Study on the aerosol optical properties and their relationship with aerosol chemical compositions over three regional background stations in China

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ABSTRACT
The special and temporal characteristics of aerosol optical depth (AOD) and Angstrom wavelength exponent (Alpha) and their relationship with aerosol chemical compositions were analyzed by using the data of CE318 sun-photometer and aerosol sampling instruments at Lin’an, Shangdianzi and Longfengshan regional atmospheric background stations. Having the highest AOD among the three stations, Lin’an shows two peaks in a year. The AOD at Shangdianzi station shows a single annual peak with an obvious seasonal variation. The AOD at Longfengshan station has obvious seasonal variation which peaks in spring. The Alpha analysis suggests that the aerosol sizes in Lin’an, Longfengshan and Shangdianzi change from fine to coarse categories. The relationship between the aerosol optical depths of the Lin’an and Longfengshan stations and their chemical compositions is not significant, which suggests that there is not a simple linear relationship between column aerosol optical depth and the near surface chemical compositions of atmospheric aerosols. The aerosol optical depth may be affected by the chemical composition, the particle size and the shape of aerosol as well as the water vapor in the atmosphere.

1. Introduction

Aerosol particles could change the earth–atmosphere radiation balance and affect the climate directly by absorbing and scattering solar radiation and the radiation of the earth–atmosphere system (Ackerman and Toon, 1981; Charlson et al., 1992). Indirectly, aerosol particles can serve as cloud condensation nuclei (CCN) or ice nuclei (IN) to change the microphysical structure, the optical properties and the precipitation efficiency of clouds, which could affect the climate (Hansen et al., 1997). The impact of aerosol on climate and environment has been a hot issue for the international scientific research community (IPCC, 2001, 2007). To fully understand the aerosol physical properties, extensive sets of both in situ and remote measurements are required (Alados-Arboledas et al., 2008). Consequently, several observation networks of aerosol optical properties have been established internationally to observe aerosol optical characterization such as AERoSol ROBotic NETwork, PHOTONS, SKYNET, AEROCAN, RIMA, AGSNet and so on (Holben et al., 1998; Goloub et al., 2008; Uchiyama et al., 2005; Bokoye et al., 2001; Campanelli et al., 2007; O’Brien and Mitchell, 2003).

Currently, the observation of aerosol optical property is deficient in China. AERONET has more than twenty stations in China, but only a few are in long-term operation (Eck et al., 2005; Xia et al., 2005). Luo et al. (2000) and Qiu and Yang (2000) have studied aerosol optical properties by using the data of ground-based direct solar radiation of China. Xin et al. (2007) has analyzed CHRNET aerosol optical characterization by using a hand-held sun-photometer. These results play an important role in the optical characterization of Chinese regional aerosol and its climatic effect.

China Meteorological Administration (CMA) has established the largest automatic observational network – China Aerosol Remote Sensing Network (CARSNET) in China since 2002. CARSNET has 37 stations at present with a CE318 sun-photometer installed at each station for the observation of regional aerosol optical property over China. Among them one is the global atmosphere watch (GAW) station (Waliguan) and six are regional atmosphere watch stations (Lin’an, Shangdianzi, Longfengshan, Shangrila, Akedala, and Jingsha). Three CE318 sun-photometers have been running continuously at Lin’an, Shangdianzi and Longfengshan stations for three years. The aim of this paper is to analyze these observation results...
and discuss the regional background aerosol optical properties over China.

2. Data and analysis method

2.1. Site distribution

The site geographical distribution is shown as Fig. 1. Lin’an regional atmospheric background station is located in northwestern Zhejiang Province (30°17’N, 119°45’E) with an altitude of 138.6 m. The observation data could represent the characteristics of aerosol optical properties in Yangtze River Delta Region (Tang et al., 2007). Shangdianzi regional atmospheric background station, southeast of Beijing, is located at 40°39’N, 117°07’E, and the altitude is about 293.3 m. The observation data at Shangdianzi can represent the basic characteristics of aerosol optical properties in northern China, especially the Beijing–Tianjin–Hebei (Jing-Jin-Ji) Metropolitan region (Tang et al., 2007). Longfengshan regional atmospheric background station is located at 44°44’N, 127°36’E with an altitude of about 330.5 m in the southeastern part of Heilongjiang province and the observation data can represent the basic situation of aerosol optical properties in northeast plain of China.

2.2. Instruments and measurement data

The CIMEL Electronique CE318 sun-photometer has eight channels, including 1020 nm, 870 nm, 670 nm and 440 nm wave bands, three 870 nm polarization channels and one 940 nm water vapor channel within a 1.2° full field-of-view (Holben et al., 1998). The bandwidth of each channel is 10 nm. Measurements at 440 nm, 670 nm, 870 nm, and 1020 nm can be used to retrieve the AOD, and measurements at 940 nm are used to obtain the total precipitable water content in centimeters (Holben et al., 1998). Aerosol size distribution, refractive index, and single-scattering albedo can be retrieved by using sky radiance almucantar measurements and direct sun measurements (Dubovik et al., 2000; Dubovik and King, 2000) as well as polarized radiances in the sun-principal plane (Li et al., 2006). The total uncertainty in optical depth is about 0.01–0.02 (Eck et al., 1999).

The instruments at the three stations are calibrated annually to assure the accuracy and reliability of the data. Sun-photometers used at Shangdianzi (n=405) were calibrated at Izana Observatory of Spain by the facilities following the AERONET calibration protocol method in 2004. Those at Lin’an (n=475) and Longfengshan stations (n=476) were calibrated at Izana in 2005. In 2007, all three instruments were inter-compared in Beijing Lingshang Mountain (115°29.76’E, 40°02.96’N, ~1517 m) with one new sun-photometer (n=584) calibrated in Izana. In 2008, all these instruments have been re-calibrated by using two CARSNET masters (n=631 and n=647). During the inter-calibration process, only the raw data during 10:00 AM to 2:00 PM (local time) on the clean and clear days were used. The AOD at 500 nm measured by the master instruments on the calibration day should be less than 0.20 and without much fluctuation. The interval of the measurements between the two masters and the instruments to be calibrated should be less than 10 s.

The sun-photometer data at Lin’an, Shangdianzi and Longfengshan regional background stations are from January of 2006 to December of 2007. The PM10 chemical composition data are from the analysis of filter samples at Lin’an and Longfengshan regional background stations from January of 2006 to December of 2006 by a portable aerosol collector (America Airmetrics Co. Ltd.). The sampling frequency is twice a week and sampling period is from 9:00 AM to 9:00 AM of the next day. Samples are continually measured at 9:00 AM to 9:00 AM of the next day. Samples are continually collected for 24 h. The ion concentrations of F−, Cl−, NO3, SO4−2, Na+, NH4+, K+, Mg2+ and Ca2+ in PM10 are tested by using an ICS3000 ion chromatography.

2.3. The calculation of aerosol optical properties

2.3.1. The calculation of aerosol optical depth


\[ I(\lambda) = \frac{I_0(\lambda)}{R^2} \exp \left( -m_r(\theta) \frac{P}{P_0} \tau_r(\lambda) - m_o(\theta) \tau_o(\lambda) - m_a(\theta) \tau_a(\lambda) \right) \]

where \( I \) is the measured irradiance (in arbitrary units), \( I_0 \) the calibration constant, \( R \) the Sun–Earth distance (in astronomical units), \( m_r \), \( m_o \), and \( m_a \) are the respective air masses for molecular scattering, ozone absorption and aerosol extinction with corresponding optical depth \( \tau_r(\lambda) \), \( \tau_o(\lambda) \), \( \tau_a(\lambda) \), \( \theta \) is the apparent solar zenith angle, \( p \) is the actual and \( p_0 \) the standard atmospheric pressure.

Aerosol optical depth can be calculated by the following formula:

\[ \tau_a(\lambda) = \frac{1}{m_a(\theta)} \left( \ln \frac{E_d(\lambda)}{E(\lambda)} \left[ \frac{m_r(\theta)}{P_0} \tau_r(\lambda) - m_o(\theta) \tau_o(\lambda) - m_a(\theta) \tau_a(\lambda) \right] \right) \]

where \( E_d \) is the observation value of the instrument, \( E_o,d \) is calibration coefficient, \( m_d(\theta) \), \( m_r(\theta) \), \( m_o(\theta) \), \( m_a(\theta) \) are calculated according to:

\[ m_a = \sin(e) + 0.0548 \times (e + 2.65)^{-1.452} \]

(Kasten, 1966)

\[ m_r = \frac{1}{\sin(e) + 0.50572(e + 6.07995)^{-1.6364}} \]

(Kasten and Young, 1989)

\[ m_o(\theta) = \frac{R + h}{\sqrt{(R + h)^2 - (R + r)^2 \cos^2(e)}} \]

(Komhyr et al., 1980); where \( e \) is the apparent solar elevation angle, \( R = 6370 \) km the mean Earth radius, \( r \) the station height above sea level in km and \( h \) the height (~22 km) of the ozone layer. The aerosol air mass is further approximated by water vapor air mass, which has similar scale height.

Rayleigh scattering optical depth is calculated using the following method:

![Fig 1. Geographical locations of Lin’an, Shangdianzi, and Longfengshan stations.](image-url)
\[ \tau_r(\lambda) = 0.00864 \times \lambda^{-\left(3.916 + 0.074\lambda + \frac{0.050}{\lambda}\right)} \times \frac{p}{1013.5} \]  

(Fröhlich and Shaw, 1980)

The optical depth of ozone absorption is calculated according to:

\[ \tau_{O_3}(\lambda) = a_{O_3}(\lambda) \times O_3 \times m_{O_3}, \]

where \( m_{O_3} \) is the air mass of Ozone, \( a_{O_3}(\lambda) \) is the absorption coefficient of ozone, \( O_3 \) is the quantity in atmosphere with unit of Dobson.

2.3.2. Calculation of Angstrom wavelength exponent

Angstrom (1929) proposed that the optical thickness of aerosol in atmosphere is decided by the following formula:

\[ \tau(\lambda) = \beta \lambda^{-\alpha} \]

where \( \alpha \) is wavelength exponent, which can reflect the size characteristics of particle diameter. The range of \( \alpha \) is about \( 0 < \alpha < 2 \) with the average of \( \alpha \) about 1.30. A smaller \( \alpha \) represents dominant coarse aerosol particles while a larger \( \alpha \) represents dominant fine particles. The range of Angstrom turbidity coefficient \( \beta \) is 0–0.50. When \( \beta \leq 0.10 \), it represents that the weather is clean, while when \( \beta \geq 0.20 \), it represents that the weather is turbid.

The optical depth calculated is Level 1.0 data, comprising the effects of clouds. In order to eliminate the effect of clouds, the cloud-screening method is implemented according to Smirnov et al. (2000). During the cloud-screening procedure, the basic principles mainly include: (1) the threshold of AOD less than –0.01 is set up to eliminate the noise may caused by the calibration, temperature correction at the wavelength 1020 nm, atmospheric pressure, and column ozone amount; (2) the variability in the triplet should be less than the maximum of either 0.02 or 0.03\( \tau_a \) (\( \tau_a \) is average of a triplet); (3) the standard deviation of the AOD at 500 nm for the entire should be less than 0.015; (4) the smoothness criteria (the first derivatives difference) should be no more than 16. Finally, the Level 1.5 AOD data for three stations are obtained.

3. Monthly and seasonal variations of AOD over three stations

Fig. 2 and 3 are the monthly and seasonal variations of AOD at the three stations, respectively. Fig. 2a shows that the AOD has bimodal distribution with great fluctuation in Lin’an in June and September. The maximum AOD at 440 nm is about 1.30, and the minimum is about 0.75. This fluctuation of AOD in Lin’an may be related to weather patterns. Cold air above the lower Yangtze River Delta Region is more active in winter. The diffusion of pollutants is accelerated because of the strong wind, which leads to a smaller aerosol optical depth in Lin’an. Moreover, Lin’an could be affected by the dust storm from North China as well as local pollution sources in spring when precipitation is relatively low. The longer residence time of aerosol particles in the atmosphere could cause large aerosol optical depth in spring. Precipitation remarkably increases in the Yangtze River Delta in July and August and decreases the concentration of atmospheric aerosol. Thus the value of the aerosol optical depth is the lower in July and August. Because the subtropical anticyclonic begins to move to north and precipitation decreases from the end of August to the beginning of September, the AOD in Lin’an slightly increases in September. Due to the large fluctuation, the seasonal variation of the aerosol optical depth is not obvious. Little seasonal differences of AOD suggest that the emission and compositions of aerosol source in this region may be relatively constant. The emission of the main source in Lin’an mainly comes from ground source, coal combustion and the emission of manufacturing industry.

Figs. 2b and 3b are the monthly and seasonal variations of AOD at the Shangdianzi station, respectively. The monthly averaged AOD at the Shangdianzi station shows a significant character of single peak distribution. The peak value appears in June with the highest AOD of about 0.90 at 440 nm. The peak AOD in June could be
possibly due to the great water vapor content in the atmosphere in this period. Pollutants can easily absorb moisture and grow. In addition to the peak AOD in June, the AOD is also relatively high in spring because the precipitation is less in north China and dust events frequently affect this area (Cheng et al., 2008; Zhang et al., 2005).

The monthly averaged AOD at Longfengshan station (Figs. 2c and 3c) also shows significant seasonal variations. The values of AOD are higher in March–June, and the maximum appears in June. AOD at 440 nm is about 0.53. The minimum AOD value appears in October with AOD at 440 nm is about 0.30. Longfengshan station is located in the northeast of China where monsoon could affect greatly in spring. This area is liable to the affection of dust aerosol from Mongolia with high concentrations of the coarse dust particles in atmosphere. The aerosol optical depth at the Longfengshan station is also high in summer, which may be related to the high water vapor in atmosphere in summer.

4. Comparison of the AOD variations over three stations

Fig. 3 shows the seasonal variations of AOD at 1020 nm, 870 nm, 670 nm and 440 nm at three stations. The AOD at the Lin’an station is the largest at all wavelengths, followed by Shangdianzi, and the Longfengshan. The high AOD at Lin’an station could be caused by high particle loading in Yangtze River Delta Region. Yangtze River Delta Region is one of the most developed areas of China with many factories and large population. Furthermore, precipitation is plentiful in this region and the water vapor content in atmosphere is high during the whole year which could contribute to the high AOD. The aerosol optical depth at the Shangdianzi station is also high. The possible reason may be that the Shangdianzi station is also located in another industrially advanced area of Jing-Jin-Ji area. The larger aerosol optical depth in this area could be caused by industrial pollution throughout the year and dust events in spring (Zhang et al., 2005). The aerosol optical depth at the Longfengshan station is the lowest because the station is located in a national forest park in northeast China. There are few industrial pollution sources near the observation site and the low population makes Longfengshan station affected by less anthropogenic activities comparing to Lin’an and Shangdianzi stations.

5. Comparison of the Angstrom parameter over three stations

Figs. 4 and 5 are the monthly and seasonal variations of Angstrom wavelength exponents and turbidity parameters at the three stations. The Angstrom wavelength exponent at the Lin’an station is larger than that at the Shangdianzi and Longfengshan stations in most months. The Angstrom wavelength exponent at the Shangdianzi station is the lowest (Fig. 4a).

Generally, the Angstrom wavelength exponent at the Lin’an station is the lowest in spring at about 1.00 in April and May, and...
the highest in autumn at about 1.60 in August. The Angstrom wavelength exponent at the Lin’an station is smaller in spring which is probably due to the long-distance transport of dust particles from north China (Gong et al., 2003). In other seasons, pollutants in this area are mainly industrial pollutants caused by man-made emission or the products of photochemical reaction. The anthropogenic activity usually produces particles smaller than dust particles.

For the Shangdianzi station, the Angstrom wavelength exponent in spring is relatively smaller than that in other seasons (Fig. 5a). The averaged Angstrom wavelength exponent is about 0.83, which suggests that aerosol particles consist of many coarse particles. Except for the affection of Jing-Jin-Ji regional industrial pollution sources, Asian dust events and local dust emission can also contribute to the coarse particles of the Shangdianzi station in spring (Xia et al., 2006; Zhang et al., 2008).

Since that there is almost no pollution sources near the Longfengshan station, aerosol particles consist mainly of fine particles which are from local natural sources or through long-distance transport. The Angstrom wavelength exponent is larger than that at the Shangdianzi station, which reflects that the aerosol particles at the Longfengshan station are smaller than those at the Shangdianzi station. Angstrom exponent at Longfengshan is small in March which could reflect the effect of dust events in spring.

Characteristics of Angstrom turbidity coefficients at three stations are shown in Figs. 4b and 5b. The Angstrom turbidity coefficients (beta) of Lin’an are larger than 0.20 throughout the year which suggests that the weather is turbid in Lin’an station. Beta in Longfengshan station shows different characteristics from that of Lin’an. The values of beta are less than 0.20 in summer, fall and winter seasons which represents that the weather of Longfengshan is relatively clean. The beta values at Shangdianzi station are larger than 0.20 in spring and summer and less than 0.20 in fall and winter which suggests the weather in spring and summer at Shangdianzi is more turbid than that in fall and winter.

6. Relationship between AOD and the aerosol chemical composition

The correlations between AOD at 440 nm and the concentrations of PM10 and soluble anions and cations such as, $\text{SO}_4^{2-}$, $\text{NO}_3^-$, $\text{NH}_4^+$ are analyzed in order to study the main influencing factor of aerosol optical properties. The linearly dependent coefficient between AOD at 440 nm and the concentration of chemical composition at the Lin’an station is smaller and the linear dependence is not significant (Fig. 6). The correlation coefficient between daily averaged AOD at 440 nm and PM10 at the Lin’an station is only about 0.29 (Fig. 6a) through the statistics of 51 samples. The correlation coefficient has not passed through the 90% significance level. But the correlation coefficient between AOD at 440 nm and the concentration of $\text{SO}_4^{2-}$ is relatively high, which is about 0.39 (Fig. 6b). The correlation coefficients among the AOD at 440 nm and $\text{NO}_3^-$ as well as $\text{NH}_4^+$ are 0.19 and 0.33, respectively, which have not passed through 90% significance level either (Fig. 6c and d).

![Fig. 5. Seasonal variations of Angstrom parameters at three stations.](image)

![Fig. 6. Correlation among AOD at 440 nm and PM10, $\text{SO}_4^{2-}$, $\text{NO}_3^-$, and $\text{NH}_4^+$ at Lin’an station.](image)
Fig. 8a shows that the relationship between the aerosol optical depth and the concentration of SO$_4^{2-}$ is positive correlation under many circumstances, which suggests sulfate could probably contributed the AOD in Lin'an.

Compared with Lin'an station, the correlations between the aerosol optical depth and PM$_{10}$ as well as ion concentration at the Longfengshan station are higher. The correlation coefficients among the AOD at 440 nm and PM$_{10}$, SO$_4^{2-}$/C$_0$, NO$_3^-$/C$_0$, and NH$_4^+$ are 0.35, 0.43, 0.23 and 0.35 respectively (Fig. 7). However, all these correlation coefficients have not passed through obvious 90% significance level. Similar to Lin'an station, the temporal variations of AOD at 440 nm and the concentration of SO$_4^{2-}$ (Fig. 8b) show that the AOD at Longfengshan station is affected more by SO$_4^{2-}$ under many circumstances. However, negative correlation also exists under some situations. This kind of negative correlation needs to be studied in the future.

From above analysis of the correlations between the aerosol optical depth and particle chemical compositions, one can conclude that the particle concentration and chemical compositions could have effects on the aerosol optical properties. This effect is very complicated but not a simple linear relationship. The AOD measured by sun-photometer represents the aerosol extinction of whole column atmosphere. However, the aerosol concentration and chemical compositions just represent the aerosol of near surface layer of the atmosphere. More studies about the vertical distribution of aerosol particles are needed on this topic.

Fig. 7. Correlation among AOD at 440 nm and PM$_{10}$, SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$ at Longfengshan station.

Fig. 8. Comparison of the variations between AOD at 440 nm and the concentration of SO$_4^{2-}$ at Lin'an and Longfengshan stations.
7. Conclusions and discussion

The following conclusions can be obtained through the analysis of the aerosol optical depths, Angstrom wavelength exponents and the chemical components of aerosol at the Lin’an, Shangdianzi and Longfengshan regional atmospheric background stations.

The monthly aerosol optical depth at the Lin’an shows a double peak variation in a year. The large AOD values appear in May, June, July and September, respectively, with no significant seasonal variations. Aerosol optical depths are similar in spring and winter. Angstrom wavelength exponents show that the aerosol particles in this area consist mainly of fine particles.

The monthly aerosol optical depth at the Shangdianzi station shows a significant seasonal variation with a maximum value in June. The highest AOD appears in summer, followed by spring, and winter. The aerosol particles in this area consist of mainly coarse particles.

The aerosol optical depth at the Longfengshan station is lower compared to those at Lin’an and Shangdianzi stations, which shows that this region is comparatively clean compared to Lin’an and Shangdianzi. Due to the effect of dust events in North China, the aerosol optical depth in spring is significantly larger than that in other seasons. The sizes of the aerosol particles in this area are between that in the Lin’an and Shangdianzi.

The AOD results in this study are cloud-screened data. Most cloud effects have been filtered by the cloud-screening algorithm. However, there are still some exceptional cases in which the cloud effects could not be filtered, e.g. under the weather conditions of stable stratus. These cloud effects could make the AOD values at the three atmospheric stations higher than they really are. The data qualities need to be made sure further by combination of cloud observation, satellite measurements and other ground observation data.

There is a positive correlation between the aerosol optical depths and their chemical components at the Lin’an and Longfengshan stations, especially to SO$_4^{2-}$ concentration. However, their correlation coefficients are too low to reach the significance level due to the negative correlation among the AOD and PM10 as well as ion concentration in some cases. The mechanism of how aerosol chemical compositions affect aerosol optical depth is still need to be studied.

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