A Sampling Approach for Evaluating Particle Loss During Continuous Field Measurement of Particulate Matter

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Abstract

A method for evaluating sample bias in field measurements is presented. Experiments were performed in the field and laboratory to quantify the bias as a function of particle size for the scanning mobility particle sizer and the aerodynamic particle sizer. Sources of bias and sample loss considered in this work were sampling line loss, instrumental differences and inlet efficiencies. Measurement of the bias and sample loss allow for correction of the data acquired in the field, so as to obtain more representative samples of atmospheric concentrations. Substantial losses of fine and ultrafine particle count were observed, with sampling line losses ranging from 10–50 %, dependent on particle size. Only minor line losses were observed for coarse particles (approximately 5 %) because the sampling line was oriented vertically.

Keywords: aerodynamic particle sizer, fine particles, number concentration, scanning mobility particle sizer, size distribution, ultrafine particles

1 Introduction

The scanning mobility particle sizer (SMPS) and the aerodynamic particle sizer (APS) are routinely employed to measure the size distribution of particulate matter (PM) in a variety of applications. A considerable amount of research applications for the SMPS, APS and other continuous aerosol samplers is in the area of atmospheric sampling (e.g., Harrison et al. [1,2], Hughes et al. [3,4], Morawska et al. [5], Shi et al. [6,7], Woo et al. [8]). However, applications of continuous samplers have broadened into other areas of research including occupational and industrial hygiene (Görner et al. [9], Thornburg and Leith [10], Zimmer and Biswas [11]), exposure assessment (Abt et al. [12], Koponen et al. [13], Miller and Nazaroff [14]), pharmaceutical product development and characterization (Mitchell et al. [15], Noble and Prather [16], Srichana et al. [17]) and process monitoring (Chisholm [18], Huang et al. [19], Ma et al. [20]).

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In many non-atmospheric applications, the particle sampling instruments are located in the immediate vicinity of the aerosol source or at a spatial proximity that is appropriate for the analysis (e.g., Noble and Prather [16], Thornburg and Leith [10], Zimmer and Biswas [11]). In contrast, atmospheric research often utilizes a permanent building, temporary shelter or mobile vehicle for location of the samplers. Temporary shelters are often designed specifically for the purpose of atmospheric sampling. As a result, the distance from the ambient atmosphere to the instrument is relatively short and, by implication, not substantially perturbing to the particles being sampled. However, permanent buildings that are employed to house instrumentation are typically not designed for the purpose of atmospheric sampling. Therefore, the outdoor ambient air sample must be drawn through a sampling line that can be several meters in length or more.

General practice for atmospheric sampling through sampling lines and manifolds include maximizing line cross section, minimizing sampling line length and minimizing sharp bends. However, in many situations practical considerations limit the options available for sampling line designs, which can lead to excessive particle loss in select size ranges as a function of particle diameter (d). Primary causes for particle losses through the sampling

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line include diffusion, impaction, sedimentation, electrostatic precipitation and thermophoresis. Additional sample biasing may occur internally in the monitoring device (Kinney and Pui [21]).

This paper describes a sampling approach designed to evaluate sample biasing during field measurements. The purpose of this paper is to demonstrate simple procedures for empirically deriving correction factors that accurately describe sample bias in field measurements of ambient PM.

2 Experimental Methods

Field experiments were performed in El Paso, TX (Noble et al. [22]) and Fresno, CA (Lawless et al. [23]). For the sake of discussion in the following section, the experimental methods and field setup at El Paso are described briefly; greater details are provided in Noble et al. [22] and Lawless et al. [23].

Using two coupled pairs of the SMPS (TSI Model 3936) and the APS (one TSI Model 3320 and one TSI Model 3310), continuous size distributions were collected for atmospheric aerosols in El Paso, TX. Sampling for the SMPS and APS was performed in 15 min windows. The SMPS measured the electrical mobility size distribution of PM over the size range of 0.02–0.7 μm; the APS measured the aerodynamic size distribution of PM over the range 0.5–20 μm.

Sampling occurred from 28 Nov 1999 through 18 Dec 1999 at two sites in urban El Paso. The first sampling site was in downtown El Paso, approximately 2 m from the nearest road. The particle sizers were located inside an instrumentation trailer owned by the Texas Natural Resource Conservation Commission (TNRCC, since renamed the Texas Commission on Environmental Quality, TCEQ). Ambient samples were brought through the trailer roof by a 3 m vertical aluminum tube (2.54 cm inner diameter) and connected directly to the SMPS and APS. The second sampling site was at Chamizal National Memorial, a park-like setting with no large buildings in the immediate vicinity. The samplers were located within a gated area, inside of a small transportable shelter. The instruments were connected to the ambient atmosphere with approximately 15 cm of conductive rubber tubing (~1 cm inner diameter). At both sites, the sampling line inlet was capped with a TSP sampling inlet for protection against rain and large objects.

In order to evaluate sample bias of the ambient field data, instrumental calibration experiments were performed in the field and in the laboratory at Research Triangle Park, NC. In the field, indoor and outdoor sampling was performed at the Downtown site, in order to determine particle loss through the sampling line. During this experiment, the two trailer doors were left open to allow airflow through the trailer.

3 Results and Discussion

There are various potential mechanisms for sample loss and biasing, as well as multiple physical variables affecting those mechanisms. Due to difficulties in theoretically accounting for sample bias, particle loss measurements may be performed experimentally (e.g., Armendariz and Leith [24], Kinney and Pui [21], Sioutas et al. [25,26], Thornburg et al. [27], Virtanen et al. [28]).

3.1 Sampling Line Losses

As mentioned, the sampling line for the instrumentation at the Chamizal site was only 15 cm long and, therefore, was considered to have a negligible effect on the measurements. However, the sampling line at the Downtown site was 3 m in length and was considered to be a potential source for sample loss.

In order to evaluate line losses, five consecutive runs were performed at 5 min intervals, alternating between indoor sampling of the trailer and outdoor sampling through the sampling line. During this test, both doors of the instrumentation trailer were open, allowing for a cross breeze to pass through the trailer. The results from these runs are shown in Figure 1 as the size-dependent transmission efficiency, E_{trans}, which is defined as

$$E_{trans} = \frac{N_{out}}{N_{in}} \tag{1}$$

where N_{out} is the number concentration as measured coming out the sampling line and N_{in} is the number concentration measured indoor, which is assumed to be at ambient conditions and, therefore, the concentration which is coming into the sampling line. The solid line on Figure 1 shows the smoothed curve that was applied to the raw data to correct for sampling line losses.

As is obvious from the uncertainty bars, which span the range of the ratio determined by these measurements, there is a substantial variability observed. This appears to be due, at least in part, to the dynamic temporal variability of ultrafine ($d < 0.1$ μm) and fine ($d < 1$ μm) particles, and the rapid rate at which these particles respond to ambient conditions. Because this sampling site was located in the immediate vicinity of a major highway, short elevations or depressions in traffic density could easily account for substantial shifts in the measured number concentrations. Likewise, immediate variations in wind speed and direction could also serve to randomly dilute PM number concentrations.

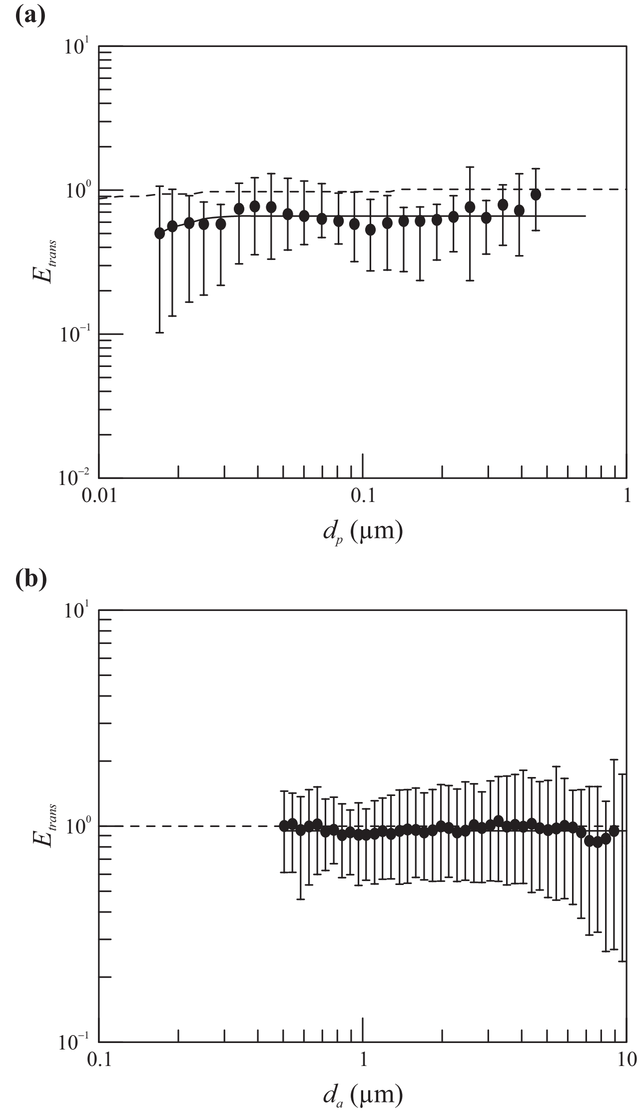

Fig. 1: Transmission efficiency through the 3 m vertical sampling line as a function of particle size. Uncertainty bars indicate the range of transmission efficiency. Solid line indicates smoothed correction applied to the ambient El Paso data. Dashed line shows the theoretical transmission efficiency based on losses due to diffusion. (a) Particle diameter measured with the SMPS. (b) Aerodynamic diameter measured with the APS.

Because the sampling line was vertically oriented with only slight bends for connection to the sampling instruments, it was anticipated that the majority of particle losses would occur for the ultrafine and fine particles through diffusion to the surface of the sampling line. Few losses for accumulation mode ($0.1\,\mu m < d < 1\,\mu m$) particles were expected. In order to complement the experimental data, a simple diffusion model was applied to compare the measured results with theoretical results (Hinds [29]). The theoretical efficiency through the sampling line, E_{theor}, is calculated as

$$E_{theor} = \frac{N_{out}}{N_{in}} \tag{2}$$

for two specific cases:

$$E_{theor} = 1 - 5.50\,\mu^{0.667} + 3.77\,\mu, \text{ for } \mu < 0.007 \tag{3}$$

$$E_{theor} = 0.819e^{-11.5\mu} + 0.0975e^{-70.1\mu} + 0.0325e^{-179\mu}, \text{ for } \mu > 0.007 \tag{4}$$

where μ is the dimensionless deposition parameter, as defined by

$$\mu = \frac{DL}{Q} \tag{5}$$

where L is the tube length, Q is volumetric flow rate and D is the particle diffusion coefficient, which is a function of temperature, particle diameter and particle slip correction factor. The results from the model are shown in Figure 1 as a dashed line.

It is obvious from the figure that the theoretical diffusion losses should be minimal for a sampling line of only 3 m in length, with transmission efficiencies close to unity over the range sampled, especially for the particles analyzed by the APS. However, experimentally, the transmission efficiency is found to be substantially lower than the theoretical value for both ultrafine and fine particles. The coarse mode ($d > 1\,\mu m$) particle transmission is only slightly lower than what is predicted theoretically. The losses greater than that which is predicted by the model may result from other loss mechanisms such as impaction, electrostatic precipitation and thermophoretic precipitation, all of which add to the complexity of the transfer loss model. Essentially, the model becomes complicated enough, that the most practical solution is simply to measure for transfer losses (Flagan [30]).

Sampling line losses also were measured in two previous field campaigns in Fresno, the results of which are shown in Figure 2. During the previous studies, sampling arrangements and setup were different from those employed in El Paso (Lawless et al. [23]). The samplers (one SMPS and one laser particle counter; Particle Measuring System, Inc. Model LASX) were located indoor in a retirement center. Outdoor sampling was performed through a horizontal sampling line of approximately 6 m in length (~1 cm inner diameter). During Fresno 1, the sampling line was made of polypropylene; during Fresno 2, the sampling line was made of a conductive rubber material.

The uncertainty bars from the two previous studies are not shown in Figure 2, but are comparable in magnitude to those calculated during the current study. Taking the uncertainty bars into account, the results from the pre-

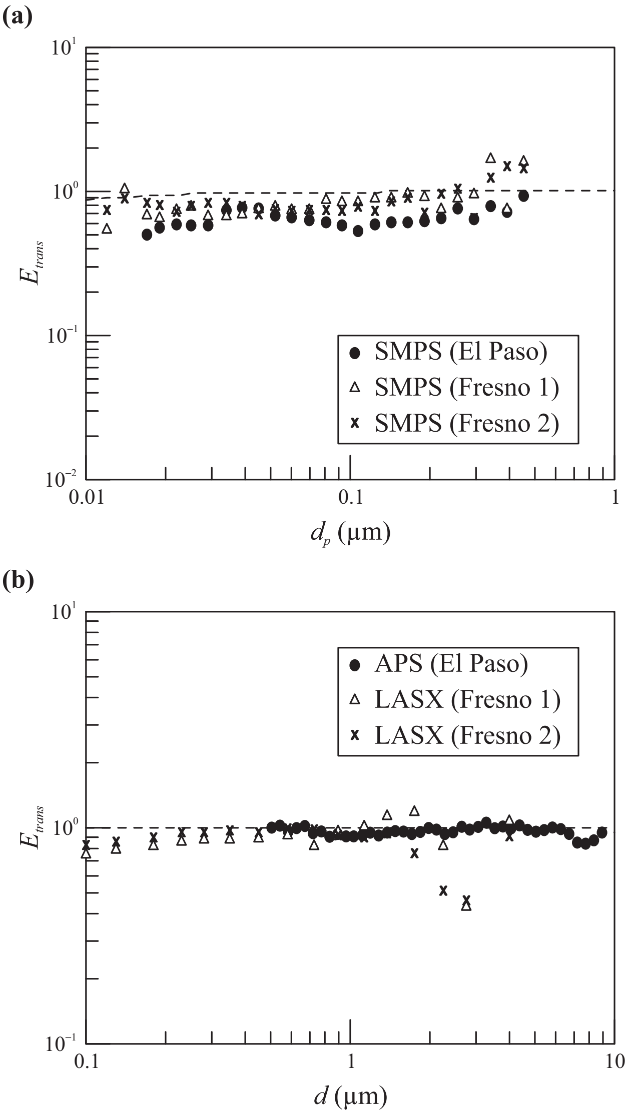

Fig. 2: Comparison of sampling efficiency during current field study in El Paso, TX, with transmission efficiencies measured during two previous field studies in Fresno, CA. Dashed line shows the theoretical transmission efficiency based on losses due to diffusion. (a) Particle diameter measured with the SMPS. (b) Aerodynamic diameter measured with the APS and light scattering diameter measured with the LASX laser particle counter. Note that the x-axis is labeled as *d* because both aerodynamic diameter and light scattering diameter are plotted on this axis.

vious measurements of sample line losses are equivalent to the current results. Though there were differences in sampling setups, several general observations can be made in comparing the three data sets. Transmission efficiency was observed to be between 0.5–0.9 for particles smaller than 0.1 μm. Transmission efficiency tends to increase for particles larger than 0.1 μm, with close to unity transmission for particles greater than 0.8 μm. Above 1 μm, particle transmission remains near unity for the vertical sampling line (El Paso) but begins to deteriorate for the horizontal sampling line (Fresno 1 and 2).

3.2 Inter-instrumental Differences

Differences in instrumental measurements can be accounted for largely by instrumental bias (one or more instruments consistently sampling higher or lower than other instruments) and also by instrumental imprecision (one or more instruments not able to provide consistent results for equivalent samples).

In order to account for inter-instrumental differences, both pairs of instruments were run side-by-side in the laboratory following the El Paso field campaign. Sixteen consecutive scans of 15 min each were performed for this comparison. The results from these runs are expressed graphically in Figure 3 as the ratio of the instrumental sampling efficiencies, E_{instr}, which is defined as:

$$E_{instr} = \frac{N_{down}}{N_{cham}} \tag{6}$$

where N_{down} is the measured number concentration of the sampler that was located at the Downtown site and N_{cham} is the measured number concentration of the sampler that was located at the Chamizal site. In Figure 3, the value for the ideal instrumental comparison, unity, is shown by the dashed line. The applied correct curve is shown in Figure 3 as a solid line.

In contrast to the line loss measurements, the uncertainty bars for the instrumental intercomparisons have a broad range (approximately one order of magnitude) of relatively low uncertainty, indicating that the instruments are very stable for those specific particle sizes. Both the SMPS and APS intercomparisons show the greatest uncertainty in the largest size channels of each instrument. For the SMPS, the instruments show significant sample-by-sample variations for particles larger than approximately 0.5 μm. For the APS, the large sample-by-sample variation occurs for particle greater than 3 μm. Both instrumental bias and imprecision can be observed from these data. In general, instrumental biasing is demonstrated by data points that substantially depart from unity; instrumental imprecision is demonstrated by large uncertainty bars.

A moderate instrumental bias is exhibited by the SMPS units for several size channels near 0.1 μm; in this size range, the SMPS from the Downtown site is consistently measuring 70–90 % of the particle count measured by the other SMPS. A more substantial instrumental biasing may be observed in the APS intercomparison in the lower and higher size channels, where E_{instr} is consistently less than and greater than unity, respectively. For the four lowest APS channels, the APS from the Down-

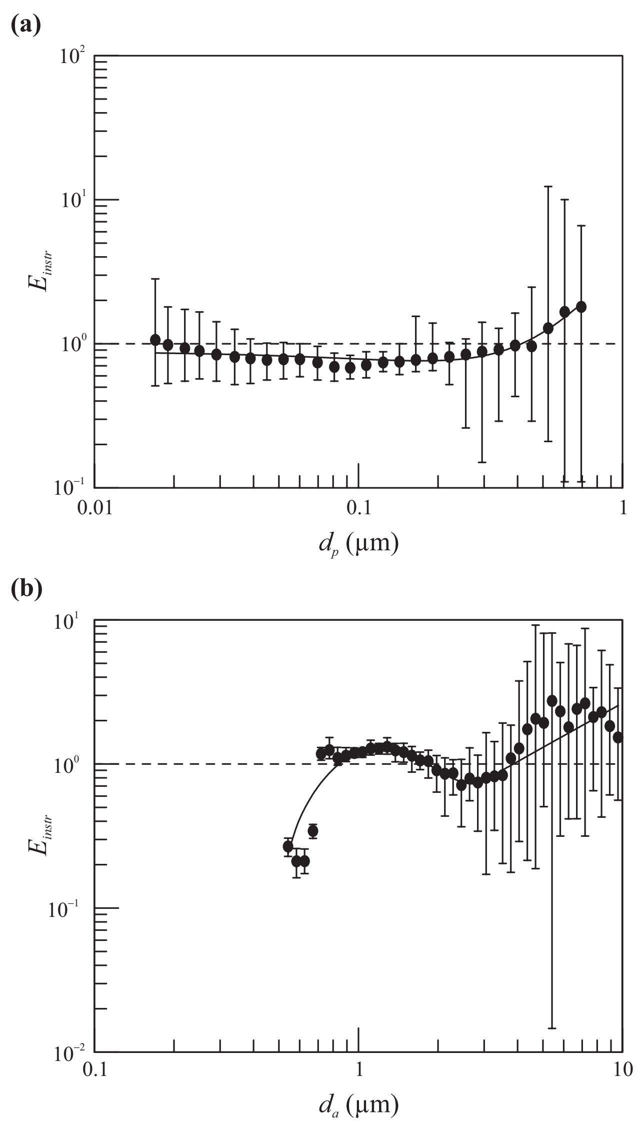

Fig. 3: Transmission efficiency intercomparison measured in the laboratory between similar instruments. Uncertainty bars indicate the range of transmission efficiency. Solid line indicates smoothed correction applied to the data. Dashed line shows the ideal transmission of unity, which would result if both instruments sampled identically. (a) Particle diameter measured with the SMPS. (b) Aerodynamic diameter measured with the APS.

town site is consistently measuring 20–40 % of the values measured by the other unit. The bias observed in these lower size bins is a repeatable (precise) bias, as indicated by the limited range of the uncertainty bars. For the size channels above 4 μm, the APS from the Downtown site is repeatedly measuring approximately 100 % higher particle count than the other instrument. Although the bias definitely exists in these higher channels, the bias is not as consistent (precise) as the bias observed in the lower size bins, as indicated by the large range of the uncertainty bars.

Instrumental imprecision is demonstrated at the high size channels by the large uncertainty bars. The magnitude of the uncertainty bars can be explained largely by the fewer number of particles counted in the higher channels, as is shown in Figure 4. The increase in uncertainty that occurs at the larger particle sizes corresponds to particle number concentrations that are 2–3 orders of magnitude less than the maximum concentration. At these lower concentrations, the detection of one additional particle per channel would account for a relative

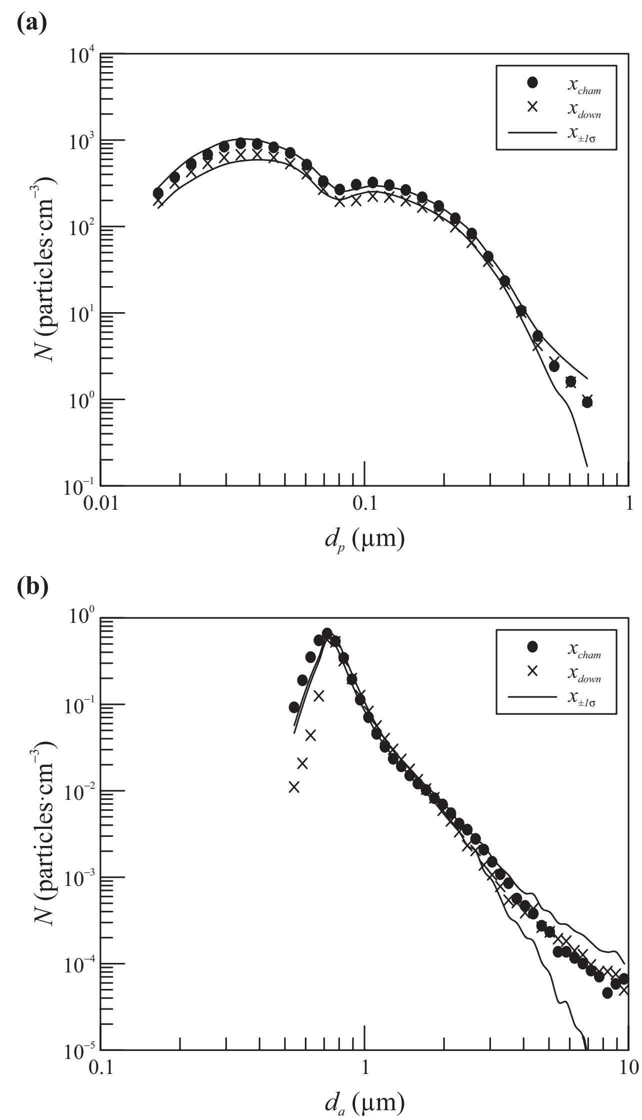

Fig. 4: Average size distributions measured in the laboratory with collocated samplers. Aerosol sampled for this intercomparison is ambient room air. Data points show the average of sixteen measurements for each instrument; x_{cham} and x_{down} are the averages, by size bin, for the samplers that were located at the Chamizal and Downtown sites, respectively. Curves ($x_{\pm 1\sigma}$) indicate the intersampler average plus and minus one standard deviation. (a) Particle diameter measured with the SMPS. (b) Aerodynamic diameter measured with the APS.

change of 100–1000 % of the measurement in comparison to the higher concentration values. Adding to this problem for the APS units is the low absolute particle count during a 15 min measurement interval. For the larger size channels which are measuring particle concentrations in the range of 10^{-4}–10^{-3} particles · cm^{-3}, these concentrations correspond to absolute counts of approximately 1–15 particles in a 15 min period. During a 15 min sampling period, measuring one particle more or less should not be surprising, yet it results in a relatively high degree of imprecision when comparing two instruments.
With corrections measured for line losses and instrumental differences, the correction for sample losses and biasing may be applied to the ambient data acquired in El Paso.

3.3 Ambient Sampling

In addition to correcting for line losses and instrumental differences, corrections to the APS data were also included for APS inlet efficiency (Kinney and Pui [21]). The results from these three corrections are applied to ambient data acquired during the field study in El Paso and are shown in Figure 5 for data sampled at the Downtown site; both uncorrected and corrected data are shown. Although there are no unexpected transformations in the data on applying the corrections, the corrected data are thought to provide results that are more representative of the atmospheric aerosol than does the raw, uncorrected data.
For the SMPS, the overall shape of the size distribution is not changed substantially by the corrections. However, the magnitude of the ultrafine mode is approximately doubled with the corrections taken into account. For the APS, both the magnitude and shape of the size distribution are observed to be changed in such a way that draws out the modality of the distribution more clearly. Specifically, both the accumulation mode peak at 0.6 µm and the coarse mode peak at 2 µm are increased in concentration, while the inter-modal region at 1 µm is lessened, thus enhancing the definition of the size distribution.
In general, the correction for the number concentration impacts the SMPS results more so than the APS results. This is largely due to the profile of the correction curve applied to the raw data. For all size channels, the correction results in higher particle counts for the SMPS; in contrast, the corrections for the APS do not result in higher particle counts for all channels. For the APS size bins near 1 µm, the correction results in a lower particle count than is measured by the instrument. (Note that this apparent depression in the correction curve is largely influenced by instrumental intercomparison; that is,

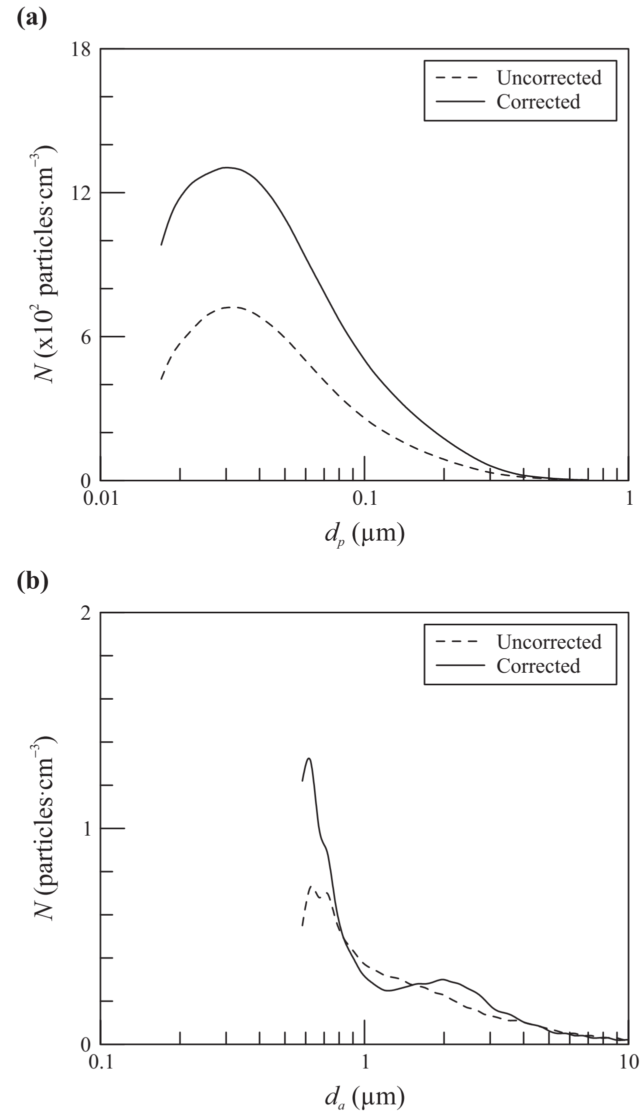

Fig. 5: Time-averaged size profile of atmospheric particles measured at the Downtown site over the entire twenty-one day sampling period. Number concentrations are corrected for line losses, instrumental differences and inlet efficiency; corrections are applied as a function of sampling channel. Both uncorrected and corrected size distributions are shown. (a) Particle diameter measured with the SMPS. (b) Aerodynamic diameter measured with the APS.

in the channels near 1 µm the values for E_{instr} are greater than unity, as shown in Figure 3.) As a result, the size-summed corrections to APS particle count do not substantially change the results in this application to ambient data.
Although not addressed in detail in this paper, the potential for phantom particle counts should also be considered in performing ambient measurements (Heitbrink et al. [31,32]). Phantom particles were not corrected for in this analysis because phantom counts only

influence time-of-flight spectrometers, such as the APS, and only in size bins where there is a relatively low particle count. In the APS sample from El Paso, particles in the size range of 5–10 µm accounted for only 5.0 % of particles (by count) in the 1–10 µm range, and only 1.6 % of total particles counted by the APS. Based on these values and dividing this correction across multiple size bins, the correction for phantom particles would change any size bin less than ~0.2 %.

4 Conclusion

Routine field sampling of ambient particulate matter can introduce substantial sample bias into the collected sample or data by means of diffusion or gravitational loss in the sampling line, inter-instrumental differences, inlet efficiencies and other loss mechanisms. Particle losses for individual experiments are highly influenced by instrumental setup, with losses varying as a function of particle size.

Data has been presented from three field campaigns, demonstrating a sampling approach to quantitate sample bias. Sample losses from SMPS measurements were observed to range from 10–50 %, dependent on particle size. Only minor particle losses were observed for the APS, at approximately 5 %. Determination of sample bias allow for correction of data acquired in the field, so as to obtain samples that are more representative of the atmosphere.

As demonstrated, the sampled size distributions (both shape and concentration) can be different than that of the ambient particulate matter. Accounting and correcting for sample biasing and losses is a potentially valuable step in the process of continuous aerosol sampling in order to obtain a more atmospherically representative sample. These corrections are especially important when attempting to correlate the PM data to other data such as gas phase concentrations, meteorological or climatological data, or health end points, such as morbidity or mortality.

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