On the Performance Parameters of PM_{2.5} and PM₁ Size Separators for Ambient Aerosol Monitoring

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ABSTRACT

The performance of particle size separators that are characterized by their cutpoint (d_{50}) and the steepness of their penetration curves (σ_g) is a critical feature of measurement methods for ambient PM_{2.5} and PM₁ monitoring. The parameters d_{50} and σ_g for PM_{2.5} size separators are regulated in the official measurement methods of many countries and regions. However, those parameters for PM₁ size separators have not been previously specified. The interaction of particle size distributions (PSDs) and the particle size separator performance characteristics has significant influence on the PM sampling results. Atmospheric PSDs for each of the four seasons covering a range of particulate pollution concentrations were measured in urban Beijing. The relative differences (RD) between estimated PM_{2.5}/PM₁ sampler concentrations and actual concentrations associated with the interaction between performance characteristics of size separator and actually measured urban PSDs, as well as for three idealized ambient PSDs, have been systematically investigated. For the PM_{2.5} size separator, when tolerance of cutpoint is $2.5 \pm 0.2 \mu\text{m}$ and σ_g is no greater than 1.3, RD values for most typical urban atmospheric aerosol conditions were within the permissible error range ($\pm 5\%$). For the PM₁ size separator, when tolerance of cutpoint and sharpness is $1.00 \pm 0.02 \mu\text{m}$ and σ_g no greater than 1.2, respectively, the RD values for typical urban atmospheric aerosol conditions are within the permissible range. Those parameters could also meet the requirements for sampling of the idealized ambient aerosol distribution.

Keywords: PM_{2.5}; PM₁; Size separator; Cutpoint; Sharpness; Particle size distribution.

INTRODUCTION

PM_{2.5} (particles with aerodynamic diameter < 2.5 micrometers [μm]) has been promulgated as an indicator in ambient air quality standards (AAQS) of many countries and regions, such as the United State of America (USA), the European Union (EU), Japan, and China (Cao *et al.*, 2013). The regulatory limits on the ambient mass concentration on annual and daily average bases have been set. To regulate PM_{2.5}, accurate measurement of its concentrations is crucial (Cheng, 2018; Lin *et al.*, 2018). Accordingly, the measurement methods for PM_{2.5} were specified along with the implementation of AAQS. Factors influencing the accuracy of the measured results include ambient aerosol concentration and composition, particle size distribution (PSD), sample volumetric flow rate, sampling time, sampler

inlet geometry, the performance of the sampler's size-selective separator, sampler internal particle losses, and all of the associated sampling and analysis procedures (Vanderpool *et al.*, 2001).

Among these factors, the performance of the particle size separator is a critical feature of the measurement method. The PM_{2.5} sampler generally utilizes a separator, such as impactor or a cyclone to remove PM greater than $2.5 \mu\text{m}$ from the airstream. The performance of the particle size separator is normally described by either a collection efficiency curve or penetration curve. The penetration curve is characterized by its cutpoint (d_{50}) and the steepness of the selection curve (σ_g). The cutpoint is the particle aerodynamic diameter at which probability of penetration through the size separator is 50%. The steepness of the penetration curve is calculated as the square root of the particle diameter associated with 16% penetration (d_{16}) divided by the particle diameter associated with 84% penetration (d_{84}). The steepness is equivalent to the geometric standard deviation of a cumulative lognormal curve fitted to the penetration data, and both are often interchanged (Kenny and Gussman, 2000; Peters *et al.*, 2001).

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The U.S. EPA (2013) requires PM_{2.5} samplers to have a cutpoint of $2.5 \pm 0.2 \mu\text{m}$, but no steepness values for the sampler are listed because there is a design standard outlined in 40CFR50, Appendix L (https://www.law.cornell.edu/cfr/text/40/appendix-L_to_part_50). The design standard specifies the construction of the WINS impactor (Peters *et al.*, 2001a, b) or the use of a very sharp cut cyclone (VSCC) (Kenny *et al.*, 2000, 2004). The VSCC has a σ_g of 1.16 (Kenny *et al.*, 2004) while the WINS impactor σ_g is 1.18 (Peters *et al.*, 2001a). The European Committee for Standardization (2014) do not specify d_{50} and steepness values, but adopts a design standard outlined in EN 12341 and specifies the use of an impactor size separator with d_{50} and σ_g around $2.5 \mu\text{m}$ and 1.2, respectively. The Japanese Industrial Standards Committee (JISC, 2008) and Ministry of Environmental Protection of China (MEP, 2013) regulated both σ_g and d_{50} values. Both stipulated the same d_{50} values as U.S. EPA. JISC (2008) required $\sigma_g = d_{20}/d_{80} < 1.5$, where d_{20} , d_{80} are the particle diameter associated with 20%, and 80% penetration, respectively. MEP (2013) regulated $\sigma_g = 1.2 \pm 0.1$, where σ_g is defined as d_{50}/d_{16} , and d_{84}/d_{50} , d_{16} , d_{50} and d_{84} were the particle diameter associated with 16%, 50%, and 84% collection efficiency, respectively. In general, d_{50}/d_{16} , and d_{84}/d_{50} in MEP (2013) are comparable in steepness.

Peters *et al.* (2001a) measured the penetration curves of five PM_{2.5} size separators widely used in the speciation samplers and integrated the curves with three idealized ambient PSDs to obtain estimates of the measured PM_{2.5} concentration and predict bias relative to the PM_{2.5} reference size separator. d_{50} and σ_g measured for five separator ranged from 2.35 to 2.72 μm , 1.25 to 1.30, respectively. Estimated PM_{2.5} concentration biases relative to the reference separator were within 5% for the idealized fine and typical ambient distributions for all separators. Bias ranged between 4% and 8% for the idealized coarse distribution, relatively higher than for the fine and typical distributions. Buser *et al.* (2007) theoretically estimated the PM_{2.5} sampling errors associated with the interaction of the PSDs and the sampler performance characteristics. In their study, d_{50} and σ_g were selected as $2.5 \pm 0.2 \mu\text{m}$, $\sigma_g = 1.3 \pm 0.03$, respectively, and PSDs were three idealized ambient PSDs and other assumed PSDs. They also found that differences between the actual sampler and the EPA idealized sampler were within 5% for the idealized fine and typical ambient distributions, while differences exceeded 5% when d_{50} was 2.5 μm and σ_g was 1.33, or d_{50} was 2.7 μm and σ_g was 1.3 ± 0.03 for an idealized coarse distribution. For assumed PSDs with mass median diameter from 5 to 20 μm and geometric standard deviation 1.5 to 2.0, differences ranged from 8% up to 1214%. These studies indicated that the interaction of the PSDs and the sampler performance characteristics had significant influences on the PM_{2.5} sampling results.

Recently, increasing frequency of PM₁ (particle with aerodynamic diameter $< 1 \mu\text{m}$) measurements have been reported (Bari *et al.*, 2015; Qiao *et al.*, 2015; Wang *et al.*, 2015; Galindo *et al.*, 2016; Kunwar *et al.*, 2016). Since PM₁ has not been adopted as a criterion in current AAQS regulations, these measurement methods including the

performance metrics of the PM₁ size separators have not yet been specified.

The objectives of this paper are: (1) Investigate the relative differences (RD) between estimated PM_{2.5} sampler concentrations and actual PM_{2.5} concentrations associated with the interaction between performance characteristics for PM_{2.5} size separator and actually measured typical urban PSDs in four seasons covering all pollution levels and thereby, further evaluate the existing performance parameters for a PM_{2.5} size separator under real world conditions of high PM_{2.5} concentrations; (2) Investigate the relative differences (RD) between estimated PM₁ sampler concentration and actual PM₁ concentrations associated with the interaction between performance characteristics for a PM₁ size separator and actually measured typical urban PSDs, and propose the performance parameters for PM₁ size separator; and (3) Evaluate the performance parameters for PM₁ size separators for the three idealized ambient PSDs developed by Vanderpool *et al.* (2001).

METHODS

Method General Description

Previous studies have elucidated the basic form of atmospheric PM size distribution (PSD), and typical PSDs showed one or more lognormal shapes (Whitby *et al.*, 1972; Liu *et al.*, 1974). In this case, the lognormal mass distribution can be expressed as:

$$f(d_p, MMD, GSD) = \frac{1}{d_p \ln GSD \sqrt{2\pi}} e^{-\frac{(\ln d_p - \ln MMD)^2}{2(\ln GSD)^2}} \quad (1)$$

where d_p , MMD and GSD is the particle diameter, mass median diameter, and geometric standard deviation of PSD, respectively (Hinds, 1999).

PM samplers have been widely used to collect information on atmospheric PM. For sampling specific PM sizes such as PM₁₀, PM_{2.5}, and PM₁, the corresponding size separators are adopted. For an ideal size separator, all particles greater than the specific size should be collected by the size separator, and all particle less than or equal to the specific size should penetrate it and be captured by the subsequent filter. The penetration curve of an ideal size separator is a step function as follows:

$$P_t(d_p, d_{50}) = \begin{cases} 1 & \text{if } d_p \leq d_{50} \\ 0 & \text{if } d_p > d_{50} \end{cases} \quad (2)$$

However, it is not possible to design a size separator with an ideal cut. In an actual PM sampler, a portion of PM greater than the specific size that should be collected by the size separator will penetrate and be collected by the filter (over-collected) while a portion of PM less than or equal to the specific size that should be captured by the filter will be collected by the size separator (under-collected). If both are equal, the actual cut is equal to ideal cut. However,

the two are usually not equal, i.e., the sampling results will deviate from the actual values. Thus, the deviations need to be estimated. Based on the introduction of Buser *et al.* (2006a, b), the methods to estimate sampler concentration and actual concentration and their relative difference were developed as follows.

The cumulative penetration efficiency of the PM sampler is defined as

$$P(d_p, d_{50}, \sigma_g) = 1 - \int_0^{d_p} \left[\frac{1}{x \ln(\sigma_g) \sqrt{2\pi}} e^{-\frac{[\ln(x) - \ln(d_{50})]^2}{2[\ln(\sigma_g)]^2}} \right] dx \quad (3)$$

where $P(d_p, d_{50}, \sigma_g)$ is the sampler penetration efficiency for particles having diameters less than d_p .

For unimodal mass size distribution, according to Eqs. (1) and (3), particle concentrations collected by the sampler (sampler concentration, C_s) can be estimated by:

$$C_s = C_m \int_0^{\infty} f(d_p, MMD, GSD) P(d_p, d_{50}, \sigma_g) dd_p \quad (4)$$

where C_m is the ambient particle concentration.

For actual concentrations, the ideal sampler should collect all PM in the size range less than d_{50} , and exclude all PM with size greater than d_{50} . According to Eqs. (1) and (2), the actual concentration C_r can be estimated by

$$C_r = C_m \int_0^{d_{50}} \frac{1}{x \ln GSD \sqrt{2\pi}} e^{-\frac{[\ln x - \ln MMD]^2}{2(\ln GSD)^2}} dx \quad (5)$$

For a bimodal mass size distribution, the sampler concentration (C_s) in each mode can be estimated separately according to Eq. (4) and then summed to give:

$$\begin{aligned} C_s &= C_{s1} + C_{s2} \\ &= C_{m1} \int_0^{\infty} f(d_p, MMD_1, GSD_1) P(d_p, d_{50}, \sigma_g) dd_p \\ &\quad + C_{m2} \int_0^{\infty} f(d_p, MMD_2, GSD_2) P(d_p, d_{50}, \sigma_g) dd_p \end{aligned} \quad (6)$$

where C_{s1} , MMD_1 , GSD_1 , and C_{m1} are the estimated sampler concentration, the mass median diameter, geometric standard deviation, and ambient particle matter concentration of accumulation-mode, respectively. C_{s2} , MMD_2 , GSD_2 , and C_{m2} are the estimated sampler concentration, the mass median diameter, geometric standard deviation, and ambient particle matter concentration of the coarse-mode, respectively.

Accordingly, the actual concentration (C_r) can be estimated by Eq. (7).

$$\begin{aligned} C_r &= C_{r1} + C_{r2} \\ &= C_{m1} \int_0^{d_{50}} \frac{1}{x \ln GSD_1 \sqrt{2\pi}} e^{-\frac{[\ln x - \ln MMD_1]^2}{2(\ln GSD_1)^2}} dx \\ &\quad + C_{m2} \int_0^{d_{50}} \frac{1}{x \ln GSD_2 \sqrt{2\pi}} e^{-\frac{[\ln x - \ln MMD_2]^2}{2(\ln GSD_2)^2}} dx \end{aligned} \quad (7)$$

where C_{r1} , and C_{r2} are the estimated actual concentration of the accumulation mode and the coarse mode, respectively.

As previously discussed, the relative differences (RD) between estimated sampler concentration and actual concentration for unimodal or bimodal mass size distribution can be also calculated as:

$$RD = \frac{C_s - C_r}{C_r} \times 100\% \quad (8)$$

Sampling of Ambient Size-resolved PM

The mass median diameter (MMD), the geometric standard deviation (GSD), and the mass concentration of the ambient PSD mode are the important input data for method calculation. To obtain those parameters, a Dekati® low-pressure impactor (DLPI, Dekati Ltd, Finland) was used to determine the size distributions of the actual ambient particulate matter. Operating at a flow rate of 10 L min⁻¹, the DLPI has 50% cut-point aerodynamic diameters of 0.027, 0.054, 0.093, 0.16, 0.26, 0.38, 0.61, 0.94, 1.59, 2.38, 3.97, 6.64, and 9.85 μm for stages 1 to stage 13, respectively. The DLPI was installed on the roof of a building approximately 25 m above the ground level on the campus of Beihang University (BHU, 39°59'N, 116°21'E) adjacent to the ambient PM samplers. The site is surrounded by educational and residential districts without major industrial sources. Major nearby roads are the North Fourth Ring Road about 900 m to the north, North Third Ring Road about 1.2 km to the south, and Xueyuan Road about 350 m to the east. The site is representative of urban Beijing.

Four sampling campaigns were conducted in autumn (September–October 2012), winter (January–February 2013), spring (March 2013), summer (July–August 2018). Sampling was conducted for 23 h per sample when the air was polluted, starting from 13:00 on that day to 12:00 on the next day. Sampling was extended to 47 h when the air quality was better, starting from 13:00 on that day to 12:00 on the next two days.

Aluminum foil was used as the collection substrates in the impactor. Before and after sample collection, all substrates were conditioned for 24 h at about 40% RH and 25°C in an air-conditioned room and weighed on a microbalance with a resolution of 1 μg.

Inversion of DLPI data were conducted using the Twomey algorithm (Twomey, 1975; Winklmayr *et al.*, 1990; Marjamäki *et al.*, 2000; Bian *et al.*, 2014). MMD, GSD, and the mass concentrations for different modes (C_m) were derived for each sample. The goodness of fit (R^2) of inversion date versus observed date ranged from 0.84 to 0.99, and averaged 0.96.

Method Application for Actual Atmospheric PM

Generally, atmospheric PSDs showed one or more mode (Whitby *et al.*, 1972, Liu *et al.*, 1974). Based on the method described in Section 2.1, the actual atmospheric PM was described by its PSD with either unimodal or bimodal distributions. These cases are discussed separately.

Unimodal PSDs

An ambient sample with a unimodal mass size distribution (accumulation mode, sampling date: 12 January 2013) was selected to apply the method for the atmospheric PSDs with one mode (Fig. 1). Open circles represent the measured PSD data. The red solid curve is the fitted unimodal PSD with MMD of 1.36 μm , GSD of 1.93, and PM concentration C_a of 665 $\mu\text{g cm}^{-3}$. A $\text{PM}_{2.5}$ sampler equipped with a size separator with parameters ($d_{50} = 2.5 \mu\text{m}$, $\sigma_g = 1.2$) was used to collect the corresponding $\text{PM}_{2.5}$ sample.

The area from the intersection of the left side of black solid line (located at 2.5 μm) and the red solid curve represents the actual $\text{PM}_{2.5}$ concentration (C_r). The area from the intersection between the red solid curve and the blue solid curve represents the $\text{PM}_{2.5}$ concentration collected by the sampler (C_s). Mass 1 (highlighted in green) and Mass 2 (highlighted in yellow) represent under-collected and over-collected PM mass, respectively. According to Eqs. (4) and (5), C_s and C_r can be estimated. Then the relative difference (RD) between these two concentrations can be estimated by Eq. (8).

PSDs with Bimodal Distribution

A sample showing a bimodal mass size distribution (accumulation mode and coarse mode, sampling date: 8–9, September 2012) was selected to illustrate the method for atmospheric PSDs with two modes (Fig. 2). The open circles represent the measured PSD data. The red solid curve

and green solid curve are the fitted PSDs. The red curve is the accumulation mode with an MMD₁ of 0.65 μm , GSD₁ of 1.74, and PM concentration C_{a1} of 77.0 $\mu\text{g cm}^{-3}$. The green curve is the coarse mode with an MMD₂ of 5.14 μm , GSD₂ of 1.89, and PM concentration C_{a2} of 55.2 $\mu\text{g cm}^{-3}$. A $\text{PM}_{2.5}$ sampler equipped with the corresponding size separator ($d_{50} = 2.5 \mu\text{m}$, $\sigma_g = 1.2$) is supposed to collect $\text{PM}_{2.5}$.

In Fig. 2(a), the area from the left side of black solid line (located at 2.5 μm) to the red solid curve represents the actual concentration (C_{r1}) in accumulation mode. The area from the red solid curve to the blue solid curve represents the concentration of accumulation mode collected by the sampler (C_{s1}). Mass_{A,1} (highlighted in green) and Mass_{A,2} (highlighted in yellow) represent under-collected and over-collected PM mass in accumulation mode, respectively.

In Fig. 2(b), the area from the intersection between the left side of black solid line (located at 2.5 μm) and the green solid curve represents actual concentration (C_{r2}) of coarse mode. The area from the yellow solid curve and the blue solid curve represents concentration of coarse mode collected by the sampler (C_{s2}). Mass_{C,1} (highlighted in green) and Mass_{C,2} (highlighted in yellow) represent under-collected and over-collected PM mass in coarse mode, respectively.

C_s and C_r can be estimated from Eqs. (6) and (7). The relative difference (RD) between these two concentrations can be estimated by Eq. (8).

RESULTS AND DISCUSSIONS

Size Distribution Characteristics of Atmosphere PM

A total of 45 samples sets were collected; 12 sets, 13 sets, 11 sets, and 9 sets of samples were collected in autumn, winter, spring, and summer, respectively.

Fig. 1. An example of unimodal lognormal PSD and sampling difference with $\text{PM}_{2.5}$ sampler.

Fig. 2. An example of bimodal lognormal PSD and sampling error with $\text{PM}_{2.5}$ sampler. (a) Sampling difference in accumulation mode; (b) Sampling difference in coarse mode.

From the $\text{PM}_{2.5}$ concentrations calculated from the measured ambient size-resolved PM and Technical Regulation on Ambient Air Quality Index issued by MEP (2012), the measured ambient size-resolved PM samples were assigned to six levels, i.e., excellent (I), good (II), slight pollution (III), moderate pollution (IV), heavy pollution (V), and severe pollution (VI). The corresponding number of samples in each level was 5, 14, 9, 6, 9, 2, respectively, covering all of the pollution levels (Table 1).

The average measured DLPI data and inversion of PSDs for the different air quality levels are illustrated in Fig. 3.

The PSDs for air quality levels I to V displayed bimodal distributions, i.e., accumulation mode and coarse mode,

while they show unimodal distribution in accumulation mode for air quality level VI. As the particulate air pollution increased, the contribution of the coarse mode to PM mass decreased and the contribution of the accumulation mode increased. The MMD (μm), GSD, and modal concentrations (C_m , $\mu\text{g}\cdot\text{m}^{-3}$) values for the unimodal or bimodal distributions for each pollution level were derived and presented in Table 1 for use in the subsequent calculations.

Evaluation of Performance Parameters of $\text{PM}_{2.5}$ Size Separator

The effect of the variations of cutpoint and sharpness for $\text{PM}_{2.5}$ size separator on the relative differences (RD)

Table 1. Mass median aerodynamic diameters (MMD, μm), standard deviation (GSD) and modal concentrations (C_m , $\mu\text{g m}^{-3}$) for ambient PSD with in different pollution levels.

Pollution level	I		II		III		IV		V		VI	
	Accumulation mode	Coarse mode	Accumulation mode	Coarse mode	Accumulation mode	Coarse mode	Accumulation mode	Coarse mode	Accumulation mode	Coarse mode	Accumulation mode	Coarse mode
MMD (μm)	0.50 $\pm$ 0.06	4.82 $\pm$ 1.06	0.49 $\pm$ 0.11	3.81 $\pm$ 1.37	0.67 $\pm$ 0.15	4.63 $\pm$ 1.26	0.65 $\pm$ 0.19	3.60 $\pm$ 0.84	0.86 $\pm$ 0.18	4.96 $\pm$ 0.84	1.27 $\pm$ 0.24	
GSD	2.11 $\pm$ 0.45	1.72 $\pm$ 0.42	1.84 $\pm$ 0.37	2.20 $\pm$ 0.24	1.90 $\pm$ 0.31	1.82 $\pm$ 0.52	1.75 $\pm$ 0.24	2.54 $\pm$ 0.42	1.94 $\pm$ 0.20	1.51 $\pm$ 0.35	2.04 $\pm$ 0.04	
C_m ($\mu\text{g m}^{-3}$)	23.29 $\pm$ 6.26	22.2 $\pm$ 8.6	40.0 $\pm$ 7.3	36.7 $\pm$ 13.4	85.2 $\pm$ 19.4	38.2 $\pm$ 13.6	91.0 $\pm$ 21.2	54.36 $\pm$ 20.2	194.7 $\pm$ 27.4	40.0 $\pm$ 27.1	508 $\pm$ 245	
PM ₁₀ fitted ($\mu\text{g m}^{-3}$)	45.52 $\pm$ 14.12		76.7 $\pm$ 17.7		123.4 $\pm$ 15.5		145.4 $\pm$ 15.6		235 $\pm$ 22		508 $\pm$ 222	
PM ₁₀ measured ($\mu\text{g m}^{-3}$)	43.48 $\pm$ 14.17		73.0 $\pm$ 16.3		120.5 $\pm$ 14.4		138.8 $\pm$ 14.1		235 $\pm$ 23		521 $\pm$ 245	
PM _{2.5} /PM ₁₀ ratio	0.61 $\pm$ 0.06		0.66 $\pm$ 0.07		0.74 $\pm$ 0.07		0.78 $\pm$ 0.03		0.79 $\pm$ 0.05		0.81 $\pm$ 0.01	

between the sampler and actual concentrations were investigated. Since the tolerance of cutpoint for PM_{2.5} size separator was nominally defined as $2.5 \pm 0.2 \mu\text{m}$ (JISC, 2008; MEP, 2013; U.S. EPA, 2013), cutpoints of 2.3 μm , 2.5 μm , 2.7 μm were considered. Values of σ_g equal to 1.1, 1.2, 1.3, 1.4, and 1.5 were examined. Based on the method in Sections 2.1 and 2.3, RD values between estimated PM_{2.5} sampler concentration and actual PM_{2.5} concentration for all sample sets were calculated. Bias values in the range of -5% to $+5\%$ are considered permissible (Kenny *et al.*, 2000, 2004). The number of samples with RD exceeding the permissible range are given in Fig. 4. The RD values as a function of cutpoint and sharpness parameters are shown in Fig. 5 for each of the 6 pollution levels ranging from excellent to extreme pollution.

When the cutpoint was 2.5 μm and sharpness σ_g was in the range of 1.1 to 1.5, the RD between estimated PM_{2.5} sampler and actual PM_{2.5} concentrations varied from -4.6% to 3.4% and are all within the permissible range (Fig. 5). Generally, the RD values increased with increasing σ_g . Higher σ_g values were then investigated. When σ_g was ≥ 1.6 , the RD value exceeded the permissible range (not shown).

The only time that the average RD values were $< -5\%$ was when σ_g was above 1.4 and the cutpoint was below ~ 2.35 for severe pollution days. When σ_g was at the high end of its range and the air quality was good, individual RD values for some of samples exceeded the permissible range. For example, when σ_g is 1.4, and 1.5, and air quality was excellent, RD values for only two samples exceeded the permissible range. However, the average RD value was within the suggested limits (Fig. 5).

These results suggest that if the cutpoint tolerance for a PM_{2.5} size separator is $2.5 \pm 0.2 \mu\text{m}$ and σ_g is ≤ 1.3 , the RD values exceeded the permissible range for only two of the 45 samples across all pollution levels. Hence, the cutpoint tolerance and σ_g values for an acceptable PM_{2.5} size separator are suggested to be $2.5 \pm 0.2 \mu\text{m}$ and ≤ 1.3 , respectively. From these results, the restriction on cutpoint tolerance (i.e., $2.5 \pm 0.2 \mu\text{m}$) and σ_g (i.e., 1.2 ± 0.1) for PM_{2.5} size separators by China MEP is reasonable (MEP, 2013).

Proposal for Performance Parameters of PM₁ Size Separator

The variations of cutpoint and σ_g for PM₁ size separator on the effect of the relative differences (RD) between the sampler concentration and actual concentration were also investigated. Cutpoint at 1.05 μm , 1.03 μm , 1.02 μm , 0.98 μm , 0.97 μm , 0.95 μm were considered along with σ_g values of 1.1, 1.2, 1.3, 1.4, and 1.5. Based on the previously described methods, the RD values between estimated PM₁ sampler and actual PM₁ concentrations for all samples were calculated. RD values in the range of -5% to $+5\%$ are again assumed acceptable by analogy with PM_{2.5} systems (Kenny *et al.*, 2000, 2004). The number of samples with RD values exceeding the permissible range are shown in Fig. 6. The contour plots showing the distributions of RD values as a function of cutpoint (d_{50}) and sharpness (σ_g) are shown in Fig. 7.

Fig. 3. Average measured and fitted particle size distributions in different air quality levels. Measured data are shown in histograms. The fitted accumulation mode and coarse mode distributions are shown in red and green dotted lines, respectively. The fitted combined two modes are shown in blue solid line.

Fig. 7 shows that there are more parameter combinations that produce unacceptable agreement between the estimated measured mass and the actual masses over the range of possible pollutant conditions. Even for the lowest level of PM pollution, the RD is out of the acceptable range for the lowest sizes and largest σ_g values. As the pollution level

increases, unacceptable average mass recoveries are obtained at the high end of the size cutoff even for low σ_g values.

When the cutpoint is at 1 μm and σ_g is in the range of 1.1 to 1.3, RD values varied from -3.95% to 0.92% and are all within the permissible range. When σ_g was 1.4 or 1.5, the RD values varied from -7.75% to 7.92% and their

Fig. 4. The number of samples with relative differences (RD) between the estimated $\text{PM}_{2.5}$ sampler and $\text{PM}_{2.5}$ actual concentrations exceeding the permissible range of $\pm 5\%$.

Fig. 5. Contour plots showing the distributions of the average RD for $\text{PM}_{2.5}$ mass values for the variations in cutpoint size (D_{50}) and sharpness (σ_g) parameters.

Fig. 6. The number of s with relative differences (RD) between the estimated PM_{10} sampler concentration and actual PM_{10} concentration exceeding the permissible range.

Fig. 7. Contour plots showing the distributions of the average RD for PM_{10} mass values for the variations in cutpoint size (d_{50}) and sharpness (σ_g) parameters.

values for nine or seventeen of the 45 samples exceeded the permissible range, respectively. Generally, for each sample, the RDs increased with increasing σ_g .

When cutpoint was at 0.95 μm and σ_g was in the range of 1.1 to 1.5, RD values between estimated PM_{10} sampler concentration and actual PM_{10} concentration varied from -11.04% to 0.38% . RD values for nine to thirty-four of 45 samples exceeded the permissible range. When the cutpoint was set at 1.05 μm and σ_g varied in the same range, RD values ranged from -5.81% to 15.28% . RD values for seven to eight of the 45 samples exceeded the permissible range (Fig. 6).

When cutpoint was 0.97 μm and σ_g was ranged from 1.1 to 1.5, RD values varied from -9.05% to 3.41% . RD values exceeding the permissible range increased with increasing σ_g . When σ_g was in the range of 1.1 to 1.2, no RD values exceeded the permissible range, while RD values exceeding the permissible range ranged up to twenty-five when σ_g increased to 1.5. When the cutpoint was at 1.03 μm and σ_g varied in the same range, RD values range from -6.55% to 12.36% . RD values for three to eleven of the 45 samples exceeded the permissible range (Fig. 7).

When cutpoint was set at 0.98 μm and σ_g is in the range of 1.1 to 1.5, RD values vary from -8.87% to 4.92% . RD values exceeding the permissible range again increased with increasing σ_g . For σ_g in the range of 1.1 to 1.2, there were no RD values exceeding the permissible range. However, there were up to twenty-two high RD values when σ_g increased to 1.5. With a cutpoint of 1.02 μm and σ_g varied in the same range, the RD values ranged from -6.94% to 10.89% . When σ_g was at 1.1, no RD values exceeded the permissible range. When σ_g was in the range of 1.2 to 1.4, only one RD value exceeded the permissible range. When σ_g increased to 1.5, ten RD values exceeded the permissible range.

From these results for typical urban atmospheric aerosols, a cutpoint tolerance for a PM_{10} sampler of $1.00 \pm 0.02 \mu\text{m}$ and σ_g no greater than 1.2 produced RD values for only one sample that exceeded the permissible range. Hence, the cutpoint tolerance for an acceptable PM_{10} size separator

is proposed to be $1.00 \pm 0.02 \mu\text{m}$ with σ_g no greater than 1.2. Compared with $\text{PM}_{2.5}$ sampler, the PM_{10} sampler is more sensitive to the variation in cutpoint and only a relatively small range of variation is allowable. For the measurements made in Beijing, 2.5 μm is closer to the minimum between the accumulation and coarse modes than 1 μm (Fig. 3). Thus, shifts in cutpoint of $\text{PM}_{2.5}$ size separator caused smaller differences in observed mass concentrations (Cao *et al.*, 2013).

Analyses for Hypothetical PSDs

Vanderpool *et al.* (2001) summarized previous ambient size distribution measurements, and numerically constructed three idealized, bimodal particle size distributions, i.e., fine, typical, and coarse ambient aerosol distributions. The three idealized distributions have been prescribed in 40 CFR Part 53 for evaluation of candidate $\text{PM}_{2.5}$ size separators (U.S. EPA, 2013). $\text{PM}_{2.5}/\text{PM}_{10}$ ratios measured in this study across four seasons and different pollution levels vary from 0.52 to 0.87 and can represent the fine ($\text{PM}_{2.5}/\text{PM}_{10}$ ratio = 0.94), and the typical ($\text{PM}_{2.5}/\text{PM}_{10}$ ratio = 0.55) ambient aerosol distributions (Vanderpool *et al.*, 2001; U.S. EPA, 2013). Our $\text{PM}_{2.5}/\text{PM}_{10}$ ratios are much greater than the idealized coarse ambient aerosol distributions ($\text{PM}_{2.5}/\text{PM}_{10}$ ratio = 0.27), which indicates that our measured PSDs cannot represent coarse aerosol distributions.

In order to evaluate the performance parameters of a PM_{10} size separator for the three idealized distributions, especially for the coarse aerosol distributions, RD values between estimated PM_{10} sampler concentration and actual PM_{10} concentration for the three idealized particle size distributions were also calculated.

The RD values between estimated PM_{10} sampler concentration and PM_{10} actual concentration for the three idealized, bimodal particle size distributions are illustrated in Fig. 8. When the cutpoint was at 0.95 μm and $\sigma_g \geq 1.2$, RD values for fine ambient aerosol distributions exceeded the permissible range. When cutpoint was at 0.95 μm and $\sigma_g \geq 1.4$, or cutpoint was at 0.97 μm and $\sigma_g = 1.5$, the RD

Fig. 8. Relative differences (%) between estimated PM_{10} sampler concentration and PM_{10} actual concentration under three idealized, bimodal particle size distributions (a) Fine distribution, (b) Typical distribution, and (c) Coarse distribution.

Fig. 8. (continued).

values for the typical ambient aerosol distribution exceeded the permissible range. When the cutpoint was at $0.95\ \mu\text{m}$ and $\sigma_g = 1.5$, the RD value for coarse aerosol distributions exceeded the permissible range. The other combinations of d_{50} and σ_g , RD values were within the permissible range. Thus, the proposed performance parameters of PM_{10} sampler for typical urban atmospheric aerosol in last section, i.e., $d_{50} = 1.00 \pm 0.02\ \mu\text{m}$, $\sigma_g \leq 1.2$ is suitable for the three idealized ambient aerosol distributions.

CONCLUSIONS

Based on the interaction between performance characteristics for $\text{PM}_{2.5}$ or PM_{10} size separators and actually measured typical urban PSDs in four seasons covering all pollution levels and three idealized ambient PSDs, the RD between estimated $\text{PM}_{2.5}$ or PM_{10} sampler concentrations and actual concentrations have been systematically investigated. For the $\text{PM}_{2.5}$ size separator, if the cutpoint tolerance is $2.5 \pm 0.2\ \mu\text{m}$ and σ_g is ≤ 1.3 , the RD values for most typical urban atmospheric aerosol conditions are within the permissible range ($\pm 5\%$).

For the PM_{10} size separator, tolerances for the cutpoint and σ_g are proposed to be $1.0 \pm 0.02\ \mu\text{m}$ and ≤ 1.2 , respectively.

Those parameters would provide acceptable performance for most typical urban atmospheric aerosol conditions and the idealized ambient aerosol distribution.

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