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Development and Evaluation of an Impactor for a $PM_{2.5}$ Speciation Sampler

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ABSTRACT

A conventional impactor for a particle speciation sampler was developed and validated through laboratory and field tests. The speciation sampler consists of the following components: a $PM_{2.5}$ conventional impactor that removes particles larger than 2.5 μm, an all-glass, coated honeycomb diffusion denuder, and a 47-mm filter pack. The speciation sampler can operate at two different sampling rates: 10 and 16.7 L/min. An experimental characterization of the impactor's performance was conducted. The impactor's collection efficiency was examined as a function of critical design parameters such as Reynolds number, the distance from the nozzle exit to the impaction plate, and the impaction substrate coating method. The bounce of particles larger than the cut point was successfully minimized by using a greased surface as the impaction substrate. Additionally, a series of field intercomparison experiments were conducted at both 10 and 16.7 L/min airflow. $PM_{2.5}$ mass and SO_4^{2-} concentrations were measured and compared with the Federal Reference Method (FRM) and found to be in good agreement. Results of the laboratory chamber tests also indicated that the impactor's performance was in good agreement with the FRM.

IMPLICATIONS

There is a growing need for methods that can be used to measure the different chemical components of $PM_{2.5}$. This paper discusses the development and characterization of a speciation sampler for sampling of atmospheric particles and gases, which can be used for the chemical composition characterization of $PM_{2.5}$. Both laboratory and field experiments were performed to determine the effect of different design parameters, such as the impaction substrate method and the distance from the nozzle exit to the nozzle diameter, on the impactor's performance. The developed speciation sampler will be used in our studies in various U.S. cities to characterize the chemical composition of ambient $PM_{2.5}$.

INTRODUCTION

Epidemiologic studies have shown significant associations between PM and increased mortality and morbidity. Recent results from various studies have indicated that the respirable fraction of PM (PM_{10} and $PM_{2.5}$) is responsible for the observed adverse health effects.[1–3] Therefore, there is a growing need for methods that can be used to measure the different chemical components of fine PM. These methods could also be used for the U.S. Environmental Protection Agency's (EPA) particle speciation network.

A speciation sampler was developed in our laboratory[4] for sampling of atmospheric particles and gases. The first stage of this speciation sampler consists of a multinozzle conventional inertial impactor, which removes particles larger than 2.1 μm. An all-glass, coated, honeycomb diffusion denuder is placed downstream of the impactor to remove organic and/or inorganic acidic gases. The denuder is a cylinder with a height of 3.8 cm containing 212 hexagonal glass tubes. This system has a higher capacity than that of an annular denuder.[5] Particulate matter can be collected downstream of the denuder on a 47-mm filter. Two denuders can be used in series. The first denuder is coated with Na_2CO_3/glycerol to collect acidic gases. A second denuder coated with citric acid can be used to collect NH_3.

An improved impactor version for this speciation sampler was subsequently developed by Sioutas et al.[6] This is a single-nozzle impactor that uses an oiled sintered disk as an impaction substrate. Although this single-nozzle design eliminates the multiple jet interaction problems,[7] particle bounce of particles larger than the cut point was found to be significant. Also, as with the previously described system, this impactor has a 2.1-mm cut point, which is lower than that of the Federal Reference Method (FRM) (2.5 μm).

To eliminate the particle-bounce problem, a new impactor for the speciation sampler was designed and tested. The impactor can be operated at either 10 or 16.7 L/min airflow rate, simply by changing only its acceleration

nozzle. As part of this development, several impaction substrate methods, including oiled metal disks of various pore size as well as greased surfaces, were tested. A parametric investigation was also performed to determine the effect of different design parameters, such as the distance from nozzle exit to the nozzle diameter and impaction substrate diameter, on the impactor's performance.

METHODS

Impactors

Impactors have been used successfully for particle collection and classification.[8–10] A conventional impactor consists of a jet of particle-laden gas impinging upon a flat surface with particles impacting on a plate. The basic mechanism for inertial deposition of particles is the specific motion of aerosol particles in the impaction zone or stagnation point. In this region, the streamlines change abruptly. Particles larger than the impactor's cut size will impact onto the plate, while smaller particles will remain in the streamlines and will not be collected.

According to impaction theory, the Stoke's number, *Stk,* is the governing parameter for impaction and is defined as follows:

$$Stk = \frac{\rho_p d_p^2 U C_c}{9\mu W} \quad (1)$$

where μ is the dynamic viscosity of the air (g/cm sec), d_p is the diameter of the particle (μm), ρ_p is the particle density (g/cm^3), W is the nozzle diameter (cm), U is the jet velocity (cm/sec), and C_c is the Cunningham slip correction factor. The slip correction factor is given by the following equation:[11]

$$C_c = 1 + \frac{2}{Pd_p}[6.32 + 2.01\exp(-0.1095Pd_p)] \quad (2)$$

where P is the absolute atmospheric pressure (cm Hg) upstream of the nozzle.

Inertial impactors have been studied extensively, both theoretically and numerically. Various guidelines for the critical design parameters were obtained from the numerical analysis of the Navier-Stokes fluid flow equations.[12,13] It has been shown that for round-nozzle impactors, the aerodynamic diameter of particles collected with 50% efficiency (d_{50}), also known as cut point or separation point, corresponds to a $\sqrt{Stk_{50}}$ of 0.5. Therefore, the theoretical d_{50} particle diameter can be calculated using eq 1 for any desired flow rate and nozzle diameter.

There are three major areas of concern associated with the design of an impactor:[14] particle bounce from the collection surface; overloading of collected particles on the impaction substrate; and interstage losses (collection of particles on surfaces other than the impaction plate). One approach frequently used for minimizing particle bounce is the application of a sticky substance such as oil or grease to the impaction plate. The use of oil as the coating medium requires the use of a porous substrate to retain the oil. Porous metal disks,[15] porous glass frits,[16] and 10-μm Teflon membrane filters have been used in various impactors.

Also, the substrate has to perform well under various loading conditions. Once a monolayer of particles is collected on the surface, incoming particles may bounce after impacting onto already collected particles. Therefore, the substrate's capacity must be large enough to maintain high collection efficiency, even under heavy loading conditions. Other factors, such as the grease coating thickness, particle hardness, and shape, can also affect bouncing behavior.[11,15,17] Another important design parameter that can significantly affect the overall impactor performance is the S/W ratio, where S is the distance between the nozzle exit and the impaction plate and W is the nozzle diameter. According to the numerical analysis of conventional impactors by Marple et al.,[18] the S/W ratio should be greater than 1 for round nozzles and 1.5 for rectangular impactors.[12,18]

Description of the Speciation Sampler

The speciation sampler (CHEMCOMB) is shown in Figure 1. Its first stage consists of a conventional inertial impactor that removes particles larger than 2.5 μm. An all-glass, coated honeycomb diffusion denuder can be placed downstream of the impactor to remove organic and inorganic acidic gases. Particulate matter is collected downstream of the denuder on a 47-mm filter. Figure 1 illustrates the four main components of the sampler: 1) the acceleration nozzle; 2) the impaction substrate; 3) the honeycomb denuder section; and 4) the filter holder. All components are made of aluminum and are carbon-impregnated Teflon-coated to minimize losses of reactive gases on the internal surfaces. The sampler can operate at two different sampling flow rates, 10 and 16.7 L/min, simply by changing only the nozzle section. The 10 and 16.7 L/min airflow rates were selected to match the airflows and cut point characteristics of the Harvard/EPA-Annular Denuder System and the FRM, respectively. Both the impactor section and the filter holder components are connected to the sampler body with spring clips. The critical impactor design parameters are shown in Table 1.

The impaction substrate section consists of a hollow aluminum disk with a height of 0.13 cm and a diameter of 2.54 cm. Two types of impaction substrates were investigated: oiled porous metal disks and greased surfaces. A porous metal disk was inserted tightly in the hollow aluminum disk and was impregnated with mineral oil (paraffin oil, NF grade, Mallinckrodt) to minimize particle

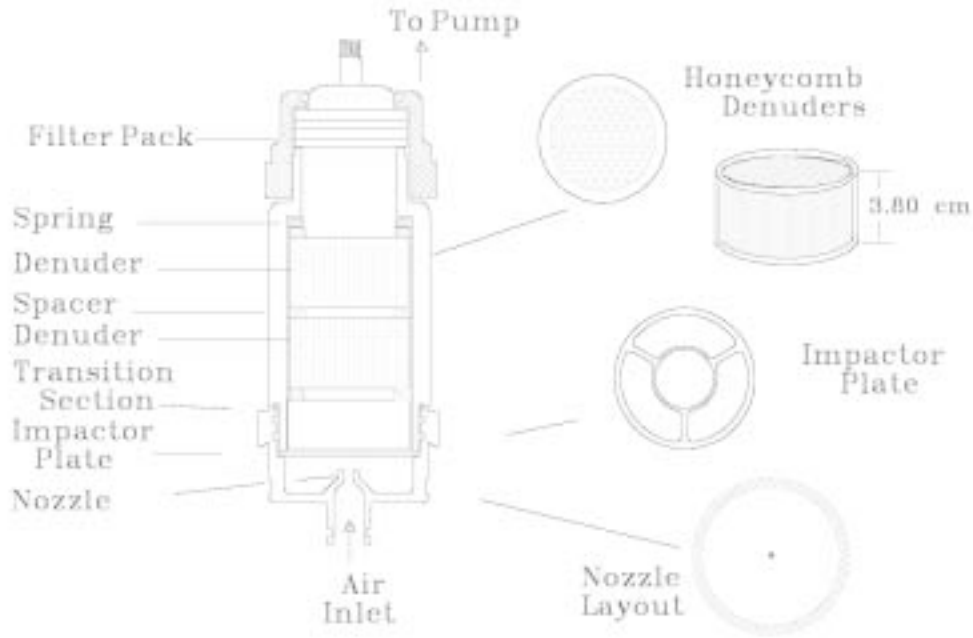

Figure 1. $PM_{2.5}$ speciation sampler (CHEMCOMB).

bounce. Metal disks of various pore sizes were tested. A greased surface was also tested as an impaction substrate by filling the cavity of the hollow aluminum disk with silicon grease. The greased surface was then smoothed to minimize turbulence in the impaction zone. The smoothness of the impaction surface was found to be important for the impactor's performance.

Laboratory Characterization of the Sampler

The experimental setup used for the characterization of the impaction section is shown in Figure 2. The sampler's particle collection efficiency was examined as a function of several design parameters, such as Reynolds number, S/W, and the substrate method. Particle concentrations were continuously monitored both upstream and downstream of the test sampler.

Polydisperse particles were generated by nebulizing an aqueous suspension of 2- to 20-μm hollow glass spheres (density: 1.1 g/cm^3) (Polysciences, Inc.) with a Retec Model X-70/N nebulizer, using filtered air at 7 psi. The aerosol was mixed using filtered room air. The test air mixture of polydisperse glass spheres then passed into the top end of a vertical cylindrical duct [95.0 cm (L) × 7.6 cm (i.d.)] made of anodized aluminum. Additional filtered room air was also added at the top of the duct. Turbulence was induced near the top using a rectangular plate, to assure uniform concentration throughout the duct. The sampler was connected at the bottom of the duct. Alternate measurements were performed upstream and downstream of the sampler. The experiments were repeated at least three times to ensure the repeatability of the calibration technique. An isokinetic probe (one for each sampling flow

Table 1. Final design characteristics of the impactors.

Flow (L/min)	Nozzle Diameter D (cm)	Jet Velocity (cm/sec)	Re	S/W	L/W	$\sqrt{Stk_{50}}$	Sampler Pressure Drop (KPa)
10	0.330	1948	4280	1.56	1.2	0.51	1.1
16.7	0.392	2306	6033	1.52	1.1	0.51	1.2

Notes: Re = Reynolds number; S = distance between the acceleration nozzle exit and the impaction substrate block (cm); L = throat length of the acceleration nozzle (cm); Stk_{50} = Stokes dimensionless number for a particle having a 50% probability of impacting; and W = characteristic dimension of the impactor which is the nozzle diameter or nozzle half width for round and rectangular nozzles, respectively (cm).

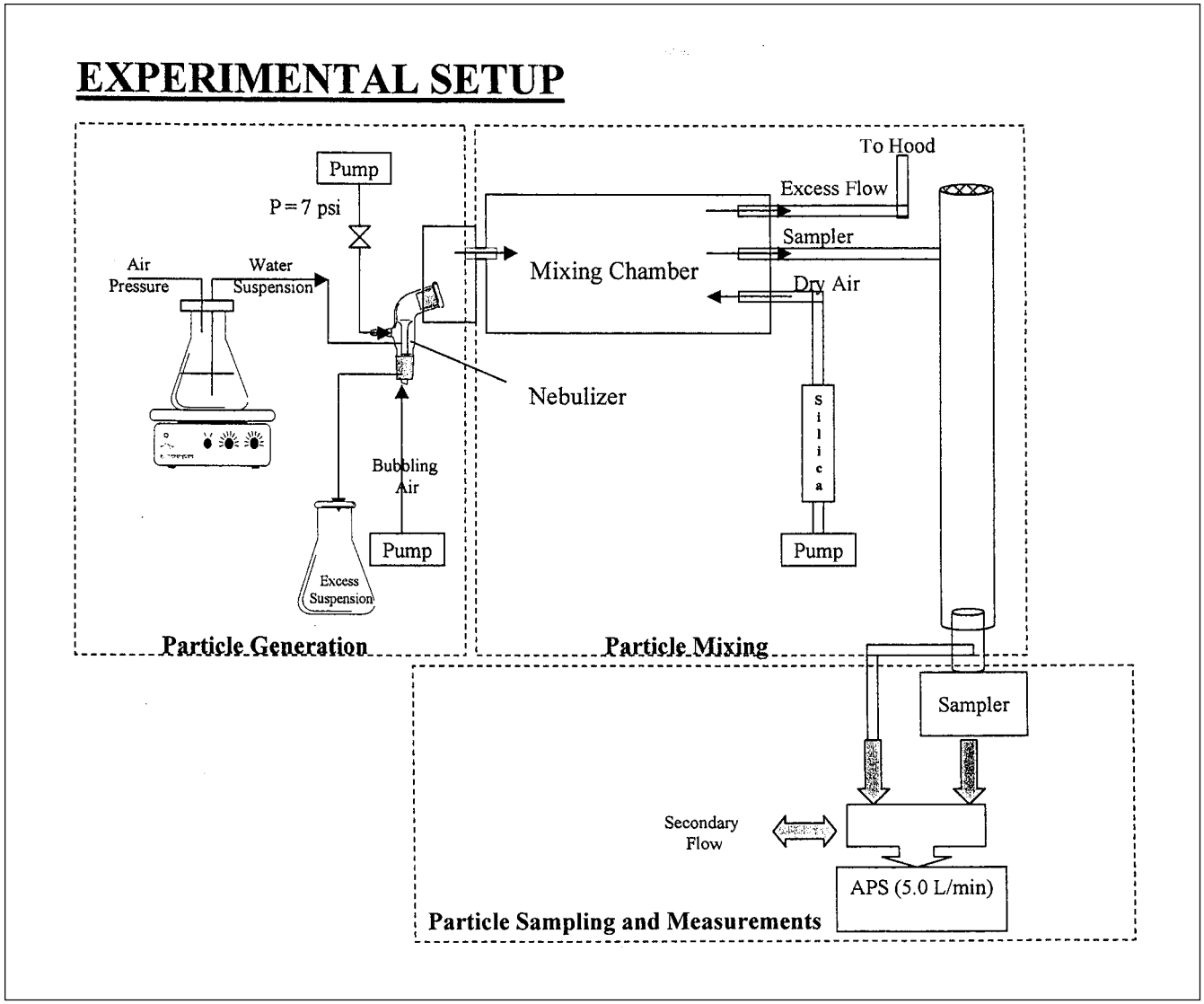

Figure 2. Experimental setup for the characterization of the sampler.

rate) was placed inside the duct ~8 duct diameters downstream from the duct entrance and was used for the upstream measurements. In each experiment, the concentration and size distribution of particles were measured for 10 min upstream, 10 min downstream, and then 10 min again upstream. An aerodynamic particle sizer (APS) (Model 3320, TSI Inc.) was used to measure particles in the size range 0.5–10 μm. At the start of each working day, the APS calibration was checked at five particle diameters (0.99, 2.13, 2.90, 4.56, and 9.14 μm) using polysterene microspheres (Polysciences, Inc.) to ensure the instrument's calibration.

For each particle size, the sampler's collection efficiency was determined as follows:

$$\text{Collection Efficiency} = \frac{C_{up} - C_{dn}}{C_{up}} \qquad (3)$$

where C_{up} and C_{dn} are the particle concentrations upstream and downstream of the impactor, respectively. Any particle losses onto the impactor's internal walls and nozzle were included in the collection efficiency measurements and calibration curves. Particle losses on the denuder section of the speciation sampler were found to be negligible.[6]

Field Validation Experiments

A series of fourteen 48-hr field validation experiments were performed on the roof of the Harvard School of Public Health in Boston, MA, between February 4 and March 27, 2000. Two FRM Well Impactor Ninety-Six systems (Rupprecht and Patashnick) were collocated with four 10-L/min and four 16.7-L/min Chemcomb speciation samplers. The samplers were assembled without the denuders. Greased impaction substrates were used in all experiments, as it was found from the laboratory tests that they perform significantly better than the oiled substrates. A razor blade was used to shave the greased surface, creating an extremely smooth surface, which, as it was also

found, significantly affects the impactor's performance (see Results section). One sampler was operated at zero flow and used as a field blank to check for any potential filter contamination during the assembly and transport of the sampler. The internal surfaces of all the samplers were cleaned with ethanol between experiments. Additionally, the sampler's greased substrates and FRM's oiled substrates were also changed between each sample. Our CHEMCOMB samplers were routinely leak-tested under a 5-in. H_2O vacuum, whereas the FRMs were periodically tested under 15-in. Hg per manufacturer's recommendations. Filters were weighed before and after sampling in accordance with EPA guidelines.[19] The Teflon filters were subsequently extracted with 0.15 mL ethanol, 0.5 mL 15-mmol NaOH, and 5 mL Milli-Q, and analyzed for SO_4^{2-} using ion chromatography (Dionex DX120).

RESULTS AND DISCUSSION

Laboratory Tests

The experimentally determined 50% cut point for 10 and 16.7 L/min flow rates is 2.2 and 2.55 μm, respectively, for the greased surface substrate. The geometric standard deviation (σ_g) of the developed impactor is 1.18 and 1.12 for 10 and 16.7 L/min, respectively. The (σ_g) parameter is an indicator of the sharpness of the collection efficiency curve. It is defined as the ratio of the aerodynamic particle diameter corresponding to 84% efficiency to the 50% cutoff point diameter.[20] The results indicate the impactor has good sharp cut characteristics ($\sigma_g < 1.2$). The effect of critical design parameters such as *S*/*W* ratio and substrate method on the impactor performance was also investigated. Two impaction substrate methods were primarily tested: oiled metal disks of various pore sizes (2, 40, and 100 μm), as well as a greased substrate.

Grease is semisolid, consisting of base oil, thickener, and additives. Base oil is critical for the impactor's performance because it wicks up by capillary action through the collected particles, providing a continuous wet surface for particle collection. Thickeners are used to increase the viscosity of the grease. Additives improve characteristics such as temperature range, corrosion resistance, adhesion, etc. A suitable grease for impaction applications has to be high in base oil, soft enough to allow particle embedding, and usable under a wide temperature range. We found that a silicone-based oil grease (Dow Corning, high vacuum grease) has an excellent performance as an impaction substrate. This particular grease, per its manufacturer, maintains its physical and chemical properties and can be used over a wide temperature range (–40 to 204 °C).

Figures 3a and b show the impactor collection efficiency as a function of the aerodynamic diameter for various impaction substrates at 10 and 16.7 L/min, respectively. A slight decrease in the collection efficiency was observed for particles larger than the impactor's cut point for oiled metal disks due to particle bounce. Bounce of particles larger than the impactor's cut point was less pronounced for larger metal disk pore sizes. Additionally, the impactor's performance was found to vary slightly with the pore size. For a pore size of 100 μm, the collection efficiency curve becomes less sharp; thus, particles smaller than the cut point appeared to be collected on the impaction substrate. The collection of particles smaller than the cut point could possibly occur because the oil was blown away from the large pores. As a result, the air streamlines penetrated into the substrate, where particles smaller than the cut point were intercepted.

The greased surface impaction substrate appears to be superior to the oiled metal disk. Collection efficiencies approached almost 100% for particles larger than the cut point. Similar results for greased surfaces were also reported by others.[21] The roughness of the greased surface was also found to be critical for the overall collection efficiency of the sampler. Two different surface types were investigated: a rough surface and a smooth surface. The rough surface was created by using an applicator to spread grease over the hollow substrate in order to make a wavy surface. A razor blade was used to shave the grease creating an extremely smooth surface. Figure 4 shows the difference in both cut point and overall collection efficiency for both 16.7 and 10 L/min flow rates for the two greased surface types. The turbulence generated due to the surface roughness is probably responsible for the losses of particles smaller than the cut point.

Any particle losses onto the impactor's internal walls and nozzle were included in the collection efficiency measurements and calibration curves. Particle losses for particles smaller than 2.5 μm as derived from the impactor collection efficiency curves are less than 10% for the final design parameters of the impactors tabulated in Table 1. The thickness of the greased surface previously has been shown to affect the overall collection efficiency of an impactor. It was found that bounce decreases with the coating thickness.[15,17] A greased coating with a thickness of a few microns can rapidly become ineffective. This is because particles accumulate on the surface and incoming particles bounce off those previously deposited.[22] A thick and smooth grease coating (0.13 cm) is a very effective impaction substrate for eliminating particle bounce, because particles can be deeply embedded into the greased surface and be wetted by the silicone oil.

A loading test was also conducted to evaluate the impactor's performance under heavy loading conditions. For this test, the previously described experimental setup and polydisperse particle generator system were used to

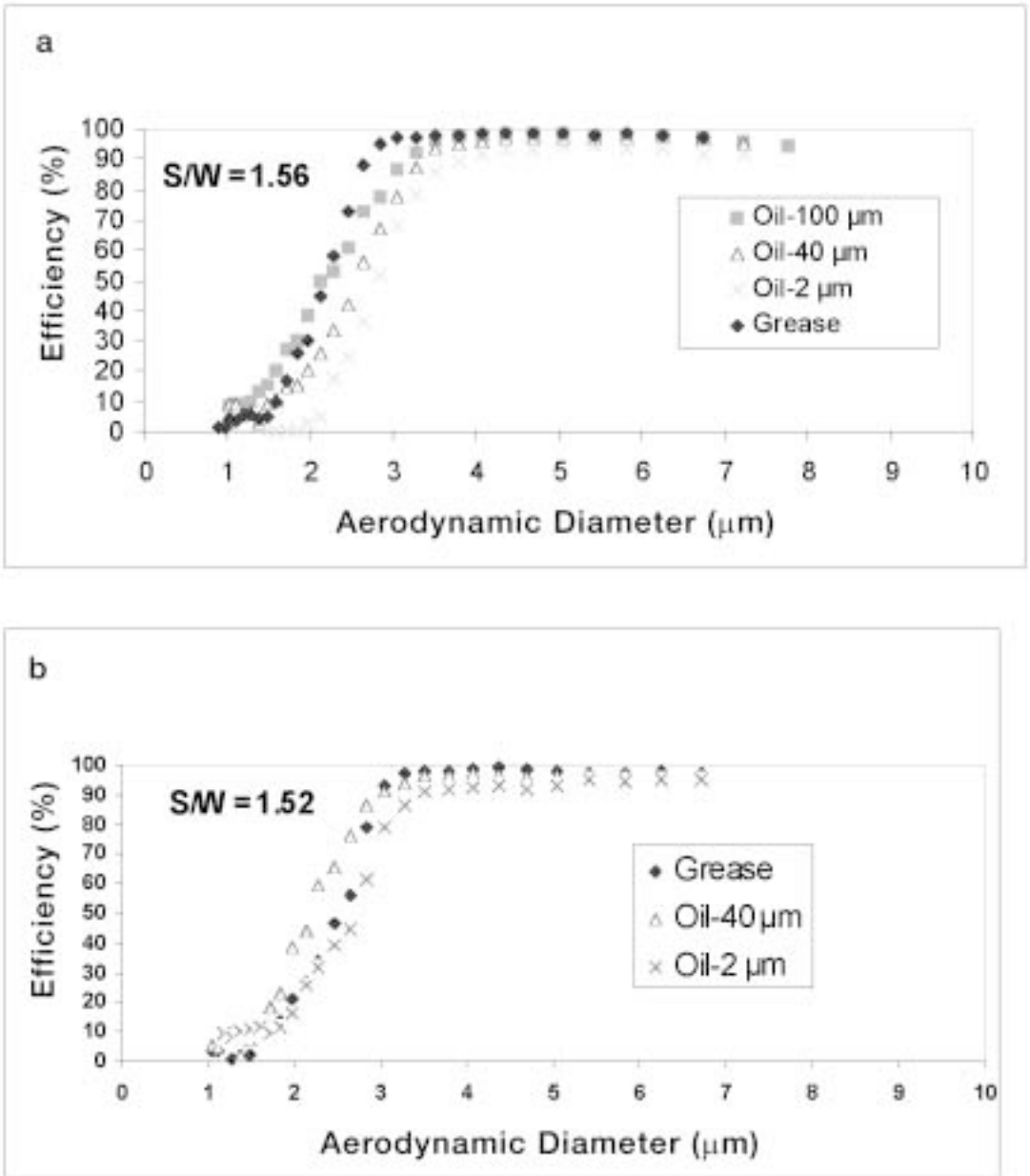

Figure 3. Collection efficiency as a function of aerodynamic diameter at (a) 10 L/min and (b) 16.7 L/min.

expose the sampler to a high particle concentration of 1400 μg/m^3 for 5 hr at a flow rate of 10 L/min. By measuring upstream and downstream of the impactor, the concentration, and the size distribution of the sampling aerosol, the amount of mass loaded on the substrate was estimated to be 3.3 mg of particles. This highly loaded greased substrate was then tested experimentally under normal loading conditions to examine the sampler collection efficiency as a function of the aerodynamic diameter. Figure 5 shows the collection efficiency of the sampler prior to and after the overloading of the impaction substrate with particles. The results showed that the overall collection efficiency and cut point of the sampler did not change after the impaction substrate was loaded. It is worth mentioning that a 100-μg/m^3 ambient concentration of particles with an aerodynamic diameter larger than the impactor's cut point would result in a 1.44-mg particle accumulation on the impaction substrate for a 24-hr sampling period, which is significantly less than the above-mentioned limit of 3.3 mg. This also implies that a 24-hr cleaning frequency and replacement of the impaction substrate is satisfactory.

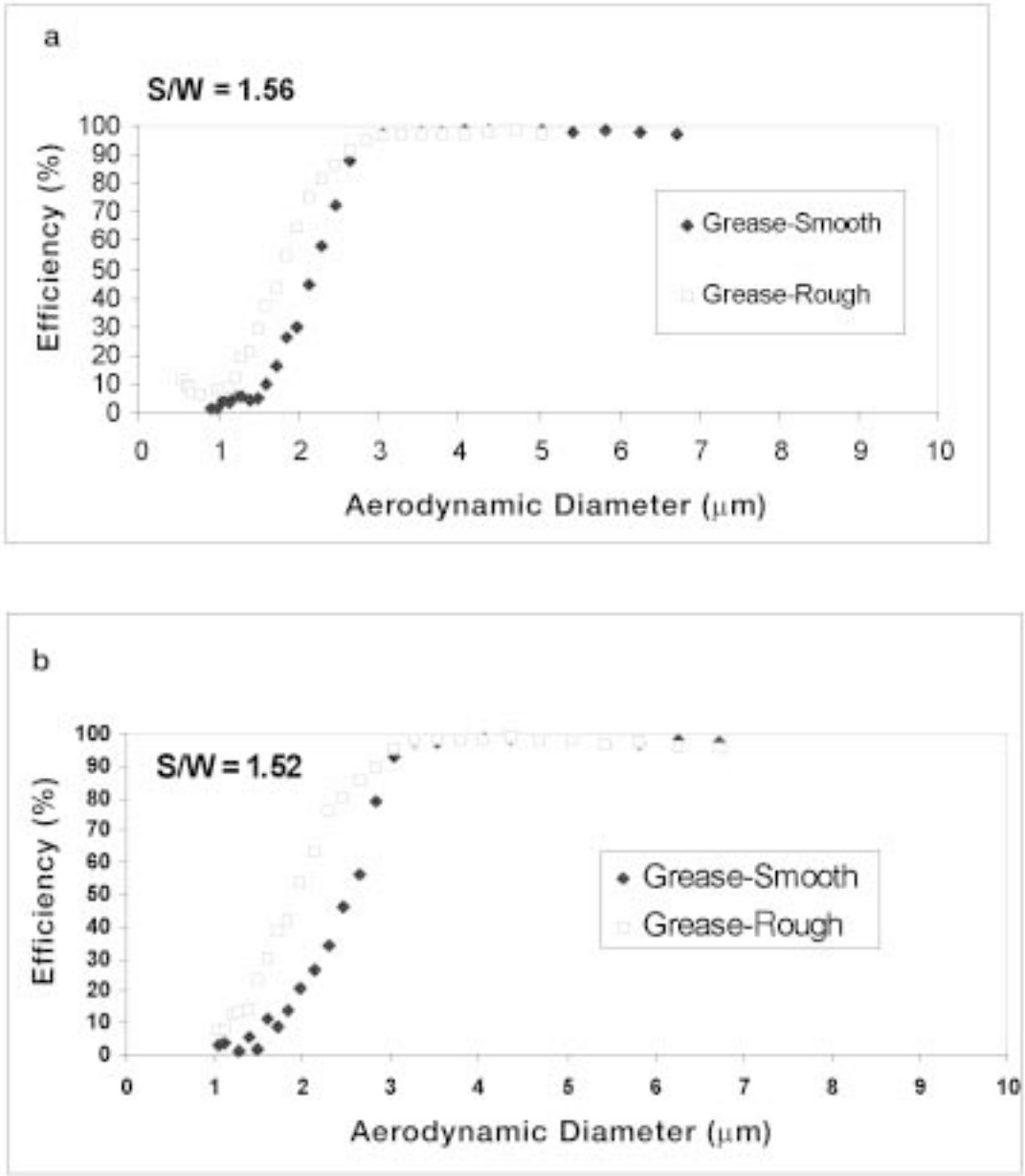

Figure 4. Collection efficiency for rough and smooth greased surface as a function of aerodynamic diameter at (a) 10 L/min and (b) 16.7 L/min.

The effect of the *S/W* ratio on sampler performance was also investigated. Figure 6 shows the collection efficiency curve for the 16.7 L/min sample for various *S/W* ratios using the greased surface impaction substrate. As can be seen, both the collection efficiency curve and particle bounce depend on the *S/W* ratio. As the distance from the nozzle exit increased, the jet dissipated before impinging the plate. Therefore, the *S/W* ratio affects both the cut point and the overall performance. In our case, it was found that an *S/W* ratio close to 1.5 would result in a sharp collection efficiency curve. Similar results were found for the 10 L/min sampler and also have been shown in other studies.[18]

Field Study

Figure 7 summarizes the $PM_{2.5}$ and SO_4^{2-} concentrations measured during the 14 field intercomparison experiments. This figure contains the following graphs: (a) $PM_{2.5}$ mass and (b) SO_4^{2-} for the 10 L/min Chemcomb, and (c) $PM_{2.5}$ and (d) SO_4^{2-} for the 16.7 L/min

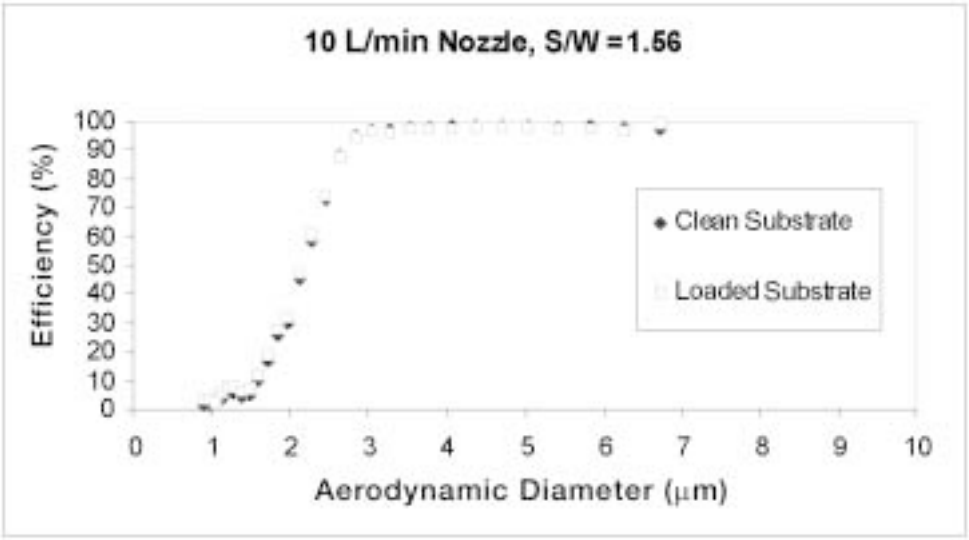

Figure 5. Collection efficiency of the 10 L/min impactor under clean and particle-loaded greased substrate.

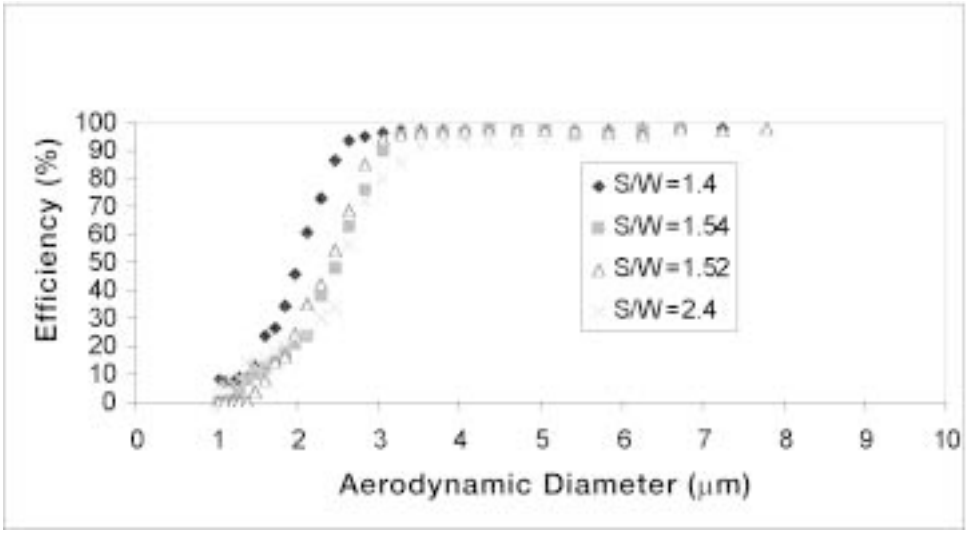

Figure 6. Collection efficiency for various *S/W* ratios.

Chemcomb. Individual Chemcomb concentrations are plotted against the average of the two FRM concentrations. Numbers in brackets indicate the 95th percentile error range for b_1 (slope) and b_o (intercept). During one of the experiments, one of the two FRMs aborted due to a large temperature difference between the filter and the inlet, whereas in the second, the oil froze, affecting collection efficiency and particle bounce. As such, the data for this experiment were not included in the regression.

Analysis of the 95th percentile error ranges indicates that for mass and SO_4^{2-}, the slope and intercept were not significantly different from unity and zero, respectively. Further, R^2 values are very close to unity. These results suggest an excellent correlation for $PM_{2.5}$ and SO_4^{2-} concentrations measured between both the 10 and 16.7 L/min Chemcombs and the FRMs.

Finally, based on the blank filter's measurements, the 24-hr limits of detection (LODs) were calculated as 3 times the standard deviation, and for the 10 L/min Chemcomb,

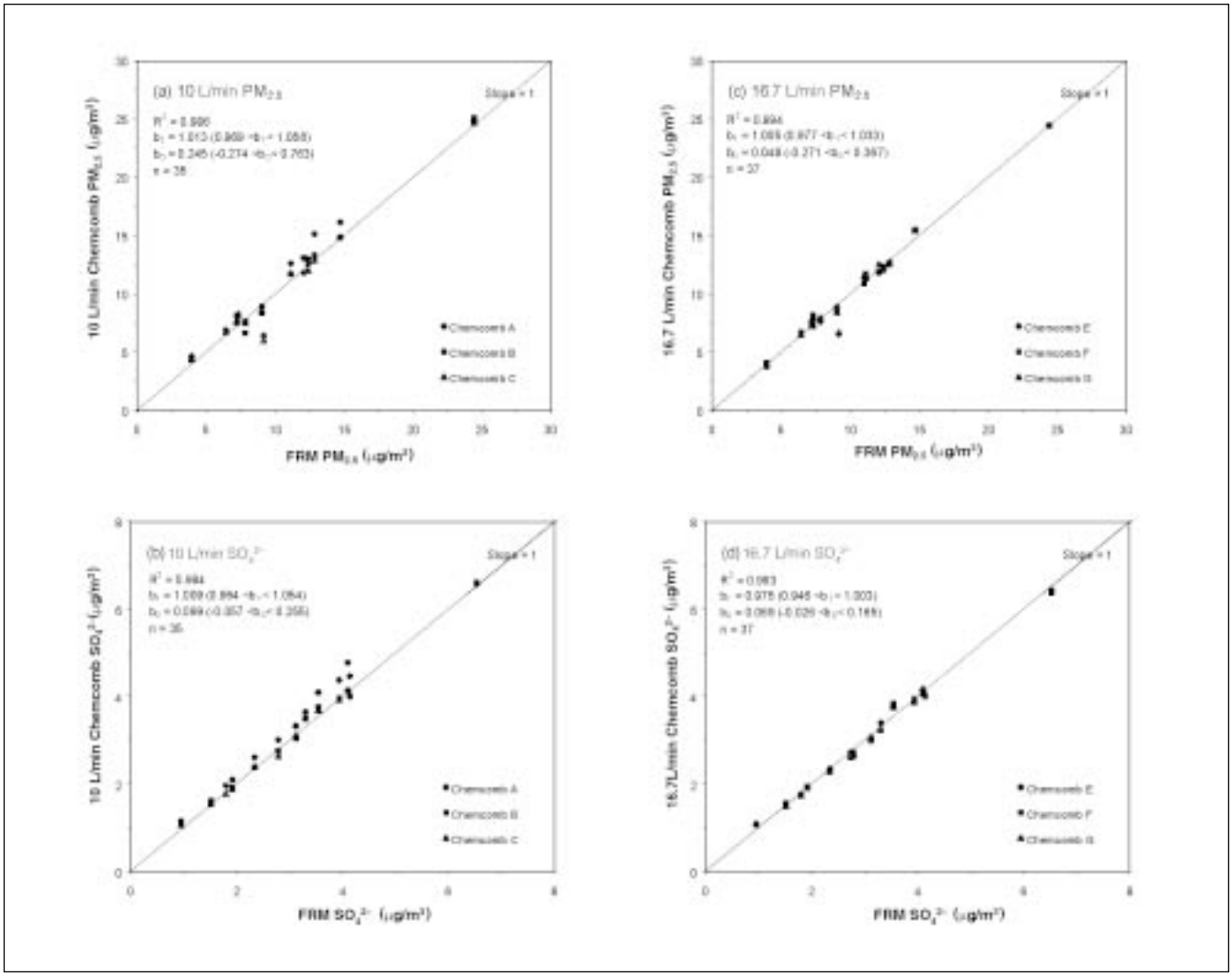

Figure 7. Graphs of $PM_{2.5}$ mass and SO_4^{2-} for both 10 and 16.7 L/min flow rates.

they were 2.53 and 0.04 μg/m³ for $PM_{2.5}$ mass and SO_4^{2-}, respectively. Similarly, for the 16.7 L/min flow rate, the 24-hr LODs were 2.10 and 0.05 μg/m³ for $PM_{2.5}$ mass and SO_4^{2-}, respectively.

SUMMARY AND CONCLUSIONS

Two conventional $PM_{2.5}$ impactors for a speciation sampler were developed and evaluated through a series of laboratory and field tests. The flow rates for these impactors are 10 and 16.7 L/min. The impactor's performance was examined as a function of critical design parameters such as Reynolds number, the distance from the nozzle exit to the impaction plate, and the impaction substrate method. The bounce of particles larger than the cut point has also been successfully minimized by using a greased surface as an impaction substrate and by optimizing the critical impactor's design parameters. The impactor efficiency curves were sharp, and particle losses were negligible. The impactors were also evaluated in field studies in which the concentrations of $PM_{2.5}$ mass and SO_4^{2-} were compared to those measured with collocated FRM samplers. The results of the field study also indicate an excellent agreement.

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