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Remote sensing of PM2.5 during cloudy and nighttime periods using ceilometer backscatter

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Abstract. Monitoring PM2.5 (particulate matter with aerodynamic diameter d≤2.5 μm) mass concentration is of more importance recently because of the negative impacts of fine particles on human health. However, monitoring PM2.5 during cloudy and nighttime periods is difficult since nearly all the passive instruments used for aerosol remote sensing are not able to measure aerosol optical depth (AOD) under either cloudy or nighttime conditions. In this study, an empirical model based on the regression between PM2.5 and the near surface backscatter measured by ceilometers was developed and tested using six years of data (2006 to 2011) from the Howard University Beltsville Campus (HUBC) site. The empirical model can explain ~ 56%, ~34%, and ~42% of the variability in the hourly average PM2.5 during daytime clear, daytime cloudy and nighttime periods respectively. Meteorological conditions and seasons were found to influence the relationship between PM2.5 mass concentration and the surface backscatter. Overall the model can explain ~48% of the variability in the hourly average PM2.5 at the HUBC site when considering the seasonal variation. The model also was tested on the four years of data (2012 to 2015) from the ARM SGP site which was geographically and climatologically different from the HUBC site. The results show that the empirical model can explain 67% and 83% of the variability in the daily average PM2.5 at the ARM SGP site and HUBC site respectively. The findings of this study illustrate the strong need for ceilometer data in air quality monitoring under cloudy and nighttime conditions. Since ceilometers are used broadly over the world, they may provide an important supplemental source of information of aerosols to determine surface PM2.5 concentrations.

Key words. PM2.5, nighttime, cloudy, remote sensing, ceilometer, air pollution

1 Introduction

The adverse impacts of high PM2.5 (particulate matter with aerodynamic diameter d≤2.5 μm) mass concentration on human health have been found from epidemiological studies around the world (Samet et al., 2000; Pope et al., 2009; Krewski et al., 2009). PM2.5 concentration was found to be associated with cardiopulmonary disease, lung cancer, and an increased morbidity and mortality (Schwartz et al., 1996; Gent et al., 2003, 2009; Dominici et al., 2006; Bell et al., 2007; Franklin et al., 2007; Slama et al., 2007; Pope et al., 2002; Miller et al., 2007; Lepeule et al., 2012). As an official norm to stand for fine particle abundance, PM2.5 mass concentrations are monitored widely by the US Environmental Protection Agency (EPA) through in situ instruments at surface monitoring sites. However, the number of EPA monitoring sites are limited. Therefore, remote sensing of PM2.5 from ground stations and satellites is desirable allowing for fuller coverage of PM2.5 concentration between the EPA surface sites.

Aerosol optical depth (AOD) plays an important role in the remote sensing of PM2.5 since it has a good relationship with PM2.5 concentration. However, most measurements of AOD which are derived from passive remote sensing techniques are only available under daytime and clear sky conditions. Remote sensing of PM2.5 during either cloudy or nighttime periods are very rare. Different from passive instruments which measure column integrated AOD, active instruments like advanced lidars have the capacity to provide the vertical distribution of aerosol backscatter coefficient even under cloudy conditions or the nighttime. However advanced lidar networks are rare due to the complexity and cost. Instead, ceilometers which are simple, automatically operating single wavelength lidars are used broadly all over the world. Ceilometers were originally developed for cloud based height retrieval. With the improvement of accuracy and power, the potential capabilities of ceilometers on detecting mixing layer height and aerosol optical properties have been explored recently (Münkel et al., 2007; Markowicz et al., 2008; Heese et al., 2010; Tsaknakis et al., 2011; Wiegner et al., 2012). Another distinct advantage of ceilometers is their small overlap distance which makes it suitable to detect aerosol information near the surface. PM2.5 concentration is an index of fine particle mass concentration near the surface while AOD is the integration of aerosol extinction in the total atmospheric column. So, using aerosol backscatter near the surface has an inherent advantage in the remote sensing of PM2.5 concentration.

There are extensive studies investigating PM2.5-AOD relationship either by the use of empirical statistical method (Engel-Cox et al., 2004; Liu et al., 2005, 2009; Gupta et al., 2006; Koelemeijer et al., 2006; Gupta and Christopher, 2008; Paciorek et al., 2008; Di Nicolantonio et al., 2009; Schaap et al., 2009; Lee et al., 2012; Sorek-Hamer et al., 2013; Strawa et al., 2013; Chudnovsky et al., 2014; Hu et al., 2013, 2014; Ma et al., 2014) or a chemical transportation model (Liu et al., 2004; Van Donkelaar et al., 2006, 2010; Kessner et al., 2013; Xu et al., 2015). In these studies, aerosol vertical distributions are estimated based on model simulation or under an assumption that aerosols are well mixed within the boundary layer and then decrease exponentially with height . Recently Li et al., (2016) developed an algorithm combining the backscatter measured from ceilometers with AOD for the PM2.5 retrieval. That work showed the capability of the ceilometer on improving PM2.5 estimation by introducing measurements of aerosol optical properties near the surface. Although there are a plenty of studies on PM2.5 estimation, studies on the remote sensing of PM2.5 during either cloudy or nighttime periods are rare due to the limitation of measurements of AOD.

In this study, to estimate PM2.5 under cloudy or during night periods, we developed a regression model based on the relationship between PM2.5 and the ceilometer backscatter under different meteorological conditions. The model is tested and validated against the 6 years (2006-2011) ground-based observations of ceilometer backscatter, PM2.5, AOD and meteorological conditions at the Howard University Beltsville Campus (HUBC) site and the 4 years (2012-2015) data from the Atmospheric Radiation Measurement (ARM) Southern Great Plains (SGP) site. The data and model are described in section 2. The results of the testing and evaluation of the model are illustrated in section 3. The discussion is given in the last section..

2 Data and model

2.1 Data

In this study, the data was obtained from the HUBC site in Beltsville, MD which is situated in a rural-suburban transition region between Washington, DC and Baltimore, MD urban centers. The site has a wide range of collocated instruments to observe atmospheric radiation, aerosol, cloud properties, meteorological conditions and air quality (Li et al., 2016), which makes the HUBC site suitable for PM2.5 investigations.

The backscatter data was provided by a Vaisala CT25k ceilometer which is a single-lens lidar system equipped with a pulsed near-infrared diode laser (905 nm). As a commercial ceilometer, the CT25k provides a range-corrected attenuated backscatter coefficient but the raw data is not available to the customer, which limits the access of the correction process. However, it was shown that the signal reduction due to the near-field problem was compensated well with the manufacturer's correction (Markowicz et al., 2008). The unique single-lens design gives full overlap of the transmitter and receiver field-of-view at an altitude of 0 m (Münkel et al., 2007), which allows CT25k ceilometers to obtain high signal-to-noise ratio for lidar return signals at a low altitude. The working wavelength of CT25k ceilometers is ~905nm where water vapor absorption exists (Wiegner et al., 2014; Wiegner and Gasteiger, 2015). However, water vapor impacts on backscatter retrieval is smaller than ~2% for 905 nm ceilometers under mid-latitude climatology (Wiegner and Gasteiger, 2015) within a short distance from the surface to the height of 150 m. Given the small attenuation within a short distance, the attenuated backscatter coefficient below 150 m can be reasonably taken as a measure of backscatter coefficient when there is no rain or fog. The vertical resolution of the CT25k is 30 m. Since we are interested in the PM2.5 concentration near the surface , we only use the first 5 layer backscatter measurements from the CT25k ceilometer to estimate PM2.5 concentrations. The choosing of 150 m is arbitrary but the sensitivity test showed that the retrieval results are quite similar for the different heights from 90 m to 300 m(Li et al., 2016).

The near surface meteorological conditions including temperature, relative humidity, pressure, wind speed and wind direction are provided by a nearby 31m micrometeorological tower and the AOD observations and cloud optical depth (COD) are retrieved from a MultiFilter Rotating Shadowband Radiometer (MFRSR). The details of the MFRSR and the corresponding retrieval algorithms are introduced in Harrison et al. (1994), Harrison and Michalsky (1994) and Min and Harrison (1996). The hourly average PM2.5 are measured by a Met One BAM-1020 (beta ray attenuation monitor) from the collocated Maryland Department of the Environment (MDE) monitor station (Li et al., 2016).

In this study, hourly average data was used for all the data sets. Precipitation and fog cases were screened out by using cloud effective radius larger than 15 μm, microwave radiometer measured liquid water path larger than 200 g/m2, ceilometer derived cloud layer lower than 200 m and relative humidity larger than 95%.

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2.2 Model

For a ceilometer, the energy observed is a function of backscattering coefficient

$$P(x) = \frac{P_0 A\eta O(x) C\Delta t}{2x^2}\beta(x)T^2(x). \tag{1}$$

Where $P(x)$ and P_0 are the received and emitted powers from a ceilometer, A and η are the area of the receiver and its efficiency respectively, and x is the range from receiver to scattering volume. $O(x)$ is overlap function, C is light speed, Δt is the laser pulse duration and $T(x)$ is the transmittance of the atmosphere between receiver and scattering volume. $\beta(x)$ is the backscattering coefficient which can be separated into two components

$$\beta(x) = \beta^m(x) + \beta^a(x). \tag{2}$$

Where $\beta^m(x)$ and $\beta^a(x)$ denote the backscattering by molecules and aerosols respectively. The aerosol backscattering can be derived from the total backscattering coefficient as the molecules scattering is well modelled by Rayleigh scattering. For the backscattering at the near-infrared wavelength, the contribution from molecules can be disregarded due to the rapidly decreased Rayleigh scattering with wavelength, so $\beta(x)$ is taken as $\sim\beta^a(x)$ in this study.

With the assumption that aerosol size distribution is bimodal lognormal and aerosol particles are spherical, Li et al., (2016) illustrated that both the extinction and PM2.5 can be expressed in terms of particle volume concentration ($c\, v_i$) for each mode as

$$ext(\lambda) = \sum_{i=1}^{2} c\, v_i\, h(R_i, \sigma_i, m, \lambda) \tag{3}$$

$$PM2.5 = \sum_{i=1}^{2} c\, v_i\, g(R_i, \sigma_i, \rho). \tag{4}$$

Where $h(R_i, \sigma_i, m, \lambda)$ and $g(R_i, \sigma_i, \rho)$ are the integral functions of volume concentration normalized aerosol size distribution, c is the total particle volume concentration, v_i is the fraction of volume concentration for each mode i, R_i, and σ_i are the geometric mean radius and the standard deviation of aerosol size distribution, respectively, λ is the wavelength, m is the refractive index, and ρ is the particle mass density. The relationship between the aerosol backscattering coefficient $\beta^a(\lambda)$ and the extinction coefficient $ext(\lambda)$at the wavelength λ is usually expressed by a lidar ratio (K)

$$K = \frac{ext(\lambda)}{\beta^a(\lambda)} \tag{5}$$

From the Eq. (3), (4), and (5) the relationship between $\beta^a(\lambda)$ and PM2.5 can be expressed by

$$PM2.5 = F\, \beta^a(\lambda). \tag{6}$$

Where

$$F = K\frac{\sum_{i=1}^{2} v_i f(R_i, \sigma_i, m, \lambda)}{\sum_{i=1}^{2} v_i g(R_i, \sigma_i, \rho)}. \tag{7}$$

The PM2.5-backscatter ratio F only depends on aerosol size and composition. Given that the variation of aerosol size and composition could be associated with the meteorological conditions and the assumption that aerosols mixed well near the surface, an empirical model based on relationship between PM2.5 and the backscatter near the surface is proposed as

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$$PM_{2.5} = a_0 + (a_1 + a_2\, f(RH) + \sum_{i=1}^{n} a_{2+i}\, M_i) \left(\int_0^z \beta(x,\lambda) dx\right)^{b_2} + \varepsilon, \tag{8}$$

Where the hygroscopic grow factor is expressed as

$$f(RH) = \frac{1}{(1-RH)^{b_1}},$$

RH is relative humidity, M_1 through M_{n} are the meteorological factors including surface temperature, wind speed, wind direction and surface pressure, z is height and a_0 through a_{2+n}, b_1 and b_2 are the regression coefficients, ε is the error term. In the following part, we will test the model performance without considering the meteorological variables. In that case, the Eq. (8) can be expressed as

$$PM_{2.5} = a_0 + a_1 \left(\int_0^z \beta(\mathrm{x},\lambda)\mathrm{dx}\right)^{b_1} + \varepsilon, \tag{9}$$

When we test the model including the impacts from observations of surface temperature (T), relative humidity (RH) and wind speed (W),the Eq. (8) can be expressed as

$$PM_{2.5} = a_0 + \left(a_1 + a_2 * \frac{1}{(1-RH)^{b_1}} + a_3\, T + a_4 * W\right) \left(\int_0^z \beta(\mathrm{x},\lambda)\mathrm{dx}\right)^{b_2} + \varepsilon. \tag{10}$$

3 Results

To test and evaluate the model, cross-validations are implemented on the 6 years of hourly average measurements at the HUBC site under the different conditions including daytime clear, daytime cloudy and nighttime periods. For the cross-validation, we randomly select 90% of the data as a training dataset and used the remaining 10% to test the modals and repeated the procedure for 100 times to avoid random bias and misleading R^2 induced by overfitting. Cross-validations are conducted for each model under each condition.

3.1 Simulation results under different sky conditions

Under daytime clear sky conditions when AOD measurements from the MFRSR are available (no cloud, daytime), the average cross-validation (CV) R^2 out of the 100 times random cross-validations for the model (Eq. 10) is 0.56 (figure 1) with RMSE is 6.12 $\mu\mathrm{g/m^3}$. This result is close to that of the non-linear model which combines both AOD and the ceilometer backscattering ($CVR^2 = 0.60$, RMSE =5.83 $\mu\mathrm{g/m^3}$) developed by Li et al. (2016) and performed much better than that of the model using AOD only ($CVR^2 = 0.40$, RMSE $= 7.14\ \mu\mathrm{g/m^3}$) (Li et al., 2016). Without considering the meteorological conditions (Eq. 9), the average CVR^2 of the model is 0.45 (figure 2) which is better than that of the model using AOD only (Li et al., 2016) but not as good as the model including meteorological variables. With the parameters (table 1, 2) from the best fitting out of the 100 independent cross-validations (10% of the total data), the correlation coefficient between all the in situ measured PM2.5 under daytime clear sky conditions and the simulated PM2.5 from the model without meteorological variables is 0.68 and increased to 0.76 when meteorological variables were included (Eq. 10).

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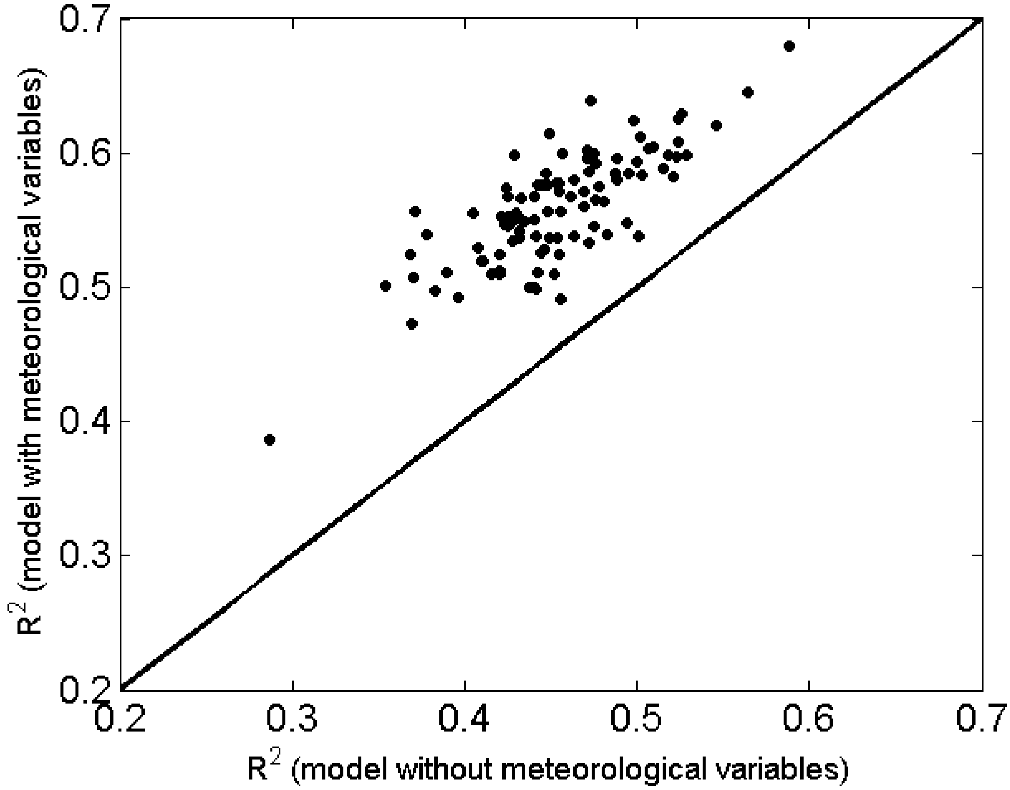

Figure 1: Comparison of R-squared out of the 100 independent cross-validations for the model without meteorological variables (Eq. 11) and the model with meteorological variables (Eq. 12) based on all the available daytime clear sky cases at the HUBC site.

Remote sensing of AOD is commonly based on the measurements of spectral extinction of solar radiation due to aerosol scattering and absorption in the atmospheric column from passive instrument. However most passive instruments cannot readily discern AOD from COD under cloudy conditions. So, any PM2.5 remote sensing method relying on passive AOD measurements cannot retrieve PM2.5 under cloudy conditions. However, measurements of backscatter under cloudy conditions are still available for ceilometers, which can help to determine the near surface aerosol extinction when the upper layer clouds exist.

Under daytime cloudy conditions, the average CVR^2 of the model without meteorological variables is only 0.11 (figure 2) which means only around 11% of the variability in the hourly PM2.5 can be explained by the model. When meteorological factors are considered, the model can explain 34% of the variability. With the parameters based on the best fitting of the 100 independent cross-validations (table 1, 2), the correlation coefficient between all the in situ measured PM2.5 under daytime cloudy conditions and the simulated PM2.5 from the model without meteorological variables is only 0.34 and it is improved to 0.59 when meteorological variables were included in the model.

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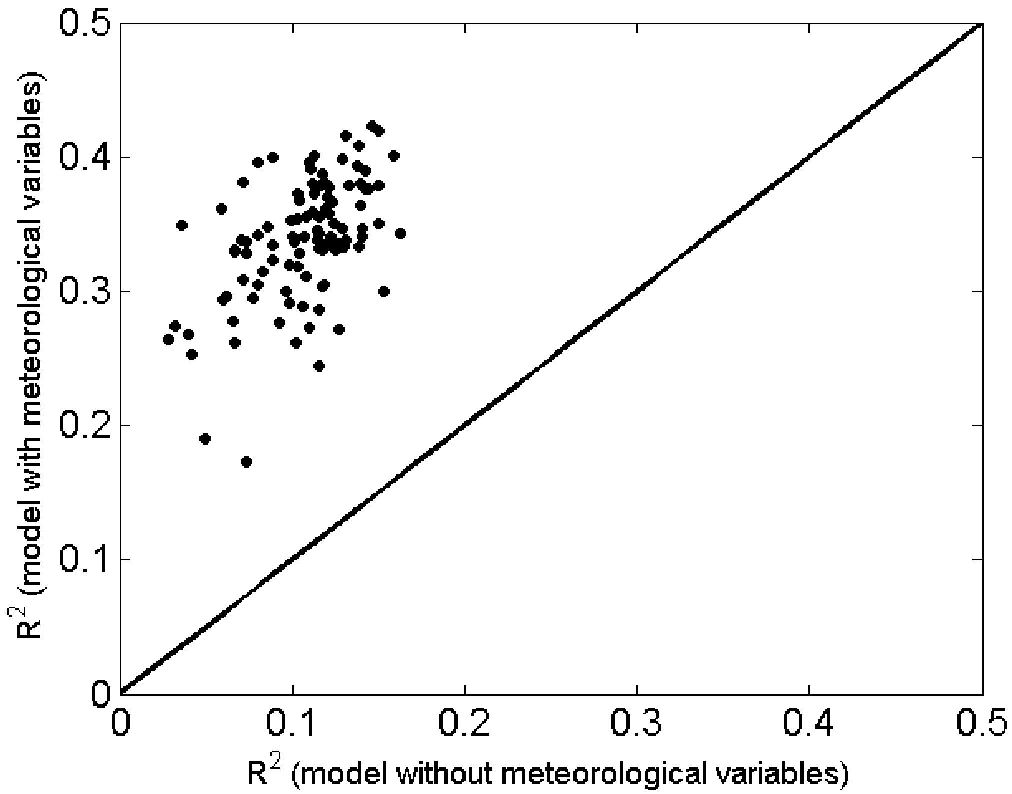

Figure 2: Same as figure 1 but just for cases under daytime cloudy conditions.

During nighttime periods, passive measurement relying on solar radiation is not available but active instruments like ceilometers are still able to measure regardless of solar radiation and have better signal-to-noise ratio because of the absence of background sunlight contamination. During nighttime periods, the average CVR^2 out of the 100 independent cross-validations for the model without meteorological variables is 0.21 while the average CVR^2 for the model with meteorological variables is 0.42 (figure 3). In this study, measurements under clear sky and cloudy sky were not separated during nighttime periods. With the parameters based on the best fitting of the 100 independent tests (table 1, 2), the correlation coefficient between all the in situ measured PM2.5 during nighttime and the simulated PM2.5 from the model without and with meteorological variables were 0.47 and 0.65, respectively.

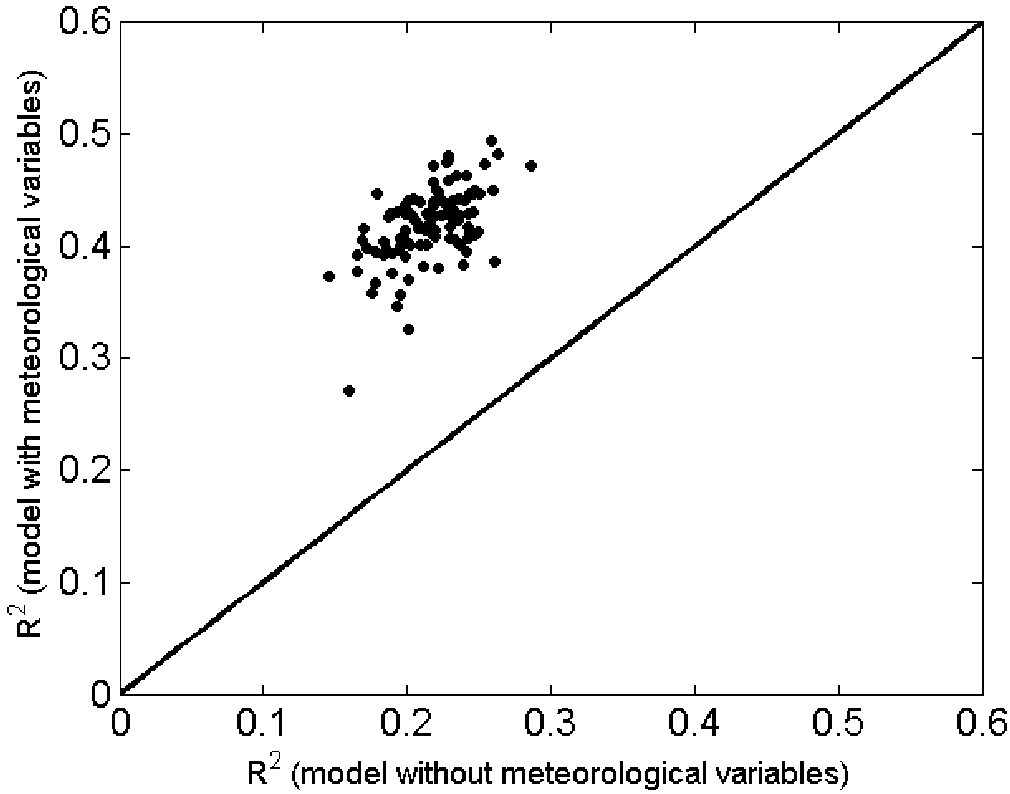

Figure 3: Same as figure 1 but just for cases during nighttime periods.

3.2 Impacts from meteorological variables

The previous results showed that without considering meteorological factors the model predicting ability largely decreased, especially under cloudy and nighttime conditions. Remote sensing of PM2.5 using backscattering coefficients is based on the relationship between PM2.5 and aerosol backscatter which is determined by aerosol physical and chemical properties. Aerosol physical and chemical characteristics are sensitive and dependent on meteorological conditions that can impact aerosol transportation, hygroscopic growth, and aerosol nucleation/creation. Therefore, meteorological conditions can be potentially used to estimate aerosol characteristics when the direct observations are not available. So taking into account the variations of meteorological conditions may largely improve the model which is based on the regression between PM2.5 and backscattering coefficients.

To investigate impacts from different meteorological factors on PM2.5 remote sensing, the relationship between each meteorological variable and PM2.5-backscatter ratio were analyzed in three data categories: daytime clear (AOD measurements are available), daytime cloudy and nighttime (figure 4-7). Among the meteorological variables, temperature

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was found to have the most prominent positive correlation with the PM2.5-backscatter ratio. The correlation coefficients equal to 0.4, 0.46 and 0.29 under daytime clear, daytime cloudy and nighttime conditions respectively (figure 4). In the eastern United States, the sulfate dominates the aerosol chemical composition (Hand et al., 2012) and the sulfate concentrations are expected to increase with increasing temperature due to faster SO2 oxidation. Fine particles have smaller backscatter coefficient due to the smaller size index based on the Mie theory (Wiscombe, 1980) compared to larger particles with the same PM2.5 mass concentration. So at the HUBC site, the increase of temperature associated with the high PM2.5-backscatter ratio could be due to the increase of fine particles.

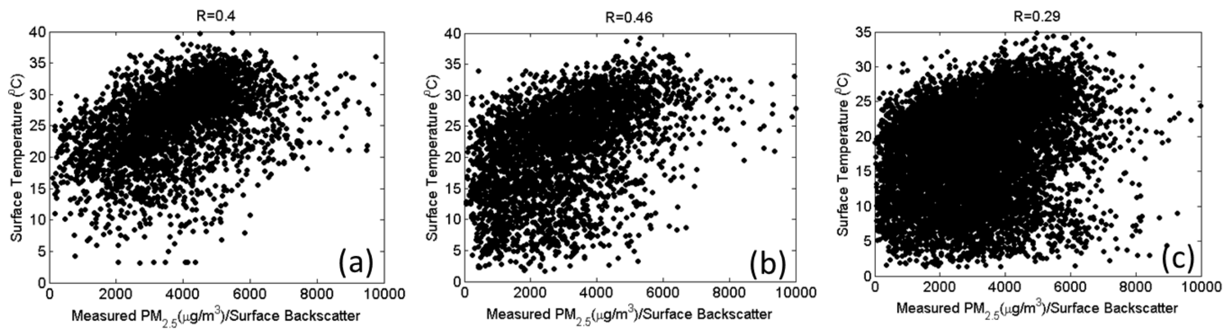

Figure 4. The relationship between surface temperature and PM2.5-backscatter ratio for (a) daytime clear sky cases, (b) daytime cloudy cases and (c) nighttime cases.

Opposite to the surface temperature, it is shown that the surface relative humidity had a prominent negative association with the PM2.5-backscatter ratio. The correlation coefficient equal to -0.12, -0.42 and -0.28 under the daytime clear, daytime cloudy and nighttime conditions respectively (figure 5). Under high relative humidity conditions there can be significant variations in the aerosol optical properties due to the aerosol hygroscopic growth effect. In the eastern United States, the dominant aerosols are composed of ammonium sulfate aerosols for which the ambient size will increase with the increase of the relative humidity due to hygroscopic growth. That can result in the decrease of the PM2.5-backscatter ratio due to the increase of the aerosol extinction cross-section while the aerosol dry mass is relatively invariant. It should be noted that the correlation coefficient is -0.12 for the cases under daytime clear conditions while it is -0.42 under the daytime cloudy condition. Chu et al., (2015) showed that the effect of hygroscopic growth on extinction is more prominent when the relative humidity is larger. Under the nighttime condition which includes both the clear and cloudy situations, the correlation coefficient is -0.28.

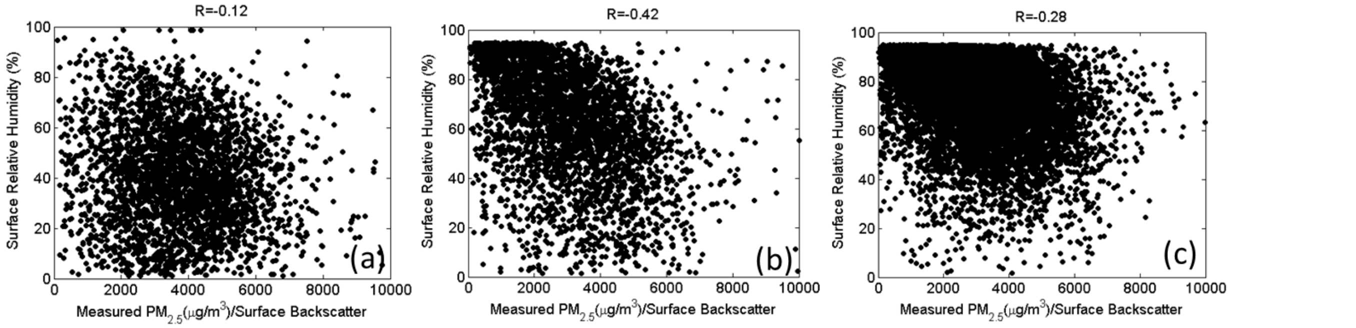

Figure 5. The relationship between surface relative humidity and PM2.5-backscatter ratio for (a) daytime clear sky cases, (b) daytime cloudy cases and (c) nighttime cases.

A negative association is also found between the wind speed and PM2.5-backscatter ratio under all the three conditions (figure 6). That may be explained by the association of higher PM2.5 concentrations with more stagnant, weaker wind conditions (Tai et al., 2010). Based on the averaged PM2.5-backscatter ratio at four wind direction ranges: east(315°to 45°), north(45°to 135°), west(135°to 225°), south(225°to 315°), the variation of the mean PM2.5-backscatter ratio at the four different wind directions was found to be small (within 10%) compared to the standard deviation (~50% of the mean value) at the HUBC site (figure 7). The association of the surface pressure with the PM2.5-backscatter ratio was found to be weak with the correlation coefficient equal to -0.05 (not shown). The distributions of PM2.5-backscatter ratio under the three conditions are shown in figure 8. Statistically, the PM2.5-backscatter ratio under daytime clear sky condition is larger than that under daytime cloudy or nighttime condition.

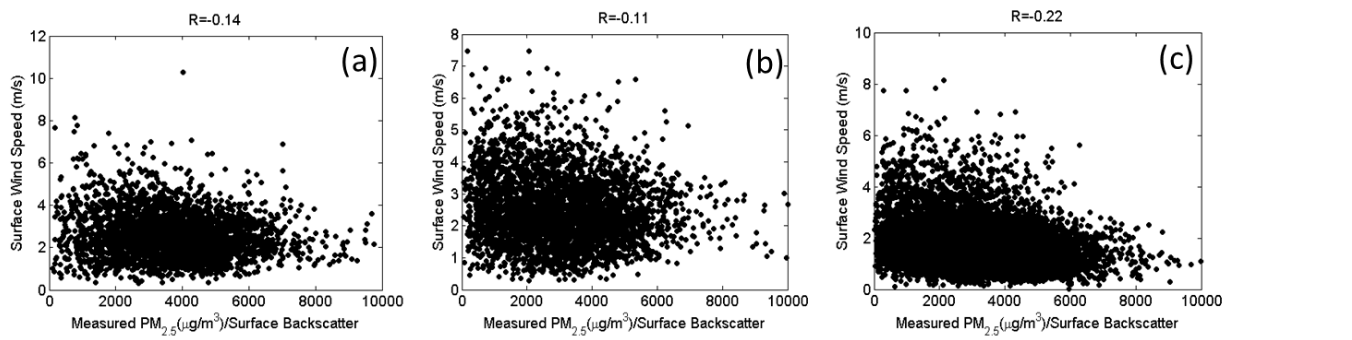

Figure 6. The relationship between surface wind speed and PM2.5-backscatter ratio for (a) daytime clear sky cases, (b) daytime cloudy cases and (c) nighttime cases.

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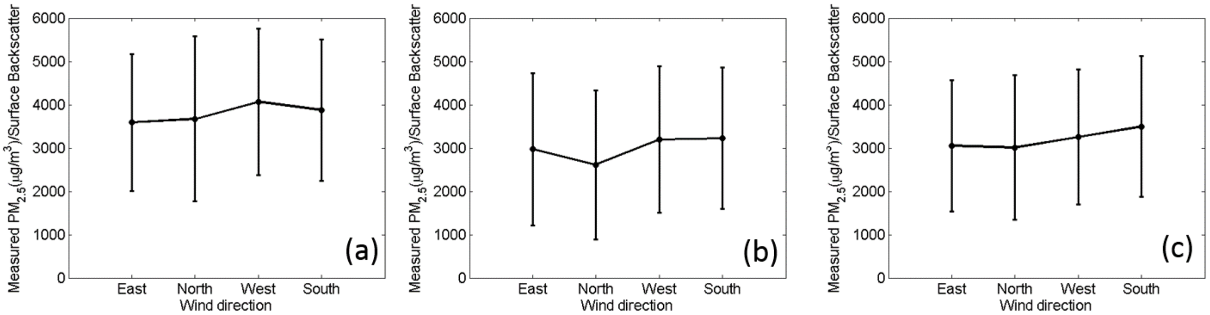

Figure 7. Average PM2.5-backscatter ratio with standard deviation at four direction ranges: east(315° to 45°), north(45° to 135°), west(135° to 225°), south(225° to 315°) for (a) daytime clear cases, (b) daytime cloudy cases and (c) nighttime cases.

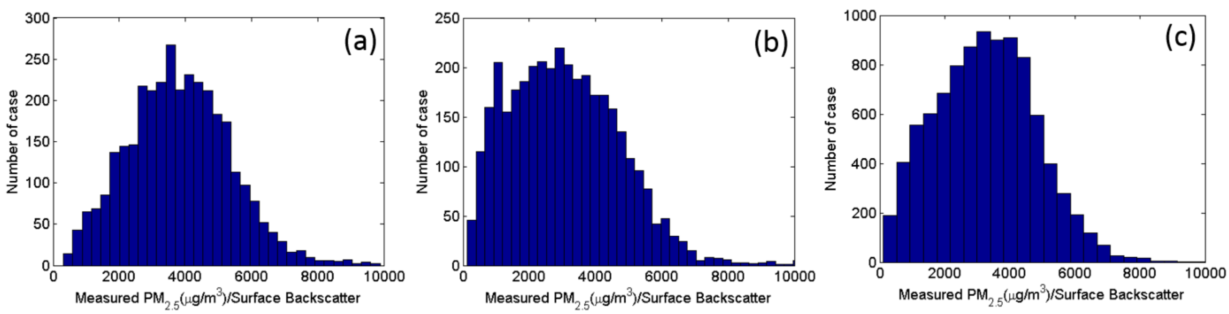

Figure 8. The number distribution of PM2.5-backscatter ratio for (a) daytime clear cases, (b) daytime cloudy cases and (c) nighttime cases.

Figures 4-7 showed the potential impacts of meteorological factors on model prediction. However, some information possibly overlapped among the different meteorological variables. To investigate the contribution of each meteorological variable on improving the model predicting power, the model was tested with different meteorological variable combinations. For each test, the cross-validation was randomly repeated 100 times based on all the available cases including the daytime clear, daytime cloudy, and nighttime periods.

Table 3 demonstrates the average CVR^2, RMSE, and the 95% confidence intervals for each test. It is shown that without the information of surface temperature, relative humidity, or wind speed the average CVR^2 of the model decreases from 0.43 to 0.37, 0.39 or 0.37, respectively. In other words, adding the variable of surface temperature, relative humidity, or wind speed in the model can bring in additional information which may improve the model prediction capability on PM2.5.

3.3 Seasonally fitting

Besides meteorological factors, the seasonal variations of aerosol physical and chemical properties could impact PM2.5-backscatter ratio and then PM2.5 retrievals. To investigate the impacts of seasonal variations on PM2.5 retrievals, we fit the model seasonally and compared that performance with the model fitted on all the data without considering seasonal variation. Same as previous section, the cross-validations were implemented for each test. The parameters of the best fitting out of the 100 independent cross-validations for each fitting method are used to calculate the correlation between the in situ measurements of PM25 and the simulated PM2.5. When meteorological variables were not considered, the simulated PM2.5 from the model with the seasonally fitted parameters had a much stronger association with the in situ measured PM2.5 (R=0.57) compared to the model with the non-seasonally fitted parameters (R=0.45)(figure 9). When meteorological variables were taken into account, the correlation coefficient between the simulation and the in situ measurements of PM2.5 for the model with the seasonally fitted parameters or non-seasonally fitted parameters are 0.69 and 0.65 respectively (figure 10) and the average CVR^2 are 0.48 and 0.43. The meteorological conditions have seasonal variation, so taking into account meteorological variables in the model can mitigate downside impacts of ignoring seasonal variations of aerosol properties on PM2.5 prediction.

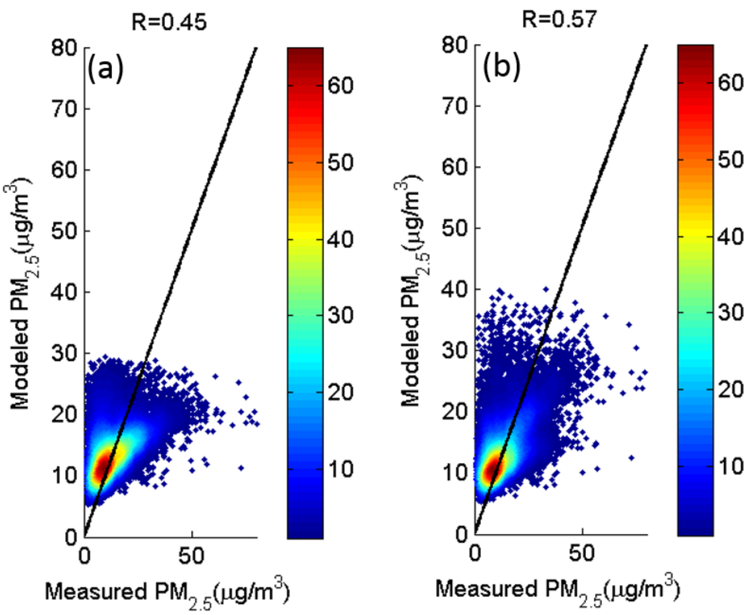

Figure 9. Comparison of measured PM2.5 and Modeled PM2.5 when meteorological variables are not taken into accounts. (a) The model is non-seasonally fitted and (b) the model is seasonally fitted. The colors stand for the number density of the points.

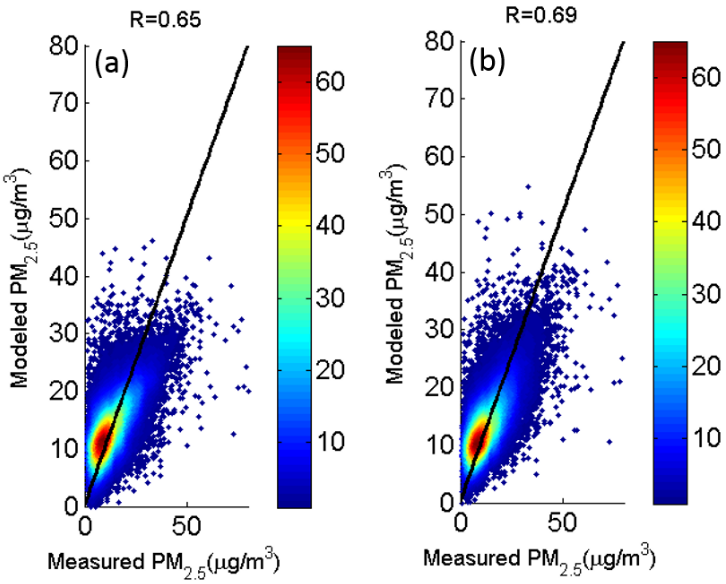

Figure 10. Same as figure 9 but all meteorological variables are taken into account.

3.4 Test in a different region

Given that aerosol types, compositions and meteorological conditions could be different in a different region, the model was tested based on the observations at the ARM SGP site which is located in Oklahoma, USA. The site is in a rural area with fewer anthropogenic aerosols compared to the HUBC and the DC area. The ARM SGP site is the largest and most extensive climate research field site in the world. In the test, we used the ceilometer backscatter and the surface meteorological conditions provided by the ARM SGP site and the FRM/FEM PM2.5 mass concentration from the nearest EPA site (36.697°N and 97.081 W) (Air Quality System Data Mart, available via http://www.epa.gov/airdata). The same cross-validation procedure was implemented on the measurements at the ARM SGP site under daytime clear, daytime cloudy and nighttime periods respectively. For the hourly average PM2.5, the cross-validation results (figure 11) show that the performance of the model with meteorological variables (Eq. 10) at the ARM SGP site were not as good as that of the HUBC site but the model without meteorological variables (Eq. 09) performed better at the ARM SGP site than at the HUBC site during daytime cloudy and nighttime periods. That could be due to the different aerosol type and composition which are associated with the hygroscopic growth of aerosols at the SGP area and the DC area. For the daily average PM2.5, the model (Eq. 12) can explain 83% and 67% of the variability in daily average PM2.5 at the HUBC site and ARM SGP site respectively (figure 12) with the fitted parameters from the best fitting out of the 100 independent cross-validations. Overall, the regression model using ceilometer backscatter performed well at both sites.

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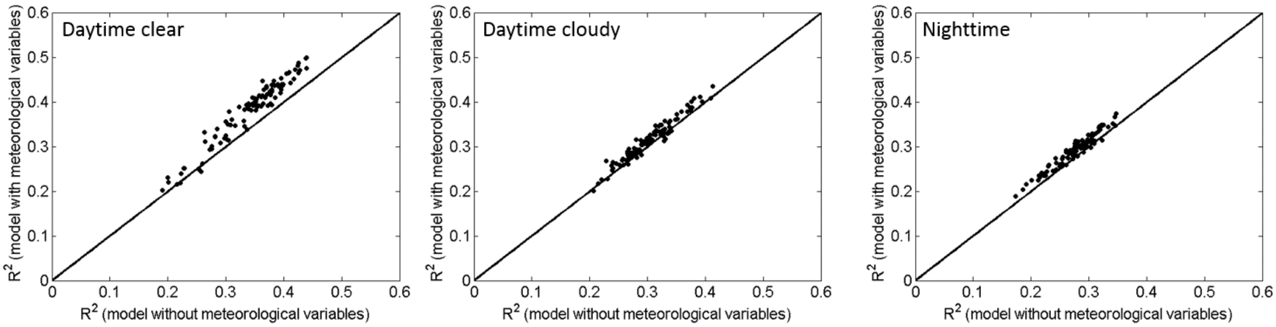

Figure 11. Comparison of cross-validation R-squared of the model without meteorological variables and model with meteorological variables during daytime clear, daytime cloudy and nighttime periods with the data from ARM SGP site.

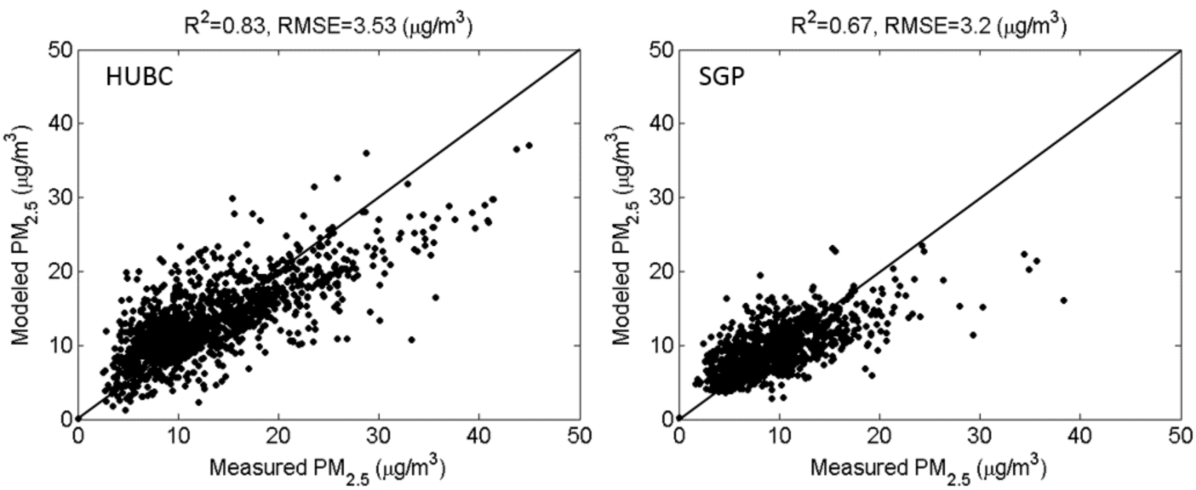

Figure 12. Comparison of daily average PM2.5 between in situ measurements and model simulation at HUBC site and ARM SGP site.

4. Discussion

Remote sensing of PM2.5 is generally based on AOD measurements due to its strong relationship with PM2.5. For nearly all the passive instruments, the measurements of AOD rely on solar radiation. Ceilometers are compact, low cost and unattended operational lidars and have been broadly used around the world. Although their laser power are relatively lower, the advantages of the small overlap distance, unattended and continuous operation make ceilometers suitable for remote sensing of aerosols near the surface. Moreover, the measurements of ceilometers don't rely on solar radiation, which makes it capable to retrieve aerosols during cloudy or nighttime periods.

In this study, an empirical model based on the regression between PM2.5 concentrations and ceilometer backscatter measurements was developed and tested with 6 years of observations at the HUBC site. The empirical model can explain ~ 56%, ~34%, and ~42% of the variability in the hourly average PM2.5 respectively during the daytime clear, daytime cloudy and nighttime periods. During the daytime clear periods the prediction capability was close to that of the model combining AOD and backscatter (explain ~60% of the variability) developed by Li et at., (2016) while during the daytime cloudy or nighttime period only the empirical model, which is independent on AOD, is available for the PM2.5 retrieval.

The impacts of meteorological conditions on the relationship between the in situ measured PM2.5 and the ceilometer measured backscatter were analyzed. The prominent positive relationship found between the surface temperature and the PM2.5-backscatter ratio could be due to the faster SO2 oxidation under higher temperature given that the dominant aerosol chemical composition is sulfate in the eastern United States. The measured relative humidity showed a significant negative association with the PM2.5-backscatter ratio, which could be due to hygroscopic growth of aerosols. The wind speed also shows a negative association with the PM2.5-backscatter ratio, but the relationship between the measured wind direction and PM2.5-backscatter ratio was found to not be obvious at the HUBC site. However, it is noteworthy that wind direction can be related to aerosol transportation and is usually associated with aerosol concentration and type. Although there was no significant association of the wind direction with the PM2.5-backscatter ratio at the HUBC site, wind direction impacts could be significant at other places where transported aerosols like dust are found near the surface. Aerosol properties usually vary seasonally due to the seasonally varied meteorological conditions, large scale transportation, and local emission of anthropogenic and natural aerosols. Taking into account the meteorological conditions in the model can to some extent mitigate the seasonal impacts on the PM2.5 retrieval and conducting the seasonally fitting can further improve the models predicting capability. Overall, the model with the seasonally fitted parameters can explain ~48% of the variability in the hourly PM2.5 including during daytime clear, daytime cloudy and nighttime periods at the HUBC site. Aerosol physical and chemical characteristics which are associated with aerosol dry mass and optical properties could be various at different locations. So a test was implemented based on the observations from the ARM SGP site which is geographically and climatologically different from the HUBC site. The results show that the impacts of meteorological conditions on the retrieval of PM2.5 using the ceilometer backscatter at the ARM SGP site is not as prominent as that at the HUBC site. That could be due to the different aerosol types at the SGP area and the DC area. Overall, the regression model using the ceilometer backscatter with meteorological variables could explain around 67% and 83% of the variability in the daily average PM2.5 at the ARM SGP site and the HUBC site respectively.

The most important objectives of this study was to develop an algorithm for remote sensing PM2.5 during cloudy and nighttime periods by using ceilometer measured backscatter. Retrievals of PM2.5 during cloudy or nighttime periods are very rare based on current remote sensing methods. A large number of ceilometers have been used over the world especially in the Europe and United States. The exploitation of the ceilometer on PM2.5 remote sensing could provide important information for air quality purpose, especially in helping to improve PM2.5 forecast over a larger area and can help fill the gaps among the EPA stations. And moreover that will largely increase the monitoring of air quality during cloudy or/and nighttime periods.

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Table 1. Parameters based on the best fitting of 100 independent tests for Eq. (11).

Best fitting parameters	$\boldsymbol{a_0}$	$\boldsymbol{a_1}$	$\boldsymbol{b_1}$
Daytime clear	-97.61	66.95	0.14
Daytime cloudy	-100.00	94.02	0.05
Nighttime	-100.00	85.70	0.08

Table 2. Parameters based on the best fitting of 100 independent tests for Eq. (12).

Best fitting parameters	$\boldsymbol{a_0}$	$\boldsymbol{a_1}$	$\boldsymbol{a_2}$	$\boldsymbol{a_3}$	$\boldsymbol{a_4}$	$\boldsymbol{b_1}$	$\boldsymbol{b_2}$
Daytime clear	-10.50	3.49	-2.92	0.06	-0.11	0.07	0.55
Daytime cloudy	-14.49	12.86	-7.20	0.10	-0.49	0.12	0.32
Nighttime	-1.38	0.74	-0.13	0.029	-0.20	0.68	0.64

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Table 3. Cross-validation tests of the model with different meteorological variables included.

Test	**R-squared (RMSE)**	**95% confidence intervals of R-squared (of RMSE)**
Test1: Model including all available meteorological variables	0.43 (6.70)	0.421-0.429 (6.672-6.736)
Test2: Model without surface temperature	0.37 (7.01)	0.367-0.375 (6.981-7.044)
Test3: Model without relative humidity	0.39 (6.91)	0.385-0.393 (6.880-6.946)
Test4: Model without wind speed	0.37 (7.01)	0.368-0.375 (6.978-7.043)
Test5: Model without wind direction	0.42 (6.71)	0.420-0.428 (6.683-6.742)
Test6: Model without surface pressure	0.42 (6.71)	0.421-0.429 (6.674-6.738)
Test7: Model not including any meteorological variable	0.21 (7.88)	0.203-0.209 (7.846-7.914)