Measurement of aerosol optical properties and their potential source origin in urban Beijing from 2013-2017

Tiantian Wang\textsuperscript{a}, Zhuofei Du\textsuperscript{a}, Tianyi Tan\textsuperscript{a}, Nan Xu\textsuperscript{a}, Min Hu\textsuperscript{a,b}, Jianlin Hu\textsuperscript{b,c}, Song Guo\textsuperscript{a,b,}\textsuperscript{*}

\textsuperscript{a} State Key Joint Laboratory of Environmental Simulation and Pollution Control, College of Environmental Sciences and Engineering, Peking University, Beijing, China
\textsuperscript{b} Collaborative Innovation Center of Atmospheric Environment and Equipment Technology, Nanjing University of Information Science & Technology, Nanjing, China
\textsuperscript{c} Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control, Nanjing University of Information Science & Technology, Nanjing, China

\textbf{ARTICLE INFO}

\textbf{Keywords:}
Aerosol optical property
Temporal variations
Meteorological dependence
Potential source contribution function (PSCF)

\textbf{ABSTRACT}

To investigate aerosol optical properties relevant to pollutant level controls in place in Beijing, particulate matters with diameter < 2.5 μm (PM_{2.5}) mass concentration, aerosol absorption (σ_{aP}) and scattering (σ_{sp}) coefficients were continuously measured from January 2014 to March 2017 at an urban site Peking University Atmosphere Environment Monitoring Station (PKUERS). Our results showed that PM_{2.5}, σ_{aP}, and σ_{sp} all decreased, especially from 2014 to 2015. Note that, a significant decrease of σ_{aP} in autumn and σ_{sp} in winter was responsible for the decrease of the year by year average. PM_{2.5}, σ_{aP} and σ_{sp} exhibited a pronounced bi-peak diurnal variation, which was attributed to the traffic emissions. The wind dependence analysis revealed that σ_{sp} was mainly influenced by local emissions but σ_{aP} was influenced by both local emissions and regional transport. The potential spatial source origins of σ_{sp} in Beijing were investigated using the potential source contribution function (PSCF). The results indicated that the potential source regions that influence σ_{sp} of Beijing were mainly from south and west of Beijing but slightly varied with different years and seasons.

1. Introduction

Atmospheric aerosols are known to perturb the earth’s radiation budget and climate directly by scattering and absorbing incoming and outgoing solar and terrestrial radiation (Charlson et al., 1992), and indirectly by acting as condensation nuclei (CCN) and/or ice nuclei (IN) in cloud formation, thus affecting the optical properties and lifetimes of clouds (Albrecht, 1989; Rosenfeld, 2000). Aerosols also play a major role in visibility impairment in urban and rural areas (Horvath, 1995) and have adverse effects on human health (Dockery and Pope, 1996). Furthermore, aerosol particles can be involved in atmospheric chemistry and thus affect concentrations of other minor atmospheric constituents like ozone (Arnold et al., 2008). Recent studies also elucidated that the enhancement of aerosol absorption by black carbon aging from anthropogenic pollution could indirectly promote the haze formation (Guo et al., 2016; Peng et al., 2016). Aerosol is considered as one of the greatest factors that are responsible for the uncertainty of the assessment of the global climate change (IPCC, 2013). Model and observation (Höller, 2003; Qian et al., 2011) studies indicated that uncertainties of the aerosol optical property measurement in China and other Asian countries contributed to the uncertainty of the estimation of aerosol radiative forcing.

To determine the influence of aerosols on climate, visibility and photochemistry, several key aerosol optical properties are required. The optical properties of aerosols are related to both their chemical composition and particle size distributions which are influenced by many processes, such as nucleation, condensation and deposition (Bergin et al., 2001). Ground based investigations on the optical properties of aerosols have been conducted in several regions of China, including Beijing (He et al., 2009; Jing et al., 2015; Tian et al., 2015), Pearl River Delta (Cheng et al., 2008; Tan et al., 2016), northern China (Ma et al., 2011; Wu et al., 2012; Yan et al., 2008) and Yangtze River Delta (Xu et al., 2012). These studies mainly focused on short-term periods, and long-term measurements of aerosol optical properties are important but rarely reported in literature.

Beijing, the capital of China, with the rapid urbanization and economic growth, particulate matter pollution has become more and more severe in Beijing (Guo et al., 2014; Hallquist et al., 2016; Hu et al., 2015). The government had taken many measures to improve the air quality. Specially, during the 2008 Beijing Olympic dramatic control measures were implemented, and the air quality was improved significantly (Guo et al., 2012, 2013). Further air pollution prevention and
control action plans were promulgated in 2013. Specific measures were introduced in S1. Thus, it is crucial to evaluate the effectiveness of these measures.

In this work, the optical properties, including scattering and absorption coefficients as well as PM$_{2.5}$ mass concentration were measured continuously for 3 years at an urban site in Beijing from 2014 to 2017. Diurnal, seasonal and interannual variations were analyzed and the influence of meteorological conditions were also analyzed and explained. Contributions of transport and regional sources to scattering coefficients ($\sigma_s$) in Beijing in different seasons and years were investigated using the receptor models (potential source contribution function and trajectory sector analysis, PSCF). The results achieved in this study would be valuable in improving our understanding of aerosol optical properties in this region and evaluating the control measurements in Beijing in recent years.

2. Experiment and methodology

2.1. Site description and instrumentation

The measurement was conducted from January 1st 2014 to March 15th 2017 at the Peking University Urban Atmosphere Environment Monito Ring Station (PKUERS, 39°59’21”N, 116°18’25”E), an urban site located in the northwest of Beijing. The detailed information about this site was described in the previous work (Guo et al., 2010; Tang et al., 2018) and showed in Fig. 1 in the supplement material. Previous studies have shown that the measurements at PKUERS can represent the air pollution in urban Beijing (Liu et al., 2017; Song et al., 2014).

Several instruments were used to measure the aerosol properties. The mass concentration was measured by a tapered element oscillating microbalance (TEOM, Thermo 1400). The TEOM provided 1 min values that were averaged to 10 min. The aerosol optical properties, i.e. aerosol absorption ($\sigma_a$) and scattering coefficients were measured by an Aurora-1000 integrating nephelometer (Ecotech Pty Ltd., Australia), and an multiangle absorption photometer (MAAP Model 5012, Thermo, Inc., Waltham, MA USA), respectively. Simultaneously, meteorological parameters, i.e. wind speed (WS), wind direction (WD), relative humidity (RH) and temperature (T), barometric pressure (BP) were measured continuously.

2.2. Data analysis

MAAP data were recorded as 5 min average, and subsequently processed to 10 min average. We compared the $\sigma_a$ of aerosol measured by MAAP and Aethalometer (Model AE-31, Magee Scientific Inc., Berkeley, Calif.) in the same period (Fig. S2). Results suggested that the data of absorption coefficient was accurate.

According to the manual, a mass absorption efficiency of 6.6 m$^2$/g was used to convert the black carbon concentrations to absorption coefficient (Petzold et al., 2002). MAAP nominally operated at a wavelength of 670 nm. However, laboratory tests revealed that the actual wavelength was 637 nm; in consequence, the light absorption coefficient and equivalent black carbon mass concentrations from the MAAP should be multiplied by a factor of 1.05 (Müller et al., 2011).

An Aurora-1000 integrating nephelometer was used to measure the 5-min average $\sigma_s$ at a single wavelength of 520 nm over scattering angle of 10°–170°. Due to Nephelometer design limitations, measurements do not cover the full (0°–180°) angular range, and scattering data need correction. It is worth noting that truncation error (angular non-idealities) dominates for large particles and for submicron aerosols is small. Considering the negligible contribution of coarse particles to light scattering (Chow et al., 2006), $\sigma_s$ can be approximately attributed to the PM$_{2.5}$, and 2.5 μm cut was employed for the Aurora-1000 measurement. In this study, for data accuracy, based on Mie scattering code and size distribution data which were independently measured by Scanning Mobility Particle Sizer (SMPS) and Aerodynamic Particle Sizer (APS), the “truncation error” almost less than 10% and even can be neglected since dust storms were uncommon in the North China Plain (Ma et al., 2006; Jung et al., 2009; He et al., 2009). The data were averaged to 10-min in this study. We compared the $\sigma_s$ of aerosol measured by Nephelometer and PAX (Droplet

Fig. 1. The location of the sampling site (PKUERS) in urban of Beijing.
Measurement Technology, Boulder, CO, USA) in the same period (Fig. S3). Results suggested that the data of scattering coefficient was accurate. Besides, the measured and calculated Visual range are illustrated in Fig. 2, which indicated that the optical property measurement was actually related well with the real optical variable, visibility. VR was regressed with PM$_{2.5}$ mass concentrations. On the basis of the relationship between PM mass concentration and VR, a non-linear relationship was found. Visibility decreased exponentially with the increase in the PM$_{2.5}$ concentration, as implied by the nonlinear fitting equation:

$$VR = 400.7[PM_{2.5}]^{-0.773} \left(R^2 = 0.80\right)$$

Where VR and PM are in the unit of km and $\mu$g/m$^3$, respectively. According to the relationship between PM$_{2.5}$ mass concentration and visual range (VR), Jung et al. (2009) proposed a nonlinear optimization formula $VR = 749.9[PM_{2.5}]^{-0.692} \left(R^2 = 0.93\right)$ (Jung et al., 2009). Xu et al. (2012) proposed $VR = 160.36[PM_{2.5}]^{-0.70} \left(R^2 = 0.86\right)$ and in this paper we proposed $VR = 400.7[PM_{2.5}]^{-0.773} \left(R^2 = 0.80\right)$. The different $R^2$ indicated that different factors contributed to visibility degradation such as aerosol chemical composition and mixture and atmospheric humidity (RH).

Single scattering albedo (SSA) was the ratio of the $\sigma_p$ over the extinction coefficient, which was the sum of $\sigma_s$ and $\sigma_p$ at a known wavelength, as indicated by the equation:

$$SSA = \frac{\sigma_p}{\sigma_s + \sigma_p}$$

The $\sigma_s$ was obtained at 525 nm and the $\sigma_p$ was measured at 670 nm. So $\sigma_s$ at 525 nm should be converted to match with $\sigma_p$ at the wavelength of 670 nm by the conversion method of Jung et al. (Jung et al., 2009).

$$\sigma_{p,670nm} = \sigma_{p,525nm} \times \left(\frac{\lambda_{670nm}}{\lambda_{525nm}}\right)^{-a}$$

where $a =$ the scattering Angstrom exponent, which was determined from spectral aerosol optical thickness (AOT) measured by an Ecotech Aurora 3000 (450, 525, 635 nm) as:

$$a = -\frac{\log(\sigma_{p,635nm}) - \log(\sigma_{p,450nm})}{\log(635nm) - \log(450nm)}$$

The correction factor was 0.55, 0.447, 0.465, 0.475 for spring, summer, fall, and winter, respectively.

2.3. Potential source contribution function (PSCF) method

Based on a statistical method called residence time analysis method (Ashbaugh et al., 1985), Malm et al. (Malm W C et al., 1986) developed the potential source contribution function (PSCF) model in 1986. PSCF was a receptor model that incorporated the aerosol chemistry and air mass transport information to determine possible source regions or dominant transport pathways. This method had been widely used to study the source regions of aerosols on regional scales and the long-range transport of air pollutants to a receptor site (Jeong et al., 2011; Sun et al., 2019; Wang et al., 2015). The PSCF at the ijth grid cell can be calculated using Eq. (1).

$$PSCF_i = \frac{m_{ij}}{n_i}$$

where $n_i$ is the total number of trajectory endpoints that fall in the ijth cell and $m_{ij}$ is the number of endpoints for the ijth cell with arrival times at the sampling site that correspond to each type of aerosol concentrations higher than an arbitrarily set criterion. The criterion values for $\sigma_p$ were chosen for the average values value for the entire period and different seasons.

Based on the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1998) at 500 m height and a time interval of 1 h for each day, air-mass back trajectories in the PSCF analysis were from the previous 72 h. Afterwards, the geographic regions covered by the air trajectories were divided into an array of $0.1^\circ 	imes 0.1^\circ$ grid cells, and for every grid cell at latitude i and longitude j, the number of the trajectory segment endpoints terminating within the cell was counted.

When calculating PSCF values, some grid cells will contain only 1 endpoint ($n_i = 1$). If this endpoint corresponds exactly to the pollution event trajectory, the PSCF for these cells will be 1, but the confidence in these PSCFs will be very low. In order to reduce uncertainties, a weight function $W(n_i)$ was used. The weight function was the following:

$$W(n_i) = \begin{cases} 
1.00 & (n_i > 80) \\
0.70 & (25 < n_i < 80) \\
0.42 & (15 < n_i < 25) \\
0.17 & (n_i < 15) 
\end{cases}$$

The $W(n_i)$ values were based on previous researches (Guo et al., 2015; Zhu et al., 2011).

3. Temporal variations of aerosol optical properties in Beijing

3.1. Year-to-year variation of the aerosol optical properties

The study summarized the statistics of the meteorological parameters was summarized in (Table S1 in the Supplement) Material. Statistics test was conducted for temperature, relative humidity, and barometric pressure. Significant test results indicated that there was no significant difference for these parameters among the three years with the significance level of 0.05.

The time series for the aerosol particle PM$_{2.5}$ mass concentration, $\sigma_p$ and $\sigma_s$ for the three years were showed in Fig. 3. The $\sigma_p$ at 532 nm and the $\sigma_p$ at 670 nm followed essentially the same trends in the PM$_{2.5}$ concentration, as implied by the nonlinear fitting equation. Table 1 presented a statistical summary of the average and standard deviation (S.D.) of PM$_{2.5}$ mass concentration, $\sigma_p$, $\sigma_s$, and SSA measured during the entire study period and the optical properties for the three years were displayed in Fig. 4. The large standard deviation for each variable reflected the large range of the measured aerosol properties. All parameters were presented annually decreasing tendency, a decrease of 32.1% with the average $\sigma_p$ the same period in
2015. It should be noted that the large decreasing tendency from 2014 to 2015, with the decline of the proportion in excess of 10%. Annual mean values (± S.D.) of SSA in 2014, 2015 and 2016 were 0.82 ± 0.16, 0.77 ± 0.13 and 0.78 ± 0.11, respectively, with the modest decrease, mainly because of the decrease of \( \sigma_a \). The \( \sigma_a \), \( \sigma_s \) and SSA in this study was compared with the measurements in previous work in Supplement.

3.2. Seasonal variations of the aerosol optical properties

Fig. 5 illustrated the seasonal variations of absorption and scattering coefficient of aerosols for the three years. Due to the heating season in Beijing, the season determination was defined in the supplement (S4). For all aerosol optical properties, the arithmetic mean values were typically different from the median values, since the probability distributions of these variables were deviate from normal distributions.
Meanwhile, the median values of $\sigma_{ap}$ and $\sigma_{sp}$ were much lower than the average values, especially in winter, implying more serious pollution in winter than other seasons. As shown in Fig. 5, the seasonal average of $\sigma_{ap}$ in winter and autumn was much higher than spring and summer, which could be related to the contributions from biomass burning (e.g., straw, corn stalks and crop waste) in the North China Plain (Cheng et al., 2013; Duan et al., 2004; Wang et al., 2007) and emissions from coal combustion in Beijing (He et al., 2009). It’s worth noting that in autumn the $\sigma_{ap}$ of three years was 38.0, 27.4, and 24.8 Mm$^{-1}$, respectively, which was higher than the annual average and was comparable to the value of winter. The existence of abundant black carbon aerosol was the main reason to cause the aerosol absorption effect (Yang et al., 2009), that might be produced from biomass burning (Yang et al., 2005).

The $\sigma_{sp}$ showed a slightly different seasonal distribution. The value of $\sigma_{sp}$ were elevated in winter and spring with the highest levels observed in winter and lower in autumn with the lowest levels in summer. As the result, relatively frequent sandstorms in spring brought the dust, which contributed to the high $\sigma_{sp}$ (Yang et al., 2009; Yu et al., 2011). Additionally, the three year’s $\sigma_{sp}$ in summer was only a half of that in winter, which could be influenced by the increased emissions from the use of coal for domestic and commercial heating, as well as the more stagnant meteorological condition (Guinot et al., 2007; Jing et al., 2015). Besides, more precipitation in summer was conducive to the removal of pollutants.

Statistical test was conducted for $\sigma_{ap}$ and $\sigma_{sp}$ among the three years (Table S2). Significant test results suggested that $\sigma_{sp}$ in spring, autumn and winter had a significant reduction while $\sigma_{ap}$ only has significant reduction in autumn both from 2014 to 2015 and from 2015 to 2016. In recent years, a series of measures, such as gradual replacement of coal combustion boilers with natural gas boilers, have been implemented to control the source emissions (Jin et al., 2016). Besides, the strict pollution control strategies were enforced for biomass burning (Zhang and Cao, 2015). The evident decline of $\sigma_{ap}$ and $\sigma_{sp}$ in autumn and winter indicated that control measures in autumn and winter were desirable, which contributes to the decline from 2014 to 2016.

### 3.3. Diurnal variation of aerosol optical properties

To determine the diurnal variation characteristics of the optical properties, a representative day with a wide range of PM$_{2.5}$ mass concentration was chosen for each season. The data was collected during the day with the most extreme values for each season (Table S1). The diurnal variation of PM$_{2.5}$ mass concentration, absorption coefficient, scattering coefficient, and SSA is shown in Fig. 4. The mean and median values were represented by the dot and line in the box, respectively, and the top and bottom of the box are the 75th and 25th percentiles, respectively. Besides, top and bottom of the whiskers and crosshatch represented the are the 90th and 10th percentiles, respectively.

#### Table 1

<table>
<thead>
<tr>
<th></th>
<th>2014 Average ± Standard deviation</th>
<th>2015 Average ± Standard deviation</th>
<th>2016 Average ± Standard deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>PM$_{2.5}$ mass concentration (µg/m$^3$)</td>
<td>74.0 ± 70.3</td>
<td>65.3 ± 66.7</td>
<td>66.3 ± 71.3</td>
</tr>
<tr>
<td>Absorption coefficient (Mm$^{-1}$)</td>
<td>35.5 ± 31.9</td>
<td>24.1 ± 28.4</td>
<td>23.5 ± 24.8</td>
</tr>
<tr>
<td>Scattering coefficient (Mm$^{-1}$)</td>
<td>290.3 ± 351.2</td>
<td>261.9 ± 286.1</td>
<td>205.7 ± 246.3</td>
</tr>
<tr>
<td>SSA</td>
<td>0.82 ± 0.16</td>
<td>0.77 ± 0.13</td>
<td>0.78 ± 0.11</td>
</tr>
</tbody>
</table>

Fig. 4. The average change of PM$_{2.5}$ mass concentration(A), absorption coefficient (B), scattering coefficient (C), and SSA(D). The mean and median values were represented by the dot and line in the box, respectively, and the top and bottom of the box are the 75th and 25th percentiles, respectively. Besides, top and bottom of the whiskers and crosshatch represented the are the 90th and 10th percentiles, respectively.
parameters, we used 10-min averages of directly measured data and indirect calculation value on aerosol optical parameters. Fig. 6 illustrated the mean diurnal pattern of PM$_{2.5}$, $\sigma_{ap}$, $\sigma_{sp}$ and SSA from 2014 to 2016 as a function of local time. There were clear diurnal variations for all variables with obvious peak-to-trough diel variation diurnal patterns that were mainly linked to both the local emission pattern and meteorological conditions such as the diel variation of the BLH and wind patterns (Cheng et al., 2008; Ma et al., 2011). The averaged diurnal cycles of ambient temperature, relative humidity and wind speed for the whole three years were presented in Fig. 7. The peak-to-trough diurnal variation was similar to those observed in other areas, such as Shanghai in winter of 2011 (Xu et al., 2012) and Shangdianzi, a suburb site in Beijing metropolis from 2003 to 2005 (Yan et al., 2008).

As shown in Fig. 6, the diurnal variation of PM$_{2.5}$, $\sigma_{ap}$, $\sigma_{sp}$ was similar with two peaks occurred at around 7:00 local time (LT) and 20:00LT. The first peak of the three parameters appeared in the early morning between 06:00LT and 08:00 LT, which corresponded with the morning rush hour traffic, during which large emissions of aerosols were released and lots of secondary aerosols formed along with meteorological conditions of low temperature, low wind speed and low planetary boundary height (BLH). It was interesting to note that, there was a delay between the $\sigma_{ap}$ and $\sigma_{sp}$ in the time of first peak during the morning traffic rush hours, which could be attributed to the time required for secondary aerosol formation through gaseous precursors while absorption was mainly caused by the primary particles (black carbon) from traffic emission. The subsequent decrease of the $\sigma_{ap}$ and

Fig. 5. Seasonal variations of absorption coefficient and scattering coefficient for the whole data set obtained at Beijing from January 1, 2014 to March 15, 2017. The mean and median values were represented by the dot and dash in the box, respectively, and the top and bottom of the box are the 75th and 25th percentiles, respectively. Besides, top and bottom of the whiskers represented the are the 90th and 10th percentiles, respectively.

Fig. 6. Average diurnal cycle of PM2.5 mass concentration(a), absorption coefficient (b), scattering coefficient (c) and SSA(d).
\(\sigma_a\) between 08:00 LT and 14:00 LT, could be attributed to the decrease in the vehicle density, the elevated planetary boundary layer (PBL), the increasing wind speed, and the increasing ambient temperature, which result in an increased vertical dispersion of aerosol (Lyamani and Alados-Arboledas, 2008; Lyamani et al., 2010). There was a difference in variability of \(\sigma_a\) and \(\sigma_p\) that the diurnal variation of \(\sigma_a\) in the noon was not as obvious as those of \(\sigma_a\), which could be related to the formation of secondary pollutants such as sulfate and nitrate that contributed the most to light scattering (Chan et al., 1999). Around noon, gaseous pollutants were converted to particulate matter which was accelerated by increasing temperatures and intensive radiation. The second peak appeared in the evening from 20:00LT to 21:00LT due to the combination of evening traffic sources and cooking sources. Those impacts presumably were exacerbated by decreasing mixing height and increased ambient temperature which led to the near-surface accumulation of pollutants. Generally, the \(\sigma_a\) was determined on a large degree by primary aerosols such as black carbon fraction contained in fresh combustion aerosols from diesel vehicles (He et al., 2009; Ruellan and Cachier, 2001). Note that, trucks with heavy pollution (such as construction waste transport trucks) only had permission to enter urban areas after 23:00LT and were not allowed to enter after 06:00LT. Thus, the relatively high \(\sigma_a\) throughout the night could be explained by the association with the formation of stable nocturnal PBL and increasing BC emissions from heavy diesel trucks. The \(\sigma_a\) and \(\sigma_p\) coefficient of the late evening peak were higher than those of morning peak, which may be partly due to the night-time truck traffic emissions, as result of Beijing local traffic regulations, which was in conflict with the study in Granada (Andreade et al., 2008; Lyamani et al., 2010). Besides, the diurnal variation of \(\text{PM}_{2.5}\) \(\sigma_a\) and \(\sigma_p\) decreased as the year increasing at every moment.

The diurnal pattern of SSA exhibited dual-dip patterns (Fig. 6d). The first dip occurred at wee hours during 04:00LT – 06:00LT and again appeared in the evening around 20:00LT. The daily maximum appeared at 14:00LT, indicating the enhancement of light scattering aerosol loading during the afternoon, which reflected the secondary aerosols formation through the photochemical processes with intensive solar radiation and high temperature conditions. It was worth noting that in the morning while the values of \(\sigma_a\) and \(\sigma_p\) was considerably high, SSA was relatively low, which could be explained by \(\sigma_p\) increased more by an order of magnitude than in the morning during traffic rush hours. In addition, contradicting to high values of \(\sigma_a\) and \(\sigma_p\), the comparatively low value of SSA, suggested that urban aerosols in the late evening contained a large fraction of absorbing material probably caused by the emission of diesel engine trucks.

The difference in the production and living activities of the cities between the weekdays (Monday to Friday) and the weekends (Saturday and Sunday) can lead to changes in the pollutant emission intensity, and in atmospheric particulate matter pollution conditions. To determine the weekly variation of aerosol optical properties, diurnal patterns of \(\sigma_a\) and \(\sigma_p\) between weekdays and weekends were displayed in Fig. 8 \(\sigma_a\) of weekends was relatively higher than that in weekdays, and reached its maximum value about 37.29 Mm\(^{-1}\) at 23:00LT, which was different from the results in Granada with lower \(\sigma_a\) in the weekend (Lyamani and Alados-Arboledas, 2008). The difference between weekend and weekdays implied that there might be an increase of traffic on weekend on account of no traffic restrictions based on the last digit of license plate numbers and non-Beijing license plate number. The diurnal variation of aerosol optical properties suggested that traffic source had a distinct impact on the aerosol optical properties and the traffic restrictions might improve the Beijing’s air quality.

4. Transport and its impact on aerosol optical properties in Beijing

4.1. Wind dependence of aerosol optical properties

Meteorological conditions have important impacts on the optical properties and especially for wind conditions, which can greatly affect the diffusion and transport of particulate matter in the air (Pateraki et al., 2012).

Fig. 9 presented the wind dependence of \(\text{PM}_{2.5}(A)\), \(\sigma_a(B)\), \(\sigma_p(C)\) and SSA(D). The parameters were plotted using polar coordinates depending on the wind speed and wind direction and the color plotted in the figure represented the magnitude of the variable. This can be seen, the maximum \(\text{PM}_{2.5}\), \(\sigma_a\) and \(\sigma_p\) occurred with calm or weak winds (wind speed < 3 m/s), independent of wind direction. This indicated that stagnant weather conditions were conducive to the accumulation and formation of pollutants. For the whole period, the minimum \(\text{PM}_{2.5}\), \(\sigma_a\) and \(\sigma_p\) occurred with wind coming from north wind while wind speed was almost higher than 4 m/s. The reason was that north winds usually occur with cold front systems and with high wind speeds (Ma et al., 2011). Much cleaner air masses significantly sweep away air pollutants so that the dilution effect led to low values of the parameters. The mean \(\text{PM}_{2.5}\) \(\sigma_a\) and \(\sigma_p\) for southerly winds were higher than that for northerly winds. When the wind blew at low wind speed, the \(\sigma_a\) value was highest and decreased with the increasing wind speed, corresponding to heavy primary emissions dominated by strong local sources. In contrast to \(\sigma_a\), although the highest \(\sigma_p\) was observed with the calm wind originating from northwest from Beijing, airflow
contained many secondary aerosols from the south sometimes caused high level of value in the $\sigma_p$. The wind dependence of SSA was displayed in Fig. 9(D), which was quite similar to that of $\sigma_p$ with high values appearing in the south wind-direction region. Meanwhile, when the wind blew from the northwest with low speed, the value of SSA was low. The results suggested the remarkable influence of neighbor pollution plumes.

Based on three-years observation data, analysis using the circular map of the aerosol optical properties to consider understand the wind dependence pattern suggested that air pollution in Beijing was attributed to the combination between local emissions and regional transports, which was similar to the result based on one-month campaign observations in summer (Garland et al., 2009; Jung et al., 2009). The correlation analysis between wind speed and PM$_{2.5}$, $\sigma_p$, and $\sigma_s$ results showed that under the confidence level of 0.01, the correlation was significant, respectively. $\sigma_p$, $\sigma_s$, and PM$_{2.5}$ mass concentration was negatively related with wind speed, respectively.

4.2. Potential source region of the aerosol in Beijing

Because $\sigma_p$ was mainly influenced by local emissions while $\sigma_s$ was attributed to both local emissions and regional transport, we used PSCF to identify the probable locations of regional emission sources contributing to Beijing $\sigma_p$ during the relatively polluted periods. The PSCF maps of the potential sources of $\sigma_p$ in Beijing between 2014 and 2016 for each year and for individual seasons were displayed in Fig. 10 and Fig. 11, combining 1-h average $\sigma_p$ and air parcel back trajectories. Due to the differences in the air flows in different years, the PSCF map distributions for different years were significantly different. The results indicated lower likelihood of high value of $\sigma_p$ resulting from local sources compared to regional transport. The largest sources which contributed to the greatest probability to Beijing $\sigma_p$ on an annual basis, with PSCF values higher than 0.5, were in the west and the south, covering the middle part of Inner Mongolia, the north of Shaanxi, the north and center of Shandong, the south of Hebei, the north of Anhui and Jiangsu, the north and east of Henan, and the west of Shandong. The results implied that the sources of $\sigma_p$ were more regional by reason of the tendency of SNA toward long-range transport. Major sources of 2014 were in the south of Hebei, especially the line of Baoding and Shijiazhuang. For 2015 the contributions of the southwest of Shandong and the northeast of Shanxi were increased. Besides the south of Hebei, the potential sources of the western and middle part of Inner Mongolia were obvious.

However, the large potential source area seasonally varied. Main sources of $\sigma_p$ in the spring were in the south of Hebei, the north and east of Henan, and for summer were the south of Hebei, the west of Shandong, and the border of Henan. Contrary to the other seasons, the potential sources in winter included, to a certain extent, focused in the vicinity of Beijing covering most areas of the south Hebei and Tianjin, which revealed that transport from the near Beijing area was more important for higher $\sigma_p$ in autumn and winter than other seasons.

5. Conclusions

Measurements of aerosol optical properties were conducted from January 2014 to March 2017 in urban Beijing China. Statistical test results indicated that the meteorological conditions among three years have no significant difference. The PM$_{2.5}$ mass concentration, $\sigma_p$, and $\sigma_s$ appeared a statistically decreasing year by year, especially from 2014 to 2015. The significant decline in $\sigma_p$ of autumn and $\sigma_s$ of winter was responsible for the decrease of the year by year average. A clear bi-peak diurnal pattern of PM$_{2.5}$, $\sigma_p$, and $\sigma_s$ suggested the traffic emissions were still the major contributors to the particle pollution in Beijing.

There was obvious dependence of optical properties on the wind speed and wind directions. When strong winds blew from the northwest of Beijing, cleaner air masses significantly scavenged air pollutants leading to the low values of the PM$_{2.5}$, $\sigma_p$, and $\sigma_s$. When the wind was from south, the air mass with secondary aerosols transported to Beijing, which caused higher $\sigma_p$. The wind dependence analysis revealed that $\sigma_p$ was mainly influenced by local emissions while $\sigma_s$ was attributed to both local emissions and regional transport. To estimate the transport pathways and regional emission sources and quantified contribution of $\sigma_p$ in Beijing, potential source contribution function (PSCF) combining the $\sigma_p$ data were applied from 2014 to 2016. The probable locations of regional emission sources were mainly in the south and the west of Beijing.
Beijing especially the line of Baoding and Shijiazhuang, which showed that the sources of $\sigma_p$ were primarily areas on the regional scale due to its tendency for long-range transport. Controlling of the source emissions in Beijing's surrounding provinces especially the south and the west i.e. Hebei, Henan, Shanxi is necessary to reduce the regional transport impact in the future.

Acknowledgments

This research was supported by National Key Research and Development Program of China (2016YFC0202000 Task 3, 2017YFC0213000 Task 3), and the National Natural Science Foundation of China (21677002, 91844301).

Fig. 10. The PSCF maps of the potential sources of scattering coefficient in Beijing during 2014–2016 for the different seasons. The red star represents Beijing. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Fig. 11. The PSCF maps of the potential sources of scattering coefficient in Beijing during 2014–2017 for the different seasons. The red star represents Beijing. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)
Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2019.02.049.

References


