Aerosol Optical Properties Observed at a Semi-Arid Rural Site in Northeastern China

Yunfei Wu1,2*, Renjian Zhang1*, Yifen Pu1, Leiming Zhang1,3, K.F. Ho4, Congbin Fu1,5

1 Key Laboratory of Regional Climate-Environment Research for Temperate East Asia, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China
2 Graduate University of Chinese Academy of Sciences, Beijing 100049, China
3 Air Quality Research Division, Science and Technology Branch, Environment Canada, Toronto, Canada
4 School of Public Health and Primary Care, The Chinese University of Hong Kong, Hong Kong, China
5 School of Atmospheric Sciences, Nanjing University, Nanjing 210093, China

ABSTRACT

Continuous in situ measurements of aerosol optical properties have been conducted since March 2010 at a semi-arid rural site in Northeastern China - Tongyu (44.56°N, 122.92°E, elevation 151 m), a reference site of Coordinate Water and Energy cycle Observation Project (CEOP). Aerosol optical properties derived from the measurements during two spring seasons in 2010 and 2011 are analyzed and presented here. Mean values (and standard deviation) of light absorption coefficient ($\alpha_{ab}$) and scattering coefficient ($\alpha_{sc}$) at 520 nm were 7.61 Mm$^{-1}$ (6.17 Mm$^{-1}$) and 89.22 Mm$^{-1}$ (88.37 Mm$^{-1}$), respectively, in spring of 2010, and were 7.01 Mm$^{-1}$ (5.60 Mm$^{-1}$) and 85.34 Mm$^{-1}$ (116.11 Mm$^{-1}$), respectively, in spring 2011. These values are lower than the majority of measurements made in other areas of China. The mean single scattering albedo ($\omega$) was 0.90, greater than values found in some developed areas in China, indicating more scattering or less absorbing aerosols at Tongyu site. Mean values (and standard deviation) of the Ångström exponent of absorption ($\alpha_{ab}$) were 1.61 (0.27) and 1.64 (0.28) for spring of 2010 and 2011, respectively, and of the scattering ($\alpha_{sc}$) were 1.48 (0.50) and 1.10 (0.50), respectively. The values of $\alpha_{ab}$ were higher than those of pure BC, implying the presence of other light absorbing materials having larger $\alpha_{ab}$ values. The moderate values of $\alpha_{sc}$ indicated that considerable amount of coarse particles presented at Tongyu site. Distinct diurnal variations of aerosol optical properties were observed. $\omega$, $\alpha_{ab}$ and $\alpha_{sc}$ had nearly opposite diurnal variations compared to those of $\alpha_{ab}$ and $\alpha_{sc}$. Dust aerosols from the northwest and anthropogenic aerosols from the south were major sources contributing to the episodic aerosol events and thus, unusual aerosol optical properties, in spring at Tongyu site. High levels of PM$_{2.5}$, if mainly caused by dust particles, do not necessarily introduce high values of $\alpha_{ab}$ and $\alpha_{sc}$.

Keywords: Light absorption; Light scattering; Semi-arid area.

INTRODUCTION

Atmospheric aerosols influence the Earth’s radiation directly through scattering and absorbing solar radiation and indirectly by acting as cloud condensation nuclei, thus affecting cloud’s optical properties and lifetime. Because of the large temporal and spatial variations in chemical composition, size distribution and mixing status of atmospheric aerosols, aerosol radiative forcing is one of the sources having largest uncertainties in climate modeling. Characterization of aerosol optical properties in various regions around the world is essential to reduce these uncertainties.

China has experienced rapid economic growth during the past three decades. Aerosol loading has thus increased dramatically in many regions of China. For example, aerosol optical depth was estimated to have increased from 0.38 in 1960 to 0.47 in 1990 (Luo et al., 2001). The increase in aerosol loading likely accounts for, at least partially, the notable decrease in sunshine duration and surface irradiance (Che et al., 2005; Liang and Xia, 2005; Qian et al., 2006). To date, a large number of studies have focused on aerosol mass concentrations and their physical and chemical properties in various regions of China (He et al., 2001; Yao et al., 2002; Cao et al., 2003, 2004; Dan et al., 2004; Guo et al., 2004; Sun et al., 2004; Yao et al., 2004; Cao et al., 2005; Duan et al., 2005; Xu et al., 2005; Yang et al., 2005; Wang et al., 2006; Meng et al., 2007; Zhang et al., 2008a; Yang et al., 2010a, b). Yet, only limited in situ measurements have been conducted on aerosol optical properties despite their critical

* Corresponding author.
E-mail address: wuyf@mail.iap.ac.cn (Yunfei Wu);
zrj@mail.iap.ac.cn (Renjian Zhang)
roles in aerosol radiative forcing and their potential impacts on climate and environment (Bergin et al., 2001; Xu et al., 2002, 2004; Yan, 2006; Li et al., 2007; Andreae et al., 2008; Garland et al., 2008; Yan et al., 2008; Che et al., 2009; Garland et al., 2009; Yang et al., 2009; Fan et al., 2010; Ma et al., 2011). Few study also showed the relationship between aerosol chemical and optical properties (Cheng et al., 2008; Huang et al., 2010; Tao et al., 2012).

In northern China, besides the anthropogenic pollutants from urbanization and industrial activities, desert dust particles from windstorms also contribute a considerable fraction to the frequently observed heavy aerosol loading. A rather complex nature of aerosol optical properties was observed in this region (Holler et al., 2003). A large area in this region belongs to semi-arid landscapes. Water and energy budgets in these areas are sensitive and can also feedback to the regional climate change. Aerosol is one of the most important components influencing the energy budget of the earth and thus, studying the aerosol optical properties in semi-arid areas is very important to the research on climate change. Several previous studies focused on the aerosol chemical components in these regions (Zhang et al., 2008a; Cheng et al., 2010; Ho et al., 2011; Shen et al., 2011), but none of these studies investigated the aerosol optical properties.

In the present study, in situ measurements of aerosol properties at a semi-arid rural site in northeastern China are analyzed. Section 2 describes the measurement site, measurement instruments, and data collection and correction procedures. Section 3 presents the characteristics and diurnal variations of aerosol optical properties and the potential causes of these variations, and also discusses in detail a few episodic cases. The conclusions and discussions are given in section 4. The analysis results presented here would be valuable in improving our understanding of aerosol effects on climate in this region.

MEASUREMENT

Site Description

Tongyu observation station (44.56°N, 122.92°N, elevation 151 m) is a reference site of the Coordinate Water and Energy cycle Observation Project (CEOP) initiated by Global Energy and Water-cycle Experiment (GEWEX). It is located in the semi-arid area in Northeast China. Routine meteorological variables have been measured since October 2002. To enhance the climate and environmental research in this region, an aerosol observation station was also established at this site in November 2007. Sporadic aerosol experiments in different regions (Bergin et al., 2001; Yan et al., 2008). However, most of these values were for urban aerosols and little was known for semi-arid rural areas. Thus, the direct method was used in the present study to obtain the aerosol absorption coefficient. Uncertainties using the direct method were expected to be smaller than those from the indirect method.

As mentioned above, Tongyu site is located in a degraded grassland area and has ‘background’ atmospheric properties to a large extent, the aerosols here are most likely aged. As suggested by Schmid et al. (2006), the value 4.4 was chosen as the multiple scattering correction factor C at reference

Absorption Measurement and Correction

An AE-31 Aethalometer (Magee Scientific, US) was used to measure aerosol absorption coefficient in real time starting March 17, 2010. The model AE-31 Aethalometer measures the optical attenuation of light from LED lamps with seven wavelengths (370, 470, 520, 590, 660, 880 and 950 nm) transmitted through the particles deposited on the quartz-filter. The differences in light transmission between those passing through the particle-laden sample spot and those of a particle free reference spot of the filter were attributed to aerosol absorption. The attenuation of light is converted to the recorded BC mass concentration using wavelength dependent calibration factors as recommended by the manufacturer (Aethalometer Manual, Magee Scientific). The aerosol light absorption coefficient can be directly calculated from the attenuation measured by the Aethalometer or indirectly calculated based on the BC concentrations recorded by the instrument (Yan et al., 2008).

The direct calculation of aerosol light absorption $\sigma_{ab}$ uses the following formula (Weingartner et al., 2003):

$$\sigma_{ab} = \frac{\sigma_{ATN}}{C \cdot R(\text{ATN})}$$

(1)

where $\sigma_{ATN}$ is the attenuation coefficient determined by the Aethalometer, the constant factor $C$ (mostly greater than unit) corrects for multiple light scattering effects within the filter, and $R(\text{ATN})$ (mostly less than unit) accounts for the ‘shadowing’ effect due to filter loading. Some previous works have been done to determine the two correction factors (Weingartner et al., 2003; Arnott et al., 2005; Schmid et al., 2006).

The indirect method to obtain the light absorption coefficient from the recorded BC concentration is based on the following equation:

$$\sigma_{ab} = \alpha \cdot [\text{BC}]$$

(2)

where $\alpha$ is the conversion factor or the BC absorption efficiency and can be determined theoretically from Mie-theory or empirically from linear regression of the Aethalometer recorded BC concentration data against the aerosol absorption coefficient measured from a reference method (Arnott et al., 2005; Yan et al., 2008).

The conversion factor in the indirect method has large variations and depends on the properties of aerosols, such as mixing status. Several empirical values were generated from aerosol experiments in different regions (Bergin et al., 2001; Yan et al., 2008). However, most of these values were for urban aerosols and little was known for semi-arid rural areas. Thus, the direct method was used in the present study to obtain the aerosol absorption coefficient. Uncertainties using the direct method were expected to be smaller than those from the indirect method.
Aerosol scattering coefficients at three wavelengths of 450, 525 and 635 nm were measured simultaneously starting March 25, 2010 using an Aurora3000 integrating Nephelometer (EcoTech, Australia) with LEDs as the light source. The scattering integration angle is from 10° to 170° as reported by the manufacturer. Zero checks were done automatically by pumping in particle-free air once every day. Because of the inconvenient conditions of the rural observation station, span checks and full calibrations were performed manually once in about every two months using particle-free HFC-R134a gas. The results and experiment records indicated that the bias was mostly less than 2 Mm⁻¹ for zero checks and less than 10% for span checks. The relative humidity in the cell of the instrument was controlled below 60% by automatic heating to prevent the influence of liquid particles. Evaporation of volatile inorganic species (such as nitrate) and volatile organic matters could be resulted from the heating. Bergin et al. (1997) reported that the decrease in scattering coefficient caused by evaporative losses of aerosols in a heating nephelometer was less than 20% for pure nitrate aerosol. Moreover, nitrate only account for a small fraction of total mass in fine particles in northern China (Zhang et al., 2009), especially in the semi-arid rural regions (Shen et al., 2011). Thus, the measured aerosol scattering coefficient due to the loss of volatile inorganic (nitrate) by the heating was estimated to be small in our research. Since a precise estimation on the loss of organic matters due to the sample heating was difficult to make because of the lack of information on organic species in the measurements, the bias due to the heating cannot be quantified.

The truncation of the integration angle can lead significant bias to the scattering coefficient, especially for large particles such as dust. In Table 2, the low values of Ångström exponent of measured scattering coefficient, which relate to the particle size, implied that large particles have a considerable contribution at Tongyu site. Thus, truncation correction was needed in this study. Many previous works have been done to correct the truncation error for the TSI 3563 integrating Nephelometer (Anderson and Ogren, 1998; Heintzenberg et al., 2006; Bond et al., 2009; Müller et al., 2009), but little was known for the EcoTech-made instrument used in the present study. However, as mentioned by Müller et al. (2009) and also by the manufacturer, the Ecotech’s Aurora model nephelometer light source has been updated to produce an angular intensity distribution function that is closer to Lambertian. Besides, the light source has an increased number of LEDs and a shorter axial dimension. All these improvements have made the light source of

### Table 1. Statistics for Ångström exponent of absorption coefficient derived from the direct correction.

<table>
<thead>
<tr>
<th>Derived ‘true’ $\alpha_{ab}$ from calculated multi-wavelength $\sigma_{ab}$</th>
<th>Mean 2010</th>
<th>Standard Deviation 2010</th>
<th>Mean 2011</th>
<th>Standard Deviation 2011</th>
<th>Median 2010</th>
<th>Median 2011</th>
</tr>
</thead>
<tbody>
<tr>
<td>Assumption values</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>of $\alpha_{ab}$ before calculated</td>
<td>1</td>
<td>1.49</td>
<td>1.52</td>
<td>0.27</td>
<td>0.28</td>
<td>1.47</td>
</tr>
<tr>
<td>calculated</td>
<td>2</td>
<td>1.73</td>
<td>1.76</td>
<td>0.28</td>
<td>0.28</td>
<td>1.59</td>
</tr>
</tbody>
</table>
measured scattering Ångström exponent (\( \text{\AA} \)) from the measured multi-wavelength scattering coefficient: total scatter from scattering Ångström exponent. which are used to calculate truncation correction factor for parameters ([nephelometer and TSI 3563. Considering that the fitting of \( a \) and \( b \) at instrument wavelengths. There is, however, a of direct source plumes, and provided recommended values size distributions covering most aerosol populations outside of direct source plumes, and recommended provided values of \( a \) and \( b \) at instrument wavelengths. There is, however, a slight difference in wavelengths between Ecotech Aurora3000 nephelometer and TSI 3563. Considering that the fitting parameters \( (a \) and \( b \) only varied slightly with wavelength, a simple linear interpolation was applied to obtain the fitting parameters at Ecotech Aurora3000 wavelengths used in our study (450, 525, 635 nm) from those proposed by Anderson and Orgen (1998) at the TSI 3563 wavelengths. The parameters used were listed in Table 3.

Air sampler inlets were placed at about 1.5 m above the roof of the monitoring room (4 m above the ground). Air was sampled at ~5 L/min passing through 0.95 cm i.d. black conductive tube into the room without any aerosol size cut-off. Simultaneous PM\(_{2.5}\) sampling was also conducted during March 27 to April 29, 2010 using an frmOMNI ambient air sampler (BGI Incorporated, USA). Each sample covered a 24 hour period from 09:00 to 09:00 the next day. The 24h-daily samples were weighted and analyzed to obtain the PM\(_{2.5}\), ionic, OC and EC concentrations at the Institute of Earth Environment, Chinese Academy of Sciences. All of the derived absorption and scattering coefficient have been manually edited to remove invalid data and data with abnormal values resulting from instrumental or sampling problems. The hourly average was obtained from the 5-minute data and the analyses below were all based on the hourly averaged data.

### RESULTS

#### Aerosol Optical Properties

The statistics of hourly-average aerosol absorption (\( \sigma_{ab} \)) and scattering (\( \sigma_{sc} \)) coefficients at 520 nm during spring of 2010 and 2011 at Tongyu station are shown in Table 4. \( \sigma_{av} \) at 520 nm was converted from that observed at 525 nm, one of the three wavelengths measured by nephelometer using the Ångström exponent of scattering. The mean (and standard deviation) values of \( \sigma_{ab} \) and \( \sigma_{sc} \) at 520 nm were 7.61 Mm\(^{-1}\) (6.17 Mm\(^{-1}\)) and 89.22 Mm\(^{-1}\) (88.37 Mm\(^{-1}\)), respectively, in spring of 2010, and 7.01 Mm\(^{-1}\) (5.60 Mm\(^{-1}\)) and 85.34 Mm\(^{-1}\) (116.11 Mm\(^{-1}\)), respectively, in spring of 2011. The corresponding median values were 5.68 Mm\(^{-1}\) and 57.08 Mm\(^{-1}\), respectively, in 2010, and 5.03 Mm\(^{-1}\) and 47.01 Mm\(^{-1}\), respectively, in 2011. The large standard deviation for each variable reflects the large range of the measured aerosol properties. Mean values of \( \sigma_{ab} \) and \( \sigma_{sc} \) measured at Tongyu site were lower compared to several measurements conducted in other areas in China (see Table 5) and reflected the properties of ‘background’ aerosols in this region. It is noticed that Tongyu had a higher mean \( \sigma_{ab} \) but lower mean \( \sigma_{sc} \), compared to a site (Yulin) near Gobi desert. This indicates that the dust components of aerosols were smaller at Tongyu than at the desert site.

The single scattering albedo (\( \omega \)) is defined as the ratio of the aerosol scattering coefficient to the extinction coefficient (sum of the absorption and scattering coefficient):

\[
\omega = \sigma_{sc} / (\sigma_{sc} + \sigma_{ab}) .
\]

It is a very critical parameter in the estimation of direct aerosol radiative forcing. For example, a small error in \( \omega \) can lead a great uncertainty in the estimation of aerosol forcing, e.g., from cooling effect to warming effect or vice versa. Mishchenko et al. (2004) suggested that \( \omega \) should be as accurate as a level within 0.03.

The mean and median values of \( \omega \) obtained at Tongyu were 0.903 and 0.908, respectively, in spring 2010, and were 0.904 and 0.906, respectively, in spring 2011 (see Table 4). These values confirmed the assumption of \( \omega \sim 0.9 \) in section 2.2 when correcting the absorption coefficient. Mean value of \( \omega \) obtained at Tongyu site was lower than the majority of \( \omega \) listed in Table 5, suggesting less absorbing (e.g., BC) or

### Table 2. Statistics for Ångström exponent of Nephelometer measured scattering coefficient.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>635–525 nm</td>
<td>1.41</td>
<td>0.98</td>
<td>0.54</td>
<td>0.59</td>
<td>1.49</td>
<td>1.11</td>
</tr>
<tr>
<td>635–450 nm</td>
<td>1.49</td>
<td>1.11</td>
<td>0.51</td>
<td>0.51</td>
<td>1.54</td>
<td>1.24</td>
</tr>
<tr>
<td>525–450 nm</td>
<td>1.59</td>
<td>1.27</td>
<td>0.53</td>
<td>0.48</td>
<td>1.59</td>
<td>1.39</td>
</tr>
</tbody>
</table>

### Table 3. The fitting parameters at different wavelengths, which are used to calculate truncation correction factor for total scatter from scattering Ångström exponent.

<table>
<thead>
<tr>
<th></th>
<th>450 nm</th>
<th>525 nm</th>
<th>635 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>( a )</td>
<td>1.365</td>
<td>1.344</td>
<td>1.314</td>
</tr>
<tr>
<td>( b )</td>
<td>-0.156</td>
<td>-0.143</td>
<td>-0.124</td>
</tr>
</tbody>
</table>
more scattering material (e.g., dust) fractions in aerosols in this area.

The Ångström exponent of absorption ($\alpha_{ab}$) and scattering ($\alpha_{sc}$) accounts for wavelength dependence of absorption and scattering coefficient. Here $\alpha_{ab}$ and $\alpha_{sc}$ were calculated using the power law fit of $\sigma_{ab}$ and $\sigma_{sc}$, respectively, and $\lambda$ in the measurement wavelength range (only 470, 520, 590, and 660 nm in the visible range for AE-31). The mean and median values of $\alpha_{ab}$ were 1.61 and 1.59, respectively, in 2010, and were 1.64 and 1.61, respectively, in 2011. These values were significantly higher than the absorption exponent of pure black carbon (BC) whose value of $\alpha_{ab}$ is suggested to equal unity (Bodhaine, 1995). This suggests that total light absorption cannot be explained by BC (or soot carbon) alone. Other light absorbing materials with high absorption Ångström exponent (i.e. relatively strong wavelength dependence) in visible spectrum, such as brown carbon (some of the organic carbon) and eolian dust, should have also presented and absorbed a lot more at shorter wavelengths than at longer wavelengths.

The mean and median values of $\alpha_{sc}$ were 1.48 and 1.53, respectively, in 2010, and were 1.10 and 1.23, respectively, in 2011. $\alpha_{sc}$ informs the size of the particles. Generally, low (or high) values of $\alpha_{sc}$ represent the domination of the large (or small) particles. Fine and coarse mode particles commonly coexist in the atmosphere. The moderate mean value of $\alpha_{sc}$ measured at Tongyu implied the coexistence of coarse mode (e.g. dust) and fine mode (e.g. sulfate) particles in this area.

Significant differences between 2010 and 2011 were found for $\alpha_{sc}$, but not for $\alpha_{ab}$. This suggests the existence of large inter-annual variations for coarse mode particles; however, the fractions of several absorbing materials in total particles seemed to change little from year to year. One possible reason causing this phenomenon might be the low contribution of dust aerosols, which contains mainly large particles, to the total light absorbing. Although the dust content in total aerosols might have large inter-annual variability, which resulted in the large variation in $\alpha_{sc}$, the absorbing properties which primary induced by BC might vary slightly.

An interesting phenomenon is, although the mean (and median) value of $\alpha_{sc}$ has significant inter-annual variation, the values of $\alpha_{ab}$ and $\alpha_{sc}$ change slightly between the two years. It means that the increased fraction of coarse mode particles, such as dust, did not change significantly the mean values of absorption and scattering coefficient.

**Table 4. Statistics of aerosol optical properties measured during spring in Tongyu station.**

<table>
<thead>
<tr>
<th>period</th>
<th>$\sigma_{ab}$ (Mm$^{-1}$)</th>
<th>$\sigma_{sc}$ (Mm$^{-1}$)</th>
<th>$\omega$</th>
<th>$\alpha_{ab}$</th>
<th>$\alpha_{sc}$</th>
<th>citation</th>
</tr>
</thead>
<tbody>
<tr>
<td>2010</td>
<td>7.61</td>
<td>89.22</td>
<td>0.903</td>
<td>1.61</td>
<td>1.48</td>
<td>Wu et al., 2012</td>
</tr>
<tr>
<td>2011</td>
<td>7.01</td>
<td>85.34</td>
<td>0.904</td>
<td>1.64</td>
<td>1.10</td>
<td></td>
</tr>
<tr>
<td>Mean</td>
<td>6.17</td>
<td>88.37</td>
<td>0.041</td>
<td>0.27</td>
<td>0.50</td>
<td></td>
</tr>
<tr>
<td>Standard Deviation</td>
<td>5.60</td>
<td>116.11</td>
<td>0.034</td>
<td>0.28</td>
<td>0.50</td>
<td></td>
</tr>
<tr>
<td>Median</td>
<td>5.68</td>
<td>57.08</td>
<td>0.908</td>
<td>1.59</td>
<td>1.53</td>
<td></td>
</tr>
<tr>
<td>Mean</td>
<td>5.03</td>
<td>47.01</td>
<td>0.906</td>
<td>1.61</td>
<td>1.23</td>
<td></td>
</tr>
<tr>
<td>Standard Deviation</td>
<td>1.23</td>
<td>90.06</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Median</td>
<td>1.23</td>
<td>90.06</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Table 5. Aerosol optical properties in spring at some other measurements in China.**

<table>
<thead>
<tr>
<th>period</th>
<th>$\sigma_{ab}$ (Mm$^{-1}$)</th>
<th>$\sigma_{sc}$ (Mm$^{-1}$)</th>
<th>$\omega$</th>
<th>$\alpha_{ab}$</th>
<th>$\alpha_{sc}$</th>
<th>citation</th>
</tr>
</thead>
<tbody>
<tr>
<td>2010</td>
<td>6 ± 11 (565 nm)</td>
<td>158 ± 193 (530 nm)</td>
<td>0.95 ± 0.05</td>
<td>1.48</td>
<td>1.10</td>
<td>Xu et al., 2004</td>
</tr>
<tr>
<td>2011</td>
<td>7.01 (520 nm)</td>
<td>18.27 ± 14.02 (525 nm)</td>
<td>0.90 ± 0.04</td>
<td>1.61</td>
<td>1.23</td>
<td></td>
</tr>
<tr>
<td>Mean</td>
<td>7 ± 11 (550 nm)</td>
<td>15 ± 160.10 (525 nm)</td>
<td>0.85 ± 0.05</td>
<td>1.64</td>
<td>1.59</td>
<td>Yan et al., 2008</td>
</tr>
<tr>
<td>Standard Deviation</td>
<td>7 ± 160.10 (525 nm)</td>
<td>5 ± 105.23 (525 nm)</td>
<td>0.27 ± 0.06</td>
<td>1.53</td>
<td>1.59</td>
<td>Yan, 2006</td>
</tr>
<tr>
<td>Median</td>
<td>6 ± 11 (550 nm)</td>
<td>15 ± 160.10 (525 nm)</td>
<td>0.81 ± 0.05</td>
<td>1.53</td>
<td>1.61</td>
<td>Li et al., 2007</td>
</tr>
<tr>
<td>Mean</td>
<td>5 ± 11 (550 nm)</td>
<td>15 ± 160.10 (525 nm)</td>
<td>0.81 ± 0.05</td>
<td>1.53</td>
<td>1.61</td>
<td>Pan, 2007</td>
</tr>
<tr>
<td>Standard Deviation</td>
<td>5 ± 160.10 (525 nm)</td>
<td>5 ± 105.23 (525 nm)</td>
<td>0.27 ± 0.06</td>
<td>1.53</td>
<td>1.61</td>
<td>He et al., 2009</td>
</tr>
<tr>
<td>Median</td>
<td>5 ± 11 (550 nm)</td>
<td>15 ± 160.10 (525 nm)</td>
<td>0.81 ± 0.05</td>
<td>1.53</td>
<td>1.61</td>
<td>Ma et al., 2011</td>
</tr>
</tbody>
</table>

The Diurnal Variation of Aerosol Optical Properties

Fig. 1 shows the diurnal cycle of the hourly averaged aerosol optical properties obtained from the measurements.
conducted in spring times. A bimodal distribution can be seen for most variables. The diurnal variation is similar to those observed in some other areas, such as Shangdianzi and Xianghe (Yan et al., 2008; Yang et al., 2009). \( \sigma_{ab} \) and \( \sigma_{sc} \) reached their major peaks in the early morning (around 06:00), bottom values in the early afternoon (13:00–14:00), and minor peaks in the late afternoon (around 18:00). The low values of \( \sigma_{ab} \) and \( \sigma_{sc} \) in the afternoon might be related to the increased turbulent intensity and thus the depth of planetary boundary layer (PBL) (Yan et al., 2008; Fan et al., 2010). Earlier studies in other regions also identified two peaks and attributed the causes to human activities (Xu et al., 2002; Wang et al., 2005; Yan et al., 2008; Yang et al., 2009; Fan et al., 2010). We suspect the same reason causing the two peaks despite the much less density of human activities in this region. However, this needs further detailed research. In general, an opposite diurnal variation for Ångström exponent \( \alpha \) was found, with two low values in the early morning and in the evening, respectively, when \( \sigma_{ab} \) and \( \sigma_{sc} \) reached their peaks. More light absorbing materials could have been produced by human activities during these time periods. Both \( \sigma_{ab} \) and \( \sigma_{sc} \) had low values in the early morning and in the evening, similar to the diurnal variation of \( \sigma \). The observed phenomena can be explained by the weaker wavelength dependence of both light absorbing and scattering during those time periods. It also suggests that weaker wavelength-dependent light absorbing materials (e.g. BC, with lower \( \sigma_{ab} \)) were often accompanied by larger particles (with lower \( \sigma_{sc} \)). \( \sigma_{ab} \) varied in a narrow range with mean values of 1.54 to 1.68 at each hour and relative smoothly (standard deviation/mean \( \approx \) 2%). This might be caused by a lack of fresh BC (or soot), which has the \( \alpha_{ab} \) value close to unit, during the study period. This seems to be the case even in the morning and in the evening times with sufficient strong light absorbing materials.

### Potential Source Influencing the Optical Properties

Hourly \( \sigma_{ab} \) and \( \sigma_{sc} \) during the time periods when PM2.5 was also sampled (March 27 to April 29, 2010) is shown in Fig. 2(a). Hourly Ångström exponent \( (\sigma_{ab} \) and \( \sigma_{sc} \)) during this period is displayed in Fig. 2(b) to infer other aerosol properties. Significant diurnal and day-to-day variations can be clearly seen for all the parameters. The variation of \( \alpha \) is highly consistent with that of \( \sigma_{sc} \). The correlation coefficient between them reaches a high value of 0.86 during this period. However, the variation of Ångström exponent was not always consistent for absorption and scattering. It illustrates that the physical and chemical properties such as compositions and size distribution of aerosols influencing this area varied with time, although they usually increased or decreased the absorption and scattering coefficient simultaneously.

As mentioned in Section 3.1, the mean value of \( \sigma_{ab} \) was about 1.60 (1.61 for 2010, 1.64 for 2011) during spring times, higher than the suggested value (~1) for BC. It suggests that a relatively high fraction of other light absorbing materials besides BC existed in the particles measured at Tongyu site. During the PM2.5 sample period, \( \sigma_{ab} \) narrowly changed around the value of 1.60, except a few spikes. The very few occurrences of the low \( \sigma_{ab} \) (close to 1) events suggest that BC was usually mixed with other materials with greater wavelength dependence in light absorbing.

![Fig. 1. Diurnal Variations of (a) absorption coefficient, (b) scattering coefficient, (c) single scattering albedo, and (d) Ångström exponent. Black lines represent 2010 and red ones represent 2011. In (d), solid lines with square symbols are for absorption and dashed lines with triangle symbols for scattering.](image)
This can be expected considering that the freshly emitted BC aerosols at the observation site was probably limited, and that most BC aerosols reaching this site were likely transported from upwind areas and were well mixed with other types of aerosols having relative greater wavelength dependence in absorbing. Compared to $\tilde{\alpha}_{ab}$, $\tilde{\alpha}_{sc}$ showed much drastic variations. The value of $\tilde{\alpha}_{sc}$ sometimes dropped to lower than one or even lower than zero. Following the relationship between aerosol optical depth and its Ångström exponent suggested by Kaufman et al. (1994), we can assume that scattering coefficient with $\tilde{\alpha}_{sc}$ greater than 1 is mainly determined by submicron aerosols, while $\tilde{\alpha}_{sc}$ less than unit is largely determined by supermicron particles. Large variation of $\tilde{\alpha}_{sc}$ indicates that considerable large particles, such as dust, frequently existed during the measurement period.

As suggested by earlier studies, pure BC (or soot) has a weak wavelength dependence in absorbing and a value of $\tilde{\alpha}_{ab}$ close to 1 in visible and near-visible spectral region (Bodhaine, 1995; Moosmüller and Arnott, 2009; Moosmüller et al., 2011). On the other hand, BC (soot) has a strong wavelength dependence in scattering due to its small particle sizes (mostly locates in the Rayleigh regime). Thus, its single scattering albedo ($\omega$) should decrease with the increasing wavelength. However, both dust and brown carbon can have greater wavelength dependence in absorption than in scattering. The values of $\omega$ for these two types of aerosols should therefore increase with the wavelength (Yang et al., 2009). From the variations of hourly $\omega$ at the three wavelengths and their difference (see Fig. 3), we can clearly find that $\omega$ usually increased with the wavelength ($\omega_{500} - \omega_{250} > 0$) during the typical sample period in spring. It indicates again that, besides BC, dust and brown carbon were also the important aerosol compositions influencing the aerosol optical properties in this area.

**Episode Case Studies**

To study the impact of different types of aerosols encountered at the observation site on the aerosol optical properties, case studies on two episodes were presented below. As seen in Fig. 2, unusual high light absorption and scattering appeared from March 29 to April 2 (Episode 1). $\tilde{\alpha}_{ab}$ and $\tilde{\alpha}_{sc}$ showed significant detachment from the mid-day of March 31, with $\tilde{\alpha}_{ab}$ increasing and $\tilde{\alpha}_{sc}$ decreasing sharply, implying the occurrence of a dust event. Comparing the absorption and scattering coefficients between March 30 and March 31, it was seen that those after the mid-day of March 31 were not very high and only had several minor peaks. This suggests that, although dust aerosols were important fractions of total aerosols, they did not necessarily result in higher values of absorption and scattering coefficient.

Mass concentration distributions of PM$_{2.5}$, Ca$^{2+}$, SO$_4^{2-}$, OC and EC (see Fig. 4) further supports this hypothesis. PM$_{2.5}$ mass concentration reached its peak value in March 31, so did those of Ca$^{2+}$ and OC. It indicated the occurrence of dust event on this day and the mixing of dust and organic matters (most likely the secondary organic aerosols, SOA). However, the peak values of absorption and scattering coefficient occurred in March 30 when the mass concentration of SO$_4^{2-}$ and EC were relatively high, rather than on March 31 when PM$_{2.5}$ reached its peak. This was likely caused by anthropogenic aerosol pollution reaching the site. The 48-hour back trajectory analysis gives a clear sense of the transition of the two type aerosols. From Fig. 5(a), it is seen that before 12:00 UTC (20:00 LTC) on March 31, the air mass influencing the observation site mainly came from the south. Moreover, the airflow always transported at the low levels. Anthropogenic aerosols from the southward polluted areas, such as the industry district around the Bohai Basin, would have been transported to the observation site. From
12:00 UTC (20:00 LTC) on March 31, air masses were mainly from the northwest direction. The descent of air masses during their transport increased the possibility of dust updraft at the national boundaries of Russia, Mongolia and China, and then brought considerable dust to downwind areas.

Another unusual high light absorption and scattering case was from April 17 to April 20 (Episode 2). Compared to Episode 1, air masses reaching the measurement site during this episode were transported from northwest to south directions (see Fig. 5(b)). \(\text{SO}_4^{2-}, \text{Ca}^{2+}, \text{OC}, \text{EC} \) and \(\text{PM}_{2.5}\) mass concentrations and absorption and scattering coefficients all increased from April 17, and reached their peak values on April 18. The magnitude of the increase in absorption and scattering coefficients was much larger than that in \(\text{PM}_{2.5}\) mass concentration. From the analysis of the two episode cases, it was concluded that the large increase in \(\text{PM}_{2.5}\) did not necessarily result in large increase in light absorption and scattering; instead, aerosol compositions could have larger impact on their optical properties. It was also seen from both of the two episodes that \(\sigma_{abs}\) and \(\sigma_{scatt}\) decreased to a low level when anthropogenic pollution affected the site, e.g. with air masses from the south (see
Aerosol optical properties measured during two spring seasons at Tongyu, a semi-arid rural site in Northeastern China, were analyzed and presented in this study. Aerosol absorption and scattering coefficients were relatively lower at Tongyu observation site than those measured at other rural or urban sites. The mean (and standard deviation) of absorption coefficient at 520 nm wavelength were 7.61 Mm$^{-1}$ (6.17 Mm$^{-1}$) in spring 2010 and 7.01 Mm$^{-1}$ (5.60 Mm$^{-1}$) in spring 2011; and the scattering coefficient were 89.22 Mm$^{-1}$ (88.37 Mm$^{-1}$) and 85.34 Mm$^{-1}$ (116.11 Mm$^{-1}$). A relatively low mean value of aerosol single scattering albedo (~0.90) was observed, suggesting the relatively weak absorption of the aerosols during spring in this area. The mean values of absorption Ångström exponent were 1.61 and 1.64 in spring of 2010 and 2011, respectively, implying that other light absorption materials besides BC were also present in aerosols. Moderate mean values of scattering Ångström exponent (1.48 in 2010 and 1.10 in 2011) indicated that large particles contributed considerable portions to light scattering.

Significant diurnal and day-to-day variations were found for all studied aerosol optical properties. Both absorption and scattering coefficients presented the bimodal diurnal variations, with one peak in the early morning and another one in the evening. Diurnal variations of single scattering albedo and Ångström exponent showed nearly opposite distributions. The two peaks of absorption and scattering coefficients might correspond to the human activities in this region, such as cooking. More light absorption materials such as BC might be produced, which resulted in the lower values of single scattering albedo and absorption Ångström exponent during those time periods. Low values of scattering Ångström exponent during the same time periods implied that absorption materials such as BC were frequently accompanied by large particles. This seems to be consistent with the fact that coal is the main fuel in northern China and that BC produced from residential coal combustion usually mixes with other aerosol compositions in relatively large particles (Yang et al., 2009). Low values of absorption and scattering coefficients in the afternoon should be attributed to the increased turbulent intensity and mixing layer depth.

Detailed analysis of the variability of hourly aerosol optical properties and two episode cases also indicates that BC was usually accompanied with large particles. Besides, highest PM$_{2.5}$ loading did not always correspond to the highest values of absorption and scattering coefficients. Under certain anthropogenic polluted conditions, high levels of second aerosol composition (SO$_4^{2-}$) and EC (BC) could introduce high light absorption and scattering, but with no so high PM$_{2.5}$ mass concentrations. Case studies also show that dust aerosols from the northwest and anthropogenic aerosols from the south were two primary sources contributing to the abnormal aerosol optical properties in spring at Tongyu site.

The results presented here were obtained from the empirical correction factors. Unfortunately, the uncertainties in obtained aerosol optical properties cannot be quantified in this study due to the inherent errors from the instruments and the lack of accurate correction factors. Although some of the results confirmed the assumption made in the data correction, more advanced and accurate instruments should be used in the future to acquire more accurate correction factors for the aerosol measurements in this area.
ACKNOWLEDGEMENTS

This research is supported by National Basic Research Program of China (2012CB955303) and National Natural Science Foundation of China (No.41175131). Prof. Jie Tang is greatly appreciated for providing the template to eliminate the abnormal values in the raw data derived by aethalometer.

REFERENCES


Atmos. Chem. Phys. 9: 8903–8915.
Xu, J., Beigin, M.H., Yu, X., Liu, G., Zhao, J., Carrico, C.M.


Received for review, November 14, 2011

Accepted, March 21, 2012