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# Reconstructed algorithm for scattering coefficient of ambient submicron particles \*

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#### ABSTRACT

Ambient submicron particles (PM1) exert significant impacts on visibility degradation during severe pollution episodes of urban China. The U.S. IMPROVE algorithms are widely used for assessing the extinction effect of atmospheric aerosols, but only suitable for fine particulate matter. A proper algorithm for PM<sub>1</sub> extinction estimation is lacking and becomes urgent, especially after the online measurement of PM<sub>1</sub> species is routine by aerosol mass spectrometers. Here we conducted three-month in-situ measurements to explore mass scattering efficiencies (MSE) of PM<sub>1</sub> major species at a supersite of eastern China. Results indicated that MSEs of ammonium sulfate and nitrate increase quickly and then keep stable with the mass accumulation, while those of organic matter keep at  $\sim 5.5 \text{ m}^2/\text{g}$  but with a large vibration in the whole mass range. The algorithm for reconstructing  $PM_1$  dry scattering coefficient was derived from the integral of the variation patterns for the three PM<sub>1</sub> species. The algorithm was then validated and compared with other empirical algorithms through separate field measurements. Good correlations between the reconstructed and measured dry scattering coefficient were observed with R square higher than 0.9 and slope of 1.01–1.05, indicating that the reconstructed algorithm can predict the dry scattering coefficient well based on PM1 chemical composition measurements in urban China. Our study is expected to provide observed insights on the variation of MSE in the wide mass range especially in the high region, as well as accurate formulas for ambient  $PM_1$  dry scattering apportionment. © 2019 Elsevier Ltd. All rights reserved.

#### 1. Introduction

Particulate matters (PM), especially submicron PM (PM with aerodynamic diameter < 1  $\mu$ m) exert significant impacts on visibility degradation and global climate change directly through optical extinction effect (Attwood et al., 2015; Cheng et al., 2008). Heavy haze pollution episodes occurred frequently in eastern China (Hu et al., 2016; Sun et al., 2013; Zhang et al., 2017). A comprehensive and accurate examination on the contribution of PM<sub>1</sub> chemical compositions to atmospheric extinction coefficients is

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crucial to control strategies towards visibility improvement.

Aerosol scattering coefficient ( $B_{sca}$ ), which is affected by multiple factors such as chemical compositions and mass concentrations, dominates the extinction effect (Watson, 2002; Gao et al., 2015). Aerosol mass scattering efficiency (MSE) is an important parameter which is used to calculate scattering coefficient ( $B_{sca}$ ) with observed mass concentration. Numerous observation studies have investigated PM<sub>1</sub> chemical mass based on high-time-resolution instruments, including High-Resolution Time-of-Flight Aerosol Mass spectrometer (HR-ToF-AMS) (Hu et al., 2017; Huang et al., 2012; Sun et al., 2016; Xu et al., 2014), Quadrupole-AMS (Sun et al., 2010), Aerodyne Aerosol Chemical Speciation Monitor (ACSM) (Sun et al., 2013) and Aerodyne soot particle-aerosol mass spectrometer (SP-AMS) (Wang et al., 2016b). Then, understanding MSEs of aerosol components is the remaining aspect to estimate their scattering coefficients and contributions to light extinction during haze





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pollution (Hand and Malm, 2007).

MSE values of aerosol chemical components are usually estimated through the theoretical calculation of Mie scattering, analysis of multiple linear regression (MLR) or the U.S. IMPROVE (Interagency Monitoring of Protected Visual Environments) algorithm. The U.S. IMPROVE algorithm, including the original and revised versions, were derived from the long-term measurements at U.S. remote sites with low aerosol loading (Pitchford et al., 2007: Watson, 2002). Due to its simplicity, the IMPROVE algorithms have been widely used in urban China such as Xi'an (Cao et al., 2012), Beijing (Han et al., 2016; Wang et al., 2015a,b), Guangzhou (Tao et al., 2014) and Nanjing (Kong et al., 2015; Wang et al., 2016a; Wu et al., 2017). The deviation between IMPROVE estimation and field measurement is  $-11\% \sim +54\%$  in the U.S. (Hand et al., 2001), while the biases significantly increase in urban China with high aerosol mass concentration (Cheng et al., 2015; Tao et al., 2014). More importantly, the MSEs in the IMPROVE algorithm are used for PM<sub>2.5</sub> rather than PM<sub>1</sub> which routine AMS/ACSM measurements covered. Multiple linear regression (MLR) method can be used to obtain the average MSEs of chemical components in PM<sub>1</sub> with measured scattering coefficients and their mass concentrations, but the regressive MSE values are unstable or unexplainable frequently (Han et al., 2016). The Mie theory usually calculates MSEs with high accuracy based on the known mass-size distribution of chemical components (Cheng et al., 2008). However, the long-term or large amount measurements of mass-size distribution for chemical components are not easy to achieve. Short-term episode campaign will affect the representativeness and applicability of the calculated MSEs. To our knowledge, the high-resolution evolution of MSE for PM<sub>1</sub> species in the wide mass range is unknown up to now, and an accurate algorithm for ambient PM<sub>1</sub> chemical scattering is also lacking.

In this study, we used a High-Resolution Time-of-Flight Aerosol Mass spectrometer (HR-ToF-AMS) to measure mass size distribution of chemical components in non-refractory submicron PM (NR-PM<sub>1</sub>), i.e., organic matter, sulfate, nitrate and ammonium at a supersite in eastern China. Hourly resolution MSEs of major chemical species (organic matter, ammonium nitrate and ammonium sulfate) were calculated based on Mie theory with HR-ToF-AMS observation data. The evolution of MSEs for the three species was investigated, as well as the relationships between MSEs and species mass concentrations within a wide mass range. In order to make the above calculations reliable, particle dry scattering coefficients were measured from a Cavity Attenuated Phase Shift ALBedo monitor (CAPS-ALB) and a Photoacoustic Extinctionmeter (PAX) simultaneously. The primary purposes of this study are to 1) better understand the highly time-resolved variations of MSEs for chemical species from clean to polluted range; 2) provide a reconstructed algorithm for scattering coefficient of ambient submicron particles.

#### 2. Materials and methods

#### 2.1. Field observation campaign

Online field observations were taken at the rooftop of building

in Shanghai Academy of Environmental Sciences (31.10°N,121.25°E), a typical urban supersite in Shanghai, the largest megacity of China. Specific location and detailed descriptions of the supersite have been stated in our previous studies (Zhu et al., 2018). The whole observations contained three periods, i.e., from August 23 to September 2, 2016, from November 28, 2016 to January 3, 2017, and from May 18 to June 4, 2017. They were distributed in the season of summer, winter and spring, respectively, and can cover almost the whole mass range of aerosol components.

The mass concentration and size distribution of NR-PM<sub>1</sub> species were measured by an HR-ToF-AMS (Aerodyne Research Inc., USA) with a time-resolution of 4 min. The principal and details of this instrument has been described in the related literatures (Canagaratna et al., 2007; Jimenez et al., 2003). Ambient air was sampled into the HR-ToF-AMS though the center of the copper at a flow rate of ~0.1 L/min. A PM URG cyclone (URG-2000-30ED) and a Naffion dryer were employed before the inlet of HR-ToF-AMS for removing coarse particles and drying aerosol particles, respectively. The primary HR-ToF-AMS measurement results were then averaged into a time resolution of 1 h. As a result a total of 1766 hourly samples were obtained. Aerosol dry scattering coefficient of PM<sub>1</sub> was measured in situ using a CAPS-ALB (Shoreline Science Research Inc., Japan;  $\lambda = 530 \text{ nm}$ ) and a PAX (Droplet Measurement Technologies Inc., USA;  $\lambda = 532 \text{ nm}$ ) with time-resolution of 1 second simultaneously. Although the total extinction coefficient was also measured by the CAPS-ALB and PAX, we only used the results of scattering coefficient as the target of this study is to investigate the mass scattering efficiency. To acquire particle size of PM<sub>1</sub> and eliminate the effects of humidity, a PM<sub>1</sub> cyclone and a diffusion dryer were installed in front of PAX and CAPS-ALB to keep relative humidity (RH) less than 40%. In order to ensure the accuracy of the dry scattering coefficient measurements, the measurement results of the two instruments (PAX and CAPS-ALB) were used for crossvalidation. The mass concentrations of PM<sub>2.5</sub> were recorded by a  $\beta$ -ray apparatus (FH62C-14 $\beta$ -ray, Thermo Scientific Co., MA) and black carbon (BC) by a three-wavelength Aethalometer (AE31, Magee Scientific Corp., USA), both with a time resolution of 5 min.

The correlation between in-situ measured concentrations of  $PM_1$  (= NR-PM\_1 + BC) and  $PM_{2.5}$  measured by  $\beta$ -ray apparatus yielded a regression slope of 0.73 and R square of 0.86 (Fig. 1a). Meanwhile,  $PM_1$  dry scattering coefficient measured by PAX agreed well with that of CAPS. The regression slope reached 1.05 with the R square as high as 0.96 (Fig. 1b). Good correlations and quantitative agreements between independent measurements support the reliability of our measurements in this study.

#### 2.2. Calculation of MSEs for PM<sub>1</sub> species

According to the definition, the MSE of individual aerosol species in  $PM_1$  (*MSE(species, PM\_1*), m<sup>2</sup>/g) can be calculated by total dry scattering coefficient due to the species divided by total mass concentration of the species in PM<sub>1</sub> size range, as shown in Equation (1) (Hand and Malm, 2007; Cheng et al., 2015).

 $MSE(\text{species}, PM_1) = \frac{\sum_{bin=1}^{D_{bin} \le 1 \mu m} 3Q_{\text{sca}}(n_{\text{species}}, D_{bin}, \lambda) / (2\rho_{\text{species}} D_{bin}) \times C_{\text{species}, D_{bin}}}{\sum_{bin=1}^{D_{bin} \le 1 \mu m} C_{\text{species}, D_{bin}}}$ 

(1)



**Fig. 1.** Linear regression between (a)  $PM_1(=NR-PM_1 + BC)$  and  $PM_{2.5}$  measurement results; (b) scattering coefficients measured by the CAPS and PAX.

In Equation (1),  $Q_{sca}$  ( $n_{species}$ ,  $D_{bin}$ ,  $\lambda$ ) represents the scattering efficiency for a single particle size channel D<sub>bin</sub> of single species. By assuming particles are externally mixed and spherical, it can be estimated using the basic Mie theory by inputting refractive index (n<sub>species</sub>), geometric mean particle diameter (D<sub>bin</sub>) and light wavelength ( $\lambda$ ) (Bohren and Huffman, 1998; Mätzler, 2002). The refractive index and density for NH<sub>4</sub>NO<sub>3</sub>, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and organic matter (OM) was derived from Pitchford et al. (2007). The refractive index  $(n_{species})$  of NH<sub>4</sub>NO<sub>3</sub>,  $(NH_4)_2SO_4$ , and OM were 1.57 + 0.0i, 1.52 + 0.0i, 1.55 + 0.0i, respectively. The density ( $\rho_{species}$ ) of NH<sub>4</sub>NO<sub>3</sub>,  $(NH_4)_2SO_4$ , and OM were 1.73 g/cm<sup>3</sup>, 1.77 g/cm<sup>3</sup> and 1.4 g/cm<sup>3</sup>, respectively. The wavelength was set as 550 nm which represents the typical length of visible spectrum. In the primary HR-ToF-AMS datasets, the aerosol size increase from 33 nm to 978 nm with total of 75 channels. The AMS reported mass concentration for each size is in the differential form of  $dM/d \log D_{va}$ , which means the necessity of converting to C<sub>species, Dbin</sub> in Equation (1) by multiplying  $\Delta \log D_{va}$  for the corresponding size channel.

It is also worth noting that the particle size unit is vacuum aerodynamic diameter  $(D_{va})$  in AMS primary datasets while the unit of  $D_{bin}$  in Equation (1) is geometric diameter. The conversion formula from  $D_{va}$  to  $D_{bin}$  is derived from DeCarlo et al. (2004) by assuming spherical particles. Finally, taking into account the transmission loss of the AMS lens (Zhang et al., 2004) and the target of PM<sub>1</sub> in this study, we unify the particle size below 1  $\mu$ m in vacuum aerodynamic meter.

#### 2.3. Evaluation of the reconstructed algorithm

The evaluation datasets, including hourly mass concentration of  $PM_1$  species by HR-ToF-AMS and hourly  $PM_1$  dry scattering coefficient by PAX, were derived from the intensive field campaigns in Shanghai Academy of Environmental Sciences from January 5–12, 2017 and in Dezhou from November 6–11, 2017. The Shanghai site was the same as the above mentioned site, while the Dezhou observation site is located in the Pingyuan Meteorological Agency (37.17°N, 116.43°E). Pingyuan is a county town located in central Dezhou, at the border of Shandong Province and Hebei Province, 320 km to the south of Beijing. The observation site was considered to be well representative to characterize air pollution in North China.

Four reconstructed algorithms (the original and revised IMPROVE algorithm, multiple regression fitting algorithm (Lan et al., 2018) and the algorithm of this study) were used for the calculation of dry scattering coefficient by inputting the mass concentration of  $PM_1$  species. Then the calculated results of four algorithms were quantitatively compared with the PAX measured results. The statistical index of deviation used for comparison was defined as the absolute bias between the calculated and measured dry scattering coefficient divided by the measured value.

#### 3. Results and discussion

#### 3.1. MSEs for major chemical species

The comparison of hourly average dry scattering coefficients between the Mie calculation and the measurements by PAX during Shanghai field campaign were shown in Fig. 2. The calculated PM<sub>1</sub> dry scattering coefficient agreed well with that measured by the PAX. Good consistencies between the calculated and measured dry scattering coefficients (slope = 1.04,  $R^2 = 0.90$ ) proved the



**Fig. 2.** Linear correlations between  $PM_1$  scattering coefficient calculated by Mie theory and measured by PAX under dry condition. The Measured B<sub>sca</sub> was measured by PAX instrument. The calculated B<sub>sca</sub> was estimated using the numerator part of Equation (1), given mass-size distribution from AMS. The data points were the hourly average calculation which obtained from the three-month field campaign in Shanghai.

Table 1	
Mass scattering efficiencies of PM1 or PM2.5 species summarize	d by the previous studies and this study.

Species	Particle size	Location	Method	MSE value (m <sup>2</sup> /g)	Reference
Organic matter	PM <sub>1</sub>	Shanghai, China	Mie theory	5.3 (min:3.6, max:7.5)	This study
	$PM_1$	Dongguan, China	MLR <sup>a</sup>	2.9 (SOA) <sup>d</sup> ; 18.2 (POA) <sup>e</sup>	Lan et al. (2018)
	$PM_1$	Beijing, China	MLR <sup>a</sup>	4.6 (SOA, NHP <sup>f</sup> ); 3(SOA, HP <sup>g</sup> )	Wang et al. (2015a,b)
	PM <sub>2.5</sub>	Shanghai, China	Mie theory	4.5 (min:3.2, max:6.3)	Cheng et al. (2015)
	PM <sub>2.5</sub>	Guangzhou, China	MLR <sup>a</sup>	2.8 (SM) <sup>h</sup> ; 4.9 (LM) <sup>i</sup>	Jung et al. (2009)
	PM <sub>2.5</sub>	Guangzhou, China	MLR <sup>a</sup>	3.0 (spring); 4.8 (summer); 6.9 (autumn); 6.5 (winter)	Tao et al. (2014)
	PM <sub>2.5</sub>	U.S. rural	IMPROVE_O <sup>b</sup>	4	Watson (2002)
	PM <sub>2.5</sub>	U.S. rural	IMPROVE_R <sup>c</sup>	2.8 (SM) <sup>h</sup> ; 6.1 (LM) <sup>i</sup>	Pitchford et al. (2007)
	PM <sub>2.5</sub>	Arizona, U.S.	MLR <sup>a</sup>	3.1	Hand and Malm (2007)
Ammonium nitrate	$PM_1$	Shanghai, China	Mie theory	4.3 (min:2.4, max:5.1)	This study
	$PM_1$	Dongguan, China	MLR <sup>a</sup>	7.0	Lan et al. (2018)
	$PM_1$	Beijing, China	MLR <sup>a</sup>	7.0 (NHP <sup>f</sup> ); 5.2(HP <sup>g</sup> )	Wang et al. (2015a,b)
	PM <sub>2.5</sub>	Shanghai, China	Mie theory	4.3 (min:2.4, max:5.8)	Cheng et al. (2015)
	PM <sub>2.5</sub>	Guangzhou, China	MLR <sup>a</sup>	2.4 (SM) <sup>h</sup> ; 4.5 (LM) <sup>i</sup>	Jung et al. (2009)
	PM <sub>2.5</sub>	Guangzhou, China	MLR <sup>a</sup>	6.1 (spring); 1.7 (summer); 4.2 (autumn); 6.7 (winter)	Tao et al. (2014)
	PM <sub>2.5</sub>	U.S. rural	IMPROVE_O <sup>b</sup>	3	Watson (2002)
	PM <sub>2.5</sub>	U.S. rural	IMPROVE_R <sup>c</sup>	2.4 (SM) <sup>h</sup> ; 5.1 (LM) <sup>i</sup>	Pitchford et al. (2007)
	PM <sub>2.5</sub>	Arizona, U.S.	MLR <sup>a</sup>	2.5	Hand and Malm (2007)
Ammonium sulfate	$PM_1$	Shanghai, China	Mie theory	4.2 (min:2.4, max:5.8)	This study
	$PM_1$	Dongguan, China	MLR <sup>a</sup>	9.5	Lan et al. (2018)
	$PM_1$	Beijing, China	MLR <sup>a</sup>	7.0 (NHP <sup>f</sup> ); 5.2(HP <sup>g</sup> )	Wang et al. (2015a,b)
	PM <sub>2.5</sub>	Shanghai, China	Mie theory	3.5 (min:2.1, max:4.7)	Cheng et al. (2015)
	PM <sub>2.5</sub>	Guangzhou, China	MLR <sup>a</sup>	$2.2 (SM)^{h}; 3.2 (LM)^{i}$	Jung et al. (2009)
	PM <sub>2.5</sub>	Guangzhou, China	MLR <sup>a</sup>	5.1 (spring); 5.7 (summer); 4.2 (autumn); 6.7 (winter)	Tao et al. (2014)
	PM <sub>2.5</sub>	U.S. rural	IMPROVE_O <sup>b</sup>	3	Watson (2002)
	PM <sub>2.5</sub>	U.S. rural	IMPROVE_R <sup>c</sup>	2.2 (SM) <sup>h</sup> ; 4.8 (LM) <sup>i</sup>	Pitchford et al. (2007)
	PM <sub>2.5</sub>	Arizona, U.S.	MLR <sup>a</sup>	2.5	Hand and Malm (2007)

<sup>a</sup> Multiple linear regression.

<sup>b</sup> Original IMPROVE algorithm.

<sup>c</sup> Revised IMPROVE algorithm.

<sup>d</sup> Secondary organic aerosol.

<sup>e</sup> Primary organic aerosol.

<sup>f</sup> Non-heating period.

<sup>g</sup> Heating period.

<sup>h</sup> Small mode.

<sup>i</sup> Large mode.

reliability of MSEs used to calculate here. Noted that the collection angle of PAX is 6°-174°, tiny different from the 0°-180° angle of Mie calculation. However, the truncation effect was estimated to be as low as ~5% and can be neglected for the comparison. The averaged MSEs of OM, NH<sub>4</sub>NO<sub>3</sub> and  $(NH_4)_2SO_4$  were  $5.3 \pm 0.5 \text{ m}^2/\text{g}$ ,  $4.3\pm1.0\ m^2/g$  and  $4.2\pm0.6\ m^2/g,$  respectively during the whole campaign. Table 1 summarized the MSEs reported in previous studies conducted in different areas all over the world. The MSEs corresponding to OM, NH4NO3 and (NH4)2SO4 for the original IMPROVE algorithm are 4, 3 and 3 m<sup>2</sup>/g, respectively. They were 40%, 43%, and 33% lower than the average MSEs in this study, respectively, implying that the dry scattering coefficient of PM<sub>1</sub> might be underestimated by applying the original IMPROVE algorithm directly. The MSEs of OM, NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> in the revised IMPROVE algorithm are 6.1  $m^2/g$ , 5.1  $m^2/g$  and 4.8  $m^2/g$  for large mode,  $2.8 \text{ m}^2/\text{g}$ ,  $2.4 \text{ m}^2/\text{g}$  and 2.2 for small mode. The maximum MSEs were 7.5 m<sup>2</sup>/g for OM and 5.8 m<sup>2</sup>/g for (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> in this study, which were both higher than the maximum values  $(6.3 \text{ m}^2/\text{g} \text{ and } 4.7 \text{ m}^2/\text{g})$  reported by Cheng et al. and the MSEs  $(6.1 \text{ m}^2/\text{g} \text{ and } 4.8 \text{ m}^2/\text{g})$  for large mode in the revised IMPROVE algorithm (Pitchford et al., 2007). The MSEs for three scattering species in PM<sub>2.5</sub> were usually lower than those in PM<sub>1</sub>, suggesting that the dry scattering coefficient of PM<sub>1</sub> probably be underestimated using existed PM2.5 empirical formulations like the IMPROVE algorithms.

MSEs for different aerosol components were not constant values. Hourly variations of MSEs for major chemical species in  $PM_1$  were shown in Fig. 3. The MSEs for each aerosol component changed dramatically during the process of pollution episodes. In

the formation phase of pollution episodes, the MSEs of three scattering species enhanced rapidly with the increase of mass concentration. During the elimination phase of the episodes, the MSEs reduced with the decrease of mass concentration. For instance, during haze pollution episodes from December 3 to 6, the highest hourly MSEs of OM, NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> can reach as high as  $6.6 \text{ m}^2/\text{g}$ ,  $6.4 \text{ m}^2/\text{g}$  and  $5.0 \text{ m}^2/\text{g}$ , and dropped to the minimum of  $4.4 \text{ m}^2/\text{g}$ ,  $2.5 \text{ m}^2/\text{g}$  and  $3.0 \text{ m}^2/\text{g}$ , respectively. This is consistent with previous studies in Guangzhou. Tao et al. (2014) reported that the MSEs of OM. NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> in winter with high aerosol mass loading were 26%, 37% and 26% higher than those in summer with low aerosol mass loadings in Guangzhou, China, respectively (Tao et al., 2014). The variations of MSEs for different aerosol components is usually determined by the integral effect of mass concentration, size distribution, morphology and mixing state (Hand and Malm, 2007). The ambient aerosol mass loading in haze pollution episodes was 2–9 times higher than that of the clean periods and the peak sizes of major chemical components were shifted to larger range during haze pollution episodes. These factors leaded to a large discrepancy between the pollution episodes and clean periods in size distribution of aerosol components, which finally affected the MSE values.

Interestingly, the MSEs did not always grow in proportion to the pollution level. For instance, the dry scattering coefficient was  $380 \text{ Mm}^{-1}$  at the pollution peak on May 25, which was 1.3 times that ( $301 \text{ Mm}^{-1}$ ) at another pollution peak on December 22. However, the corresponding MSEs (OM: 7.31, NH<sub>4</sub>NO<sub>3</sub>: 5.56, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>: 5.43 m<sup>2</sup>/g) at the latter pollution peak were actually higher than those (OM: 4.93, NH<sub>4</sub>NO<sub>3</sub>: 3.63, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>: 3.34 m<sup>2</sup>/g)



Fig. 3. Temporal variation of (a) aerosol mass concentration for major chemical components; (b) aerosol mass scattering efficiency calculated by Mie theory and scattering coefficient measured by PAX.

at the former peak. Some previous studies suggested that in addition to mass concentrations, the diversities in MSEs for chemical species are also due to the differences in their size distributions (Cheng et al., 2015; Seinfeld and Pandis, 2016). Wang et al. (2015a,b) investigated the evolution of chemical species during the heating and non-heating periods, and then used the MLR method to obtain the mass extinction efficiency of each chemical species. The MSEs of NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> during the heating period were lower than those during the non-heating period, while the mass concentration of each species during the heating period was about 1.5–2.2 times higher than that during the non-heating period (Table 1). Their explanation is that the particle size shifted to a smaller size range during the heating period (Wang et al., 2015a,b).

In order to further understand the influence of size distribution on the MSE, the relationship between the MSE and size distribution of different species was examined (Fig. 4). With the increase of MSEs for three scattering species, their particle sizes increase gradually, indicating that higher MSEs correspond to mass-size distributions with larger particle sizes. The small particle size (<200 nm) for three species contributed slightly to the enhancement of MSEs, while the MSEs increased from  $1 \text{ m}^2/\text{g}$  to  $6 \text{ m}^2/\text{g}$  as the particle size shifted from 200-300 nm to 600-700 nm. It is worth noting that when particle sizes larger than 700 nm, the range of MSEs were being narrow again. Calculations using the Mie theory suggest that when the incident wavelength of visible light is 550 nm, the maximum MSE of the monodispersed aerosol particles usually appears at a geometric diameter of 500–600 nm (Seinfeld and Pandis, 2016). These measurement results of this study also indicated that the closer the peak size of mass distribution to 500–600 nm, the higher MSE values of PM species.

#### 3.2. Relationship between the MSEs and the mass concentrations

The variation in MSEs associated with the mass concentrations of different species were shown in Fig. 5. The overall trend of OM was different from that of  $NH_4NO_3$  and  $(NH_4)_2SO_4$ . The MSE of OM

dispersed between 4 and  $6.5 \text{ m}^2/\text{g}$  under low mass range and gathered at  $5.2-5.5 \text{ m}^2/\text{g}$  when the mass concentration was higher than  $25 \,\mu\text{g}/\text{m}^3$ . The significant vibration of MSE under low concentration was perhaps due to the complexity of organic sources and molecular structure. Fig. 6 showed that the peak particle size of OM mass distribution was at ~500 nm, and has almost no change with the total mass increase, resulting in the MSE at a gathered stable level of  $5.2-5.5 \text{ m}^2/\text{g}$  in the high mass range.

The relationship between MSEs of NH<sub>4</sub>NO<sub>3</sub> and its mass concentration was similar to that of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>. The MSEs enhanced rapidly with the increase of mass concentrations when mass concentrations were low, and tended to be constant after the mass concentration reached a certain value. As the mass concentration below  $12 \,\mu g/m^3$ , the MSE of NH<sub>4</sub>NO<sub>3</sub> linearly increased from  $1.77 \, m^2/g$  to  $5.63 \, m^2/g$ . The increase rate of MSE declined with the increase of the mass concentration. When the mass concentration was over  $30 \,\mu g/m^3$ , the MSE of NH<sub>4</sub>NO<sub>3</sub> floated in a small range around  $5.4 \, m^2/g$ . The MSE of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> had the same increasing behavior but at a smaller mass concentration range. The corresponding MSE increased from  $2.44 \, m^2/g$  to  $4.56 \, m^2/g$  with the mass concentration of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> increasing from  $2.1 \,\mu g/m^3$  to  $9.6 \,\mu g/m^3$ . The increasing rate of MSE for (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> slowed down in the mass concentration ranges of  $10-20 \,\mu g/m^3$ . With the mass concentration further increasing, the MSE of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> floated around  $5.2 \, m^2/g$ .

As the mass concentration of NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> increased, the peak particle size range gradually shifted from 300-400 nm to 600–700 nm (Fig. 6), resulting in the notable increase of MSEs (Fig. 5). This is the reason why the MSE of NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> increases significantly in the low mass concentration range. However, the higher mass concentrations (NH<sub>4</sub>NO<sub>3</sub> > 25 µg/m<sup>3</sup>; (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> > 20 µg/m<sup>3</sup>) did not correspond to larger peak sizes of mass distributions for NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, in accordance with that mentioned in MSEs variations with mass concentrations.

To deeply investigate the MSEs of chemical components, we used formula fitting to explore the relationship between MSEs and the concentrations of OM, NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>. The mass



**Fig. 4.** Relationship between mass scattering efficiency and size distribution for (a) organic matter (OM), (b) NH<sub>4</sub>NO<sub>3</sub> and (c) (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>. The plots are colored by  $dM/dlog_{10}d_{va}$  (unit: µg/m<sup>3</sup>) of OM, NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, respectively.

concentrations of chemical species were divided into ten segments with a certain step, and the average concentration and MSE corresponding to the segments were calculated. The dynamic variability of MSEs for NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> with mass concentrations was in accordance with the logistic curve, that is, it increases exponentially first, then becomes saturated, and the rate becomes slower (Fig. 5). The relationship was then fitted using the sigmoid function to obtain the following formulas:

$$MSEOM = 5.5 \left( 1 + e^{0.1[OM] - 7.8} \right)^{-1}$$
(2)

$$MSE_{NH_4NO_3} = 5.5 \left(1 + e^{-0.2[NH_4NO_3] - 0.4}\right)^{-1}$$
(3)

$$MSE_{(NH_4)_2SO_4} = 5.2 \left( 1 + e^{-0.2 \left[ (NH_4)_2 SO_4 \right] - 0.1} \right)^{-1}$$
(4)

where [OM], [NH<sub>4</sub>NO<sub>3</sub>] and [(NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>] were the mass concentrations of organic matter, NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, respectively.

Both R<sup>2</sup> values were 0.96 for formulas fitting of MSEs against



**Fig. 5.** Relationship between mass scattering efficiency (MSE) and mass concentration for major chemical components. The line is the formula fitting. The total number of data points is 1766. The average mass scattering efficiency in each segment are illustrated and the error bars in the y-axis were the standard deviations of mass scattering efficiency.

NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, indicating that the formulas can well estimate the MSEs with the mass concentration of NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>. For OM, the R<sup>2</sup> value was 0.27, which indicated that the relationship between MSE and mass concentration is not a simple logistic function. The low R<sup>2</sup> was mainly caused by the notable diversity of MSE under low mass range. Future studies on the specific reason of the dispersed MSEs and how to classify them to regular clusters are necessary and expected.

In summary, the calculation formula for dry scattering coefficient of  $PM_1$  was summarized as follows. Note that this formula only considered the major scattering species and the minor species such as metal elements are usually negligible for  $PM_1$ .

$$\begin{split} B_{sca} &= 5.5 \times \left(1 + e^{-0.2[NH_4NO_3] - 0.4}\right)^{-1} \times \left[NH_4NO_3\right] + \\ &5.2 \times \left(1 + e^{-0.2[(NH_4)_2SO_4] - 0.1}\right)^{-1} \times \left[\left(NH_4\right)_2SO_4\right] + \\ &5.5 \times \left(1 + e^{0.1[OM] - 7.8}\right)^{-1} \end{split}$$

(5)



**Fig. 6.** Mass-size distributions under different total mass concentrations for major chemical components. Different mass concentration intervals are marked by lines of different forms and colors, respectively.

## 3.3. Comparison of the reconstructed algorithm with other algorithms

The dry light-scattering coefficients of Shanghai and Dezhou, two cities with quite different particle characteristics, were calculated based on PM1 species of AMS from local field campaigns. Four methods were applied in these case studies, including original IMPROVE algorithm, revised IMPROVE algorithm, multiple regression fitting MSEs (Lan et al., 2018) and the reconstructed algorithm of this study. All the calculated dry scattering coefficients are then compared with the results of in-situ measurements (Fig. 7 and Fig. 8). The dry scattering coefficients by the four calculation methods and measurements had good consistency during the observation period. There was a strong linear relationship between the calculated and measured data in Shanghai. Regression R<sup>2</sup> value of the original IMPROVE algorithm, the revised IMPROVE algorithm, multiple regression and this study reached as high as 0.93, 0.91, 0.95 and 0.93 respectively. The correlation between the calculated and measured values was slightly worse in Dezhou, but all the R<sup>2</sup> values were still above 0.88.

However, quantitative bias was much different among the four algorithms in spite of the good correlation. For the multiple regression fitting by Huang et al. (Lan et al., 2018), the sum of calculated dry scattering coefficients of major chemical species was much higher than the results measured in Shanghai and Dezhou (Fig. 7), due to the large MSEs of the three species obtained by MLR fitting. Compared with the measured scattering coefficients, the results calculated based on the original IMPROVE algorithm agreed well under the low dry scattering region (<100 Mm<sup>-1</sup>), which was likely due to the MSEs of chemical species for PM<sub>2.5</sub> in the original IMPROVE algorithm were approximate to those minimum MSEs for PM<sub>1</sub> in this study (Table 1). However, they had low consistency under high dry scattering region for the original IMPROVE algorithm. This phenomenon was also observed in the revised IMPROVE algorithm. The dry scattering coefficients calculated by the revised IMPROVE algorithm were lower than measured values, which is consistent with that the MSEs in large mode for the revised IMPROVE algorithm are lower than those maximum MSEs in this study. Good consistencies of the results between the calculated and measured scattering coefficients under low and high scattering regions in both Shanghai and Dezhou proved the reliability of fitting formula in this study.

Quantitative deviations between the calculated and measured dry scattering coefficients for four methods were also estimated. In Shanghai, the reconstructed dry scattering coefficient showed a 37% overestimation when using MSEs fitting by Huang et al. The estimated dry scattering coefficients using the original IMPROVE algorithm and the revised IMPROVE algorithm were 48% and 17% lower than the measurements, respectively. The dry scattering coefficients calculated by the formula in this study were only 4% lower than the measured values (Fig. 8a). In Dezhou, the calculations were overestimated by 56% for multiple regression fitting. For the original IMPROVE algorithm, the calculated dry scattering coefficient was underestimated by 60% (Fig. 8b). However, the application deviations for Dezhou area using the revised IMPROVE algorithm and the formula fitted in our study were 4% and 7%, respectively. In general, the fitting formula in this study could largely reduce the integrated bias, and predict the dry scattering coefficient well based on chemical composition data in typical urban areas of China.

#### 4. Conclusions

Mass scattering efficiencies (MSEs) of aerosol chemical species are important for assessing the radiation effect of atmospheric aerosols. We investigated the mass-size distributions of major chemical species in PM<sub>1</sub> during spring, summer, winter in urban Shanghai with an Aerodyne AMS. The hourly variation in MSEs of major chemical species were estimated based on Mie theory with the observation data. The average estimated MSEs of three scattering species, i.e., organic matter (OM), ammonium nitrate  $(NH_4NO_3)$  and ammonium sulfate  $((NH_4)_2SO_4)$ , were  $5.3 \pm 0.5 \text{ m}^2/\text{g}$ ,  $4.3 \pm 1.0 \text{ m}^2/\text{g}$  and  $4.2 \pm 0.6 \text{ m}^2/\text{g}$ , respectively. As the mass concentrations of NH<sub>4</sub>NO<sub>3</sub> and  $(NH_4)_2SO_4$  were lower than 12  $\mu$ g/m<sup>3</sup>, their MSEs enhanced largely with the increasing of mass concentration. When their mass concentrations were higher than  $12 \mu g/$ m<sup>3</sup>, the MSEs of NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> fluctuated in a very narrow range  $(5.2-5.4 \text{ m}^2/\text{g})$ . However, the MSE of OM showed little variation with the increase of mass concentration. The formulas were applied to fit the MSEs of OM, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>NO<sub>3</sub> with their mass concentration and reconstructed new dry scattering coefficients. Reconstructed dry scattering coefficients versus in-situ measurement results had good correlations and small deviations in both Shanghai ( $R^2 = 0.95$ , slope = 1.10) and in Dezhou ( $R^2 = 0.90$ , slope = 1.10).



Fig. 7. Temporal variation of calculated scattering coefficient in (a) Shanghai and (b) Dezhou. MLR is the abbreviation of multiple linear regression. The color blocks mark the calculated scattering coefficient. The black solid line presents the measured scattering coefficient. The black line disconnection is caused by missing data due to instrument failure.

Our findings indicate that establishing formula for the mass scattering efficiencies of  $PM_1$  major chemical species under high mass loadings is necessary to supplement the calculation of the dry scattering coefficient of particulate matter in urban China. The empirical formula for  $PM_1$  dry scattering coefficient obtained in this study is expected to be applicable during both polluted and clean periods for other urban regions of China, except for special pollution events such as sandstorms.



**Fig. 8.** Linear correlations between PM<sub>1</sub> calculated scattering coefficient and measured values under dry condition in (a) Shanghai and (b) Dezhou.

#### **Declaration of interests**

- The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.
- The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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#### Appendix A. Supplementary data

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