Ambient Atmospheric Aerosol Extinction Coefficient Reconstruction from PM_{2.5} Mass Concentrations and Application to Haze Identification in China

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ABSTRACT

This study developed a method of reconstructing the aerosol extinction coefficient based on hourly observations of the fine-particle (PM_{2.5}) mass concentration, relative humidity (RH), and visibility at 9 stations in China between 2014 and 2015. First, we applied κ -Köhler theory to evaluate the number concentration distribution of the fine particles under ambient conditions from the PM_{2.5} mass and then used Mie theory to calculate the aerosol extinction coefficient. Second, we established the reconstruction model and identified reference values for the relevant parameters. After sensitivity tests confirmed good agreement between the extinction coefficients obtained through combinations of various values and those resulting from the reference values, linear regression was employed to reduce the discrepancy between the reconstructed and the observed coefficients. A closure study enabled us to determine the threshold of the extinction ratio (β/β_{Obs}) and identify haze and fog weather phenomena at the stations. Finally, we assessed the bias in the predicted number of hours with haze for 61 stations in China by comparing the estimates derived from different values for the model's parameters with those derived from the reference values and found a relative bias of less than 15% for approximately 99.8% of the stations, indicating the feasibility of our approach for detecting haze.

Keywords: Aerosol extinction reconstruction, Haze and fog, Identification

1 INTRODUCTION

Low-visibility events have frequently occurred in many regions in China due to intense industrial activities and the high population density during recent years, and as a result, high aerosol loadings cause a series of problems affecting human health, economics and climate (Charlson *et al.*, 1987; Tegen *et al.*, 2002; Yu *et al.*, 2002; Chan and Yao, 2008; Zhang *et al.*, 2015). Aerosol particles can directly degrade the visibility through the extinction effect in the visible light spectrum (Yu *et al.*, 2014). However, low-visibility episodes (e.g., with a visibility less than a few hundred meters) can also be caused by water droplets or ice crystal or aerosol hygroscopic growth under high-humidity conditions (Chen *et al.*, 2012; Deng *et al.*, 2016). For example, particles with a high hygroscopicity have been observed in the North China Plain (NCP) (Meier *et al.*, 2009; Liu *et al.*, 2011; Wu *et al.*, 2017; Liu *et al.*, 2020). It has also been revealed that the occurrence of severe haze events is

OPEN ACCESS

Received: July 8, 2020

Revised: January 21, 2021

Accepted: February 2, 2021

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Publisher:

Taiwan Association for Aerosol
Research

ISSN: 1680-8584 print

ISSN: 2071-1409 online

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closely related to hydrophilic inorganic water-soluble ions, including sulfate, nitrate and ammonium, in the Yangtze River Delta (YRD) region (Leng *et al.*, 2016). A high ambient humidity can increase the aerosol liquid water content and enlarge the aerosol particle diameter, thereby enhancing the extinction effect and degrading the visibility in the Pearl River Delta (PRD) region (Deng *et al.*, 2016). On the basis of the aerosol hygroscopic observations mentioned above, it is necessary to investigate the statistical relationship between the atmospheric aerosol concentration and visibility under the influence of humidity.

Closure studies can be applied for the estimation of the uncertainty of measurement techniques or numerical models. Many studies exist on the reconstruction of light extinction from aerosol measurements, and a relatively good agreement has been achieved with direct observations. For example, Ma *et al.* (2011) calculated the dry aerosol scattering coefficient from particle number size distribution (PNSD) observations under different mixing state assumptions with a modified Mie model and reported good correlation coefficients ($R > 0.98$) between their calculations and direct measurements by nephelometers. Eichler *et al.* (2008) derived the ambient PNSD from dry PNSD and size-dependent particle hygroscopic growth factors and found that compared to Raman Lidar retrievals, the extinction coefficient under the ambient relative humidity (RH) was suitably estimated with a mean bias less than 3%. Regression equations of the light extinction were also developed on the basis of the aerosol volume concentration and RH with a good agreement between the regression calculations and visibility measurements (Chen *et al.*, 2012).

In addition, there are many other studies on aerosol extinction coefficient reconstruction from the aerosol mass concentration. The Interagency Monitoring of Protected Visual Environments (IMPROVE) method is one basic protocol to reconstruct aerosol light extinction properties from the aerosol chemical composition, although a certain bias exists with direct observations (Lowenthal and Kumar, 2003; Tao *et al.*, 2012; Fu *et al.*, 2016). Focusing on the over- or under-prediction of extinction for different haze periods with the IMPROVE method, many revised algorithms have been developed, and a better agreement has been achieved (Ryan *et al.*, 2005; Wang *et al.*, 2016; Wang *et al.*, 2017; Pitchford *et al.*, 2019). A notable correlation (e.g., $R^2 = 0.8$ for the NCP) between the extinction coefficient and aerosol mass concentration was observed by Luan *et al.* (2018). Studies have used the mass extinction efficiency to establish quantitative relationships between the ambient extinction coefficient (visibility) and fine-particle ($PM_{2.5}$) mass concentration (Cheng *et al.*, 2017). Many different models have also been developed to estimate the long-term historical or spatial-temporal $PM_{2.5}$ concentration from ground visibility monitoring data or satellite retrieval products (Han *et al.*, 2016; Liu *et al.*, 2017; Zeng *et al.*, 2018) since Kastex *et al.* (1969) introduced the parameter fit equation between the aerosol scattering hygroscopic growth factor $f(RH)$ and RH. In this study, the aerosol extinction coefficient is reconstructed from $PM_{2.5}$ and RH observations based on statistical methods under assumptions for PNSD and aerosol density and hygroscopic growth, and aerosol extinction closure estimation has been conducted. The results of this study will provide a quantitative relationship between the ambient visibility, $PM_{2.5}$ mass concentration and RH, and offer a practical method to distinguish haze and fog.

In this paper, a brief introduction of the data and method for aerosol extinction coefficient reconstruction is presented first, followed by the sensitivity analysis and its correction method for reconstructed ambient atmospheric extinction coefficients; and the closure study of the aerosol extinction method and its application for haze identification are given next. Summary and discussions are given in the end.

2 DATA

Hourly $PM_{2.5}$ and PM_{10} mass concentration data from 2014 to 2016 were obtained from the Air Quality Monitoring Network of the Ministry of Ecology and Environment and the China Atmosphere Watch Network (CAWNET) of the China Meteorological Administration (CMA) (Wang *et al.*, 2015; Gao *et al.*, 2020; Zhang *et al.*, 2020). The data in 2016 are used for method evaluation. High-quality hourly meteorological parameters are acquired from the National Meteorological Information Center of the CMA (<http://idata.com>). Most meteorological and aerosol monitoring stations are not collocated. Hence, only the aerosol monitoring stations with corresponding meteorological observations within 3 km are considered in this study. Strict data quality control

was performed for the analysis. Routine maintenance and calibration procedures for aerosol equipment are referenced according to relevant station logging files, and the nonlinear relationship between the visibility, $PM_{2.5}$ and RH is also considered for station selection, referring to Chen *et al.* (2012) (shown in Fig. 1), while data with precipitation are removed. In addition, meteorological and aerosol observations at 61 stations in China are also adopted to evaluate the applicability of the method.

Data were also screened to eliminate the influence of sand weather. It has been reported that the mean mass concentration ratio of fine particles $PM_{2.5}$ to PM_{10} ($PM_{2.5}/PM_{10}$) is above 0.5 at most stations in China except for Lhasa in Tibet (0.41) from 2006–2014 (Wang *et al.*, 2015). The $PM_{2.5}/PM_{10}$ ratio gradually decreases to approximately 0.2 when sand/dust storms occur (as indicated in Table 1). Thus, observations with $PM_{2.5}/PM_{10}$ ratios lower than 0.5 are treated as affected by sand/dust storms and removed in this study.

3 METHOD FOR AEROSOL EXTINCTION COEFFICIENT RECONSTRUCTION

Observational studies of aerosol optical properties and aerosol size distributions in China suggest that $PM_{2.5}$ contributes more than 80% of the aerosol extinction coefficient under polluted conditions, and the aerosol extinction coefficient can be suitably estimated from the $PM_{2.5}$ mass concentration (Ma *et al.*, 2011). In this study, a method based on the Mie model (Wiscombe, 1979, 1980) is applied to reconstruct the extinction coefficient from the $PM_{2.5}$ mass concentration under assumptions of the aerosol size distribution and hygroscopic growth. The introduction to the related parameters and reconstruction method are as below.

Fig. 1. The site locations of the 9 aerosol monitoring stations in China (blue dot) and scatter plot of the visibility (V) against RH- $PM_{2.5}$ at each station from 2014–2015. The number (Num) in each panel denotes the validated data (in hours) after precipitation and sand/dust hours were removed. The red triangles denote the site locations of the 61 stations for method evaluation in China. And the lime plus denotes the location of SDZ.

Table 1. The PM_{2.5}/PM₁₀ ratio during sand/dust events and on non-sand/dust days in different places in China.

City	Non-sand/dust days	Sand/dust storms	Period	Reference
Beijing	0.38–0.88	~0.3	2000 Apr 25	Xie <i>et al.</i> (2005)
	> 0.6	< 0.2	2013 Feb 27	Xu <i>et al.</i> (2016)
	> 0.6	< 0.2	3013 Mar 8–9	Xu <i>et al.</i> (2016)
Shanghai	~0.58	~0.19	2007 Apr 2	Fu <i>et al.</i> (2010)
Xi'an	~0.55	~0.32	2010 Apr	Wang <i>et al.</i> (2013)
Wuhan	~0.5	0.1–0.5	2013 Mar 9–10	Cao <i>et al.</i> (2015)
Guangzhou	0.73–0.79	0.30	2009 Apr 26–28	Wu <i>et al.</i> (2011a)
Chengdu	~0.72	~0.40	2013 Mar	Chen <i>et al.</i> (2015)

3.1 Aerosol Complex Refractive Index

The dry aerosol complex refractive index ($\tilde{M}_{dry}$) is obtained from studies in northern China (Chen *et al.*, 2012; Ma *et al.*, 2015). The “wet” aerosol complex refractive index ($\tilde{M}$) under ambient conditions is calculated based on the two-component (dry aerosol and water uptake) aerosol model (Chen *et al.*, 2012; Ma *et al.*, 2015):

$$\tilde{M} = \frac{D_{dry}^3}{D_{wet}^3} \times \tilde{M}_{dry} + \frac{D_{wet}^3 - D_{dry}^3}{D_{wet}^3} \times \tilde{M}_{water} \quad (1)$$

where D_{dry} is the diameter of dry aerosol particles, and D_{wet} is the diameter of wet aerosol particles (Wex, 2002; Ma *et al.*, 2011).

3.2 Aerosol Diameter under Wet Conditions

The calculation of ambient aerosol extinction coefficient required the size of wet aerosol. The size parameter (x) of wet aerosol at wavelength of incident light (λ) is expressed as:

$$x = \frac{\pi D_{wet}}{\lambda} \quad (2)$$

where ambient aerosol particle diameter D_{wet} can be calculated through the κ -Köhler theory (Köhler, 1969; Petters and Kreidenweis, 2007) as described in Eq. (3):

$$S = \frac{D_{wet}^3 - D_{dry}^3}{D_{wet}^3 - D_{dry}^3 (1 - \kappa)} \exp \left(\frac{4\sigma_s/a M_w}{RT \rho_w D_{wet}} \right) \quad (3)$$

where S is the water vapor saturation ratio, which is equivalent to RH under unsaturated conditions (i.e., particles are not activated); κ is the hygroscopicity parameter; σ_s/a is the surface tension coefficient of water; M_w is the mole weight of water; R is the gas constant; T is the ambient temperature; and ρ_w is the density of water. Iterative method is adopted to solve the value of D_{wet} .

The total aerosol volume concentration (V) can be obtained from the aerosol mass concentration (M):

$$M = \rho V \quad (4)$$

Aerosol density is affected by aerosol chemical compositions and their mixing state. Aerosol density for PM_{2.5} can be ranged from 1.2–1.8 in China (Hu *et al.*, 2012; Fan *et al.*, 2018).

The diameter of dry aerosol D_{dry} can be calculated from V when the aerosol size distribution is assumed. Field observations conducted in some regions in China, including the NCP, YRD, and PRD regions, show that the aerosol volume distribution mainly has two modes with one major mode (at about 300–500 nm) and a second mode ($> 3 \mu\text{m}$) (Chen *et al.*, 2006; Yue *et al.*, 2013; Chen *et al.*, 2014; Peng *et al.*, 2014; Kuang *et al.*, 2016). The major mode mainly contains accumulation-mode particles, which are the most important contributor to aerosol light extinction, while the

second mode contains coarse-mode particles whose contribution to light extinction can generally be neglected (D'Andrea *et al.*, 2015; Ma *et al.*, 2015; Qi *et al.*, 2015). Thus, for simplicity, it is feasible to use a one-mode distribution model to represent the aerosol volume size distribution as:

$$\frac{dV}{d\log D_{dry}} = \frac{V}{\sqrt{2\pi} \log \sigma_g} \exp\left(-\frac{(\log D_{dry} - \log D_g)^2}{2\log^2 \sigma_g}\right) \quad (5)$$

where D_g and σ_g are the geometric mean and standard deviation, respectively, for dry aerosol particles. From previous studies, D_g ranges from 300 nm to 500 nm, and σ_g ranges from 1.4 to 2.2 in China (Peng *et al.*, 2014; Ma *et al.*, 2015).

3.3 Aerosol Hygroscopicity Parameter (κ)

The value of κ depends on the aerosol chemical composition. It can be calculated according to the simple mixing rule by the volume fraction (ϵ_i) and hygroscopicity parameter (κ_i) of the aerosol chemical composition (Petters and Kreidenweis, 2007):

$$\kappa = \sum_i \epsilon_i \kappa_i \quad (6)$$

For high hygroscopic aerosol component such as sulfate, κ_i can be above 0.5. For hydrophobic aerosol component such as black carbon, organic carbon or sand dust, κ_i can be below 0.1. Previous studies on the value of κ in China are summarized in Table 2. In general, the value of κ is approximately 0.3 in different regions in China. For example, using a κ value of 0.3 with a standard deviation of 20% can suitably describe the observed cloud condensation nuclei (CCN) concentration in Guangzhou (Rose *et al.*, 2010).

4 SENSITIVITY ANALYSIS AND EXTINCTION CORRECTION

Based on the analysis above, the values of κ , ρ , D_g and σ_g are all within a certain range. A set of recommended values of the parameters are given in Table 3, i.e., they are set to be 0.3, 1.5 g cm⁻³, 400 nm and 1.8 in the reconstruction model and are designated as *recommended parameters*. Hereafter, the reconstruction method with the recommended parameters is referred to as the “reference calculation.” The sensitivity of these parameters will be examined in the following sections.

4.1 Test for Hygroscopicity Parameter

When κ is equal to be 0.3, D_{wet} can be obtained from the κ -Köhler theory (Eq. (3)). Then, the size growth factor of aerosols $g(RH)$ at different RHs (e.g., 85–95%) can be calculated with Eq. (7) (Liu *et al.*, 1978):

$$g(RH) = D_{wet}/D_{dry} \quad (7)$$

Table 2. Previous studies on the aerosol hygroscopicity parameter (κ) in different regions in China.

Region	κ value (diameter in nm)			Reference
Beijing	0.25 (< 50)	0.45 (50–2000)	0.3 ± 0.1 (mean)	Gunthe <i>et al.</i> (2011)
	0.16 (50)	0.19 (100)	0.26 (200)	Wu <i>et al.</i> (2016)
Tianjin	0.25 (50)	0.28 (100)	0.32 (200)	Liu <i>et al.</i> (2011)
Shanghai	0.25 (50)	0.28 (100)	0.31 (200)	Ye <i>et al.</i> (2013)
	0.24 (50)	0.28 (100)	0.31 (200)	Ye <i>et al.</i> (2011)
Nanjing	0.25 (< 430)	0.40 (430–2100)	0.25 (2100–1000)	Zhang <i>et al.</i> (2017)
Guangzhou	0.25 (50)	0.40 (200)	0.30 (mean)	Rose <i>et al.</i> (2010, 2011)
Numerical model	0.27 ± 0.21 (mean over land)			Pringle <i>et al.</i> (2010)
	0.72 ± 0.24 (mean over ocean)

Table 3. Constants in this study.

Constant	Recommended value (units)
Aerosol complex refractive index for dry aerosols ($\tilde{M}_{dry}$)	1.55–0.04i
Aerosol complex refractive index for water ($\tilde{M}_{water}$)	1.33–0.00i
Hygroscopicity parameter (κ)	0.3
Surface tension coefficient of water ($\sigma_{s/a}$)	7.42×10^{-2} (N m ⁻¹)
Mole weight of water (M_w)	18×10^{-3} (kg mol ⁻¹)
Gas constant (R)	8.3145 (J mol ⁻¹ K ⁻¹)
Ambient temperature (T)	283.15 (K)
Water density (ρ_w)	1.0×10^3 (kg m ⁻³)
Aerosol density (ρ)	1.5×10^3 (kg m ⁻³)
Geometric mean aerosol diameter (D_g)	400 (nm)
Standard deviation of aerosol diameter (σ_g)	1.8
Wavelength of incident light (λ)	550 (nm)

Comparisons of the calculated curves of $g(RH)$ at different RHs with field observations in different cities, including Beijing (Massling *et al.*, 2009), Tianjin (Liu *et al.*, 2011), Shanghai (Ye *et al.*, 2011), and Guangzhou (Eichler *et al.*, 2008), are shown in Fig. 2. The calculated $g(RH)$ values are consistent with the field observations in most cases. However, the calculations are lower than the observations when RH is high and the aerosol particle size is large.

Furthermore, we adopt the observations at Lanzhou (LZ) station from July to December 2015, to investigate the sensitivity of calculated β_{Mie} to parameter κ . The PM_{2.5} mass concentration ranged from a few $\mu\text{g m}^{-3}$ to more than 500 $\mu\text{g m}^{-3}$ at this station, which can represent the normal PM_{2.5} situations in China. β_{Mie} is calculated with different κ values (0.1–0.5), as shown in Fig. 3. The mean β_{Mie} values are 544, 612, and 679 Mm^{-1} for κ values of 0.2, 0.3 and 0.4, respectively, and the correlation coefficients between β_{Mie} ($\kappa = 0.3$) and β_{Mie} ($\kappa = 0.2$ or 0.4) are 0.99 at a confidence level of 95%. Even for the nearly hydrophobic ($\kappa = 0.1$) or mostly hydrophilic aerosols ($\kappa = 0.5$), the correlation coefficients are also larger than 0.90. This means that the value of κ mainly affects the absolute values of β_{Mie} .

4.2 Test for Aerosol Volume Size Distribution

To investigate the impact of implementing the single-mode aerosol volume size distribution in aerosol extinction calculations, β_{Mie} is determined using the bimodal log-normal volume size

Fig. 2. Aerosol particle diameter variations of $g(RH)$ under different RH conditions (85–95%). The curves denote the calculated $g(RH)$ when κ is equal to 0.3, and the scatter points denote the field observations obtained in different cities in China.

Fig. 3. Correlations between β_{Mie} calculated for $\kappa = 0.3$ (reference calculation) and other κ values (0.1–0.5). The asterisk denotes that the correlation coefficient has passed the t -test at a confidence level of 95%.

Fig. 4. Correlations between β_{Mie} calculated using the reference parameters and bimodal log-normal volume size distribution or single fine-mode distribution with different D_g and σ_g values at Lanzhou (LZ) station from July to December 2015. The asterisk indicates that the correlation coefficient has passed the t -test at a confidence level of 95%.

distribution assumption with a second coarse-mode distribution in addition to the reference fine-mode volume distribution, and parameter values of $D_g = 3 \mu\text{m}$ and $\sigma_g = 1.6$ are assumed for the second (or coarse) mode (Ma *et al.*, 2015). Hence, the PM_{10} mass concentration is also adopted to calculate the coarse-mode volume concentration. In this test calculation, other parameters, such as κ and ρ for both modes and D_g and σ_g for the fine mode, are the same as those in the reference calculation. The results and a comparison of β_{Mie} calculated with the bimodal distribution to the reference calculation (single fine-particle mode) at the LZ station are shown (in gray dashed line) in Fig. 4.

β_{Mie} calculated with the bimodal normal volume size distribution attains a very high correlation coefficient (approximately 1.00) with that calculated using the single fine mode. This implies that

Table 4. Mean values of β_{Mie} (unit: Mm^{-1}) calculated with different D_g and σ_g values and correlation coefficients (R) between the calculated β_{Mie} values and those obtained with the reference parameters at LZ station from July to December 2015. The asterisk denotes that the correlation coefficient has passed the t -test at a confidence level of 95%.

D_g (nm)	$\sigma_g = 1.4$		$\sigma_g = 1.6$		$\sigma_g = 1.8$		$\sigma_g = 2.2$	
	Mean	R	Mean	R	Mean	R	Mean	R
100	176.73	0.99*	197.51	0.99*	217.68	1.00*	247.85	1.00*
200	438.61	0.99*	443.65	0.99*	441.25	1.00*	425.27	1.00*
300	639.66	1.00*	603.66	1.00*	568.10	1.00*	511.50	1.00*
400	721.95	1.00*	662.18	1.00*	612.01	1.00*	543.75	1.00*
500	716.38	1.00*	655.60	1.00*	608.79	1.00*	549.01	1.00*
600	662.21	0.99*	616.44	1.00*	583.45	1.00*	541.69	1.00*

the contribution of coarse-mode particles is very small and can be neglected in terms of the aerosol extinction coefficient during periods without dust weather influences. The single fine-mode normal distribution can be reliably applied to describe the volume size distribution of atmospheric aerosol particles under polluted or haze weather conditions.

Another aspect of uncertainty stems from the variation in D_g and σ_g values, which may vary over a wide range in different regions and seasons. Thus, β_{Mie} is also calculated using the single fine-mode log-normal distribution with different D_g (100–600 nm) and σ_g (1.4–2.2) values, where κ and ρ remain the same as the reference calculation. The results are shown in Fig. 4 and Table 4.

The mean value of β_{Mie} calculated with different D_g and σ_g values ranges widely from 176.73 to 721.95 Mm^{-1} (Table 4). However, the correlation coefficient is greater than 0.99. This is consistent with previous observation studies (Chen *et al.*, 2012).

4.3 Test for Aerosol Density (ρ)

The sensitivity of the aerosol density is also evaluated. The mean β_{Mie} value calculated at LZ station from July to December 2015 is 540.59 Mm^{-1} with ρ equal to 1.7 $g\ m^{-3}$ and the other parameters remain the same as the reference parameters. The correlation coefficient between the β_{Mie} values for ρ equal to 1.7 $g\ m^{-3}$ and 1.5 $g\ m^{-3}$ is approximately 1.00.

4.4 Linear Regression

From the discussions above in this section, it is found that the reconstructed extinction coefficients with the reference parameters may differ from the real values due to the site dependence of the parameters. The influence of aerosol size distribution against the reference parameters on β_{Mie} ranged from –71.1% to 18.0%. The influence of the maximum and minimum κ value against the reference κ on β_{Mie} ranged from –22.2% to 21.9%. When aerosol density increased from 1.5 to 1.7 $g\ m^{-3}$, β_{Mie} decreased by –11.7%. However, the correlations of β_{Mie} between the reference calculation and the sensitivity test calculation for the different parameters agree quite well. These results suggest that the biases of the reconstructed β_{Mie} can be corrected according to the site observations. Accordingly, the linear regression method is adopted to correct the reconstructed aerosol extinction coefficients based on visibility site observations. The regression correction method is described as follows.

Firstly, from Koschmieder's equation (Koschmieder, 1924), the observed ambient atmospheric extinction coefficient (β_{Obs}) can be obtained.

Generally, it is recognized that when RH is below 80%, even considering RH measurement errors and humidity fluctuation, fog droplets are hardly formed in the atmosphere, i.e., the light extinction of the atmosphere is only caused by aerosol particles (including the enhancement via aerosol hygroscopic growth) and air molecules. This is consistent with the guidance for haze observations issued by many organizations and researchers (Wu, 2005; WMO, 2008; CMA, 2010). Thus, the observed atmospheric extinction coefficients with $RH < 80\%$ and visibility < 10 km, which are equivalent to the ambient aerosol extinction coefficients, are used to build a correction equation for β_{Mie} . The slope (a) and interception (b) of the linear regression equation for β_{Obs} with β_{Mie} are determined through the least-squares fitting method. Therefore, corrected values of the reconstructed aerosol extinction coefficient β can be obtained by:

$$\beta = a \cdot \beta_{Mie} + b \quad (8)$$

With the use of the data at the 9 stations in China from 2014–2015, the slope, intercept, and reconstructed and corrected aerosol extinction coefficients β_{Mie} and β , respectively, at each station are obtained, as shown in Fig. 5.

It is shown (Fig. 5) that the mean correlation coefficient (R) of the 9 stations is 0.82, with a maximum value of 0.92 at YCG station and a minimum value of 0.59 at PY station. This implies that the extinction coefficient reconstruction method performs quite well at the stations. Low correlation coefficient at PY may be related to the matching problem of $PM_{2.5}$ and visibility observations. The mean biases between the reconstructed aerosol extinction coefficients and corresponding observations with and without linear regression correction at the 9 stations are -35 Mm^{-1} (a relative bias of approximately 5%) and -87 Mm^{-1} (a relative bias of approximately 18%), respectively. There are differences in the values of linear slope and intercept among different stations, which implies different size distribution or hygroscopicity characteristics at these stations. Considering the measurement errors in $PM_{2.5}$, RH and visibility, it can be concluded that linear regression correction has effectively improved the reconstructed aerosol extinction calculation.

5 AEROSOL EXTINCTION CLOSURE AND APPLICATION FOR HAZE IDENTIFICATION

According to the World Meteorological Organization handbook for weather observations (WMO, 2008), haze is defined as the degradation of visibility by particles, which means that under haze conditions, light extinction of the atmosphere is mainly caused by aerosols and the enhancement of light extinction due to aerosol hygroscopic growth. Fog is another low-visibility weather phenomenon. However, under fog weather conditions, light extinction of the atmosphere is generally dominated by fog droplets, and their contributions to visibility degradation are much greater than those from aerosol particles (Elias *et al.*, 2009). Considering that uncertainties exist in the observations and aerosol light extinction calculations, it is reasonable to assume that when the contribution of light extinction from aerosols to atmospheric extinction exceeds a certain threshold, light extinction of the atmosphere can be regarded as mainly caused by aerosols. Under such an assumption, the threshold of the contribution of light extinction from aerosols can be derived through an aerosol optical extinction closure study.

Fig. 5. Mean aerosol extinction coefficients at the 9 stations in China from 2014–2015. β_{Mie} and β are the reconstructed and corrected aerosol extinction coefficients, respectively. β_{Obs} is the aerosol extinction coefficient calculated from the visibility observations. R is the correlation coefficient between the calculated and observed aerosol extinction coefficients.

5.1 Optical Extinction Closure under Low-RH Conditions and the Extinction Ratio

The observations from 2016 are used to study the extinction closure under low-RH conditions. The method for light extinction coefficient reconstruction is described in the previous sections (the third section and the fourth part of the fourth section). The relationship between the calculated and observed extinction coefficients at 9 stations under $RH < 80\%$ is shown in Fig. 6. The correlation coefficient is 0.84, and the slope is approximately 0.70.

Since low-visibility events (i.e., visibility < 10 km) with $RH < 80\%$ are mainly caused by atmospheric aerosol particles, the calculated and corrected light extinction coefficients β should be close to the observations β_{Obs} under haze conditions, but much smaller than β_{Obs} under fog conditions. Therefore, the ratio of β to β_{Obs} (β/β_{Obs}) could be used to distinguish haze from fog. The ratio of haze identified by setting different β/β_{Obs} thresholds ($Haze$) against the direct observations ($Haze_{Obs}$), with $RH < 80\%$ and visibility < 10 km is shown in Fig. 7(a). The probability distribution function (PDF) of β/β_{Obs} for the data is also shown in Fig. 7(b). The $Haze/Haze_{Obs}$ ratio decreases with increasing β/β_{Obs} ratio threshold (Fig. 7(a)). When β/β_{Obs} ratio threshold increases from 0.7 to 1.0, $Haze/Haze_{Obs}$ decreased from approximately 0.9 to less than 0.6. When we set β/β_{Obs} ratio threshold to be 0.8, about 84% of haze out of the total haze samples can be identified, with the highest value 92% at LZ station and the lowest value 79% at QD.

Ideally, if the contributions of atmospheric extinction were only from fine particles and air molecules, and the aerosol model and parameters were correct, β/β_{Obs} should be approximately 1.00 when $RH < 80\%$ and visibility < 10 km. However, in actual cases, only 65% and 79% of the total observation samples have β/β_{Obs} ratios higher than 0.9 and 0.8, respectively (Fig. 7(b)). This is related to the uncertainties caused by the assumptions on aerosol model and parameters used in the extinction coefficient reconstruction method and observation errors, such as RH observation errors, and so on. This will be discussed in the following sections. Thus, we recommend a β/β_{Obs} ratio of 0.8 as the threshold value based on both identification ability and observation facts.

Conclusively, adopting the recommended β/β_{Obs} ratio threshold 0.8, about 16% of haze hour in average could not be identified, with the maximum value 20% and the minimum value 7%, implying that the uncertainty of the method can be about 16%.

5.2 Application for Haze Identification under High-RH Conditions

Based on the discussions above, we can apply the method and parameters described above to identify the haze weather phenomenon when RH is high, i.e., $80\% \leq RH < 95\%$, according to the

Fig. 6. Relationship between the calculated (β) and observed (β_{Obs}) extinction coefficients at $RH < 80\%$ at the 9 stations in 2016.

Fig. 7. (a) Relationship between the $Haze/Haze_{Obs}$ and β/β_{Obs} ratios and (b) the probability distribution of β/β_{Obs} at $RH < 80\%$ and visibility < 10 km at the 9 stations in China from 2014–2015.

ratio of β/β_{Obs} higher than 0.8. The haze and fog hour at the 9 stations are shown in Fig. 8. Different stations have different proportion of haze to fog hour, which may be related to the regional climate conditions and pollutant emission distributions. HZ, XJH, and YCN have higher probabilities of fog/mist than other stations. This is consistent with previous studies that reported more fog/mist days in these locations (Wu *et al.*, 2011b). And this indirectly proves the reliability of the method. Fog mainly concentrated in southeast coastal regions (such as HZ and XJH) and Sichuan Basin but the least in northwest region, the Qinghai-Tibet Plateau and Inner Mongolia (Sun *et al.*, 2008). Besides, mass concentration at HZ and XJH are lower than that at other stations. The reason why YCN had comparable haze and fog hour may be related to its specific terrain-related low-altitude temperature inversion (Deng *et al.*, 2018). YCG, CP, QD, LZ, YJ, and PY have higher

Fig. 8. Haze and fog/mist hours, $PM_{2.5}$ mass concentration, and RH at the 9 stations at $80\% \leq RH < 95\%$ in China from 2014–2015.

Table 5. Variation in the parameters used in sensitivity tests.

Parameters	Values
D_g	250, 400 and 550 nm
σ_g	1.5, 1.8 and 2.1
κ	0.2, 0.3 and 0.4

probability of haze hours. Firstly, this may be related to higher aerosol loadings at these stations. After all, the essence of haze is the pollution process caused by high aerosol loadings. PY has relatively low $PM_{2.5}$ concentration, but has relatively high probability of haze. This may be related to its relatively high PM_1 loading and its hygroscopicity growth (Yue *et al.*, 2013).

5.3 Sensitivity of All Parameters to Haze Identification

To analyze the uncertainties of haze identification using the method presented above, we apply several combinations of parameters with different values, which are likely under real atmospheric conditions, to a series of sensitivity tests in terms of haze identification at the 61 stations. The variations in the parameters are listed in Table 5.

The relative bias of the haze hours identified with the different parameter values against the reference calculation is shown in Fig. 9. The mean value of the bias is -0.87% for the 26 sensitivity tests over 61 stations, with the $D_g\text{--}\sigma_g\text{--}\kappa$ combination 250 nm–2.1–0.2 has the highest negative bias -7.9% , and the combination 550 nm–1.8–0.4 has the lowest positive bias 3.4% .

Uncertainties of the method varies from different stations. The maximum relative bias is -16.8% for 61 stations. A total of 63.6% of all stations has a relative bias smaller than 5%, and 94.3% of all stations has a relative bias smaller than 10%, while 99.8% of all stations has a relative bias smaller than 15%. This implies that the method is feasible for haze identification. Over all, mean haze hour bias by all parameters is approximately -0.87% .

5.4 Uncertainties by RH and $PM_{2.5}$ Observation Errors

Monte Carlo method is adopted to estimate uncertainties brought by RH observation errors. RH, visibility and $PM_{2.5}$ observations at 9 stations during 2015–2016 are used. RH errors between -5% and 5% are randomly added to the RH observations when $90\% \leq RH < 95\%$ (12,025 samples). On the basis of the reference test, 10 groups of tests have been carried out. Therefore, there are 120,250 sets of samples used. Total haze hour at 9 stations are calculated. Compared with the reference test, the mean haze hour bias is -5.2% with a standard deviation 5.5%. Thus, haze hour uncertainty brought by RH observation errors can be estimated at approximately -5.2% .

Fig. 9. Haze hour bias ratio identified for the different parameters ($D_g\text{--}\sigma_g\text{--}\kappa$) against the reference parameters (400 nm–1.8–0.3) from 2014–2015 at 61 stations in China.

Fig. 10. Haze hours identified by method used in this study (M_{Mie}), M80 and M90 at the Beijing Shangdianzi station during 2015–2016.

Ambient aerosol particles may be enlarged under high-RH conditions. And particles with $D_{wet} > 2.5 \mu\text{m}$ will be cut off by the $\text{PM}_{2.5}$ inlet. The influence of the size cut-off by $\text{PM}_{2.5}$ inlets was estimated adopting the extinction coefficient reconstruction method proposed above. Aerosol extinction coefficients were calculated under different RH conditions with the same dry aerosol size distribution. The influence of the cut-off on aerosol extinction coefficient is estimated at $< 0.4\%$ under low-RH conditions ($\text{RH} < 80\%$). It is between 0.6–2.5% under high-RH conditions ($\text{RH} > 80\%$).

5.5 Comparisons of Different Haze Identification Methods

Traditional haze identification methods are mainly based on visibility and relative humidity. Main difference between these methods is in the selection of RH threshold. For example, one haze identification method widely used in observation service in some provinces in China is defined as follows: eliminating the low-visibility phenomena caused by precipitation, sand/dust or blowing snow; $\text{RH} < 80\%$; and horizontal visibility $< 10 \text{ km}$. We name the method M80. And another haze identification method widely used in scientific research is similar to the M80, except for $\text{RH} < 90\%$, which is named M90. The method proposed in this work is named M_{Mie} .

Comparison of the haze identification results at the Beijing Shangdianzi station (SDZ; Station ID: 54421; location shown in Fig. 1) during 2015–2016 is shown in Fig. 10. It can be found that M80 identified the least haze hour and M90 identified the most. And the haze hour identified by M_{Mie} is in the middle of the two methods.

6 SUMMARY AND DISCUSSION

This study established a model for reconstructing the aerosol extinction coefficient based on hourly observations of the fine-particle ($\text{PM}_{2.5}$) mass concentration, relative humidity (RH), and visibility at 9 stations in China. We obtained the particle number concentration distribution of the dry aerosol from the $\text{PM}_{2.5}$ mass and then calculated it for the wet aerosol using κ -Köhler theory. Finally, we evaluated the aerosol extinction coefficient by employing Mie theory.

A closure study conducted under ambient conditions allowed us to derive the threshold of the extinction ratio by comparing the estimated light extinction values to the observed ones (which were determined from the measured visibility). When the RH was low, the method proposed in this study accurately detected more than 80% of the hours during which haze occurred. When the RH was high ($80\% \leq \text{RH} < 95\%$), however, this approach more accurately differentiated between haze and fog, which suggests that combining $\text{PM}_{2.5}$ observations with visibility and RH data can enhance haze identification during operational monitoring at meteorological stations.

Although our technique for identifying haze relies on multiple assumptions about the aerosol properties, e.g., the size distribution, hygroscopicity, density, and refractive index, applying linear regression to the model predictions effectively shrank the bias, reducing the overall estimated uncertainty to approximately 16% for the data we analyzed.

ACKNOWLEDGMENTS

This work was performed under the auspices of the National Key Research and Development Program of China (2017YFC1501702) and the National Natural Science Foundation of China (41705133 and 91644223).

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