Estimation of $PM_{2.5}$ from fine-mode aerosol optical depth

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Abstract: The correlation between fine Particulate Matters ($PM_{2.5}$) and Fine-mode Aerosol Optical Depth (AOD_f) is established. AOD_f is obtained from product of Aerosol Optical Depth (AOD) and fine-mode fraction at Beijing site belonging to the AErosol RObotic NETwork (AERONET) in January 2013. And then we compare estimation with observation of $PM_{2.5}$ from 1 to 15 February, 2013. The results show that the developed correction method is effective to estimate $PM_{2.5}$ during haze, with root-mean-square error of 50 $\mu g/m^3$ at a mean level of 85 $\mu g/m^3$. The relationship between AOD_f and $PM_{2.5}$ is obviously better than that of AOD and $PM_{2.5}$. It is also found that when the relative humidity is higher than 80%, the humidity correction on AOD-$PM_{2.5}$ correlation is limited, and the vertical correction cannot improve the correlation during haze.

Key words: aerosol optical depth, $PM_{2.5}$, fine mode fraction

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1 INTRODUCTION

Haze, defined as a large number of small particles suspended in the air, is an air phenomenon leading to the horizontal visibility less than 10 km generally (China Meteorological Administration, 2003). The haze has been widely known as an atmosphere pollution phenomenon of fine particulate matter at the surface due to increasing human activities in urban areas (Wu, 2012). In the past 30 years, with the fast-paced economic development, the rapid growth of the number of vehicles, air pollutions caused by atmospheric fine particles happen frequently (He, et al., 2002). Atmospheric fine particles can cause many diseases, especially respiratory diseases such as chronic bronchitis. Therefore, more attentions are paid to the monitoring of atmospheric fine particulate matter no matter what fields such as environmental protection, public health and scientific research. Use of remote sensing measurement to estimate $PM_{2.5}$ can contribute to monitoring and management of haze pollutions.

There are three methods to estimate fine particulate matter mass concentration at the surface, including the linear regression, the multivariate statistical regression, as well as the model inversion coupled the observation data. Linear regression method has been widely used due to the simplicity and strong extrapolation capability (Wang and Christopher, 2003; Hutchison, et al., 2003; Li, et al., 2003a,b,c, 2005; Engel-Cox, et al., 2004; Koelemeijer, et al., 2006; Xu, et al., 2006; Kumar, et al., 2007; Liu, et al., 2007; Liu, 2007; Schaap, et al., 2009; He, et al., 2009; Van Donkelaar, et al., 2010; Zheng, et al., 2011). In order to build the relationship between AOD and $PM_{2.5}$, the humidity correction, as well as the vertical correction is needed to deal with remote sensing observation in the environment condition (Liu, 2007; He, et al., 2009; Zheng, et al., 2011). Although aerosol optical depth from remote sensing is primarily caused by fine particles, the impact of the fine-mode fraction on the correlation between AOD and $PM_{2.5}$ cannot be ignored. Di Nicolantonio, et al. (2007) estimated fine-mode aerosol optical depth and built the correlation using fine-mode fraction from MODIS inversion. Fine-mode fraction from MODIS is sensitive to complex surface albedo model. And thus inversion stability of fine-mode fraction is poor and the results in their study are not very good. Fine-mode fraction can be obtained not only from satellite observations, but also from ground-based remote sensing observations. Due to the small surface effect in process of the ground-based observation, the fine-mode fractions are more stable and accurate. O'Neill, et al., (2001a, 2001b, 2002, 2003) presented the Spectrum Deconvolution Algorithm (SDA) of fine-mode fraction calculation using multi-band aerosol optical depth.

In this study, we explore the relationship between fine-mode aerosol optical depth AOD_f and $PM_{2.5}$ in the period of haze p

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ollution in January 2013 at Beijing site. Fine-mode aerosol optical depth is estimated using multiband aerosol optical depth, meanwhile, the correlation between AOD_f and $PM_{2.5}$ is established to improve the capacity for monitoring and estimating fine particulates near the surface.

2 DATA AND METHODOLOGY

2.1 Data

Table 1 Date sources and the site location

Data	Location	Time
AOD	AERONET at Beijing site (116.38°E, 39.98°N, 92.00m, #41, 3 wavelength) (116.38°E, 40.00°N, 59.00 m, #350, 5 wavelength)	2013-01
Mixture layer height	NCEP Global reanalysis data (116.47°E, 39.8°N)	2013-01
Relative humidity	Southern observatory 54511 (116.47°E, 39.8°N, 31.3 m)	2013-01
$PM_{2.5}$	U.S. embassy in Beijing (116.43°E, 39.91°N, 45 m)	2013-01

We use the data of aerosol optical and microphysics properties from AERONET (Aerosol Robotic Network) at Beijing site, and observation instrument is Sun-Sky radiometer (CE318). Two instruments are used in Beijing, and then aerosol optical depths are obtained at three wavelengths (#41, 870 nm、675 nm、440 nm) and five wavelengths (#350, 870 nm、675 nm、500 nm、440 nm、380 nm) respectively (listed in Table 1). At the same time, sky light and principal plane scans are carried out. Using these data, aerosol properties can be retrieved, including the whole layer of the atmosphere information, such as Ångström exponent, aerosol volume distribution and fine-mode fraction. We mainly use AOD at 500 nm from AERONET Lev 1.5 interpolated by AOD at 440 nm, 675 nm and 870 nm, fine-mode fraction, aerosol volume distribution and complex refractive index.

In order to estimate $PM_{2.5}$, we compare and analyze AOD from #41 and #350 instruments, and find hourly AODs are well consistent, with averaged error less than 0.04. Therefore, AODs from the two measurements are complemented each other in this study.

$PM_{2.5}$ is derived from the U.S. embassy in Beijing (unit: $\mu g/m^3$) and instrument is MetOne BAM 1020 with hourly averaged output of $PM_{2.5}$. Meteorological data are CMA southern automatic meteorological observation station located at 116.46°E, 39.8°N. The data of Mixture Layer Height (MLH) was from National Centers for Environmental Prediction (NCEP) and Final Operational Analysis data (FNL) global reanalysis data, with the horizontal resolution of 1° × 1°, and time step of 6 h. Mixture layer height is extracted at location of 116.46°E, 39.8°N and we perform temporal interpolation of the data using the sine function.

2.2 Correction method of fine-mode aerosol

Atmospheric aerosol optical depth can be obtained from the integral of the aerosol extinction coefficient over the vertical profile. Fine-mode AOD in the column can be expressed as

$$AOD_f = \int_0^{\infty}\int_0^{f} Q_{ext,amb}(r,z)\, n_{amb}(r)\, \pi r^2 \mathrm{d}r\mathrm{d}z \quad (1)$$

where $Q_{ext,amb}(r,z)$ is particles extinction coefficient in ambient; $n_{amb}r$ is particles size distribution in ambient; f denotes fine-mode; r is the radius of the aerosol particles; z is the vertical height. Therefore, the relationship between AOD and AOD_f can be expressed as

$$AOD_f = AOD \cdot \eta \quad (2)$$

where η is the fraction of fine-mode aerosol optical depth to the total AOD.

2.3 Calculation of fine-mode fraction (η) using SDA method

Aerosol fine-mode fraction is obtained according to study of O'Neill, et al., (2001b, 2002). The basic theory is that AOD can be divided into two parts of fine-mode and coarse-mode AODs i.e. $\tau = \tau_f + \tau_c$. Using the expression $\eta = \frac{\tau_f}{\tau}$, we can get the first-order logarithmic derivative of AOD as below.

$$\alpha = -\frac{\mathrm{dln}\tau}{\mathrm{dln}\lambda} = \frac{\alpha_f\tau_f + \alpha_c\tau_c}{\tau} = \alpha_f\eta + \alpha_c(1-\eta) \quad (3)$$

And thus,

$$\eta = \frac{\alpha - \alpha_c}{\alpha_f - \alpha_c} \quad (4)$$

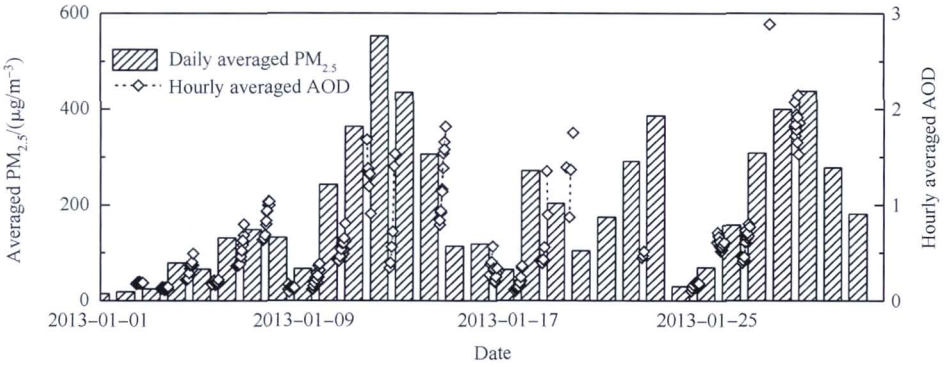
where α is Ångström exponent; α_f is fine-mode Ångström exponent referred to the solution from O'Neill, et al., (2001b) and Zhang, et al., (2013); α_c is coarse-mode Ångström exponent assuming to be a constant value (−0.15) which is counted from a large number of statistics (O'Neill, et al., 2001b, 2002). This assumption is equivalent to setting the bimodal size distribution of coarse-mode part, namely the slope of coarse-mode aerosol optical depth to be constant. With Eq. (2) and Eq. (4), we can proceed with optical measurements, and then calculate fine-mode aerosol optical depth. O'Neill, et al., (2001b) compared η from SDA method and that from Mie theory calculation, and the root mean square error was approximately 0.1. Hansen (1974) indicated that the scattering efficiency of the particle was evident when the wavelength was close to the radius of particles. Therefore, it is accepted that the approximate particles size of 1.0 μm has significant extinction efficiency at 500 nm. Although truncation diameter is not clear, to a certain extent, AOD_f can represent the fine particle optical contribution.

3 METHOD OF $PM_{2.5}$ ESTIMATION USING AOD_f AND VALIDATION

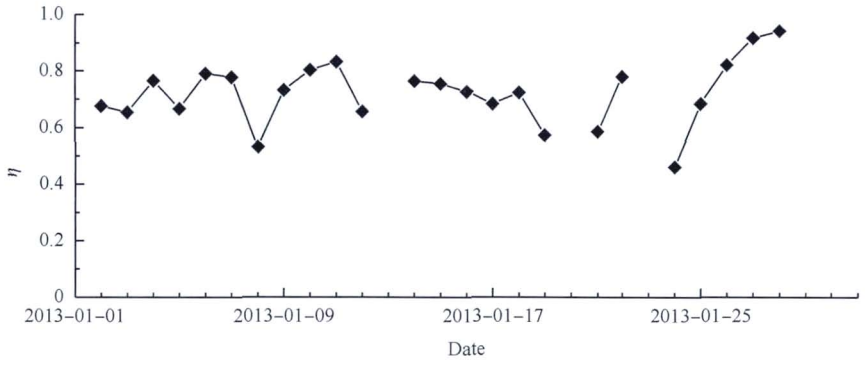
3.1 Data analysis

In January 2013, five processes of serious haze pollutions occurred, the second in which was the most serious (January 10 to 16, 2013) with daily averaged $PM_{2.5}$ of more than 500 μg/m³. Fig. 1(a) shows the hourly averaged AOD at 500 nm and daily averaged $PM_{2.5}$ in January. There is only one measurement (Lev 1.5) at 11:00 on January 27, 2013, and the value of AOD is large. According to the automatic meteorological station

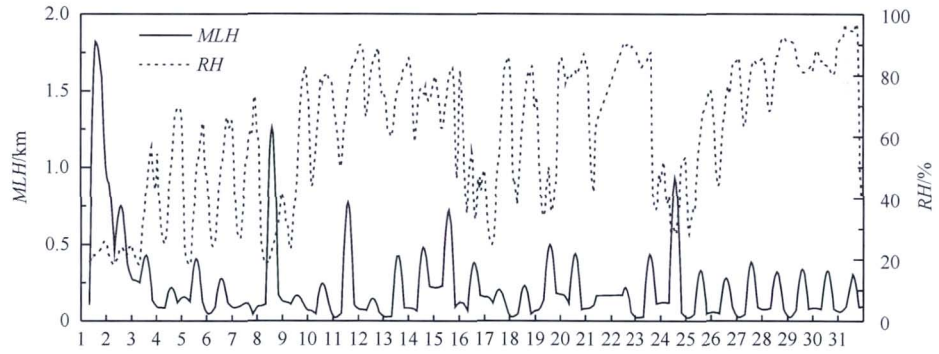
(54511) monitoring, cloud covered all-sky during 00:00—12:00 on January 27, 2013. The variety of AOD at 500 nm and $PM_{2.5}$ data in Fig. 1(a) shows that the mean AOD value is 0.52, in a range from 0.09 to 1.85, and the averaged $PM_{2.5}$ concentration in the haze pollution is 201 $\mu g/m^3$. The $PM_{2.5}$ is significantly higher than results from the study of Yang, et al. (2002) from 1999 to 2001 (about 109.6 $\mu g/m^3$ at Qinghuayuan Station), which is less serious than in Beijing.



(a) Hourly averaged AOD and daily averaged $PM_{2.5}$



(b) Daily averaged fine-mode AOD



(c) Mixture layer height and relative humidity

Fig. 1 Observation in January of 2013 at Beijing site

Fig. 1(b) shows daily averaged fine-mode fraction in January of 2013, which has in the same trend as the $PM_{2.5}$. The maximum and minimum of fine-mode fraction reach 0.94 and 0.46 respectively. The maximum of $PM_{2.5}$ equals to 203 $\mu g/m^3$ when daily averaged η is less than 0.6 in January, and AOD and AOD_f are 1.35 and 0.57 (on January 19, 2013). It should be noted that during the third and fourth haze pollution from January 19 to 21, 2013, a rainfall event occurs. It is found that the fine particles extinction can be effectively obtained by η which can be used as identification of pollution level.

Fig. 1(c) shows the changes of mixture layer height and relative humidity in January 2013. From Fig. 1(c), the mixture layer height is relatively lower. The mean mixing layer height is just 207 m, with the minimum of 21 m. The relative humidity keeps higher, with the averaged relative humidity of 60% and the maximum of 97%. Due to larger relative humidity and lower mixing layer height, atmospheric stratification appears more stable and lasts longer, leading to fog forming easily which contributes to accumulation of particles near the surface. Hourly averaged parameters are listed in Table 2.

Table 2 Hourly averaged value of AOD, $PM_{2.5}$, η, *MLH*, and *RH* in January 2013 at Beijing site

	AOD	$PM_{2.5}$/(μg/m^3)	η	*MLH*/m	*RH*/%
Mean	0.52	199	0.75	229	61
Maximum	1.86	>500	0.97	1822	97
Minimum	0.09	7	0.31	21	18

3.2 Effectiveness of aerosol fine-mode correction

Eq. (2) indicates that, the impact of coarse-mode AOD can be effectively removed from total AOD using fine-mode aerosol optical depth to estimate $PM_{2.5}$. Fig. 2(a) shows the changes of fine-mode aerosol optical depth, total aerosol optical depth and $PM_{2.5}$ January18 to19, 2013. The relative humidity on January 18, 2013 is higher than that on January 19. The wind speed on January 18, 2013 is lower than that on January 19, 2013, with daily averaged relative humidity of 65% and 50%, and daily averaged wind speed of 1.2 m/s and 2.2 m/s, respectively (Fig. 2(b)(c)). The fine-mode aerosol optical depth decreases, while the total aerosol optical depth increases, with relative humidity decreasing and wind speed increasing after 13:00 on January 19, 2013. The fine particle extinction decreases on January 19, 2013, but coarse particles extinction leads to the increase of total aerosol optical depth. Daily averaged $PM_{2.5}$ is reduced from 272 μg/m^3 on January 18, 2013 to 203 μg/m^3 on January 19, 2013. Especially, $PM_{2.5}$ decreases evidently after 13:00 on January 19, 2013 while the total AOD increases obviously. The relationship between AOD and $PM_{2.5}$ presents negative correlation in this period. However, fine-mode aerosol optical depth as well as the $PM_{2.5}$ presents the descending trend. In other words, fine-mode aerosol optical depth can better express fine particulate matter mass concentration. In the serious haze period (e.g., January 2013), the correlation between AOD_f and $PM_{2.5}$ is established to obtain a uniform fitting formula to estimate $PM_{2.5}$ as below.

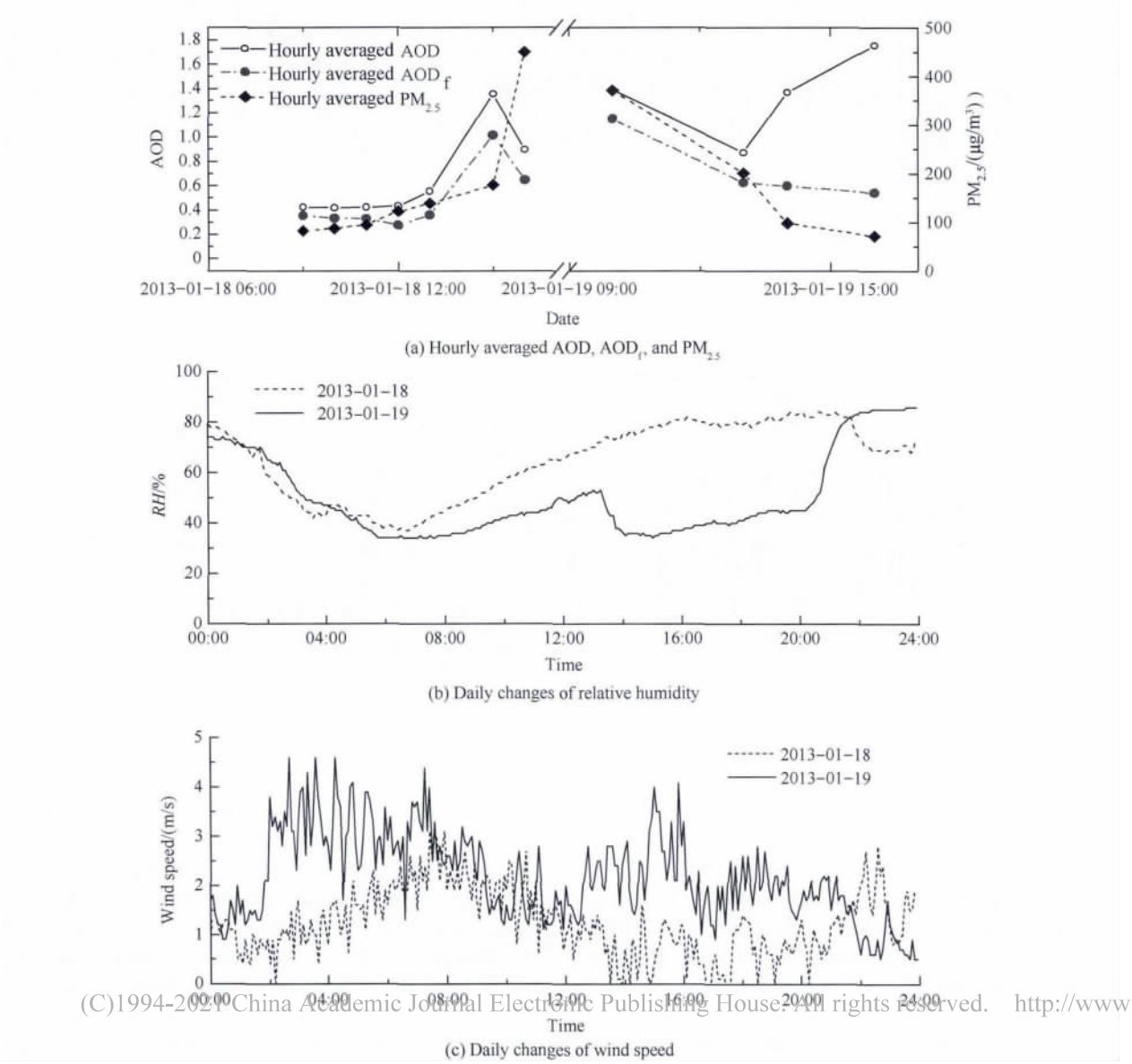


Fig. 2 Comparison of observation on January 18 and 19, 2013 at Beijing site

3.3 Estimation of $PM_{2.5}$

On the basis of spectrum deconvolution algorithm, aerosol optical depth is calculated with fine-mode fraction in order to obtain the contribution from the fine particle extinction. Fig. 3 shows the relationship between AOD and $PM_{2.5}$. In January 2013, there are 139 samples selected in the statistical regression analysis, and mean value of $PM_{2.5}$ is 112 $\mu g/m^3$. The correlation between aerosol optical depth and $PM_{2.5}$ is shown in Fig. 3(a), with $R^2 = 0.69$. The correlation of linear fitting function is obtained, and the slope and intercept are 204.49 and 5.14, the standard deviation of 11.65 and 8.13, respectively. When the AOD is transformed into AOD_f in the relationship, correlation is improved with R^2 up to 0.77 (Fig. 3(b)). The slope and intercept of the linear fitting function with AOD_f are 244.48 and 10.63, and the standard deviation of intercept is decreased from 8.13 to 6.70. It is indicated that the discrete degree of scatter is reduced after correction with fine-mode fraction. The Root Mean Square (RMS) error from directly established the relationship by AOD and $PM_{2.5}$ is 61 $\mu g/m^3$, and RMS from that after fine-mode fraction correction is 53 $\mu g/m^3$. Tian, et al. (2010) focused on the $PM_{2.5}$ near the Ontario area, Canada. They reported that there was larger error to estimate $PM_{2.5}$ using AOD from MODIS. It is indicated that RMS is up to 7.3 $\mu g/m^3$ when the averaged $PM_{2.5}$ equals to 7.8 $\mu g/m^3$. While the averaged $PM_{2.5}$ equals to 10.3 $\mu g/m^3$, RMS can be up to 9.7 $\mu g/m^3$. In their research, the frequency for the condition that $PM_{2.5}$ is more than 60 $\mu g/m^3$ was very few. Compared with the results in this study, although there was lower root mean square error in the paper of Tian, et al. (2010), the averaged value is too low, about 10 $\mu g/m^3$. The mean value of $PM_{2.5}$ is 112 $\mu g/m^3$, 10 times larger than Tian's, and root mean square error is bigger. In the period of haze pollution, root mean square error is decreased obviously with the regression relations between fine-mode aerosol optical depth and $PM_{2.5}$. Therefore, use of AOD_f to replace AOD obviously improves the correlation of $PM_{2.5}$, and the linear fitting Equation is shown below.

$$PM_{2.5} = 244.48AOD \cdot \eta + 10.6 \quad (5)$$

Compared the scatter points in Fig. 3(a)(b), discrete degree of scatter is reduced after correction in the case of the large AOD and small $PM_{2.5}$. Therefore, the correlation between fine-mode aerosol optical depth and $PM_{2.5}$ can improve the estimation accuracy of $PM_{2.5}$ by remote sensing.

3.4 Validation

The relationship between AOD_f and $PM_{2.5}$ is established during a period of serious haze pollution in January 2013. In order to verify the reliability of the relationship, validation is performed based on the data on February 1 to 15, 2013. The haze pollutions are also very serious in February, especially in 5 to 6, 8 and 9, and 12 to 13. The value of $PM_{2.5}$ is up to 365 $\mu g/m^3$, and AOD_f up to 0.97. The averaged value of $PM_{2.5}$ is 84 $\mu g/m^3$ in February, less than that of 112 $\mu g/m^3$ in January. It is noted that fine particulate matter pollution decreases in February.

Fine particulate matter is estimated using Eq. (5) over a half month of February, and compared with observations (Fig. 4).

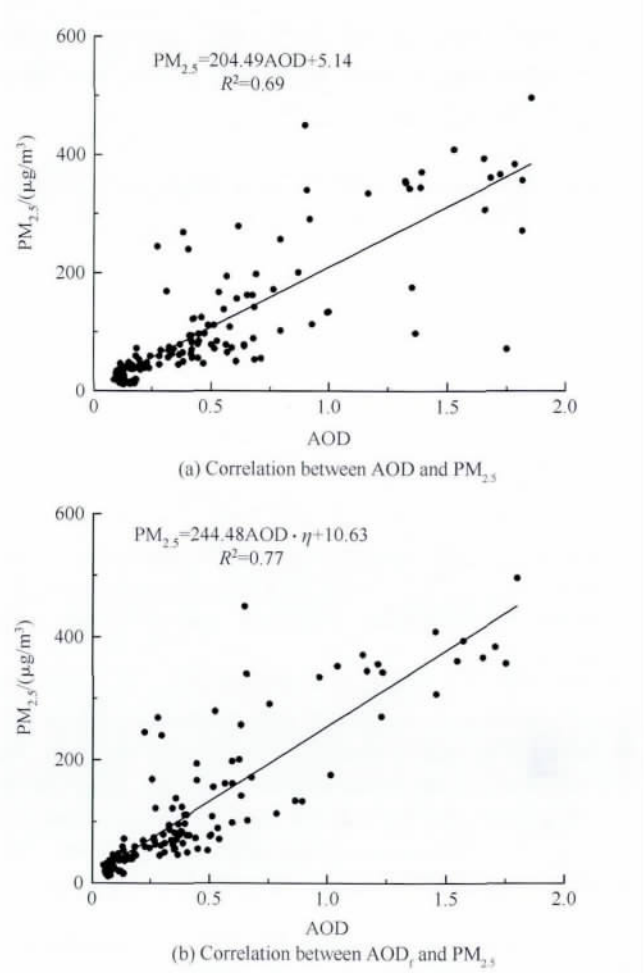


Fig. 3 The correlation analysis between AOD and $PM_{2.5}$ (The standard deviation of the slope and intercept in linear fitting formula in parentheses)

Fig. 4 shows that estimation of $PM_{2.5}$ is close to observations, with the mean value of 85 $\mu g/m^3$ and the root mean square error of 50 $\mu g/m^3$. The latter two haze processes are reproduced using the relationship to estimate $PM_{2.5}$. However, the haze pollution on February 5 to 6 is not reproduced correctly due to lack of effective aerosol optical depth measurements.

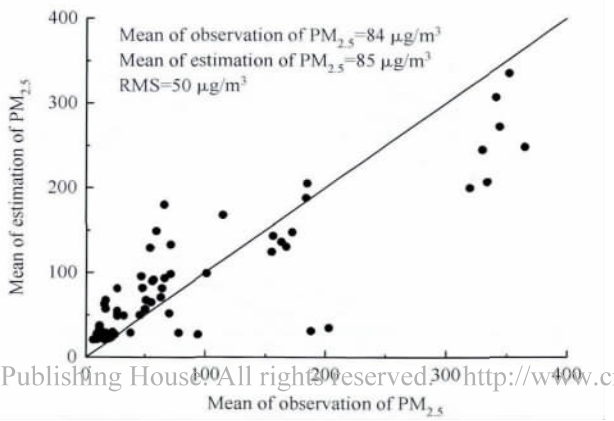


Fig. 4 Comparison between estimation and observation of $PM_{2.5}$ during February 1 to 15, 2013 at Beijing site

4 INFLUENCES OF MIXTURE LAYER HEIGHT AND RELATIVE HUMIDITY ON ESTIMATION OF $PM_{2.5}$

4.1 Influence of mixture layer height on estimation of $PM_{2.5}$

We extract mixture layer height of Beijing site four times every day (2:00, 8:00, 14:00, 20:00) in January 2013 by using global reanalysis data NCEP FNL. In order to get the hourly mixture layer height, samples at four times (see above) are interpolated into 24 h by sine function. Aerosol extinction coefficient $k_{ex}(z)$ satisfies the exponential distribution when the height changes (Zhang & Zhou,1995)

$$k_{ex}(z) = \int_0^{\infty} \sigma_{ex}(r,z_0)\, e^{-\frac{z}{MLH}} dr \quad (6)$$

where $\sigma_{ex}(r,z_0)$ represents aerosol extinction at the surface, *MLH* is mixture layer height, r is aerosol particle radius. The relationship between aerosol extinction coefficient at the surface and total AOD is:

$$k_{ex}(z_0) = \mathrm{AOD}/MLH \quad (7)$$

According to the LiDAR data, the average mixture layer height during haze is about 500 m (Zhang, et al., 2013). The changes of aerosol extinction coefficient with height do not follow exponential distribution during the severe haze events (Lv, et al., 2013). Therefore, the relationship between AOD and $PM_{2.5}$ obtained by Eq. (7) is unsatisfactory, with R^2 of 0.28.

4.2 Influence of relative humidity on estimation of $PM_{2.5}$

According to the relationship between relative humidity and aerosol scattering coefficient near the surface, aerosol scatter coefficient increases with relative humidity (Fig. 5). We simulate aerosol scattering coefficient affected by hygroscopic growth factor by $f(RH) = (1 - RH/100)^{-1}$ with initial value of $RH = 10\%$. When dry aerosol scattering coefficient equals 1.0 and $RH > 80\%$, aerosol scattering coefficient increases rapidly. When $RH = 90\%$, aerosol scattering coefficient can be 10 times of initial value.

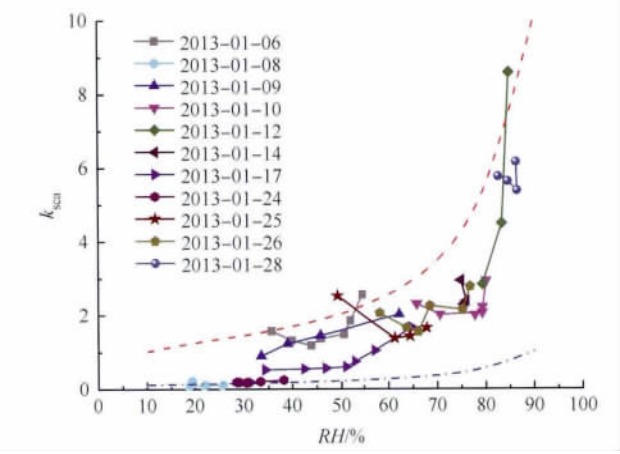


Fig. 5 The correlation of relative humidity and aerosol scattering coefficient in January, 2013
(dash line and dot-dash line denoted as aerosol scattering coefficient of 0.1 and 1.0 when $RH = 10\%$, respectively)

In our study, when relative humidity exceeds 80%, AOD is lower than 2.0. In general, when relative humidity is lower than 80%, aerosol scattering coefficient follows hygroscopic growth curve. Fig. 5 shows the relationship between scattering coefficient at the surface and relative humidity, according to the method proposed by Liu (2008). In Fig. 5, aerosol scattering coefficient increases rapidly when $RH > 80\%$. On January 12, 2013, the scattering coefficient varies with relative humidity which do not agree with the equation of hygroscopic growth factor introduced by Kotchenruthe (1999), due to the phase of hygroscopic matters, such as NaCl, NH_4Cl, $(NH_4)SO_4$, changed when relative humidity is close to 80% (Wu, 2013). Therefore, the hygroscopic matter only affects aerosol scattering when relative humidity between 40%—80% and when $RH >$ 80% the effect of hygroscopic growth factor is not obvious. The relationship b etween aerosol scattering coefficient and relative humidity meet the hygroscopic growth when fine particulate matter mass c oncentration lower than 200 $\mu g/m^3$ and $RH < 80\%$. Calculating aerosol scattering coefficient at the surface needs mixture layer height, and thus the uncertainty of mixture layer height can affect the relationship. When $PM_{2.5}$ is in high concentration, hygroscopic growth factor correction method cannot achieve good results. After correction of hygroscopic growth factor, the relationship between AOD_f and $PM_{2.5}$ becomes worse, R^2 is only 0.35, and some scatters located in region with lower AOD and higher $PM_{2.5}$.

5 CONCLUSIONS

Aerosol fine mode fraction can be simply obtained using SDA method, and then the contribution from fine particle extinction is obtained. We perform a correlation analysis between AERONET Lev 1.5 data and United States Embassy in situ measurements of $PM_{2.5}$ mass concentration in January 2013 at Beijing. It is found that fine-mode correction can improve the correlation. At the same time, from analyzing the effects of mixture layer height and relative humidity on the correlation between AOD and fine-mode fraction, we conclude that:

(1) The contribution from fine particle extinction can be obtained from fine-mode fraction η. During haze events in January 2013, the correlation coefficient from regression between AOD_f and $PM_{2.5}$ is larger than that between total AOD and $PM_{2.5}$, which means that fine-mode particle is the major contribution to aerosol extinction;

(2) $PM_{2.5}$ can be estimated better by using the regression of AOD_f. The root-mean-square error is reduced in this study and the linear fitting formula in this study is:

$$PM_{2.5} = 244.48\mathrm{AOD} \cdot \eta + 10.6, R^2 = 0.77;$$

(3) Hygroscopic growth factor of aerosol scattering coefficient is useful when relative humidity is between 40%—80%; while its effect is limited when $RH > 80\%$. Aerosol vertical profile disagrees from exponent distribution in heavy haze days, and thus the correlation of AOD_f-$PM_{2.5}$ using vertical factor is failed.

In this paper, we establish the relationship between AOD_f and $PM_{2.5}$ using aerosol fine-mode correction, and improve the remote sensing method of estimating $PM_{2.5}$. However, hygroscopic growth and vertical profile corrections are still challeges during

hazewhich needs to study in depth in the future.

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利用细模态气溶胶光学厚度估计 $PM_{2.5}$

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摘 要: 本文利用 2013 年 1 月 AERONET (Aerosol Robotic Network) 北京站的气溶胶光学厚度 AOD (Aerosol Optical Depth)、细颗粒物光学厚度占总光学厚度的比例即气溶胶细模态比例 η 以及地面监测的细颗粒物 $PM_{2.5}$ (Particulate Matter 2.5) 质量浓度数据建立气溶胶细模态光学厚度 AOD_f (fine-mode Aerosol Optical Depth) 与 $PM_{2.5}$ 的线性回归关系,并利用 2013 年 2 月 1 日—15 日的数据验证该方法。结果表明,利用 2013 年 1 月建立的回归方法能够有效估算灰霾期间 $PM_{2.5}$,获得 $PM_{2.5}$ 的均值为 8 5 $\mu g/m^3$,均方根误差为 50 $\mu g/m^3$。利用气溶胶细模态订正方法估算的 AOD_f 与 $PM_{2.5}$ 的相关系数大于 AOD 与 $PM_{2.5}$ 的相关系数,这表明灰霾期间以 $PM_{2.5}$ 为代表的细模态颗粒物成为气溶胶消光的主体,且 AOD 与 $PM_{2.5}$ 的关系转化为 AOD_f 与 $PM_{2.5}$ 的相关关系时,相关程度提高。垂直分布修正在灰霾时对改善 AOD 与 $PM_{2.5}$ 相关关系作用不明显; 当相对湿度大于 80% 时,湿度订正效果受到较大限制。

关键词: 气溶胶光学厚度, $PM_{2.5}$, 气溶胶细模态比例

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1 引 言

霾被定义为大量极细微的干尘粒等均匀地浮游在空中,使水平能见度小于 10 km 的空气普遍混浊现象(中国气象局,2003)。而灰霾在近十年的研究中被广泛用于特指人类活动增加导致的城市区域近地层大气的细颗粒物污染现象(吴兑,2012)。过去 30 年,由于经济增长和机动车数量增加造成大气细颗粒物组成的灰霾污染频发(He 等,2002)。更重要的是,大气细颗粒物可引起多种疾病,尤其是呼吸道疾病如慢性支气管炎等([2011 – 10 – 12] https://openknowledge.worldbank.org/handle/1098612136)。大气细颗粒物的监测越来越受到重视,无论是在环境保护、公众健康还是科研领域。利用遥感手段监测并估算近地面细颗粒物质量浓度对灰霾的监测和治理有重要意义。

遥感估算近地面颗粒物质量浓度最主要的方法有: 线性回归法,包含复杂因子的多元回归法,耦合观测数据的模型反演法等。由于线性回归法复杂程度低、外推性强而受到广泛使用(Wang 和 Christopher, 2003; Hutchison, 2003; 李成才 等, 2003a, 2003b, 2003c, 2005; Engel-Cox 等, 2004; Koelemeijer 等, 2006; 徐祥德 等, 2006; Kumar 等, 2007; Liu 等, 2007; 刘勇, 2007; Schaap 等, 2008; 何秀 等, 2009; VanDonkelaar 等, 2010; 郑卓云 等, 2011)。建立 AOD 与 $PM_{2.5}$ 之间的相关关系需要对遥感观测获得环境湿度下的整层气溶胶光学厚度进行湿度订正和垂直订正(刘勇 等,2007,何秀 等,2009,郑卓云 等,2011)。虽然遥感手段获得的 AOD 主要源自细颗粒物的消光作用,但细粒子消光在总光学厚度中所占的比例对 AOD 与 $PM_{2.5}$ 相关关系的影响也是不可忽视的。因此,Di Nicolantonio 等人(2007) 利用 MODIS 反演的气溶胶细模态比例估算

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得到 AOD_f 与细颗粒物的相关关系。但是,MODIS 反演的气溶胶细模态比例在复杂地表上空敏感性很强,反演稳定性较差,因此其相关关系并不理想。气溶胶细模态比例不仅可在卫星观测反演结果中获得,地基观测也可获得该物理量,且由于地基观测中,地表影响小,使结果更加稳定准确。O'Neill 等人(2001a, 2001b, 2002, 2003)阐述了光学厚度光谱退卷积方法,该方法仅利用多波段光学厚度即可获得 AOD_f 在 AOD 中的比例。

本文探讨 2013 年 1 月灰霾污染严重时期北京地区气溶胶光学厚度与细颗粒物污染之间的相关关系。利用多波段光学厚度计算的气溶胶细模态比例估计气溶胶细模态光学厚度,并与细颗粒物质量浓度之间建立相关关系,提高遥感手段监测和估算近地面细颗粒物质量浓度的能力。

2 数据及理论基础

2.1 数据介绍

本文使用 AERONET (Aerosol Robotic Network) 北京站的气溶胶光学和微物理特性数据,观测仪器为 CE318 型太阳光度计。由于北京测站拥有两台太阳光度计仪器,分别可获得 3 个波段(870 nm、675 nm、440 nm)和 5 个波段(870 nm、675 nm、500 nm、440 nm、380nm)的气溶胶光学厚度(见表 1),同时进行各波段天空光的主平面和平纬圈扫描。利用这些数据可反演整层大气中气溶胶特性,例如 Ångström 指数、粒子体积谱分布以及细粒子比例等。本文主要使用 Lev1.5 的 500 nm 的 AOD(针对 3 个波段仪器测量结果进行插值获取 500 nm 的 AOD)和反演的细模态比例,以及天空光反演的粒子体积谱分布和复折射指数等数据。

表 1 数据来源及站点位置

数据	站点	观测时间
AOD	AERONET 北京站 (116.38°E, 39.98°N, 92.00 m, #41 仪器, 3 个波段) (116.38°E, 40.00°N, 59.00 m, #350 仪器, 5 个波段)	2013-01
混合层高度 *MLH*	NCEP 全球再分析资料 (116.47°E, 39.8°N)	2013-01
相对湿度 *RH*	南郊观象台 54511 站 (116.47°E, 39.8°N, 31.3 m)	2013-01
$PM_{2.5}$	美国驻京大使馆 (116.43°E, 39.91°N, 45 m)	2013-01

由于 AERONET 北京站有两台仪器同时观测,因此本文将两台仪器观测数据进行对比分析。由于估算 $PM_{2.5}$ 需要两种仪器的光学厚度小时平均值数据具有一致性,因此选取光学厚度小时平均值对比。#41 与#350 仪器 AOD 小时平均值具有很好的一致性,平均误差为 0.04。因此,本文认为两台仪器测量 AOD 的小时平均值在估算 $PM_{2.5}$ 的计算中可以相互补充。

$PM_{2.5}$ 数据来自美国驻京大使馆,监测仪器为 MetOne BAM 1020,输出数据为每小时平均值。气象资料来自中国气象局南郊观象台自动观测站,观测站位于 116.46°E, 39.8°N。混合层高度数据使用的是 NCEP (National Centers for Environmental Prediction) 的 FNL (Final Operational Analysis data) 全球再分析资料([2013-02-12] http://rda.ucar.edu/datasets/ds083.2),其水平分辨率为 1°×1°,时间间隔为 6 h。提取 116.46°E, 39.8°N 位置处的边界层高度,利用正弦函数将其插值为时间分辨率为一小时的数据序列。

2.2 气溶胶细模态订正方法

大气气溶胶的光学厚度表示气溶胶消光系数在垂直方向上的积分。在单位面积的空气柱里,细颗粒物的光学厚度可表示为:

$$\mathrm{AOD_f} = \int_0^{\infty}\int_0^{f} Q_{\mathrm{ext,amb}}(r,z)\, n_{\mathrm{amb}}(r)\, \pi r^2 \mathrm{d}r\mathrm{d}z \quad (1)$$

式中,$Q_{\mathrm{ext,amb}}(r,z)$ 为环境状况下颗粒物的消光系数,$n_{\mathrm{amb}}(r)$ 为环境状况下颗粒物尺度谱分布,f 即为细颗粒物空气动力学直径,r 为气溶胶粒子的半径,z 为垂直高度。由此 AOD 与 AOD_f 的相关关系可表示为:

$$\mathrm{AOD_f} = \mathrm{AOD}\cdot\eta \quad (2)$$

2.3 光谱退卷积方法获气溶胶细模态比例 η

气溶胶细模态比例可根据 O'Neill 等人(2001b, 2002)的研究求得,其基础理论是将光学厚度 τ 分为细模态光学厚度 τ_f 和粗模态光学厚度 τ_c 两部分,即 $\tau = \tau_f + \tau_c$,进而对光学厚度进行求导计算。$\eta = \dfrac{\tau_f}{\tau}$,可以得到光学厚度的一阶对数导数为:

$$\alpha = -\frac{\mathrm{d}\ln\tau}{\mathrm{d}\ln\lambda} = \frac{\alpha_f\tau_f + \alpha_c\tau_c}{\tau} = \alpha_f\eta + \alpha_c(1-\eta) \quad (3)$$

同时有:

$$\eta = \frac{\alpha - \alpha_c}{\alpha_f - \alpha_c} \tag{4}$$

式中,α为Angstrom指数,可利用光学厚度对波长求导获得。α_f为细模态Angstrom指数,本文借鉴O'Neill等(2001b)和Zhang等(2013)的方法求解。α_c为粗模态Angstrom指数,假设其为恒值(-0.15)。该假设等价于设定了双峰粒子谱分布的粗模态谱部分,即粗模态气溶胶光学厚度的斜率被假设为不变量,该假设由大量实例统计而来(O'Neill 等 2001b, 2002)。结合式(2)和式(4),即可从光学测量入手,计算获得气溶胶细模态光学厚度。O'Neill等(2001b)将η与Mie理论计算结果(截断直径为1.2 μm)对比,均方根误差近似为0.1。Hansen和Travis(1974)指出波长与粒子半径相当时,粒子散射效率最高。因此,500 nm 波长处消光最显著的颗粒物尺度约在1.0 μm。虽然 AOD_f 并没有明确的截断直径,但在一定程度上可代表细颗粒物的光学贡献。

3 AOD_f 估算 $PM_{2.5}$ 方法分析及验证

3.1 资料分析

2013 年 1 月经历了 5 次严重的灰霾过程,以第 2 次最为严重(1 月 10 日—16 日),12 日 $PM_{2.5}$ 日平均值超过500 μg/m^3。图1(a)显示2013年1月500 nm AOD小时平均值与$PM_{2.5}$日平均值的时间序列图。图1中可以看到1月27日仅11时Lev1.5

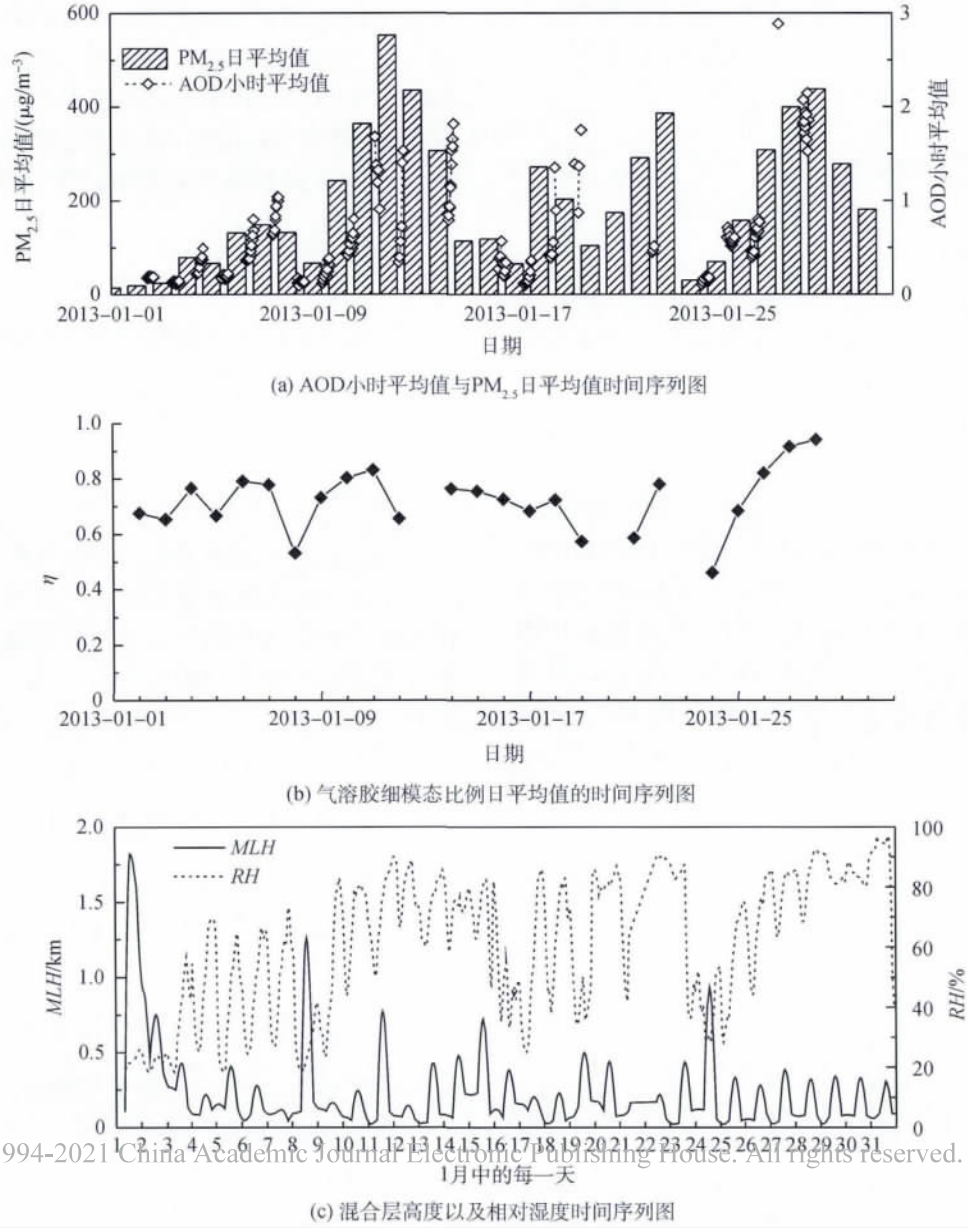


图1 2013 年 1 月观测量时间序列图

有观测值,且 AOD 值较大。根据南郊观象台自动气象站 54511 监测的云量数据表明,2013 年 1 月 27 日 00:00—12:00,全天空高云覆盖。统计 500 nm 的 AOD 和 $PM_{2.5}$ 数据可知,在 2013 年 1 月观测到的 AOD 平均值为 0.52,范围在 0.09—1.85,观测到的本次灰霾污染 $PM_{2.5}$ 平均浓度为 201 $\mu g/m^3$。本文观测结果明显高于杨复沫等人(2002)在 1999 年—2001 年的 $PM_{2.5}$ 年平均值(清华园观测站约 109.6 $\mu g/m^3$),这说明北京地区细颗粒物污染严重。

图 1(b)显示了 2013 年 1 月份的细模态比例日平均值,从图中可以看出,细模态比例日平均值的变化与 $PM_{2.5}$ 日平均值变化趋势一致,其极大值与极小值分别为 0.94 和 0.46。在多次经历严重灰霾的 1 月中,细模态比例的日平均值小于 0.6 时,$PM_{2.5}$ 浓度最大值仅为 203 $\mu g/m^3$,相应的 AOD 日平均值为 1.35,AOD_f 日平均值仅为 0.57(1 月 19 日)。1 月 19—21 日恰为第 3 次和第 4 次灰霾的间歇期,历经一次降水过程。这说明,通过细模态比例可有效获得细颗粒物的消光,也可作为辨识灰霾污染程度的参考量。

图 1(c)显示了 2013 年 1 月的混合层高度和相对湿度变化的情况。从图 1(c)可知,混合层高度在整个 1 月份都处于较低水平,平均混合层高度仅为 207 m,极小值仅为 21 m。而相对湿度始终较高,1 月平均相对湿度为 60%,极大值高达 97%。由于 2013 年 1 月相对湿度较大,混合层高度较低,大气层垂直结构稳定且持续时间长,导致雾霾易形成不易消散,大量颗粒物在近地面积累。各参量的小时平均统计结果列于表 2。

表 2 2013 年 1 月小时平均气溶胶光学厚度、细颗粒物浓度、细模态比例、混合层高度和相对湿度的统计列表

	AOD	$PM_{2.5}$/($\mu g/m^3$)	η	MLH/m	RH/%
平均值	0.52	199	0.75	229	61
极大值	1.86	>500	0.97	1822	97
极小值	0.09	7	0.31	21	18

3.2 气溶胶细模态订正有效性分析

由式(2)可知,细颗粒物引起的消光与 AOD_f 相对应,利用 AOD_f 估算 $PM_{2.5}$ 可有效去除气溶胶粗模态对 AOD 的影响。图 2(a)给出了 2013 年 1 月 18 日—19 日 AOD_f 和 AOD 以及相应的 $PM_{2.5}$ 变化情况。1 月 19 日相对湿度明显低于 18 日,1 月 18 日风速低于 19 日,日平均相对湿度分别为 65% 和 50%,日平均风速分别为 1.2 m/s 和 2.2 m/s(图 2(b)、图 3(c))。由图 2 可知,2013-01-19 13:00 以后相对湿度降低,风速增加,AOD_f 降低,而 AOD 增加。这说明,2013 年 1 月 19 日细颗粒物消光减弱,而粗颗粒物消光增加造成 AOD 增加。$PM_{2.5}$ 日平均值从 18 日的 272 $\mu g/m^3$ 下降到 19 日的 203 $\mu g/m^3$。尤其在 2013-01-19 13:00 以后,$PM_{2.5}$ 浓度减小明显,但 AOD 总量增加明显,使该时段的 AOD 与 $PM_{2.5}$ 呈现负的相关关系。而 AOD_f 与 $PM_{2.5}$ 趋势一致,呈现下降趋势。也就是说,气溶胶细模态光学厚度能更好的表达细颗粒物质量浓度的下降。在灰霾严重时期(如 2013 年 1 月),利用 AOD_f 与 $PM_{2.5}$ 建立的相关关系能够获得统一的拟合公式,提高相关性,且更有效的估计细颗粒物质量浓度。

3.3 通过气溶胶细模态订正估算 $PM_{2.5}$

利用光谱退卷积方法获得的细模态比例作为 AOD_f 的计算基础,以此求取细颗粒物消光贡献。图 3 为 AOD 与 $PM_{2.5}$ 的相关关系。2013 年 1 月,本文选取 139 个样本点进行统计回归分析,$PM_{2.5}$ 的均值为 112 $\mu g/m^3$。AOD 与 $PM_{2.5}$ 的相关关系如图 3(a)所示,R^2 为 0.69,利用线性拟合得到的相关关系函数,其斜率和截距分别为 204.49 和 5.14,标准偏差分别为 11.65 和 8.13。当 AOD 与 $PM_{2.5}$ 的关系变为 AOD_f 和 $PM_{2.5}$ 的关系时,相关程度提高。(如图 3(b)),R^2 可达 0.77,斜率和截距有一定变化,分别为 244.48 和 10.63,截距的标准偏差从 8.13 减小到 6.70,这说明拟合后散点的离散程度减弱。计算以上两种情况下的均方根误差可知,由 AOD 与 $PM_{2.5}$ 直接建立的拟合关系均方根误差为 61 $\mu g/m^3$。而利用细模态气溶胶光学厚度 AOD_f 与 $PM_{2.5}$ 建立拟合公式的均方根误差为 53 $\mu g/m^3$。Tian 和 Chen(2010)针对美国安大略湖附近的 $PM_{2.5}$ 进行了研究,结果表明利用 MODIS 反演的 AOD 估算 $PM_{2.5}$ 结果存在较大误差,他们指出,$PM_{2.5}$ 均值为 7.8 $\mu g/m^3$ 时,均方根误差达 7.3 $\mu g/m^3$;当均值为 10.3 $\mu g/m^3$ 时,均方根误差可达 9.7 $\mu g/m^3$。在他们的研究中 $PM_{2.5}$ 大于 60 $\mu g/m^3$ 的频率很少。与本文的研究结果相比,虽然 Tian 和 Chen(2010)的研究结果呈现较小的均方根误差,但其均值也较小,仅为 10 $\mu g/m^3$ 左右,而本文着重给出严重灰霾期间细颗粒物质量浓度估计值,其实测结果均值为 112 $\mu g/m^3$,是

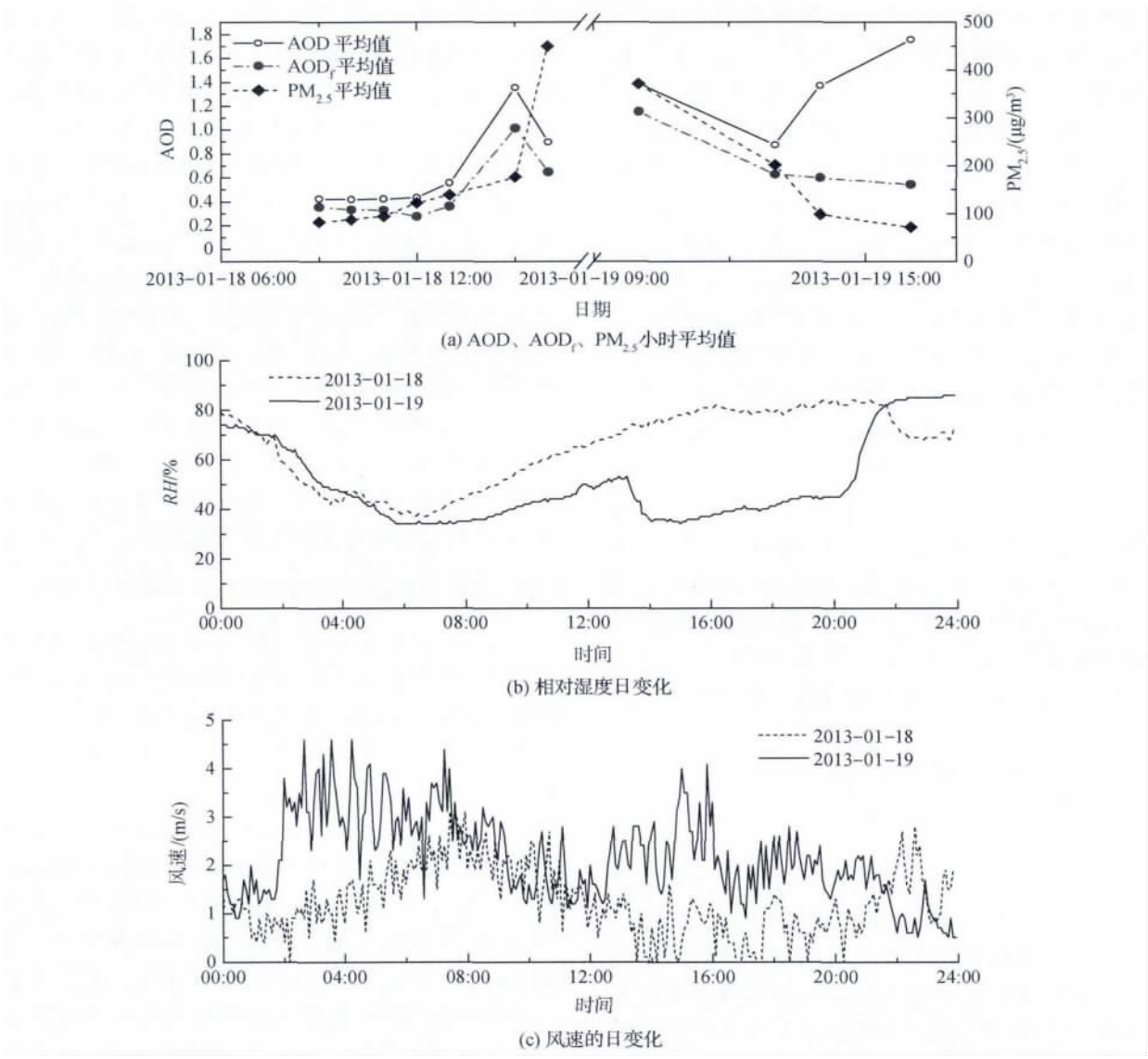


图2 2013 年 1 月 18、19 日观测对比图

Tian 和 Chen(2010) 等结果的 10 倍,因而均方根误差同样较大。但在灰霾较严重时期,用 AOD_f 与 $PM_{2.5}$建立的回归关系,均方根误差明显减小,因此应用 AOD_f 对提高 AOD 与 $PM_{2.5}$的相关关系效果明显,获得线性拟合公式为:

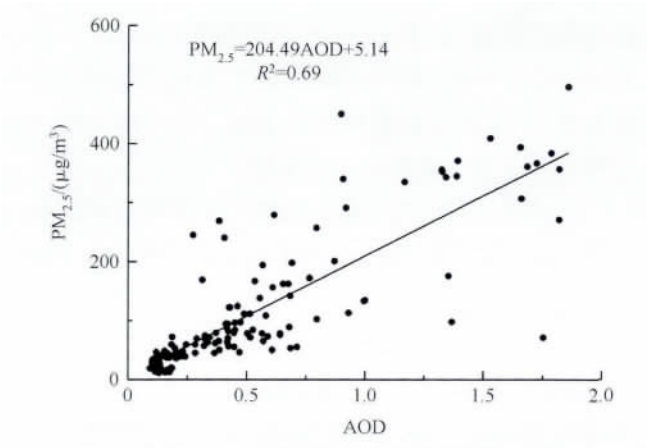
$$PM_{2.5} = 244.48AOD \cdot \eta + 10.6 \quad (5)$$

对比图 3(a) 和图 3(b) 中的散点发现,当 AOD 较大而 $PM_{2.5}$较小的散点经过气溶胶细模态纠正后离散性减弱,相关性提高。因此,建立 AOD_f 与 $PM_{2.5}$的相关性对于遥感估算 $PM_{2.5}$有改进作用。

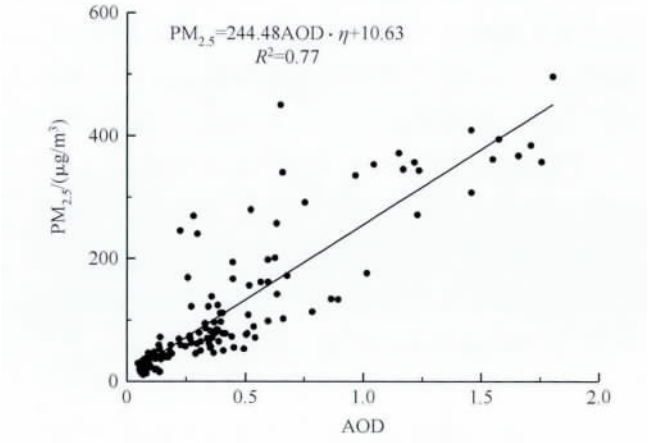
3.4 验 证

本文利用 2013 年 1 月严重灰霾期间的 AOD_f 与 $PM_{2.5}$建立了相关关系。为了验证该关系的可靠性,选取 2013 年 2 月 1 日—15 日的数据进行验证计算。该期间灰霾天气仍很严重,在 5 日—6 日、8 日—9 日以及 12 日—13 日均出现污染。2 月 12 日—13 日,$PM_{2.5}$值可达 365 μg/m³,AOD_f 可达 0.97。2 月 1 日—15 日,$PM_{2.5}$均值为 84 μg/m³,较 1 月的统计平均值 112 μg/m³ 有所减小。这说明,2 月细颗粒物污染有所减弱。

利用式(5) 对 2 月前半月的 $PM_{2.5}$进行估算,并与观测值对比(如图 4 所示)。由图 4 可知,观测的 $PM_{2.5}$与估算值接近,估算的 $PM_{2.5}$均值为 85 μg/m³,其均方根误差为 50 μg/m³。估算获得的 $PM_{2.5}$很好的再现了后两次灰霾过程。而 2 月 5 日—6 日的灰霾过程未能很好的再现,这主要是由于未能获得有效的光学厚度观测值造成的。



(a) AOD与$PM_{2.5}$的相关关系图



(b) AOD_f与$PM_{2.5}$的相关关系图

图3 AOD 与 $PM_{2.5}$ 的相关性分析图

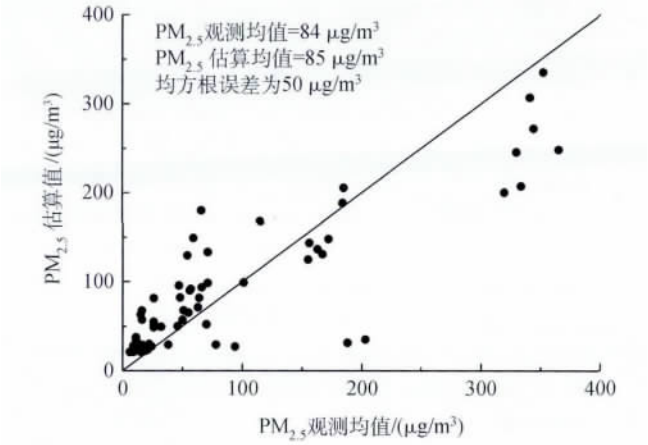


图4 2013 年 2 月 1 日—15 日期间 $PM_{2.5}$ 估算值与 $PM_{2.5}$ 实测值的对比图

4 混合层高度和相对湿度对估算 $PM_{2.5}$ 的影响

4.1 混合层高度对估算 $PM_{2.5}$ 的影响

本文利用 NCEP FNL 全球再分析资料提取 2013 年 1 月北京站的混合层高度每天 4 个时刻(2:00, 8:00, 14:00, 20:00)的数据。为了获取每小时的混合层高度,本文利用正弦函数将每天 4 时刻的混合层高度插值到 24 h。假设混合层内气溶胶消光系数 $k_{ex}(z)$ 随高度 z 的变化遵循指数分布(章澄昌 等, 1995):

$$k_{ex}(z) = \int_0^{\infty} \sigma_{ex}(r, z_0)\, e^{-\frac{z}{MLH}} dr \qquad (6)$$

式中,$\sigma_{ex}(r, z_0)$ 为气溶胶近地面消光,z_0 为近地面高度,MLH 为混合层高度,r 为气溶胶半径。因此,近地面气溶胶消光系数与整层光学厚度存在以下关系:

$$k_{ex}(z_0) = AOD/MLH \qquad (7)$$

激光雷达实测资料表明,2013 年 1 月混合层高度平均值仅为 500 m 左右(张婉春 等,2013),且根据激光雷达探测获得的后向散射廓线可知,严重灰霾期间气溶胶随高度的变化并非遵循指数分布(吕阳 等,2013)。因此,利用式(7)进行垂直订正获得的 AOD 与 $PM_{2.5}$ 相关关系不理想,R^2 仅为 0.28。

4.2 相对湿度对估算 $PM_{2.5}$ 的影响

由相对湿度小时平均数据与近地面气溶胶散射的关系可知,随着相对湿度的增加,气溶胶散射系数 K_{sca} 也随之增长(图 5)。以相对湿度为 10% 时的散射系数作为起始值模拟了吸湿增长因子 $f(RH) = (1 - RH/100)^{-1}$ 对气溶胶散射的影响。当干气溶胶散射系数为 1.0,相对湿度大于 80% 时,气溶胶散射系数急速增长,甚至当相对湿度达到 90% 时,散射系数可达到初始值的 10 倍。

在 2013 年 1 月的实测数据中,相对湿度超过 80% 时,气溶胶光学厚度均小于 2.0。总体看来,当相对湿度小于 80% 时,基本符合吸湿增长因子的作用曲线。图 5 为近地面散射系数与相对湿度的关系图,借鉴 Liu 等人(2008)充分利用环境湿度的日变化进行气溶胶吸湿散射的研究,假设气溶胶化学成分在一天中的变化很小,对每天散射系数日变化进行研究分析。由图 5 可知,当相对湿度大于 80% 时,散射消光急剧增长(如 2013 年 1 月 12 日)。2013 年 1 月 28 日相对湿度也较大,其散射系数与相对湿度的关系同样不满足 Kotchenruther 等人(1999)提出的吸湿订正因子。这主要是因为大气气溶胶中的吸湿性物质 NaCl、NH_4Cl、$(NH_4)SO_4$ 等在环境相对湿度接近 80% 时发生相变(吴兑,

2004)。因此,在2013年1月,吸湿增长因子仅在相对湿度介于40%—80%时,对气溶胶消光增长有着明显作用;而当相对湿度过大时($RH>80\%$),该吸湿增长因子订正效果受到限制。然而,当相对湿度小于80%时,仅在细颗粒物质量浓度小于200 $\mu g/m^3$ 时散射系数与相对湿度的关系满足吸湿增长关系。在计算近地面散射系数时,需用到混合层高度信息,混合层高度测量不准确也会对散射系数产生影响。由于2013年1月颗粒物质量浓度持续较大,因此目前的吸湿增长因子订正方法不能获得较好的订正效果。利用吸湿增长因子订正后,AOD_f 与 $PM_{2.5}$ 相关关系变差,R^2 仅为0.35。湿度订正后,部分散点集中在光学厚度较小而 $PM_{2.5}$ 值很大的区域内。

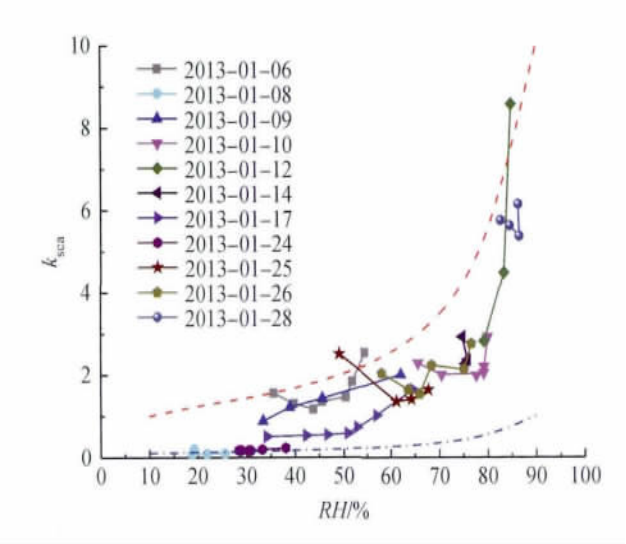


图5 2013年1月相对湿度与气溶胶散射系数的相关关系(其中,虚线和点划线分别表示 RH 在10%时,散射系数为0.1和1.0作为起始值的干气溶胶散射系数在吸湿增长因子作用下的散射消光变化曲线)

5 结 论

光谱退卷积方法可简便的求得气溶胶细模态比例,进而获得细颗粒物消光的贡献。本文利用2013年1月AERONET的Lev1.5气溶胶光学厚度数据与美国大使馆在位测量的近地面 $PM_{2.5}$ 浓度进行相关性分析。在AOD-$PM_{2.5}$ 相关性分析过程中,使用气溶胶细模态订正可有效提高其相关系数。同时,分析混合层高度和相对湿度对气溶胶光学厚度与细颗粒物质量浓度相关关系的影响,并得到以下结论:

(1)通过气溶胶细模态比例可有效获得细颗粒物的消光贡献;在2013年1月严重灰霾时期,估算的 AOD_f 与 $PM_{2.5}$ 的相关系数大于AOD与 $PM_{2.5}$ 的相关系数,这表明灰霾期间以 $PM_{2.5}$ 为代表的细模态颗粒物成为气溶胶消光的主体;

(2)建立 AOD_f 与 $PM_{2.5}$ 的相关关系式可更好地估算 $PM_{2.5}$,且可有效减小均方根误差。本文获得线性拟合公式为:

$$PM_{2.5}=244.48AOD\cdot\eta+10.6,R^2=0.77;$$

(3)气溶胶散射系数的吸湿增长因子仅在相对湿度介于40%—80%时作用明显;当相对湿度过大时($RH>80\%$),该吸湿增长因子订正效果受到限制;在严重灰霾时期,气溶胶垂直分布不满足指数分布假设,因此 AOD_f 的垂直订正对改善 AOD_f 与 $PM_{2.5}$ 相关关系作用不明显。

严重灰霾期间,本文利用气溶胶细模态订正方法建立气溶胶细模态光学厚度与细颗粒物质量浓度的相关关系,改进了遥感估算 $PM_{2.5}$ 的方法。但气溶胶吸湿增长与垂直分布仍是一个亟待解决的问题,这些问题将在以后进一步深入研究。

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