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

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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°N, 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 (σ_{ab}) and scattering coefficient (σ_{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 (ω) 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 (α_{ab}) were 1.61 (0.27) and 1.64 (0.28) for spring of 2010 and 2011, respectively, and of the scattering (α_{sc}) were 1.48 (0.50) and 1.10 (0.50), respectively. The values of α_{ab} were higher than those of pure BC, implying the presence of other light absorbing materials having larger α_{ab} values. The moderate values of α_{sc} indicated that considerable amount of coarse particles presented at Tongyu site. Distinct diurnal variations of aerosol optical properties were observed. ω, α_{ab} and α_{sc} had nearly opposite diurnal variations compared to those of σ_{ab} and σ_{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 σ_{ab} and σ_{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, b, 2009, 2010a, b). Yet, only limited in situ measurements have been conducted on aerosol optical properties despite their critical

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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 measurements were conducted in the following two years. Continuous measurements of aerosol optical have been conducted since March 2010.

Tongyu observation station lies in a degraded grassland area and beside a small town with no major emission source except scattered, sparse anthropogenic activities. Measurements at Tongyu are expected to represent the 'background' atmospheric aerosol properties in this region. Aerosol properties are likely to be decided by the air masses transported from upwind.

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 (Aethalomter Manual, Magee Scientific). The aerosol light absorption coefficient can be directly calculated from the attenuation measured by the Aethalomater or indirectly calculated based on the BC concentrations recorded by the instrument (Yan *et al.*, 2008).

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

$$\sigma_{ab} = \frac{\sigma_{ATN}}{C \cdot R(ATN)} \tag{1}$$

where σ_{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 $R(ATN)$ (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:

$$\sigma_{ab} = \alpha \cdot [BC] \tag{2}$$

where α 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.

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

wavelength (532 nm) for internally mixed aerosol (e.g., aged ambient aerosol) with single scattering albedo (ω_{550}) close to 0.90. The correction factor C at the wavelengths of AE-31 Aethalometer (here only 470, 520, 590, 660 nm in the visible range were used) could be converted using the following formula (Schmid *et al.*, 2006):

$$\frac{C}{C_{ref}} = \frac{\lambda^{A\cdot\ln\lambda+B}}{\lambda_{ref}^{A\cdot\ln\lambda_{ref}+B}} \quad (3)$$

$$A = 0.102\alpha_{ab}^2 - 0.187\alpha_{ab} - 0.141 \quad (4)$$

$$B = -1.275\alpha_{ab}^2 + 2.564\alpha_{ab} + 1.827 \quad (5)$$

where subscript 'ref' refers to the reference wavelength (532 nm) and α_{ab} is the absorption Ångström exponent of the aerosol. α_{ab} can be calculated from the following equation:

$$\alpha_{ab} = -\frac{\ln(\sigma_{ab}^{\lambda_1} / \sigma_{ab}^{\lambda_2})}{\ln(\lambda_1 / \lambda_2)} \quad (6)$$

where $\sigma_{ab}^{\lambda 1}$ and $\sigma_{ab}^{\lambda 2}$ are absorption coefficient at two different wavelengths λ_1 and λ_2. Analogous to α_{ab}, the Ångström exponent of scattering (α_{sc}) is defined by replacing σ_{ab} by σ_{sc}. The loading correction R is given by the equation (Schmid *et al.*, 2006):

$$R(ATN) = (\frac{1}{f} - 1)\frac{\ln ATN - \ln 10}{\ln 50 - \ln 10} + 1 \quad (7)$$

with $f = 1.2$ for ω_{532}~0.9. R is set to unity while attenuation (ATN) is less than 10.

Because α_{ab} was unknown before data analysis, mean values needed to be first assumed for Eqs. (4) and (5) to calculate σ_{ab}. Yang *et al.* (2009) reported values of α_{ab} measured at a site (Xianghe) 70 km east of Beijing. The mean value of α_{ab} during the whole experiment was 1.47. The value was only 1.89 even with dust storm events. Considering that α_{ab} for pure BC has a value of 1, the mean value of α_{ab} measured at Tongyu site should be in the range of 1 to 2. Thus, three mean values of α_{ab}, i.e., 1, 1.5 and 2, were assumed in the calculation. The 'true' α_{ab} value could then be calculated from the multi-wavelength absorption coefficient using the power-law relation. The statistics of the derived 'true' α_{ab} are listed in Table 1. Obviously, the assumed value of 1.5 was more reliable than the other two in our study here. Thus, all the analyses in the following sections were based on the assumed mean α_{ab} value of 1.5.

Scattering Measurement and Correction

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.*, 2003), 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.

		Derived 'true' α_{ab} from calculated multi-wavelength σ_{ab}					
		Mean		Standard Deviation		Median	
		2010	2011	2010	2011	2010	2011
Assumption values	1	1.49	1.52	0.27	0.28	1.47	1.49
of α_{ab} before	1.5	1.61	1.64	0.27	0.28	1.59	1.61
calculated	2	1.73	1.76	0.27	0.28	1.71	1.74

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

	Mean		Standard Deviation		Median	
	2010	2011	2010	2011	2010	2011
635–525 nm	1.41	0.98	0.54	0.59	1.49	1.11
635–450 nm	1.49	1.11	0.51	0.51	1.54	1.24
525–450 nm	1.59	1.27	0.53	0.48	1.59	1.39

Ecotech nephelometer more similar to the TSI 3563. So the truncation correction method proposed by Anderson and Ogren (1998) for TSI 3563 instrument can also be used here to derive more reliable scattering coefficient.

The formula for the truncation correction is:

$$\sigma_{sc} = C \cdot \sigma_{neph} \tag{8}$$

where σ_{sc} is the corrected ('true') scattering coefficient, σ_{neph} is the nephelometer measured scattering coefficient, and C is the correction factor. C can be estimated by the measured scattering Ångström exponent (α_{neph}) calculated from the measured multi-wavelength scattering coefficient:

$$C = a + b \cdot \alpha_{neph} \tag{9}$$

where a and b are the fitting parameters. Anderson and Orgen (1998) compared the TSI 3563 measured scattering coefficient to the numerical values calculated from Mie theory using aerosols with different refractive indices and size distributions covering most aerosol populations outside of direct source plumes, and provided recommended 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.

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

	450 nm	525 nm	635 nm
a	1.365	1.344	1.314
b	–0.156	–0.143	–0.124

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 (σ_{ab}) and scattering (σ_{sc}) coefficients at 520 nm during spring of 2010 and 2011 at Tongyu station are shown in Table 4. σ_{sc} 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 σ_{ab} and σ_{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 σ_{ab} and σ_{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 σ_{ab}, but lower mean σ_{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 (ω) is defined as the ratio of the aerosol scattering coefficient to the extinction coefficient (sum of the absorption and scattering coefficient):

$$\omega = \sigma_{sc} / (\sigma_{sc} + \sigma_{ab}) . \tag{10}$$

It is a very critical parameter in the estimation of direct aerosol radiative forcing. For example, a small error in ω 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 ω should be as accurate as a level within 0.03.

The mean and median values of ω 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 ω~0.9 in section 2.2 when correcting the absorption coefficient. Mean value of ω obtained at Tongyu site was lower than the majority of ω listed in Table 5, suggesting less absorbing (e.g., BC) or

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

	Mean		Standard Deviation		Median	
	2010	2011	2010	2011	2010	2011
$^{*}\sigma_{ab}$ (Mm^{-1})	7.61	7.01	6.17	5.60	5.68	5.03
$^{*}\sigma_{sc}$ (Mm^{-1})	89.22	85.34	88.37	116.11	57.08	47.01
$^{*}\omega$	0.903	0.904	0.041	0.034	0.908	0.906
$^{**}\alpha_{ab}$	1.61	1.64	0.27	0.28	1.59	1.61
$^{***}\alpha_{sc}$	1.48	1.10	0.50	0.50	1.53	1.23

$^{*}\sigma_{ab}$, σ_{sc} and ω correspond to wavelength of 520 nm.
$^{**}\alpha_{ab}$ is from the power law fit of σ_{ab} and λ at 4 wavelengths of AE-31 Aethalometer at visible range (470, 520, 590, and 660 nm).
$^{***}\alpha_{sc}$ is from the power law fit of σ_{sc} and λ at 3 wavelengths of Aurora-3000 Nephelometer (450, 525, and 635 nm).

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

	period	σ_{ab}	σ_{sc}	ω	citation
Yulin	1999	6 ± 11 (565 nm)	158 ± 193 (530 nm)	0.95 ± 0.05	Xu *et al.*, 2004
Shangdianzi	2003 & 04	18.27 ± 14.02 (525 nm)	154.09 ± 160.10 (525 nm)	0.85 ± 0.05 (525 nm)	Yan *et al.*, 2008
Lin'an	2004	44.3 ± 19.7 (532 nm)	229.4 ± 104.8 (525 nm)	0.82 ± 0.03 (525 nm)	Yan, 2006
Xianghe	2005	65 ± 75 (550 nm)	468 ± 472 (550 nm)	0.81–0.85 (550 nm)	Li *et al.*, 2007
Beijing (Urban)	2006	33.1 ± 29.2 (532 nm)	278.0 ± 248.1 (525 nm)	0.87 ± 0.05 (525 nm)	Pan, 2007
Beijing (Urban)	2005 & 06	45 ± 39 (532 nm)	243 ± 255 (525 nm)	0.81 ± 0.10 (525 nm)	He *et al.*, 2009
Wuqing	2009	47 ± 38 (637 nm)	280 ± 253 (550 nm)	0.82 ± 0.05 (637 nm)	Ma *et al.*, 2011
Tongyu	2010&11	7.28 ± 5.87 (520 nm)	86.99 ± 105.23 (520 nm)	0.90 ± 0.04 (520 nm)	This study

more scattering material (e.g., dust) fractions in aerosols in this area.

The Ångström exponent of absorption (α_{ab}) and scattering (α_{sc}) accounts for wavelength dependence of absorption and scattering coefficient. Here α_{ab} and α_{sc} were calculated using the power law fit of σ_{ab} and σ_{sc}, respectively, and λ 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 α_{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 α_{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 α_{sc} were 1.48 and 1.53, respectively, in 2010, and were 1.10 and 1.23, respectively, in 2011. α_{sc} informs the size of the particles. Generally, low (or high) values of α_{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 α_{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 α_{sc}, but not for α_{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 α_{sc}, the absorbing properties which primary induced by BC might vary slightly.

An interesting phenomenon is, although the mean (and median) value of α_{sc} has significant inter-annual variation, the values of σ_{ab} and σ_{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.

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). σ_{ab} and σ_{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 σ_{ab} and σ_{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 ω was found, with two low values in the early morning and in the evening, respectively, when σ_{ab} and σ_{sc} reached their peaks. More light absorbing materials could have been produced by human activities during these time periods. Both α_{ab} and α_{sc} had low values in the early morning and in the evening, similar to the diurnal variation of ω. 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 α_{ab}) were often accompanied by larger particles (with lower α_{sc}). α_{ab} varied in a narrow range with mean values of 1.54 to 1.68 at each hour and relative smoothly (standard devation/mean ≈ 2%). This might be caused by a lack of fresh BC (or soot), which has the α_{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 σ_{ab} and σ_{sc} during the time periods when $PM_{2.5}$ was also sampled (March 27 to April 29, 2010) is shown in Fig. 2(a). Hourly Ångström exponent (α_{ab} and α_{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 σ_{ab} is highly consistent with that of σ_{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 α_{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 $PM_{2.5}$ sample period, α_{ab} narrowly changed around the value of 1.60, except a few spikes. The very few occurrences of the low α_{ab} (close to 1) events suggest that BC was usually mixed with other materials with greater wavelength dependence in light absorbing.

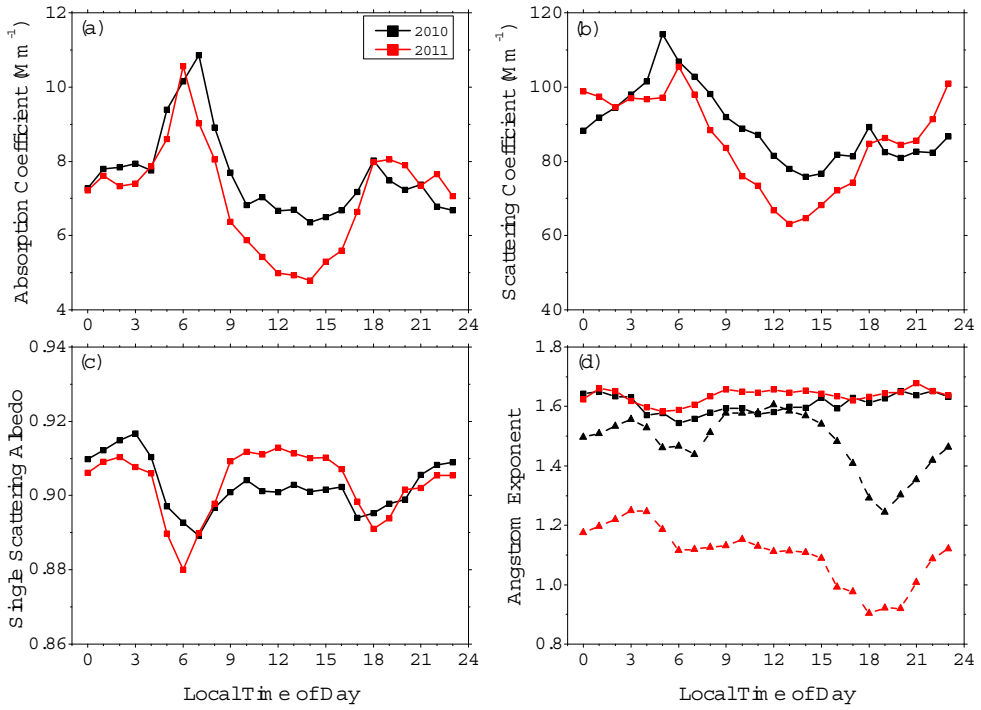

Fig. 1. Diurnal Variations of (a) absorption coefficient, (b) scattering coefficient, (c) single scattering albedo, and (d) Ångström exponent. Black lines represent 2010 and red ones represent 2011. In (d), solid lines with square symbols are for absorption and dashed lines with triangle symbols for scattering.

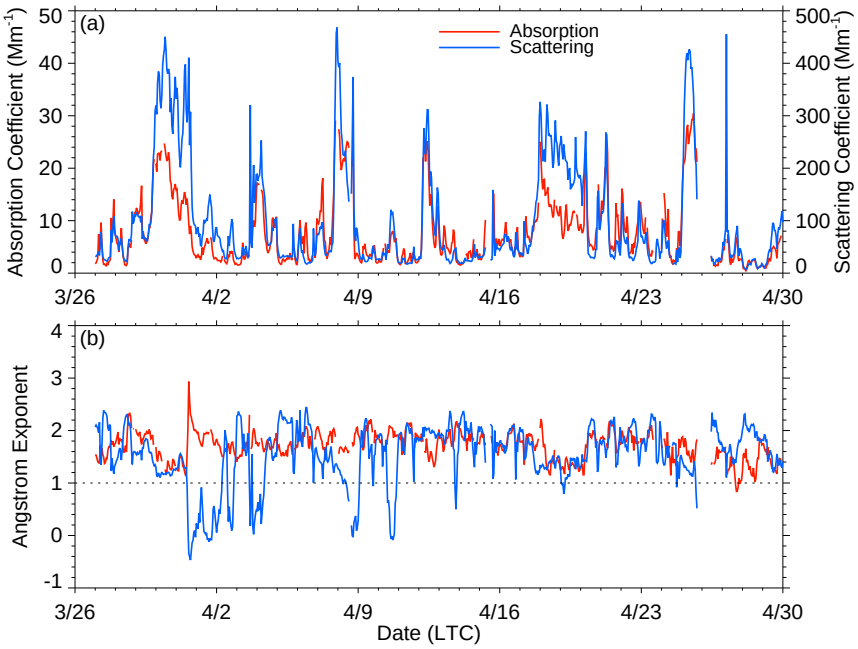

Fig. 2. Time series of hourly (a) absorption and scattering coefficient at 520 nm wavelength, and (b) Ångström exponent during the sampling period (March 27 to April 30, 2010). Red lines represent absorption and blue ones represent scattering.

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 α_{ab}, α_{sc} showed much drastic variations. The value of α_{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 α_{sc} greater than 1 is mainly determined by submicron aerosols, while α_{sc} less than unit is largely determined by supermicron particles. Large variation of α_{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 α_{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 (ω) 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 ω for these two types of aerosols should therefore increase with the wavelength (Yang *et al.*, 2009). From the variations of hourly ω at the three wavelengths and their difference (see Fig. 3), we can clearly find that ω usually increased with the wavelength ($\omega_{660} - \omega_{470} > 0$) 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). α_{ab} and α_{sc} showed significant detachment from the mid-day of March 31, with α_{ab} increasing and α_{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

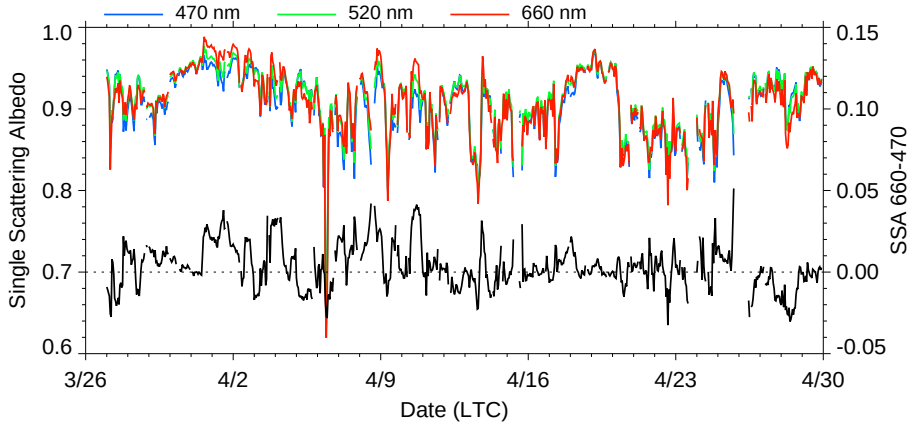

Fig. 3. Time series of hourly single scattering abledo at different wavelengths: 470 nm (blue), 520 nm (green), 660 nm (red), and the difference between single scattering abledo at 470 nm and 660 nm (black) during the sampling period.

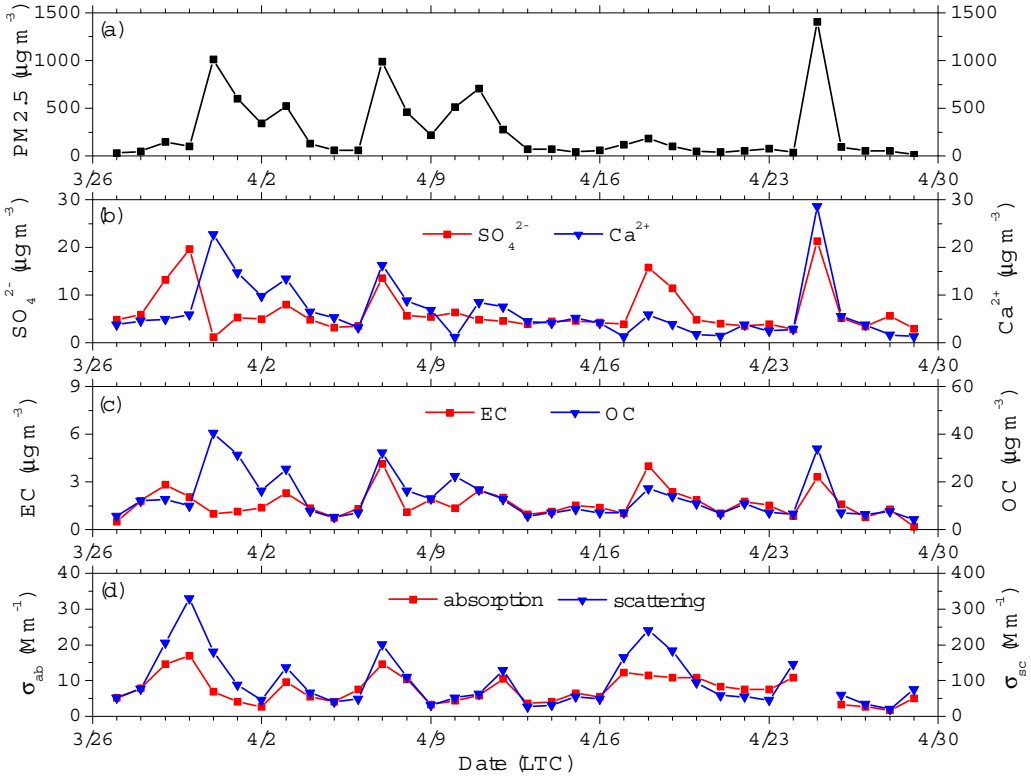

Fig. 4. Variability of daily mass concentration of (a) $PM_{2.5}$, (b) SO_4^{2-} (red) and Ca^{2+} (blue), (c) EC (red) and OC (blue), and (d) absorption (red) and scattering (blue) coefficient. Daily absorption and scattering coefficient is averaged of hourly values from 09:00 to 09:00 (LTC) of next day.

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)). SO_4^{2-}, Ca^{2+}, OC, EC and $PM_{2.5}$ 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 $PM_{2.5}$ mass concentration. From the analysis of the two episode cases, it was concluded that the large increase in $PM_{2.5}$ 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 α_{ab} and α_{sc} decreased to a low level when anthropogenic pollution affected the site, e.g. with air masses from the south (see

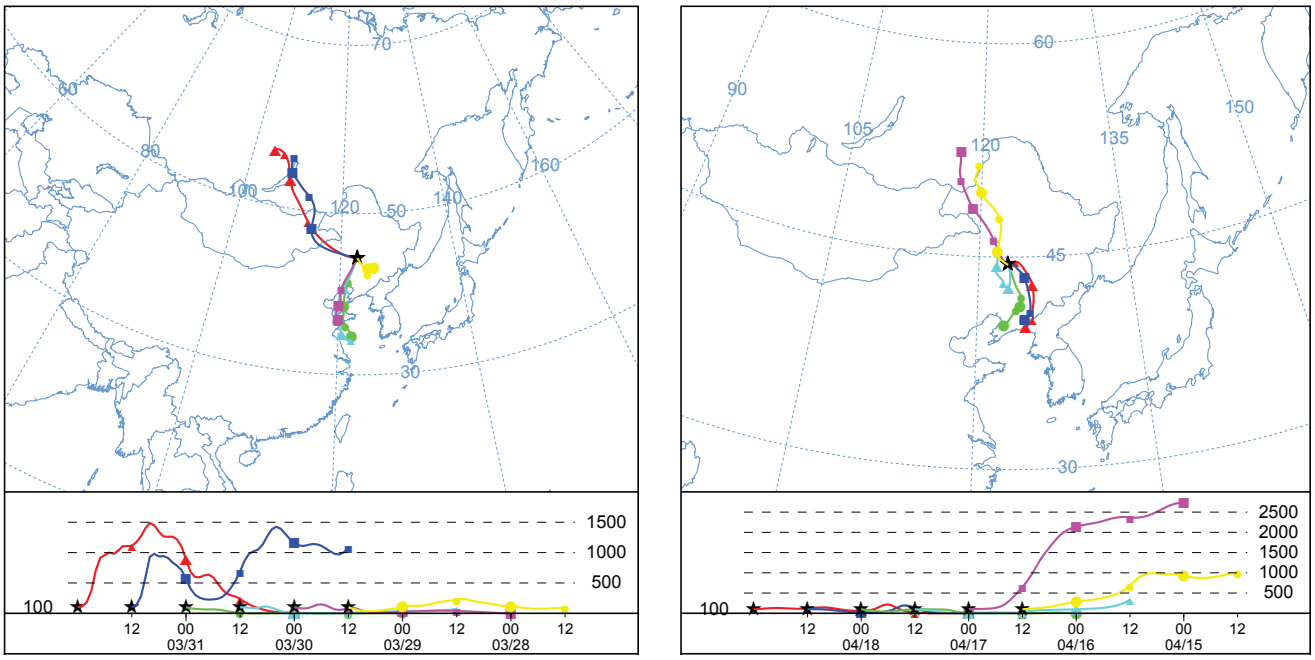

Fig. 5. 48-hour back trajectories of two cases: (a) end at 00:00 (UTC) April 1; (b) end at 00:00 (UTC) April 19.

Fig. 2(b)). This suggests that BC, which has low value of α_{ab}, was always accompanied with relative large particles having low value of α_{sc} in this area. Thus, BC measured at Tongyu site should be well mixed with other aerosol compositions.

CONCLUSION AND DISCUSSION

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 high 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.

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