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1 **Satellite-based PM2.5 estimation using fine-mode aerosol optical**

2 **thickness over China**

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Abstract

Accurate estimation of ground-level $PM_{2.5}$ from satellite-derived aerosol optical thickness (AOT) presents various difficulties. This is because the association between AOT and surface $PM_{2.5}$ can be affected by many factors, such as the contribution of fine mode AOT (FM-AOT) and the weather conditions. In this study, we compared the total AOT and FM-AOT for surface $PM_{2.5}$ estimation using ground-based measurements collected in Xingtai, China from May to June 2016. The correlation between $PM_{2.5}$ and FM-AOT was higher (r = 0.74) than that between $PM_{2.5}$ and total AOT (r = 0.49). Based on FM-AOT, we developed a ground-level $PM_{2.5}$ retrieval method that incorporated a Simplified Aerosol Retrieval Algorithm (SARA) AOT, look-up table–spectral deconvolution algorithm (LUT-SDA) fine mode fraction (FMF), and the $PM_{2.5}$ remote sensing method. Due to the strong diurnal variations displayed by the particle density of $PM_{2.5}$, we proposed a pseudo-density for $PM_{2.5}$ retrieval based on real-time visibility data. We applied the proposed method to determine retrieval surface $PM_{2.5}$ concentrations over Beijing from December 2013 to June 2015 on cloud-free days. Compared with Aerosol Robotic Network (AERONET) data, the LUT-SDA FMF was more easily available than the Moderate Resolution Imaging Spectroradiometer (MODIS) FMF. The derived $PM_{2.5}$ results were compared with the ground-based monitoring values (30 stations), yielding an R^2 of 0.64 and root mean square error (RMSE) = 18.9 ug/m^3 (N = 921). This validation demonstrated that the developed method performed well and produced reliable results.

Keywords: $PM_{2.5}$; MODIS; fine mode fraction; AOT

1. Introduction

Particulate matter with an aerodynamic diameter less than 2.5 μm ($PM_{2.5}$) has adverse effects on human health (Anderson et al., 2012). Numerous studies have demonstrated that $PM_{2.5}$ is associated with mortality, respiratory system problems, and lung cancer (Pope III et al., 2002; Brook et al., 2010; Kloog et al., 2013). More specifically, a decrease of 10 μg/m^3 in the concentration of $PM_{2.5}$ was associated with an estimated increase in life expectancy of approximately 6 months (Pope III et al., 2009). Thus, it is important to accurately assess the distribution of $PM_{2.5}$ concentrations and exposures to guide control measures for mitigating health impacts (Bell et al., 2011; Luo et al., 2014).

Monitoring site measurements of ground-level $PM_{2.5}$ concentrations offer a convenient but rudimentary option for pollution and health studies; their spatial coverage is sparse and limited (Han et al., 2015). Satellite remote sensing has been used in many studies to obtain large-scale $PM_{2.5}$ distributions (Liu et al., 2009; Lee et al., 2011; Chudnovsky et al., 2014; Kloog et al., 2015). The common satellite product for estimating ground-level $PM_{2.5}$ concentration is aerosol optical thickness (AOT). Researchers have determined the relationships between AOT and $PM_{2.5}$ concentration through various approaches, including empirical statistical models (Engel-Cox et al., 2004; Gupta et al., 2006; Schaap et al., 2009; Guo et al., 2014), chemical transport models (Wang et al., 2010; van Donkelaar et al., 2010; Liu et al., 2011; Xu et al., 2013), and physical models (Kokhanovsky et al., 2009).

Most of the recent studies using satellite AOT to derive $PM_{2.5}$ have focused on the total AOT for estimation of ground-level $PM_{2.5}$ (Eeftens et al., 2012; Lee et al., 2012; Luo et al., 2014). However, some researchers reported that fine mode AOT (FM-AOT) has a higher correlation with ground-level $PM_{2.5}$ (Nicolantonio et al., 2007; Zhang and Li, 2013). Van Donkelaar et al. (2011) successfully applied FM-AOT for $PM_{2.5}$ estimation in Moscow with high accuracy. Compared with using total AOT to estimate $PM_{2.5}$, Nicolantonio et al. (2007) reported a significant improvement in using FM-AOT; correlation coefficients increased from 0.59 to 0.74 for measurements collected over north Italy in June. Zhang and Li (2013) reported that the relationship

90   between FM-AOT and PM$_{2.5}$ was significantly closer than that between total AOT and

91   PM$_{2.5}$ under hazy weather conditions. Although the correlations with PM$_{2.5}$ are

92   improved by using FM-AOT, there have been few studies comparing total AOT and

93   FM-AOT for estimating PM$_{2.5}$. However, because FM-AOT is used in PM$_{2.5}$ retrieval

94   models, the fine mode fraction (FMF), which is used to separate contributions from

95   smaller and larger particles in AOT, is becoming an increasingly important parameter.

96   Zhang and Li (2015) proposed an expression between Aerosol Robotic Network

97   (AERONET) FMF and volume-to-extinction ratio of fine particulates (VE$_f$) for PM$_{2.5}$

98   retrieval, and applied the expression using Moderate Resolution Imaging

99   Spectroradiometer (MODIS) FMF. However, AERONET FMF has a very different

100   calculation method than MODIS FMF (Gasso et al., 2006). Levy et al. (2007)

101   reported that MODIS FMF had a poor correlation with AERONET FMF, and this was

102   also reported by Zhang and Li (2015). Therefore, MODIS FMF may not be suitable

103   for models based on AERONET FMF.

104   Despite promising recent progress in surface PM$_{2.5}$ estimation from satellite AOT,

105   uncertainties still exist due to several factors. Toth et al. (2014) found that both the

106   quality of satellite-based AOT retrieval as well as the surface-to-column

107   representativeness of aerosol particles affects the estimation of PM$_{2.5}$ concentration.

108   In addition, the planetary boundary layer height (PBLH) was reported to have a

109   significant impact on the AOT-PM$_{2.5}$ relationship (Gupta and Christopher, 2009); this

110   is because greater PBLH is more favorable for dilution and diffusion of pollutants,

111   which produces low concentrations of surface PM$_{2.5}$ despite high AOT (Qu et al.,

112   2016). Relative humidity (RH) was found to be another key factor that could result in

113   discrepancies between AOT and surface PM$_{2.5}$ (Wang, 2003; Paciorek et al., 2008).

114   Wang et al. (2010) reported a significant improvement in surface PM$_{2.5}$ estimation

115   when a RH correction was incorporated (R^2 increased from 0.35 to 0.66). Furthermore,

116   recent studies have used the MODIS AOT products (MOD04) for PM$_{2.5}$ monitoring,

117   but its spatial resolution is 10 km, which is unsuitable for exposure estimates in urban

118   areas (Jerrett et al., 2005). Chudnovsky et al. (2013) reported that the spatial

119   resolution of AOT also affected PM$_{2.5}$ accuracy: the correlation between AOT and

PM$_{2.5}$ decreased significantly because the AOT resolution was degraded. Although MODIS recently released a 3-km AOT product, the accuracy was less reliable than that of 10-km AOTs (Munchak et al., 2013; Yan et al., 2016).

Yan et al. (2017) proposed a look-up table–spectral deconvolution algorithm (LUT-SDA) method for satellite FMF that showed a high correlation with AERONET FMF. In addition, Bilal et al. (2013) developed a Simplified Aerosol Retrieval Algorithm (SARA) to derive AOT from MODIS, which provided up to 500-m spatial resolution AOT and has been validated in Hong Kong and Beijing (Bilal et al., 2014). Thus, in this study, we used field data to compare the total AOT and FM-AOT with the surface particulate matter concentration. Then, we developed a ground-level PM$_{2.5}$ retrieval model for satellite data based on FM-AOT, which incorporated the LUT-SDA (Yan et al., 2017), SARA (Bilal et al., 2013), and the PM$_{2.5}$ remote sensing method (Zhang and Li, 2015).

2. Data and Methods

2.1 Ground-based experiment at Xingtai

To compare the correlations between total AOT and FM-AOT with surface PM$_{2.5}$, we performed a ground-based experiment in Xingtai city, Hebei province, China, (37.18°N, 114.36°E, Figure 1). Xingtai is an important transportation hub and center of heavy industry (Chen et al., 2017). It also has high raw coal consumption of more than 3,000 kt (Cheng et al., 2017). The key industries in Xingtai are construction, coal, and the metallurgy, all of which produce high emissions of air pollutants (Wang et al., 2015).

The surface particulate matter concentration was obtained using a GrayWolf 6-channel handheld particle/mass meter (PC-3016A). As a stand-alone meter, the GrayWolf PC-3016A can measure particle concentration (μg/m^3) with a size range of 0.3-10.0 μm (https://www.wolfsense.com/). Thus, besides PM$_{2.5}$, we also collected PM$_1$ concentration data in this study. Here the PM data is to compare the relationship between total AOT, FM-AOT and PM$_{2.5}$ at Xingtai.

The columnar aerosol data were measured using a CE318-DP sun-sky radiometer

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150 (AOT uncertainties are 0.01-0.02, Li et al. (2016)). This polarized version CE318

151 belongs to the Sun-sky radiometer Observation NETwork (SONET) and the collected

152 data had been uploaded to AERONET SONET_Xingtai (Li et al., 2016). The aerosol

153 data used in this research is corresponding to the level 1.5 of the AERONET (Holben,

154 et al., 1998).

155   Meteorological parameters including temperature (T, uncertainties are 0.2℃), RH

156 (uncertainties are 1%-5%), wind speed (WS, uncertainties are 0.5 m/s -1 m/s), wind

157 direction (WD, uncertainties are 5°-10°) and visibility (VIS, uncertainties are 20%)

158 were simultaneously recorded by an automatic meteorological station at the same

159 location as the study area (WMO, 2008). All measurements in Xingtai were collected
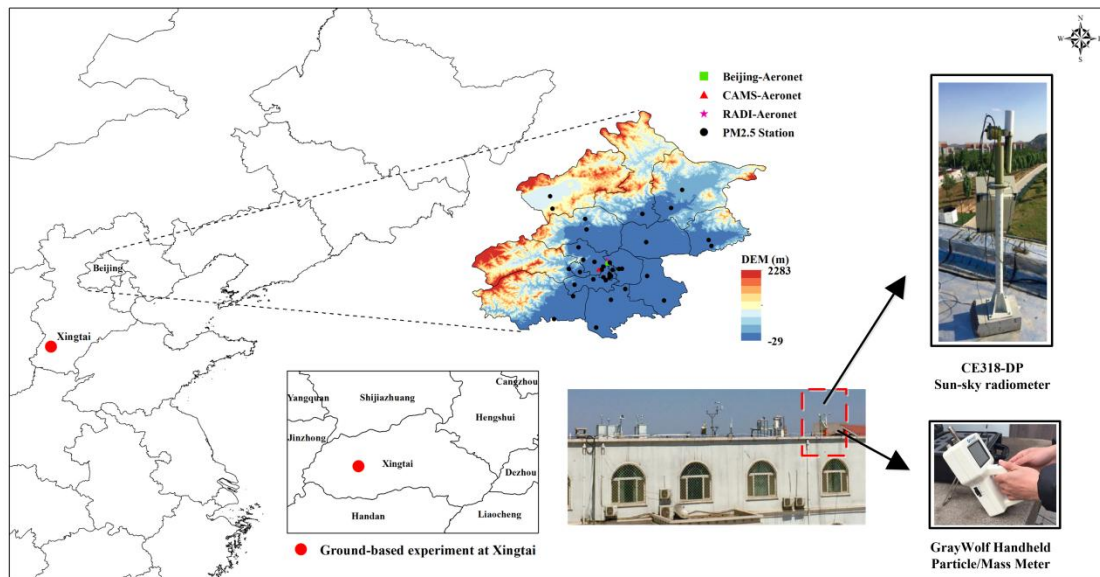
160 from 1 May to 1 Jun 2016.

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162 Figure 1. Ground-based experiment in Xingtai and satellite validation in Beijing.

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164 **2.2 Satellite AOT**

165   The MODIS MOD02HKM, MOD03, and MOD09 cloud-free data were acquired

166 (https://ladsweb.nascom.nasa.gov) for AOT retrieval, as shown in Table 1. In addition,

167 Collection 6 MODIS aerosol products (C6 MOD04) were obtained for this study, and

168 the C6 cloud mask data (Aerosol_Cldmsk_Land_Ocean) were extracted from MOD04

169 and used for cloud screening in aerosol retrieval algorithm. The SARA was applied in

170 the Beijing area due to its high accuracy. AOT derived by SARA was validated in

Beijing and the results showed a close correlation with AERONET AOT (0.97-0.99), with a small root mean square error (RMSE) 0.067-0.133 (Bilal et al., 2014). In this study, 550 nm AOT was retrieved by SARA and its comparison with AERONET was showed in supplementary Figure S1. It shows the majority of the observations (81%) are within the error range $\pm$ (0.05+0.15$_{\text{AERONET AOT}}$), which indicates that the retrieved AOT values are of good quality.

Table 1 The MODIS data used in this study

Day	Month	Year	Day	Month	Year
11	12	2013	13	11	2014
12	12	2013	17	11	2014
14	12	2013	22	11	2014
26	12	2013	1	12	2014
28	12	2013	3	12	2014
30	12	2013	17	12	2014
1	1	2014	24	12	2014
3	1	2014	31	12	2014
22	1	2014	2	1	2015
4	2	2014	11	1	2015
2	5	2014	6	1	2015
7	5	2014	27	1	2015
18	5	2014	30	1	2015
3	6	2014	5	2	2015
12	6	2014	17	2	2015
27	6	2014	26	2	2015
28	6	2014	3	3	2015
10	7	2014	11	3	2015
12	7	2014	24	4	2015
15	8	2014	26	4	2015
25	8	2014	4	5	2015
3	9	2014	7	5	2015
8	9	2014	19	5	2015
9	9	2014	26	5	2015
15	9	2014	2	6	2015
16	10	2014	8	6	2015
17	10	2014	18	6	2015

2.3 AERONET

The AERONET program is a federation of ground-based remote sensing aerosol networks (Holben et al., 2001). The AERONET collaboration provides globally distributed observations of spectral AOT, inversion products, and other AOT-dependent products. AOT data are computed for three data quality levels: Level 1.0 (unscreened), Level 1.5 (cloud-screened), and Level 2.0 (cloud-screened and quality-assured). In this study, 550 nm AOT (interpolated by 675 nm and 440 nm) and FMF were collected at the AERONET sites in Beijing (Level 2), CAMS (Level 2), and RADI (only Level 1.5 available) from December 2013 to June 2015 for SARA and validation purposes.

2.4 Beijing $PM_{2.5}$

Hourly ground-measured $PM_{2.5}$ data over the Beijing region from December 2013 to June 2015 were acquired from the Beijing Municipal Environmental Monitoring Center (http://zx.bjmemc.com.cn) for the proposed model validation. The $PM_{2.5}$ monitoring sites, shown in Figure 1, are mainly located in urban areas, with a few in rural areas. Measurements of $PM_{2.5}$ concentration were based on the Chinese National Ambient Air Quality Standard (GB3095-2012), using the tapered element oscillating microbalance method (TEOM) and the beta-attenuation method (Li et al., 2015).

2.5 LUT-SDA FMF

As described in Yan et al. (2017), the LUT-SDA has been developed for satellite images based on only two wavelengths of AOT to solve the FMF problem. This method is based on the SDA currently used in AERONET. Thus, the outcome of the LUT-SDA is a good match to that of AERONET and is suitable as an input parameter for the AERONET FMF-based model. The LUT-SDA builds a LUT to retrieve the FMF using satellite-derived AOT and Ångström exponent, the detailed algorithm is shown in supplementary Figure S2. To build a LUT, a set of hypothetical FMF (η) and Ångström exponent derivative (α') values are imported to the SDA along with the

satellite-determined Ångström exponent (α) to derive the Ångström exponent of fine mode aerosols (αf). The LUT-SDA successfully applied to MODIS data not only verified the application to the urban scale (Beijing), but also its expansion to large areas. The retrieved FMF images represent the spatial distribution of the fine aerosol contribution to the total AOT with complex surface types. Therefore, in this study, we used the LUT-SDA for the retrieval FMF for FM-AOT and correlated it with $PM_{2.5}$ estimations.

2.6 PBLH and RH

The Weather Research and Forecasting (WRF) model (ver. 3.6.1) was applied to produce the required PBLH and RH values. The WRF model is a mesoscale numerical weather prediction system that has been validated for producing accurate simulation of meteorological data (Skamarock et al., 2005; Grguric et al., 2014). Initial and boundary conditions were from the National Centers for Environmental Prediction (NCEP) Final (FNL) Operational Global Analysis data (http://rda.ucar.edu/datasets/ds083.2/). NCEP FNL is provided globally at 1 degree resolution every 6 hours. The physics options selected for the WRF simulation for this study were the same as in Zheng et al. (2015), which is also shown in supplementary Table S1.

2.7 Ground-level $PM_{2.5}$ retrieval model

Zhang and Li (2015) developed a $PM_{2.5}$ remote sensing method for ground-level $PM_{2.5}$ estimation, which they validated in Jinhua city, China (Li et al., 2016). This method proposed a relationship between AOT and $PM_{2.5}$ concentration based on AERONET data, which included FM-AOT conversion, fine particle volume calculation, and PBLH and RH correction. The surface $PM_{2.5}$ concentration can be obtained by the following equation (Zhang and Li, 2015):

$$PM_{2.5} = AOT \cdot \frac{FMF \cdot VE_f \cdot \rho_{f \cdot dry}}{PBLH \cdot f(\mathrm{RH})} \quad (1)$$

where AOT is retrieved from satellite, FMF is fine mode fraction calculated by

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LUT-SDA, VE_f is the columnar volume-to-extinction ratio of fine particulates, $\rho_{f \cdot dry}$ is density of dry $PM_{2.5}$, PBLH is the planetary boundary layer height and f (RH) is the optical hydroscopic growth function. VE_f can be calculated by (Zhang and Li, 2015):

$$VE_f = 0.2887 FMF^2 - 0.4663 FMF + 0.356 \quad (0.1 \le FMF \le 1.0) \quad (2)$$

In this study, f (RH) is based on Chen et al. (2015):

$$f(\text{RH}) = \begin{cases} 1.02 \times (1 - \text{RH}/100)^{-0.21 \times RH/100} & (\text{RH}/100 < 0.6) \\ 1.08 \times (1 - \text{RH}/100)^{-0.26 \times RH/100} & (\text{RH}/100 \ge 0.6) \end{cases} \quad (3)$$

$\rho_{f \cdot dry}$ is assumed to have a constant value of 1.5 g/cm^3 in Zhang and Li (2015) and Li et al. (2016). However, in this study, we assumed that $\rho_{f \cdot dry}$ varies daily (further discussion is in Section 4.2), and then proposed a pseudo-density of $PM_{2.5}$ (ρ_{pseudo}) to substitute for the daily-varied $\rho_{f \cdot dry}$, which can be calculated by ground-based measurements. Thus based on Eq.(1), the pseudo-density of $PM_{2.5}$ is computed as follow:

$$\rho_{pseudo} = AOT \cdot \frac{FMF \cdot VE_f \cdot PM_{2.5}^{\text{V}}}{PBLH \cdot f(\text{RH})} \quad (4)$$

where AOT and FMF are obtained from AERONET stations corresponding to satellite overpass, and PBLH and f (RH) are produced by WRF.

In Eq (4), $PM_{2.5}^{\text{V}}$ is calculated from visibility (VIS) data. Previous studies indicated that $PM_{2.5}$ can be calculated from VIS using the following power function (Leung et al., 2008; Bai et al., 2016):

$$PM_{2.5}^V = A \cdot x^B \quad (5)$$

where x is visibility (VIS) (km), which was obtained from weather station for Beijing (https://www.wunderground.com/). The A and B parameters in this equation are given in Table 2 (Chen et al., 2010). Chen et al. (2010) indicated that it easily causes fog when RH>90%, thus it is not suitable for the estimation of $PM_{2.5}$ from VIS under this weather condition.

Table 2 Power function for $PM_{2.5}^{V}$ (x is VIS in km)

Relative humidity	Power function for $PM_{2.5}^{V}$ (mg/m³)
RH < 70%	$PM_{2.5}^{V} = 0.6977x^{-0.9517}$
70% ≤ RH ≤ 80%	$PM_{2.5}^{V} = 0.3628x^{-1.028}$
80% < RH ≤ 90%	$PM_{2.5}^{V} = 0.2957x^{-0.9463}$

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268   A schematic diagram of the ground-level PM$_{2.5}$ retrieval method used in this

269 study is shown in Figure 2. The method consists of two main steps: (i) calculation of

270 ρ_{pseudo} using VIS data using Eq (4); and (ii) incorporation of ρ_{pseudo} in Eq (1) with

271 SARA-AOT, LUT-SDA FMF, and the PBLH and RH correction for surface PM$_{2.5}$

272 retrieval. As mentioned in Zhang and Li (2015), the result of Eq. (1) is in mg/m³, and

273 should be divided by 1,000 for comparison with the *in situ* measurements in μg/m³.

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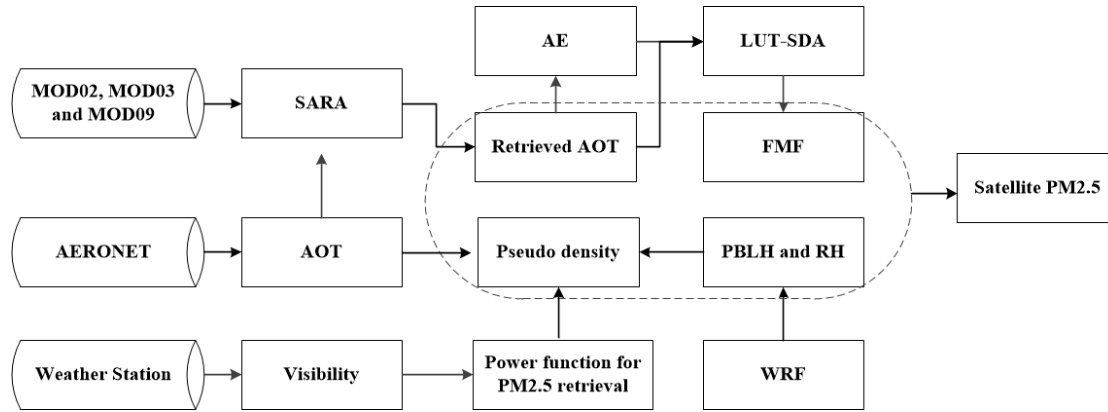
Figure 2. Schematic diagram of the ground-level PM$_{2.5}$ retrieval model.

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3. Ground-based experiment in Xingtai

3.1 Dependence of PM on meteorological parameters

280   Descriptive statistics for the data obtained in Xingtai are presented in

281 supplementary Table S2. The PM and Meteorological data are recorded each 10

282 minutes, and thus there are totally 19414 data obtained; however, VIS N is finally

283 19394 since some measurements are missing. Due to the cloud-screened process, the

aerosol data are 1196. Although the mean PM_1 and $PM_{2.5}$ were 28.87 μg/m^3, and

45.14 μg/m^3, respectively, indicating a moderate health risk level, their standard

deviations were 21.65 μg/m^3, and 32.19 μg/m^3, respectively, which indicates

significant variation in the surface PM concentration during the study period. Figure 3

shows the time series of PM_1, $PM_{2.5}$, T, RH, and VIS from 1 May to 1 June 2016 in

Xingtai. It can be seen that there was a heavy pollution event from 9 to 12 May 2016

(highlighted in gray). On those days, the $PM_{2.5}$ and PM_1 rose as high as 290 μg/m^3,

and 135 μg/m^3, respectively, and VIS decreased significantly to 4,868 m.

The correlations between $PM_{2.5}$ and meteorological data are shown in Figure 4.

$PM_{2.5}$ was significantly correlated with WS, RH, and VIS. However, T changes did

not significantly affect $PM_{2.5}$ concentrations, and we observed a small negative

correlation (r = -0.107). Dawson et al. (2007) proposed that $PM_{2.5}$ levels display an

almost negligible response to T changes in summer, largely due to increases in sulfate

canceling out any decrease in nitrate and organics. RH was highly correlated with

$PM_{2.5}$ (r = 0.637), which is in agreement with the findings of Luo et al. (2014).

Furthermore, Elminir (2005) reported that an increase in humidity reflects an updraft

of boundary layer air masses to 3 km, which may cause higher levels of air pollution.

Thus, RH should be taken into account as an important factor in surface $PM_{2.5}$

estimation models.

VIS and $PM_{2.5}$ had the most significant anti-correlation (r = -0.840). The main

reason for this is that fine particulate matter can scatter and absorb sunlight easily (Liu

et al., 2014). Zhao et al. (2011) reported that, based on the negative correlations

between VIS and $PM_{2.5}$ concentration, aerosols could cause a greater degree of VIS

impairment in the summer months. We also observed a similarly strong negative

correlation, which due to the WS being low during the study period, as shown in

supplementary Table S2 (mean = 3.74 m/s, Q3 = 4.70 m/s). Wang et al. (2006)

reported that a WS of less than 7.0 m/s produced a high correlation between VIS and

$PM_{2.5}$, with the correlation increasing with decreasing WS. Thus, it is appropriate to

use VIS to estimate surface $PM_{2.5}$ concentration, as reported by Chen et al. (2010).

The use of VIS to improve the retrieval accuracy of $PM_{2.5}$ will be discussed further in
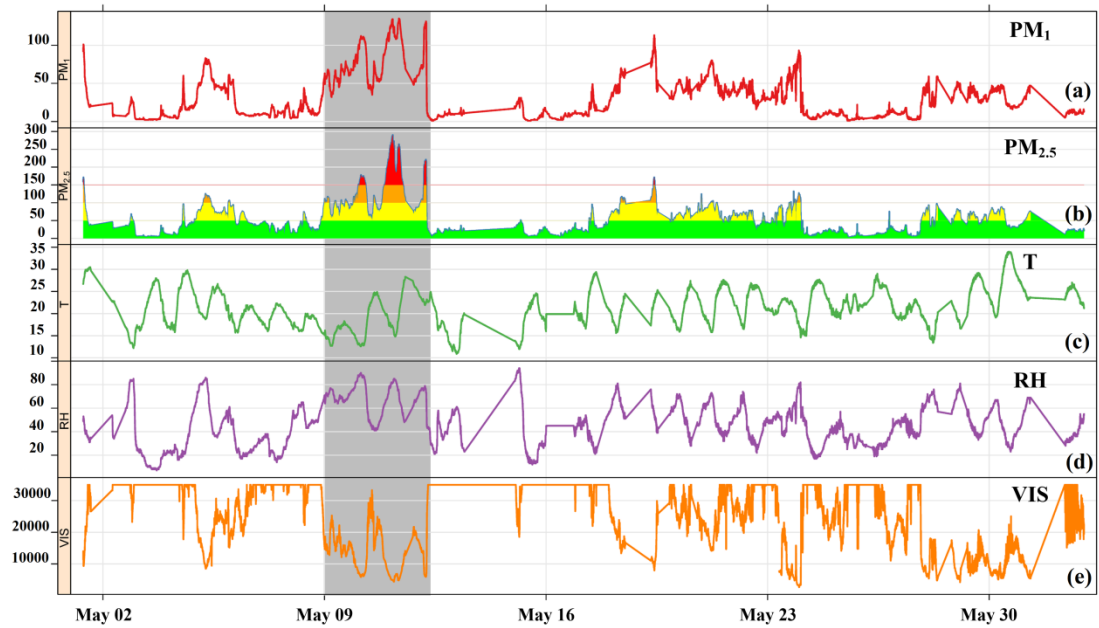
314   Section 4.2

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317   Figure 3. Time series of (a) PM_1, (b) $PM_{2.5}$, (c) temperature (T), (d) relative humidity

318   (RH), and (e) visibility (VIS) in Xingtai, China. The gray highlighted area is the most

319   heavily polluted during the experiment. The shaded colors in (b): the green is 0-50

320    $\mu g/m^3$, the yellow is 50-100 $\mu g/m^3$, the orange is 100-150 $\mu g/m^3$, and the red is greater
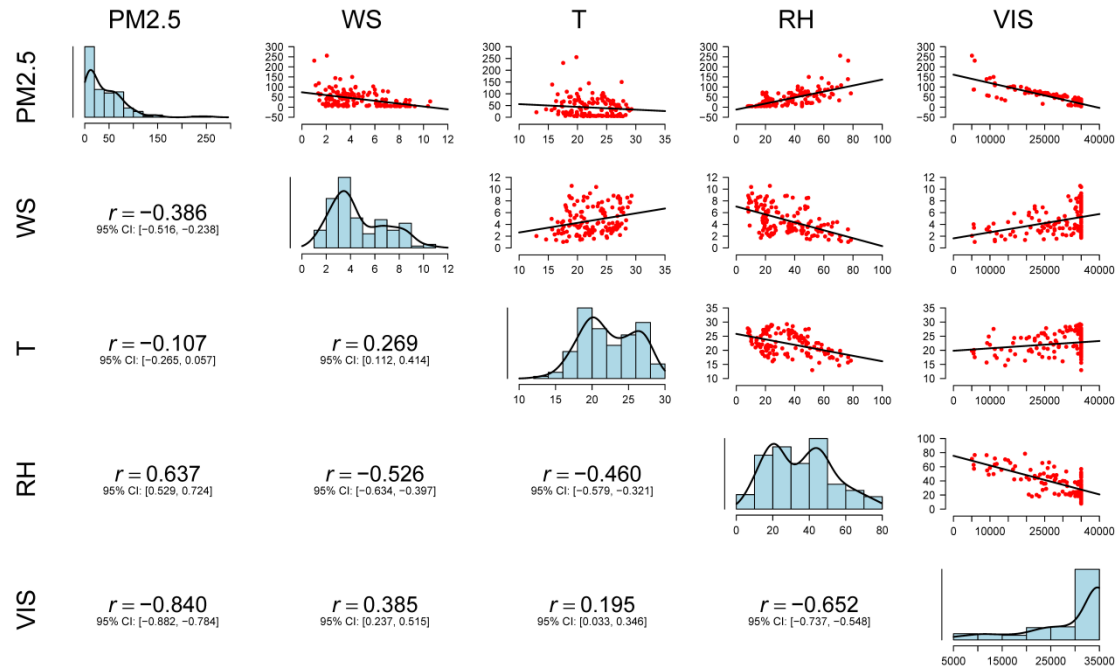
321   than 150 $\mu g/m^3$.

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3.2 Comparison between total AOT and FM-AOT for PM$_{2.5}$ estimation

Figure 5 shows time series of total AOT, FM-AOT, and FMF. The orange-shaded areas shows when the total AOT was much higher than FM-AOT, which indicates that coarse mode AOT accounted for a major proportion of the total AOT. However, on the days around 8 May 2016, the FM-AOT was very similar to total AOT, and the FMF was approximately 0.93, indicating that FM-AOT accounted for most of the total AOT on those days (indicated by the rectangle with broken red line). The time series of FMF in Figure 5 also reveals significant variation in the ratio between FM-AOT and total AOT among different days. Kim et al. (2007) observed that the monthly mean FMF varied from 0.60 (December) to 0.77 (July), which reflected a marked difference between FM-AOT and total AOT, associated with different air pollution conditions.
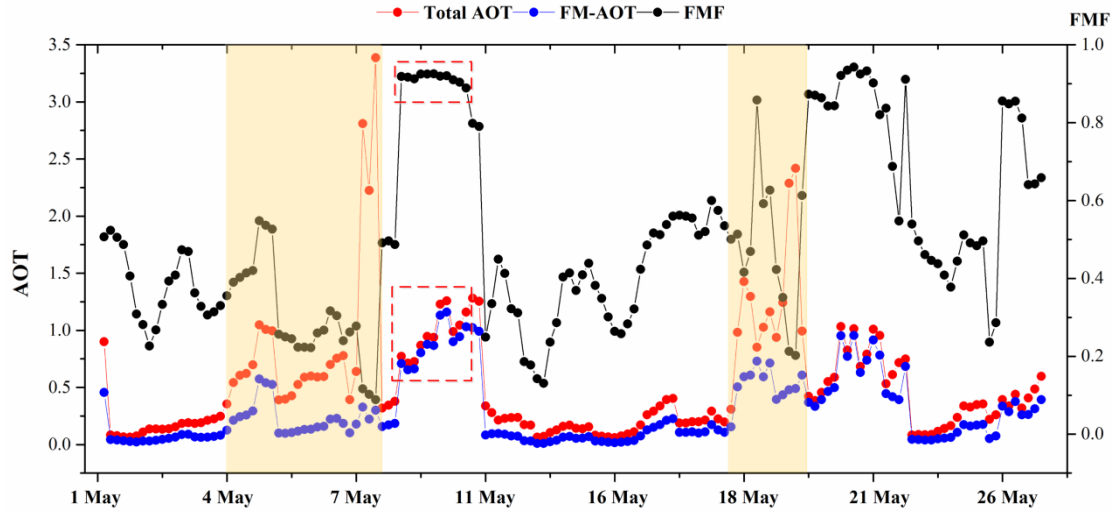
Figure 5. Time series of total aerosol optical thickness (AOT), fine mode AOT (FM-AOT), and fine mode fraction (FMF) in Xingtai, China. The shaded areas present that the total AOT is much higher than FM-AOT. The red dotted rectangles show that the FM-AOT, especially thoes with high FMF values, is very similar to total AOT.

The comparison between the PM$_{2.5}$ retrieved from total AOT and that from FM-AOT is shown in Figure 6. In total, 146 hourly data values were collected for statistical correlation analysis. The correlation between total AOT and PM$_{2.5}$ was r = 0.49, with three obvious outliers highlighted by colored circles in Figure 6. However, the correlation between FM-AOT and PM$_{2.5}$ (r = 0.74) was clearly higher than that between total AOT and PM$_{2.5}$. When the FMF correction was incorporated, the three outliers disappeared and the data were close to the fitted line. The comparison between total AOT and FM-AOT with PM$_1$ is also presented in Figure 6. It can be seen that FM-AOT was significantly more closely correlated with PM$_1$ than was total AOT with PM$_1$ (r increased from 0.50 to 0.82), and the correlation was higher than with PM$_{2.5}$ (r = 0.74). This is because the range of the upper limits of the radius for FM-AOT in AERONET is 0.439–0.992 μm, which contains all of the fine particles of PM$_1$ (Wang et al., 2015). Thus, these comparisons indicate that the FM-AOT is more suitable for fine particulate matter estimation, which is in agreement with the research of Nicolantonio et al. (2007), and Zhang and Li (2013).
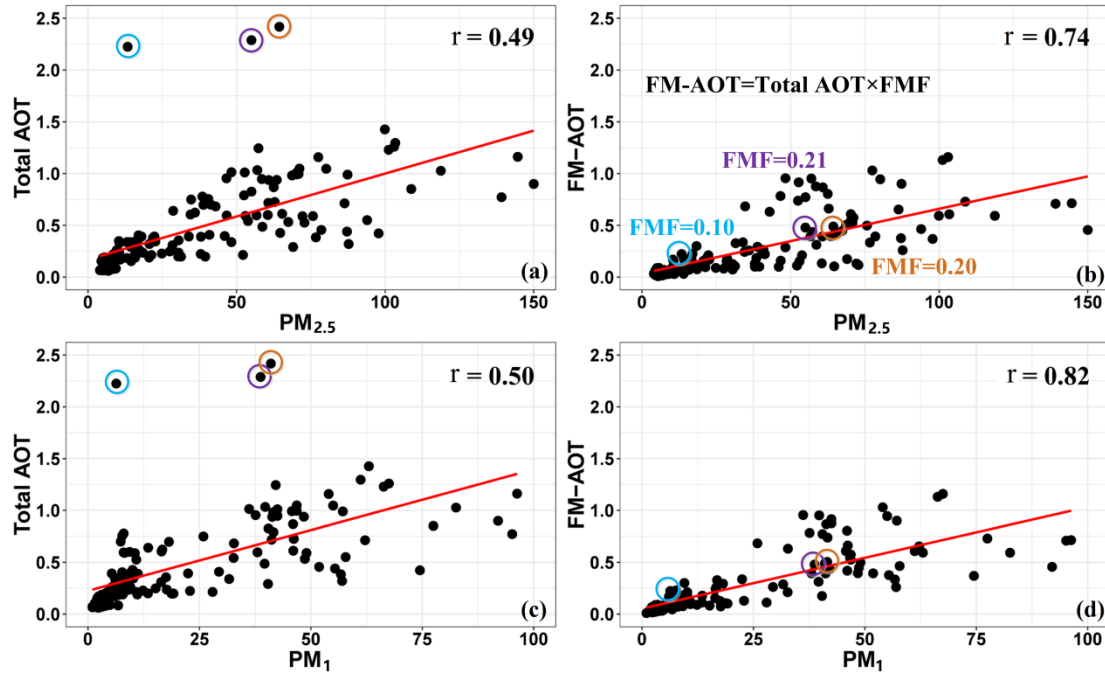
4. Ground-level $PM_{2.5}$ estimation based on satellite FM-AOT

4.1 LUT-SDA FMF

As shown in Eqs. (1) and (2), FMF is an important parameter that can directly affect not only VE_f but also final $PM_{2.5}$ outcomes. Thus, reliable FMF values are necessary for the $PM_{2.5}$ retrieval model. Although the MODIS aerosol products provide the FMF data, its retrieval accuracy over land is highly uncertain (Levy et al., 2010). Thus, in this study, LUT-SDA was applied to improve FMF. Comparisons of FMF values calculated from the LUT-SDA versus those calculated from AERONET and MODIS FMF are presented in Figure 7. It can be seen that the LUT-SDA FMF values are in close agreement with the AERONET FMF values. The mean absolute errors (MAE) between LUT-SDA FMF and AERONET FMF at the three selected stations were 0.146 (RADI), 0.149 (Beijing), and 0.159 (CAMS). However, MODIS

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FMF data were missing for most of the study period. Levy et al. (2010) considered MODIS-retrieved FMF over land to still be an experimental product with little physical validity. Li et al. (2016) also mentioned limitations relating to the accuracy of MODIS retrieval parameters with respect to the application and validation of Eq. (1). Although there are still limitations for satellite retrieval FMF, as reported in Yan et al. (2017), it provides a spatial view of FMF for each pixel of the satellite image, whereas field measurements only provide point scale outcomes.
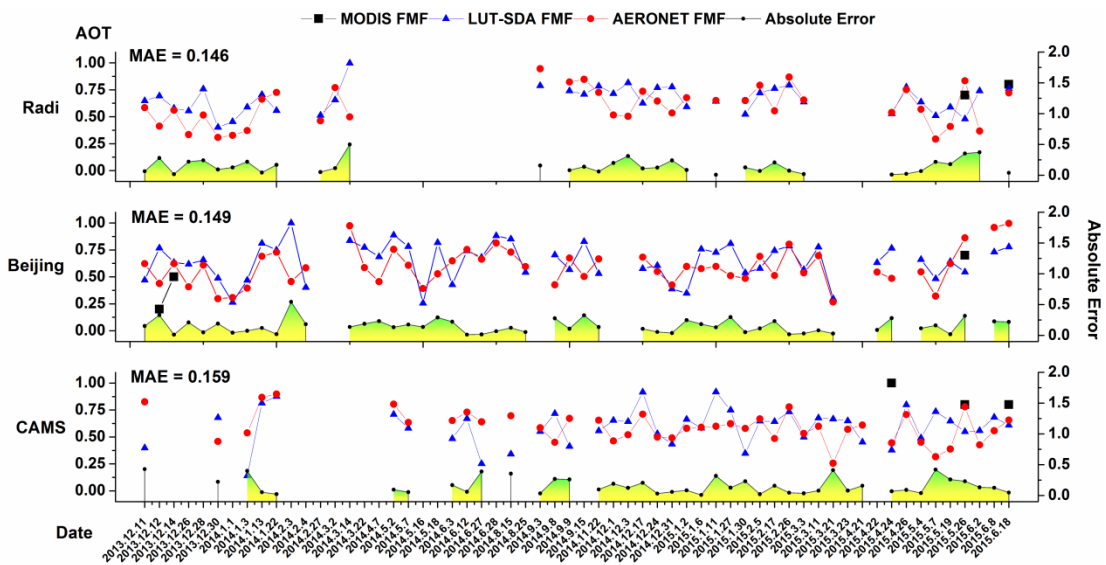

Figure 7. Comparisons among AERONET FMF, MODIS C6 FMF, and LUT-SDA FMF for different AERONET stations. The MAE is the mean absolute error. The green and yellow shadings show the absolute error between LUT-SDA FMF and AERONET FMF.

4.2 Pseudo-density for PM$_{2.5}$

Several studies have reported strong seasonal and diurnal variations in the particle density of PM$_{2.5}$ (Zhao et al. 2013; Wang et al., 2014; Liu et al., 2015). As shown in Table 4, the density of PM$_{2.5}$ in the warm season is always higher than that in the cold season. On average, the density of PM$_{2.5}$ in Beijing can increase from 1.68 g/cm^3 in the cold season to 1.81 g/m^3 in the warm season, due to the contribution from organic matter during the cold season (Liu et al., 2015). Thus, it is problematic to use

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a constant empirical $PM_{2.5}$ density value in Beijing and it is necessary to use an estimate of ambient particle density for $PM_{2.5}$ to improve the accuracy of surface $PM_{2.5}$ retrieval. Therefore, in this study, we proposed a pseudo-density to address this issue. Based on Chen et al. (2010), $PM_{2.5}$ can be directly calculated from the VIS through the power functions under different RH conditions, as shown in Table 2. And then using Eq. (4) and (5), we calculated the daily pseudo-density of $PM_{2.5}$ during the study period. As shown in Figure 8, the pseudo-density of $PM_{2.5}$ showed significant diurnal variations due to changes in weather and pollution conditions. The maximum daily $PM_{2.5}$ pseudo-density could be greater than 2.5 g/cm^3, and the minimum could be less than 0.5 g/cm^3. The results from statistical analyses of $PM_{2.5}$ pseudo-density are shown in Figure 9. The mean value was 1.02 g/cm^3, with a confidence interval of 0.97 to 1.06 g/cm^3, and the maximum standard deviation was 0.62. Figures 8 and Figure 9 show that assuming daily variability in $PM_{2.5}$ density in the retrieval model is more appropriate than assuming constant density.

Table 4 Density of $PM_{2.5}$ in Beijing from the literature

| Reference	Density (g/cm^3)	Sampling time
Liu et al. (2015)	1.60 ± 0.43	July to September 2014
	1.41 ± 0.40	November 2013 to January 2014
Zhao et al. (2013)	1.66 ± 0.74	January to February 2010
	1.82 ± 0.33	July to August 2009
Gao et al. (2007)	1.5	April to August 2005
Cao et al. (2012)	1.79 ± 0.23	June to July 2003
	1.65 ± 0.37	January 2003
Wang et al. (2014)	1.72 ± 0.94	December 2002 to February 2003
	1.82 ± 0.47	July to August 2002
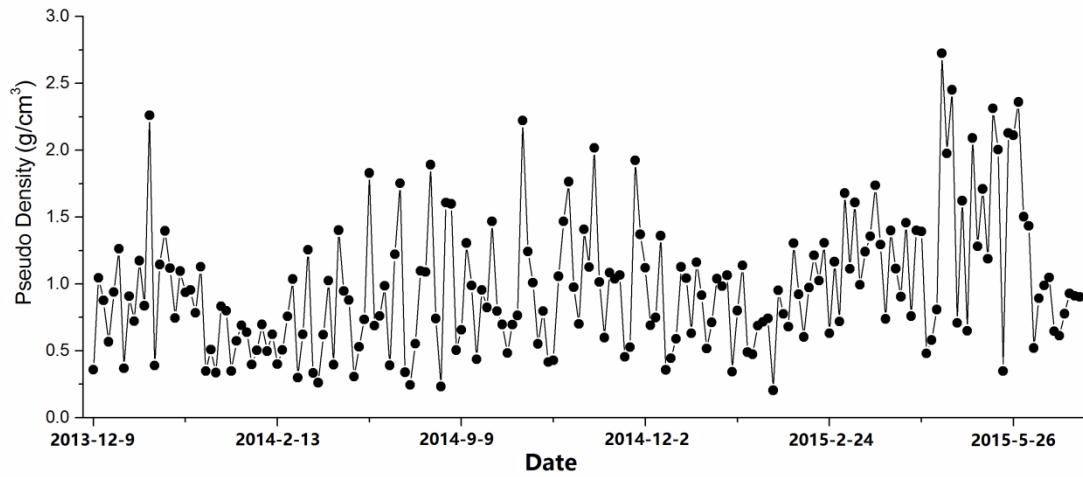
Figure 8. Variation in $PM_{2.5}$ pseudo-density (g/cm^3) from December 2013 to June 2015.
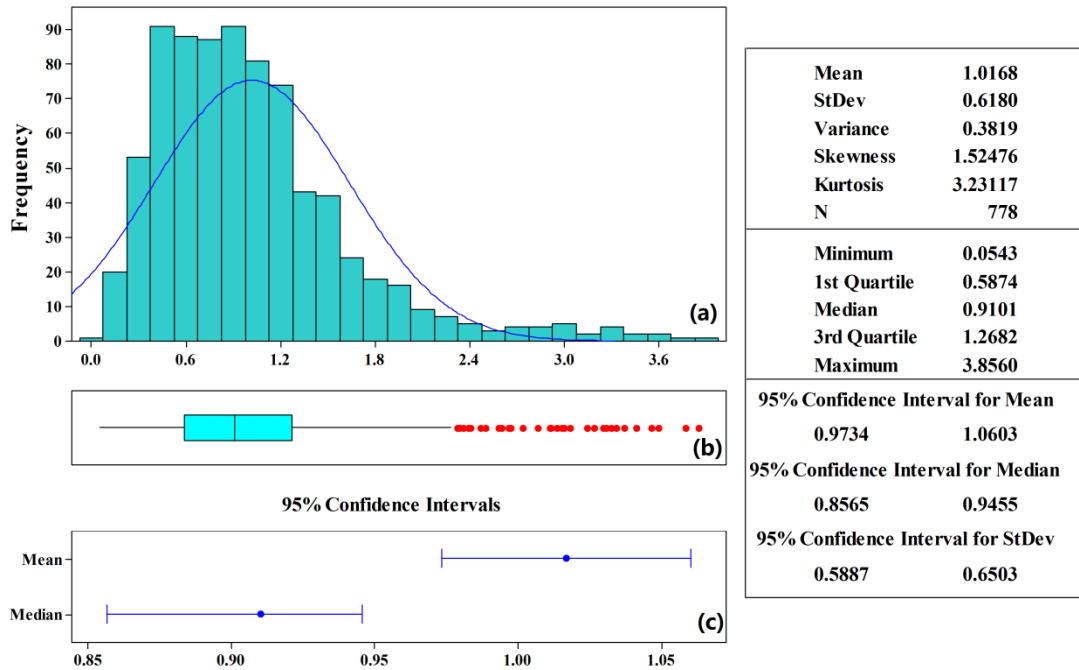

Figure 9. Statistical analyses of $PM_{2.5}$ pseudo-density (g/cm^3) during the study period. Figure (a) is the histogram of $PM_{2.5}$ pseudo-density, (b) is the boxplot of $PM_{2.5}$ pseudo-density, and (c) is the interval plot for mean and median of $PM_{2.5}$ pseudo-density.

4.3 $PM_{2.5}$ retrieval results and validation

To illustrate the outcomes of the $PM_{2.5}$ retrieval model proposed in this study, we

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443   used two retrieval results as examples. Figure 10 shows estimated $PM_{2.5}$

444   concentrations on 6 January 2015. It can be seen that the AOT retrieved using the

445   SARA ranged from 0.05 to 0.27, and its highest values were always centered in the

446   south of Beijing (Figure 10A). The FMF derived from the LUT-SDA presented high

447   values in central and south-eastern urban areas that have continually high traffic flow

448   and dense populations (Figure 10B). These areas with high FMF indicate a significant

449   contribution of fine particles to the total AOT. The PBLH and RH produced by the

450   WRF are shown in Figure 10 C and E. There is a marked difference in the spatial

451   coverage of PBLH on this day (There is also a significant difference in the spatial

452   distribution of PBLH in other study days, as shown in Figure S1-S4). In the north of

453   Beijing, PBLH reached 605–746 m, and in most central areas was more than 474 m,

454   whereas in the east and west of Beijing, PBLH was 366–417 m. Figure 10E shows

455   that the RH decreased gradually from the north-west (0.29–0.33) to the south-east

456   (0.18–0.21) of Beijing. The f (RH) corresponding to RH is shown in Figure 10D. It

457   can be seen that its spatial pattern is similar to that in Figure 10E. Finally, the derived

458    $PM_{2.5}$ is presented in Figure 10F. The spatial distribution pattern of $PM_{2.5}$ spatial

459   distribution pattern is similar to that of AOT shown in Figure 10A. High levels of

460    $PM_{2.5}$ in southern and eastern urban areas are clearly visible, indicating that

461   anthropogenic pollution was heavy in these areas. However, the $PM_{2.5}$ mass

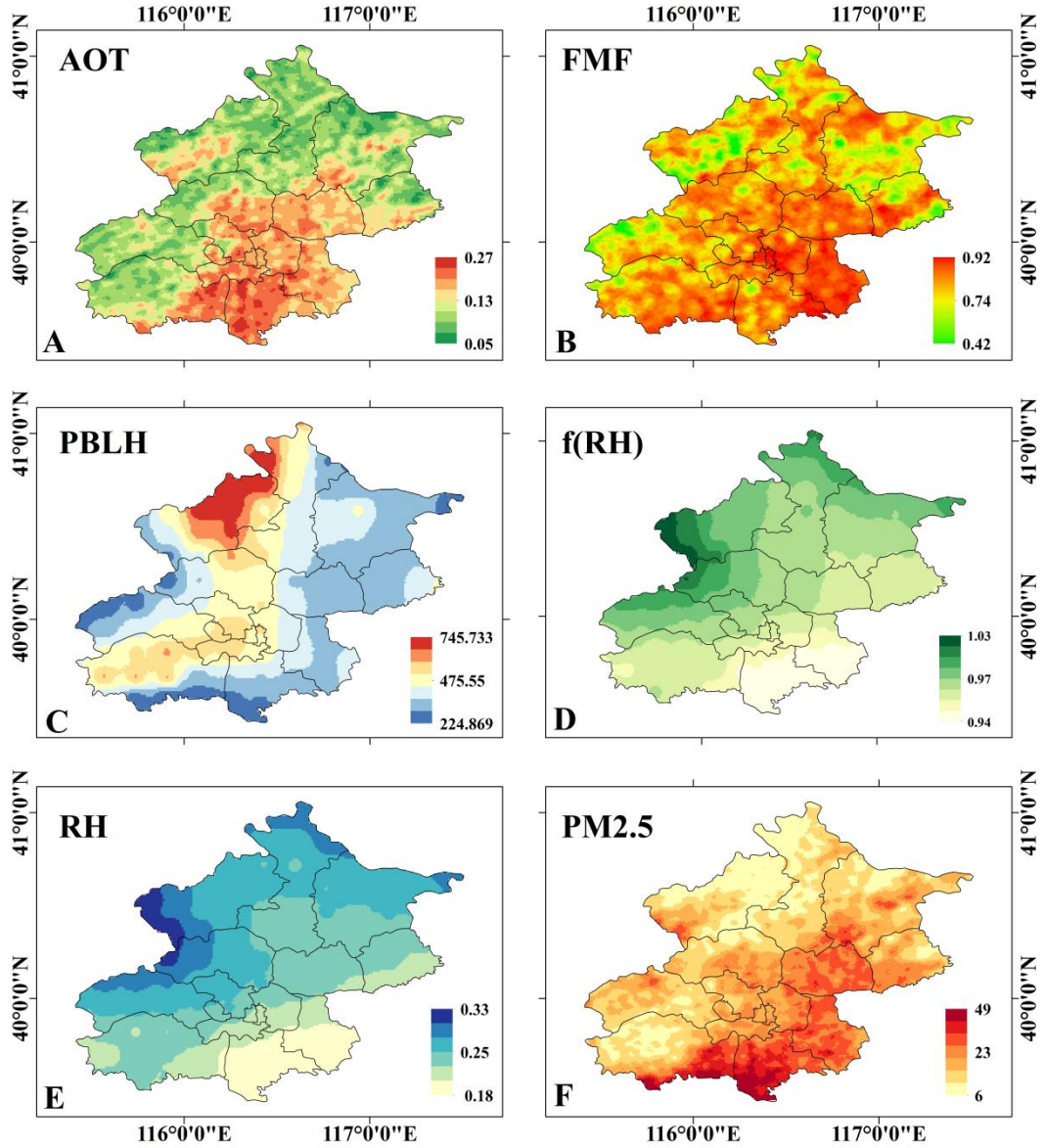462   concentration was low (6-23 μg/m^3) in northern rural areas.

463

To test the performance of the PM$_{2.5}$ retrieval model developed in this study, we extended the PM$_{2.5}$ retrieval to the areas surrounding Beijing. The test data were obtained on 16 October 2014 and the derived results are shown in Figure 11. High AOT was observed in the south of Beijing, east of Tianjin, and south-west and east of Hebei (Figure 11A). From Figure 11B, it can be seen that FMF values were high in

475 the center of Beijing, south of Hebei and east of Tianjin. The PM$_{2.5}$ retrieval results

476 are shown in Figure 11C. It can be seen that high-PM$_{2.5}$ polluted regions occurred in

477 the south of Beijing and Hebei, and also in the east of Tianjin. These results are

478 consistent with Zhang and Li (2015), Ma et al. (2014), and Zheng et al. (2016).

479 Comparing Figure 11 A and C, the derived PM$_{2.5}$ has a similar spatial pattern to AOT,

480 but with some differences due to the incorporation of FMF, RH, and PBLH

481 corrections, such as in the area of Beijing where the AOT was low, but the PM$_{2.5}$

482 concentration was high.
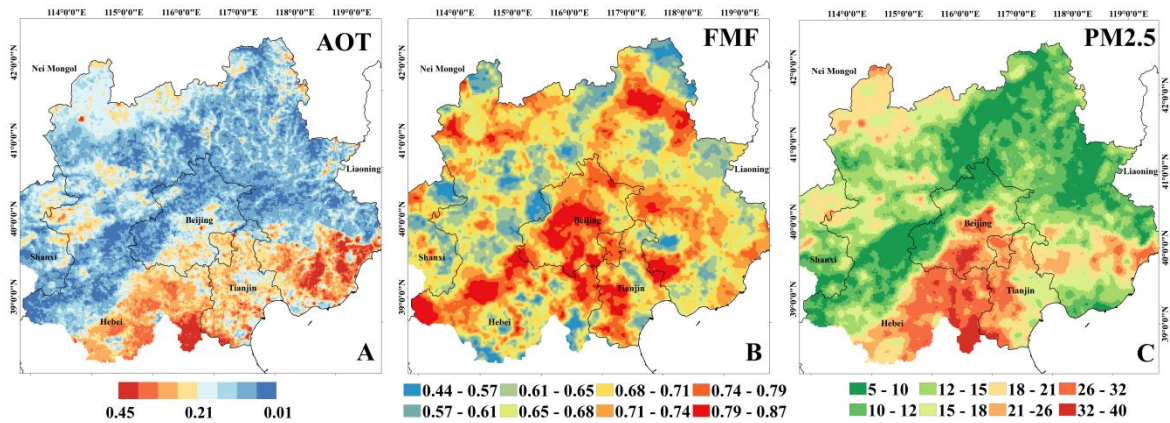
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485 Figure 11. Extension of PM$_{2.5}$ retrieval to the Beijing neighborhood. A is the AOT

486 retrieved from SARA, B is the FMF by LUT-SDA, and C is the estimated PM$_{2.5}$ by Eq.

487 (1) (units: μg/m^3).

488

489 Figure 12A shows the mean PM$_{2.5}$ concentration from satellite-retrieved data and

490 ground-based measurements from December 2013 to June 2015. High PM$_{2.5}$ levels

491 can be seen clearly in the southern urban region. PM$_{2.5}$ concentrations in these areas

492 were 42–44 μg/m^3 from satellite retrieval, and 42–48 μg/m^3 from ground-based

493 measurements. In contrast, PM$_{2.5}$ levels were significantly lower in the rural areas

494 north and west of Beijing, where the population is sparse and there are fewer vehicles.

495 Figure 12A shows that there is close agreement between the satellite-retrieved data

496 and *in situ* measurements, and the derived PM$_{2.5}$ spatial distribution was similar to

497 that reported by Li et al. (2015). Figure 12B presents the validation results of derived

PM$_{2.5}$ with 921 *in situ* PM$_{2.5}$ measurements. Linear regression gave a slope of 0.67 and an intercept of 9.5, with a coefficient of determination (R^2) of 0.67 (N = 921) and RMSE of 18.9 μg/m^3. The validation results showed that the derived PM$_{2.5}$ agreed closely with *in situ* observations, indicating that the retrieval model performed well.
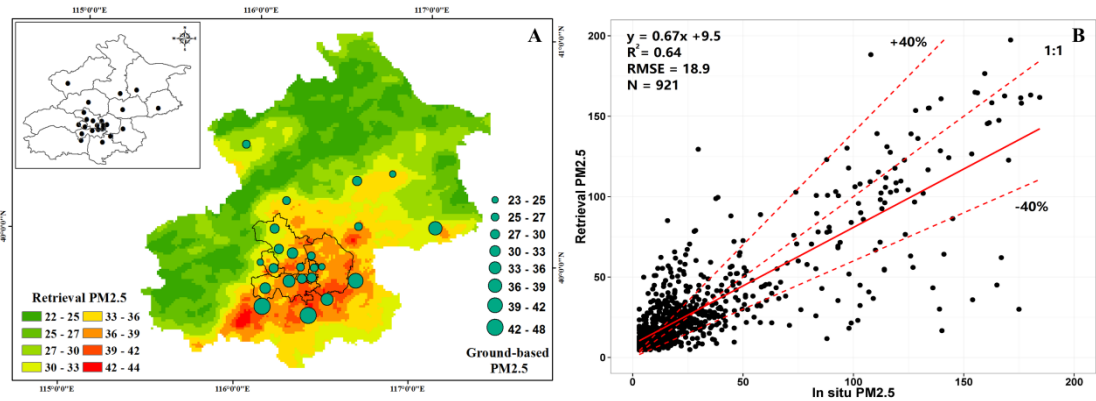

Figure 12. Mean PM$_{2.5}$ from December 2013 to June 2015. (A), and validation (B). Units: μg/m^3. Dotted red lines show the estimated error envelope line $\pm 0.4 \cdot PM2.5_{in\ situ}$, and the solid red line shows the 1:1 line.

4.4 Discussion

In this study, we found that FM-AOT was more closely correlated than total AOT with ground-level PM$_{2.5}$ concentration. In contrast, Wang et al. (2015) reported a significantly lower correlation when total AOT was replaced by FM-AOT for PM$_{2.5}$ estimation. This inconsistency may be because the PM$_{2.5}$ and AOT data were not collected from the same locations: the straight-line distance between the sites where the data on the two variables were collected was about 3,000 m in Wang et al. (2015). However, in this study, the PM$_{2.5}$ and AOT data were collected from the exact same location. For the comparisons of FMF between MODIS, LUT-SDA and AERONET, we observed that the MODIS FMF data had significant uncertainties and large amounts of data were unavailable. Because of this issue, the MAE of the PM$_{2.5}$ retrieval results in Zhang and Li (2015) was 64 μg/m^3 using MODIS FMF, which is quite large. One possible reason for this is that the relationship between VE$_f$ and FMF is based on the inversion method AERONET data in Eq (2); however, the FMF from

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the MODIS C6 product is derived using a different method and has a different definition. There can be highly significant uncertainties in the VE_f calculation using the MODIS C6 FMF relative to the AERONET FMF. Therefore, this can be a significant limitation for the application of Eqs. (1) and (2). To address this problem, we used the LUT-SDA FMF in this study, which is more reliable, as shown in Figure 7. In addition, assuming a constant density is inappropriate for real-time $PM_{2.5}$ estimation due to distinct diurnal and seasonal variations (Liu et al., 2015). Hoff and Christopher (2009) suggested that satellite AOT should be combined with ground-based measurements to improve PM estimation from space. Therefore, using VIS data from weather stations, the intraday pseudo-density was calculated for each satellite image. Although this provides only rough density estimation, it is important for real-time surface $PM_{2.5}$ retrieval and produces an accurate $PM_{2.5}$ with a RMSE of 18.9 μg/m^3.

5. Conclusion

Based on ground-based measurements obtained in Xingtai city, China during May–June 2016, surface Particulate Matter$_{2.5}$ (PM$_{2.5}$) was found to have a closer correlation with the Fine Mode-Aerosol Optical Thickness (FM-AOT) (r = 0.74) than with the total AOT (r = 0.49). We established a $PM_{2.5}$ retrieval model incorporating look-up table–spectral deconvolution algorithm (LUT-SDA) fine mode fraction (FMF) (Yan et al., 2017), SARA AOT (Bilal et al., 2013), and the $PM_{2.5}$ remote sensing method (Zhang and Li, 2015). Compared with Aerosol Robotic Network (AERONET) data, the LUT-SDA FMF was found to be more reliable than the MODIS FMF. The mean absolute errors (MAEs) between AERONET FMF and LUT-SDA FMF were 0.146 in RADI, 0.149 in Beijing, and 0.159 in CAMS. Due to significant diurnal variation in the density of $PM_{2.5}$, we proposed a pseudo-density to enhance $PM_{2.5}$ estimation based on real-time visibility (VIS) data. The standard deviation of the $PM_{2.5}$ pseudo-density was 0.62, which also varied diurnally due to changing weather and pollution conditions; this finding is consistent with previous studies (Zhao et al. 2013; Wang et al., 2014; Liu et al., 2015). The performance of the ground-level $PM_{2.5}$

retrieval model developed in this study was tested over Beijing from December 2013 to June 2015. The retrieval results compared to 30 *in situ* sites showed that the model performed well ($R^2 = 0.64$, root mean square error (RMSE) = 18.9 μg/m^3 [N = 921]). Furthermore, the retrieval results demonstrated that the developed model was able to monitor air pollutants in the Beijing regions with full spatial coverage and temporal resolution.

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