized cylindrical aluminum dilution tube (150 [H] cm × 7.62 [O.D.] cm). Particle free and dry dilution air was also introduced into the top of the dilution tube. To assure a well-mixed aerosol distribution inside the tube, a round plate was placed inside the tube, downstream of the input flows. Each stage of the HVCI was evaluated separately. The sampler for each stage was mounted at the bottom of the dilution tube. The particle number concentration and size distribution were measured for a period of 10 min upstream and downstream of the sampler (impaction stage) using isokinetic probes. This sequence was repeated twice. An aerodynamic particle sizer (APS 3320, TSI, Inc., St. Paul, MN) and a scanning mobility particle sizer (SMPS, TSI Inc, St. Paul, MN) were used to measure particles with diameters from 0.5 to 20 μm and from 0.02 to 0.5 μm, respectively. A condensation particle counter (CPC 3010A, TSI Inc., St. Paul, MN) was employed to measure the particle number concentration. Even though the nominal minimum mass median sizes for both the hollow and solid spheres were 2 and 3 μm, respectively, the number concentrations for both types of spheres for sizes down to 0.02 μm were sufficient to perform SMPS experiments. Particle losses to the sampler walls and jet nozzles were measured using the tested impaction stage without the impaction substrate.

The collection efficiency for a given particle size, $E(d_a)$, for either the impaction substrates or the backup filter, was

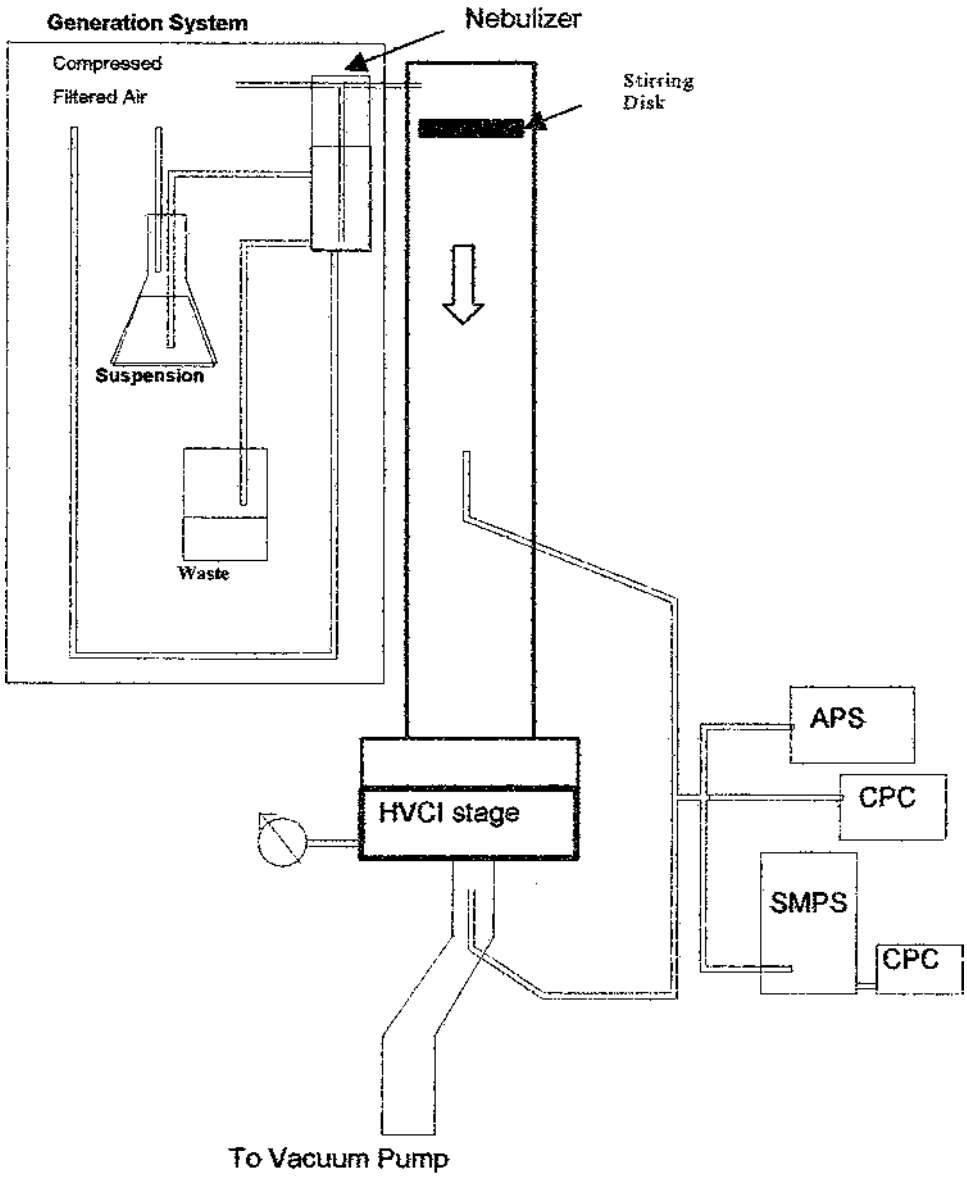


Figure 3. Impactor test experimental apparatus.

calculated as follows:

$$E(d_a) = \frac{C(d_a)}{T(d_a)} = \frac{T(d_a) - P(d_a)}{T(d_a)}, \quad [4]$$

where $T(d_a)$, $C(d_a)$, and $P(d_a)$ are the total, collected, and penetrated particles of aerodynamic diameter of d_a, respectively. The collection efficiency data were fitted using the Boltzmann sigmoidal algorithm (Origin, MicroCal Software Inc.) as follows:

$$E(d_a) = \frac{A_1 - A_2}{1 + e((d_a - x^o)/dx)} + A_2, \quad [5]$$

where x^o is the median aerodynamic diameter, dx is the width of the fitting, and A_1 and A_2, are the coefficients determined by the algorithm. The sharpness (s) of the collection efficiency curve was calculated using the following equation:

$$s = \sqrt{\frac{d_{84.1}}{d_{15.9}}}, \quad [6]$$

where $d_{84.1}$ and $d_{15.9}$ are the sizes of particles having collection efficiencies of 84.1 and 15.9%, respectively (Hinds 1999).

RESULTS AND DISCUSSION

Impactor Calibration Experiments

The collection efficiency curves and the interstage losses for the 4 cascade impactor stages are shown in Figure 4. The

experimentally calculated cutpoints (d_{50}, μm), the collection efficiency sharpness (s), and the pressure drop (ΔP, kPa) are presented in Table 1.

The first stage is designed to remove large particles ($da >$ 10 μm) (Table 1). The experimentally determined cutpoint for this stage is 9.90 μm, which corresponds to $\sqrt{Stk} = 0.7$. The collection efficiency curve sharpness is 1.17 (Table 1), which indicates an excellent separation of particles larger than the cutpoint from the airstream (Figure 4). The $\sqrt{Stk}$ number and the impactor sharpness are similar to those previously reported for impactors with rectangular nozzles (Kavouras et al. 2000; Marple et al. 1993). Pressure drop is only 0.25 kPa.

The experimentally determined cutpoint of the second impactor stage is 2.46 μm ($\sqrt{Stk} = 0.53$, $s = 1.43$) (Table 1). For particle above 4.0 μm the collection efficiency was higher than 98% (Figure 4). This suggests that coarse particle bounce-off and re-entrainment losses were insignificant. The experimentally determined cutpoint of this stage is smaller than that theoretically calculated ($d_a = 4.20$ μm) for a flat rigid surface impaction substrate, assuming a $\sqrt{Stk} = 0.7$ (Table 1). However, it is similar to that obtained for rectangular nozzles using polyurethane foam as an impaction substrate (Kavouras et al. 2000). This cutpoint decrease was recently reported for both rectangular and round nozzles and was explained by penetration of some air streamlines into the porous polyurethane foam surface (Kavouras et al. 2000; Kavouras and Koutrakis 2000). The pressure drop is only 0.75 kPa.

The experimental cutpoint and $\sqrt{Stk}$ for the third stage are 1 μm and 0.51, respectively. The Stokes number is similar to that of the second stage (Table 1; Figure 4a). The sharpness of the collection efficiency curve ($s = 1.44$) is adequate to effectively collect (up to 97%) particles above 1.5 μm (Table 1; Figure 4a). The pressure drop is 1.25 kPa.

The experimental cutpoint and $\sqrt{Stk}$ for the fourth stage are 0.1 μm and 0.34, respectively (Table 1; Figure 4a). The particle acceleration velocity is 18,600 cm/s (Table 1), thus the pressure drop across this stage is dramatically increased to 19.9 kPa (Table 1). The collection efficiency curve sharpness is 1.33, and as indicated by Figure 4, collection of particles with diameters from 0.13 to 1.00 μm was higher than 99%.

Inertial particle losses within the HVCI may be caused by the turbulent flow in the acceleration nozzle region, at flow turns, and at the flow exit/entrance between stages. Particle losses inside each stage were determined by measuring particles up- and downstream of the stage, with the substrate removed. It is worth mentioning that the collection efficiencies for each stage illustrated in Figure 4a include particles collected on both the impaction substrate and particles lost on stage walls and the acceleration nozzle. Figure 4b illustrates the particle losses in each stage as a function of the particle aerodynamic diameter. Coarse particle losses (2.5–10 μm) in the first stage increased from 5% for 4 μm particles up to 20% for particles with a diameter larger than 8 μm. Fine particle losses in the second stage are minimal (<5%). Particle losses in the third and fourth stages were higher (about 10%) due to the increased diffusional deposition of these small particles. Similar results were previously reported for other cascade impactors (Kavouras et al. 2000; Marple et al. 1991).

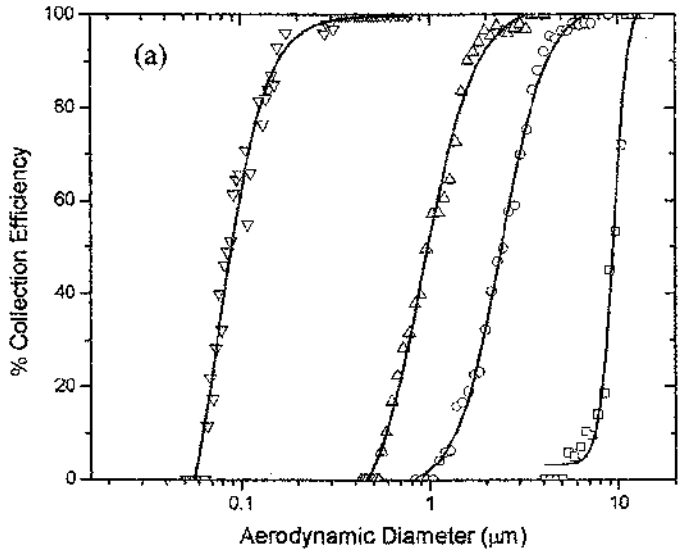


Figure 4a. Collection efficiency curves of HVCI impactor stages.

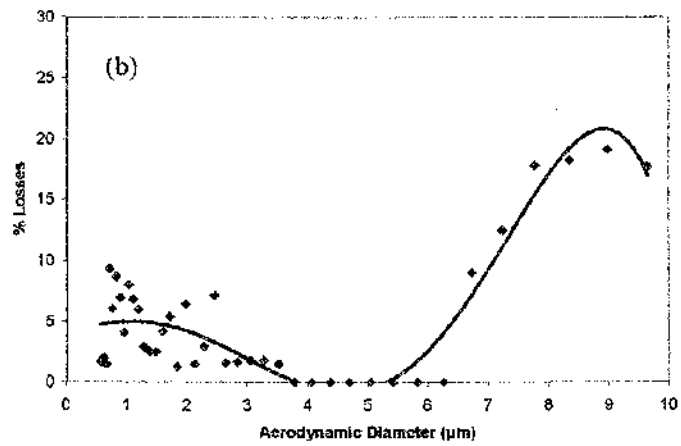
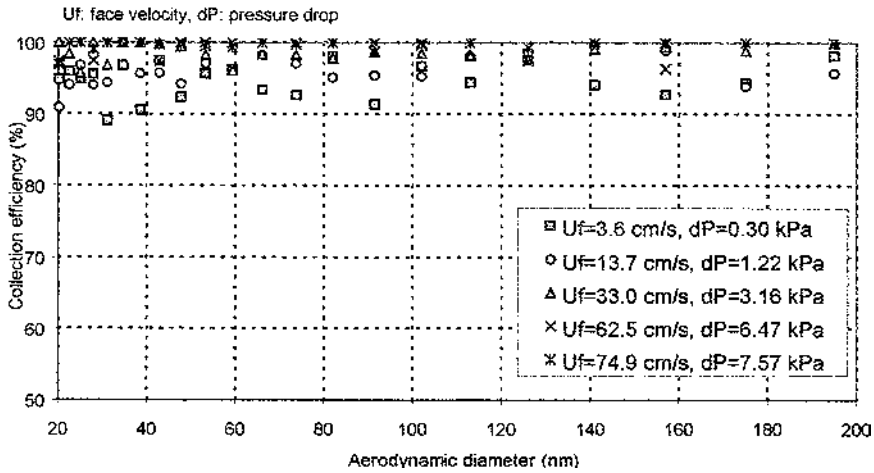


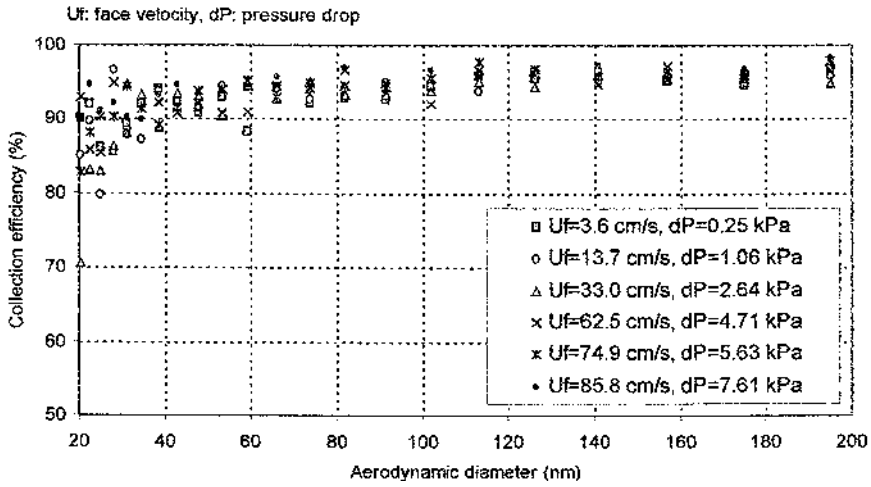
Figure 4b. Interstage particle losses.

Backup Filter. The performance of various filter materials to effectively collect ultrafine particles downstream of the last stage was evaluated. Filter materials included polypropylene filter (Monadnock, Grade 5300), glass fiber filter (Gelman, type A/C), and Teflon coated quartz filter (Pallflex, Emfab-TX40). Their collection at the corresponding face velocities and pressure drops are illustrated in Figure 5.

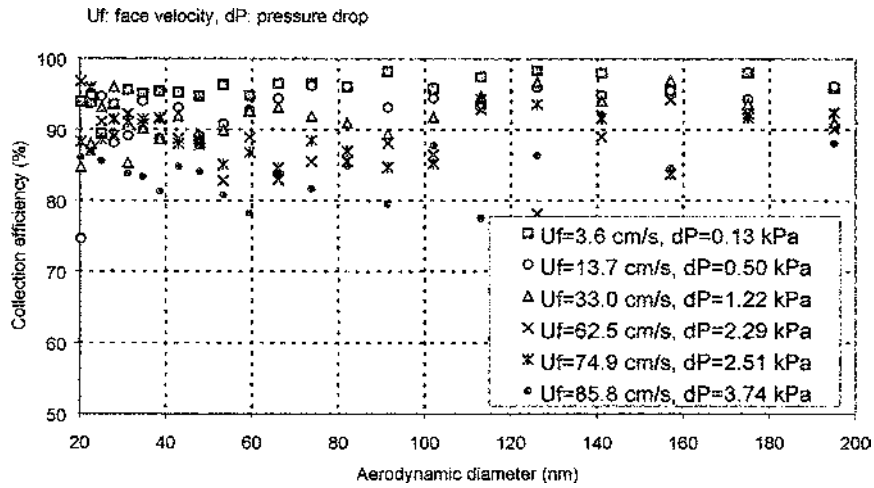
In order to use filters for toxicological and chemical studies, it is necessary to chemically clean and autoclave them at 120°C. The cleaning procedure, including cleaning solvents used, was described in detail by Salonen et al. (2000). The particle collection efficiency and pressure drop were measured prior to



(a)

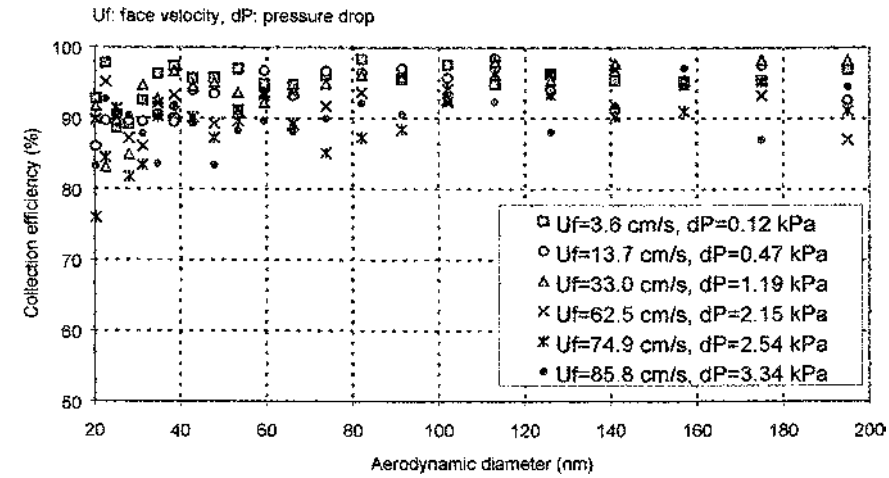


(b)

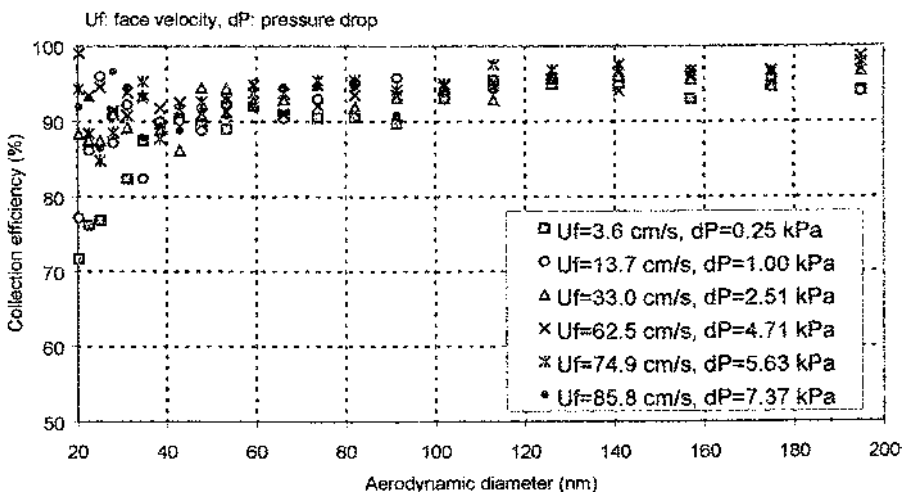


(c)

Figure 5. Untreated backup filter collection efficiency at different face velocities and pressure drops. (a) Glassfiber filter A/C, no treatments; (b) TX40, no treatments; (c) Polypropylene 5300, no treatments.



(a)



(b)

Figure 6. Treated (autoclaved and chemically cleaned) backup filter collection efficiency at different face velocities and pressure drops. (a) Polypropylene filter (5300); (b) TX-40 filter.

and after filter treatment (no treatment was made for the glass fiber filters). The results shown in Figures 5 (untreated) and 6 (treated) suggest only minor differences in collection efficiency and pressure drop. For all tested filter materials, collection efficiencies decreased with face velocity. As expected, the highest efficiency was observed for the lowest velocity of 3.6 cm/s. In order to achieve this face velocity for a total flow rate of 900 L/min, it would be necessary to use a filter diameter of 76.4 cm, which is significantly bigger than the stage O.D. of 15 cm. The corresponding face velocity for a 15 cm diameter is 85 cm/s. At this face velocity, all tested filter materials had collection efficiencies >70%, with the polypropylene filter presenting the minimum, <4 kPa, pressure drop. It is important to keep the pressure drop this low to minimize volatilization losses of semi-volatile particles. Therefore the polypropylene filter was selected as the most appropriate collecting medium. For a face velocity of 85 cm/s, its corresponding collection efficiency was >85% and the corresponding pressure drop was 3.3 kPa.

Stage Response Factor and Mass Loading

The conversion of integrated impactor measurements to a continuous size distribution is a rather complicated task that requires the use of an inversion algorithm. An inversion algorithm developed by Rader et al. (1991) to estimate the stage response factor and the collected mass for the Andersen Mark III and Marple personal impactors was used. This algorithm takes into account the collection efficiency measurements, as well as interstage and inlet losses. Briefly, the collected mass (M, μg) was calculated as follows:

$$M = V \cdot \int K_i(d_a) \cdot T_i^*(d_a) d(d_a), \qquad [7]$$

where $K_i(d_a)$ is the response factor of a particle with diameter d_a for the stage i, $T_i^*(d_a)$ is the corrected mass concentration (μg/m^3) entering the size selective inlet of particles between d_a, and $d_a + d(d_a)$ and V is the air sample (m^3). The mass concentration entering the cascade impactor was corrected for wall stage losses as follows:

$$T_i^*(d_a) = T_i(d_a) \cdot L_0(d_a) * (1 - L(d_a)), \quad [8]$$

where $T_i(d_a)$, $L_0(d_a)$, and $L(d_a)$ are the total mass concentrations, inlet losses, and interstage losses for a given particle size, respectively. The correction of the particle size distribution is an acceptable assumption and provides a good agreement between the theoretical and experimental mass loadings (Rader et al. 1991).

The response factor (K_i) for a stage I is the ratio of the particle mass concentration collected that entered the i stage. The stage response factor for the first stage is equal to the collection efficiency

$$K_1(d_a) = E_1(d_a), \quad [9]$$

whereas for the rest of the stages the response factor is calculated using the following equation:

$$K_i(d_a) = E_i(d_a) \cdot (1 - E_{i-1}(d_a)) \cdot \cdots \cdot (1 - E_1(d_a)), \quad [10]$$

where $i = 1, 2, 3, \ldots, n$, the number of stage. The response factor for the backup filter was also calculated using the above equation.

For our calculations, the particle size distribution data obtained in Boston, MA, were used (Long 2000). These data served as input for Equations (7) and (8) to calculate the collected mass for each stage. Figure 7 shows the calculated mass distribution using HVCI stage response factors and the corresponding size distribution of particles using continuous instrumentation (APS 3310 and SMPS/CPC) in Boston for 3 samples. In the first case (Figure 7a), the distribution was bimodal. The first mode was associated with very small particles ($d_a < 0.99$ μm). The second mode was observed in the coarse fraction ($2.5 < d_a < 10.0$ μm). The mass concentrations of the 2 modes were 28.3 and 6.3 μg/m^3 (Figure 7a). The predicted size distribution maxima was calculated at the 4th (0.09–0.99 μm) stage (27.1 μg/m^3), while predicted concentrations for the 2nd (2.5–10 μm) and 3rd (1.0–2.5 μm) stages were similar (6.2 and 6.0 μg/m^3, respectively). The estimated mass of 27.1 μg/m^3 for the 4th stage was in agreement with that profile measured using continuous analyzers (Figure 7a). The particle size distribution for the other 2 cases (Figures 7b and c) exhibited a predominant occurrence in the size fraction between 0.1 and 1.0 μm with mass concentrations of 17.0 and 23.9 μg/m^3, respectively. The corresponding mass concentration calculated using the stage response factor for these days showed a similar profile with maximum at the 4th stage of 16.2 and 23.2 μg/m^3, respectively.

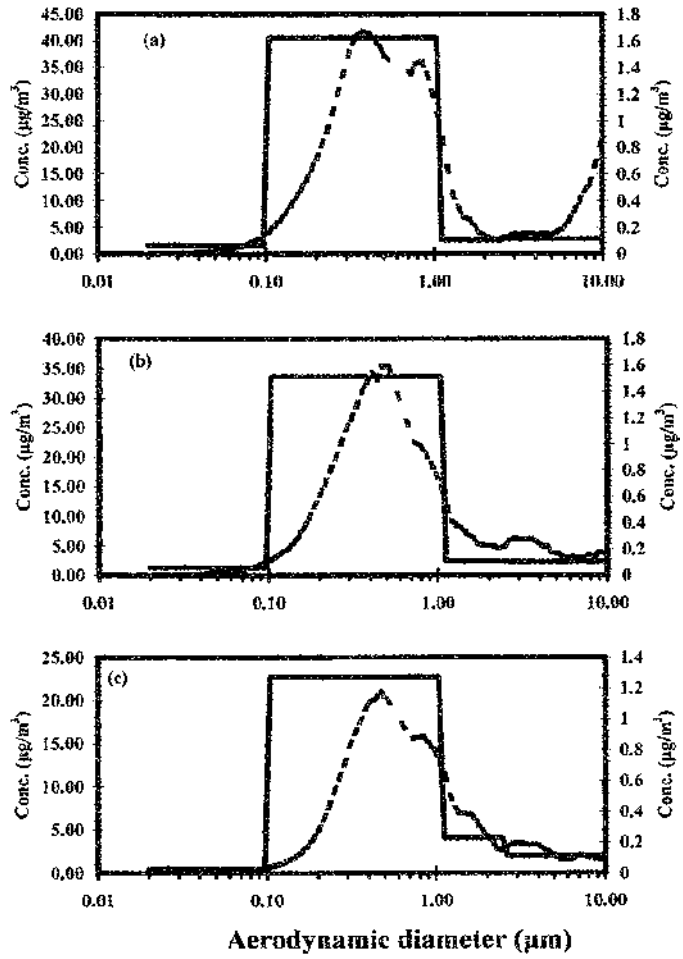


Figure 7. Comparison of the size distribution of particles measured with continuous analyzers with that estimated using HVCI for 3 different cases.

CONCLUSIONS

A High Volume Cascade Impactor (HVCI) was developed. This sampler operates at a flow rate of 900 L/min and consists of 4 stages and a backup filter. The size cutpoints of the 4 stages are 10, 2.5, 1.0, and 0.1 μm, respectively. The versatile design of the HVCI allows for the inclusion of additional stages. The use of polyurethane foam as an impaction substrate eliminates the use of adhesives such as grease or mineral oil, while it eliminates particle bounce-off and re-entrainment losses. Finally, because of its high capacity, the HVCI can be used to collect large quantities of particles (mg–g levels) for toxicological, biological, and chemical characterization studies.

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