Aerosol Optical Properties Retrieved from a Prede Sky Radiometer over an Urban Site of Beijing, China

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Abstract

SKYNET is an international research network of ground-based Prede sky radiometers for the observation and monitoring of aerosol–cloud–radiation interactions in the atmosphere. The algorithm developed by SKYNET is SKYRAD.pack, which can be used to process the measurement data of Prede instruments. In this study, the latest SKYRAD.pack software (Version 5.0) has been used to retrieve the aerosol optical properties measured by a SKYNET Prede sky radiometer over an urban site of Beijing, China. Continuous data have been processed over a two-year period, and inversion products, including aerosol optical depth (AOD), Ångström exponent (\(\alpha\)), volumes of different aerosol particle size distributions, and single-scattering albedos (SSA), have been analyzed.
### 1. Introduction

Aerosol particles can change the Earth’s atmospheric radiation balance and affect the climate directly by absorbing and scattering solar radiation and radiation from the Earth’s atmosphere (Ackerman and Toon 1981). In order to estimate the radiative effect of atmospheric aerosols within the climate system, it is of the utmost importance to accurately determine their optical and radiative properties in the atmospheric column (Holben et al. 2001). When estimating aerosol parameters, the sun–sky radiometric technique is the most accurate and widely used (Holben et al. 1998).

SKYNET (Takamura and Nakajima 2004; Nakajima et al. 2007) is a research network located principally in Asia and Europe. Currently, it is composed of nearly 70 sites, which use a globally standardized sky radiometer (Prede Co. Ltd., Japan) to make measurements and collect data. The radiative and optical properties of aerosols can be retrieved from the combination of sun-direct and sky-diffuse radiation measured by the sky radiometer through the application of inversion algorithms (Nakajima et al. 1983, 1996). Retrieved properties include single-scattering albedos (SSA), phase functions, real and imaginary parts of the refractive index, and the volumes of different aerosol particle size distributions. The effective radius and asymmetry parameters can be also determined from the volumes of different aerosol size distributions and phase functions. Aerosol optical properties obtained from SKYNET have been used to investigate temporal and spatial characteristics of aerosols for climate and environmental studies and to validate satellite remote sensing results (Higurashi and Nakajima 2002; Kim et al. 2005; Sohn et al. 2007; Pandithurai et al. 2009; Campanelli et al. 2010; Khatri et al. 2010; Takenaka et al. 2011).

Nakajima et al. (1996) implemented an original algorithm called SKYRAD.pack. This program was developed to be applied to Prede radiometers and constitutes the starting point for the SKYNET international network. Several different versions of the inversion algorithms have been proposed over the years. Currently, the SKYNET uses the SKYRAD.pack algorithm version 4.2 (Takamura and Nakajima 2004). Some researchers have used the SKYRAD.pack version 4.2 algorithm to retrieve aerosol optical properties (Che et al. 2008; Estellés et al. 2012). In version 4.2, the kernel functions are computed with the nonsphericity effects (Kobayashi et al. 2010). The latest version, the SKYRAD.pack version 5.0, uses \textit{a priori} estimation (Hashimoto et al. 2012).

The high aerosol burden regions, such as areas in China and India, are still not well characterized in terms of microphysical and optical properties. Beijing is a typical example of high aerosol burden regions in China because of the increasing number of motor vehicles and energy consumption. Therefore, aerosol optical property observation in this area is of common interest within the scientific community (Che et al. 2014). The aim of this study is to analyze approximately two years’ worth of continuous observations made by the Prede sky radiometer in an urban site of Beijing using the latest version of the SKYRAD.pack, version 5.0. The results of this study may be taken more widely as indicative of aerosol optical properties due to the heavy anthropogenic aerosol loading in urban areas of China and other regions of East Asia.

### 2. Instrumentation and retrieval method

#### 2.1 Instrumentation and data

There are two types of Prede sky radiometers, POM-01 and POM-02. The POM-01 model uses a
silicon photodiode sensor to measure direct solar irradiance and sky radiance within a 1.0° full field-of-view for seven wavelengths of 315, 400, 500, 675, 870, 940, and 1020 nm at every 10 or 15 minutes (Uchiyama et al. 2005). Sky radiance is measured at 24 predefined scattering angles and at regular time intervals. The POM-02 model is equipped with an InGaAs detector, thus extending the wavelength to 11 wavelengths, including 315, 340, 380, 400, 500, 675, 870, 940, 1020, 1627, and 2200 nm, for cloud applications. The precision of the in situ method has been estimated to be within 1 %–2.5 % depending on the wavelength (Campanelli et al. 2004).

A Prede POM-02 sky radiometer was installed in May 2008 at the Chinese Academy of Meteorological Sciences (CAMS, 116.317 E, 39.933 N, 105 m) in the northern urban area of Beijing to measure aerosol optical properties and has been continuously running since then. In this study, data from 5 channels at 400, 500, 675, 870, and 1020 nm from June 2008 to May 2010 at Beijing were used to retrieve aerosol optical properties, including the aerosol optical depth (AOD), Ångström exponent ($\alpha$), the volumes of different aerosol particle size distributions ($dV/dlnr$), and single-scattering albedos. In this study, the four seasons considered are spring (March to May inclusive), summer (June to August inclusive), autumn (September to November inclusive), and winter (December to February inclusive), as part of the investigations into the seasonal variations in aerosol optical properties. Continuous measurement data for April 2010 is unavailable because of a gap in the data due to the atmospheric conditions prevailing at the time and subsequent problems with the instrument.

2.2 Retrieval methods

In this study, aerosol optical properties were retrieved using SKYRAD.pack version 5.0. SKYRAD.pack is an open source software package released on the OpenCLASTR web page (http://157.82.240.167/~clastr/), which consists of two different modules called “dtform” and “sproc.” The “dtform” module formats the Prede sky radiometer data so it can be processed by the “sproc” module. AODs, SSA, complex refractive indices, and aerosol particle size distributions were derived using a radiative transfer code as well as a linear and nonlinear inversion scheme (Nakajima et al. 1996).

In SKYRAD.pack version 5.0, the MLM method defined by Rodgers (2000) was used, based on the Bayesian theory:

$$p(x|f) = \frac{p(f|x)p(x)}{p(f)}$$  (1)

where $p$ is the probability density function and is defined as the Gaussian distribution, and $x$ and $f$ denote state and measurement vectors, respectively. In the MLM method, $x$ is chosen so that the posterior probability $p(x|f)$ is maximized under the conditions wherein a priori information has already been supplied. Organizing this nonlinear equation such that $p(x|f) = \max$, the following equation was obtained in tangential space for solution according to the Newtonian method:

$$x_{k+1} = x_k + \left(U_k^T S_k^{-1} U_k + S_a^{-1}\right)^{-1}$$

$$\left[U_k^T S_k^{-1} (f - f_k) - S_a^{-1} (x_k - x_a)\right]$$  (2)

where $x_k$ is the solution at the $k$-th iteration step, $f_k = f(x_k)$ is an observation modeled using $x_k$, $x_a$ is the a priori value of $x$, $S_k$ is the measurement error covariance matrix, $S_a$ is the covariance matrix defined by a priori and state values, $S_n = [(x - x_0)(x - x_0)^T]$, and $U$ is the Jacobi matrix, $\partial f/\partial x$.

Thus, the retrieval algorithm used in version 5.0 allows for the rigorous retrieval of both the aerosol size distribution and the spectral complex refractive index.

Version 5.0 also uses an a priori volume size distribution function of a bimodal log-normal function,

$$v(r) = \sum_{n=1}^{2} C_n \exp \left[-\frac{1}{2} \left(\frac{\ln r - \ln r_{mn}}{\ln S_n}\right)^2\right]$$  (3)

where $r_{m1} = 0.1 \mu m$, $r_{m2} = 2.0 \mu m$, $S_1 = 0.4$, $S_2 = 0.8$, $C_1 = 1.0 \times 10^{-12}$, and $C_2 = 1.0 \times 10^{-12}$, each following reported climate values (Higurashi et al. 2000). For a priori estimates of the real part ($n$) and the imaginary part ($m$) of the refractive index, $n$ is set as 1.50 and $m$ is set as 0.005, both of which are spectrally independent values.

3. Results and discussion

3.1 Temporal variation in AOD, $\alpha$, and SSA values in Beijing

The monthly AOD values at 400, 500, 675, 870, and 1020 nm over the two years of measurements from June 2008 to May 2010 in Beijing are shown in Fig. 1. The AOD value was found to vary from 0.13 (5th percentile) to 1.44 (95th percentile) with a median of 0.43 at 400 nm, 0.11 (5th percentile) to 1.14 (95th percentile) with a median of 0.34 at 500 nm, 0.08 (5th percentile) to 0.84 (95th percentile)
with a median of 0.24 at 675 nm, 0.07 (5th percentile) to 0.67 (95th percentile) with a median of 0.20 at 870 nm, and 0.06 (5th percentile) to 0.52 (95th percentile) with a median of 0.16 at 1020 nm. Temporal variations in Beijing indicate that the AOD increases from mid-winter to early summer and then decreases from mid-summer to early winter. The maximum (minimum) value of the AOD was recorded during May and June (December and January).

In this study, the AODs obtained at 500 and 870 nm have been used to calculate $\alpha$ as a qualitative indicator of aerosol size distributions (Toledano et al. 2007; Kaskaoutis et al. 2007; Masoumi et al. 2010). As Fig.1f shows, monthly $\alpha$ values were found to vary from 0.38 (5th percentile) to 1.43 (95th percentile) with a median of 1.00. The maximum and minimum values were ~1.80 and near zero or even negative, respectively.

SSA is defined as the ratio of the scattering coefficient to the extinction coefficient of a particle at a specific wavelength and varies between 0 and 1. Its value is mostly dependent upon the chemical composition, shape, size distribution, and matter concentration of aerosol particles (Jacobson 2000). Aerosols that absorb more light have a lower SSA than purely light-scattering ones, after SSA values have been standardized (CCSP 2009). Monthly variations in SSA values at 400, 500, 675, 870, and 1020 nm are shown in Fig. 2. The SSA value displays a high degree of uncertainty when the AOD at 500 nm is lower than 0.40 (Dubovik and King 2000). In this study, all SSA values with an AOD <0.40 at 500 nm have been discounted. SSA values range from 0.88 (5th percentile) to 0.97 (95th percentile) with a median of 0.93 at 400 nm, 0.90 (5th percentile) to 0.98 (95th percentile)
with a median of 0.96 at 500 nm, 0.89 (5th percentile) to 0.99 (95th percentile) at 675 nm, 0.84 (5th percentile) to 0.99 (95th percentile) at 870 nm, and 0.83 (5th percentile) to 0.99 (95th percentile) with a median of 0.94 at 1020 nm.

3.2 Frequency distribution of AOD, α, and SSA values in Beijing

The frequency distribution of AOD values at 500 nm and the corresponding α values are shown in Figs. 3 and 4. The bin interval for the AOD and α values was 0.10. Frequency histograms of AOD values at 500 nm showed obvious single-peak distributions. The average annual mode of readings within the range 0.00–0.50 was 72.4 %, most likely reflective of local background atmospheric conditions in Beijing (Fig. 3a). The frequency distribution between 0.00 and 0.50 accounts for 69.5 %, 59.2 %, 76.2 %, and 79.0 % of the total readings in spring, summer, autumn, and winter, respectively. The high aerosol burden in the atmosphere, such as the dust transported from northwest China during the spring (Wang et al. 2008b) and secondary organic aerosol particles prevalent in summer (Sun et al. 2010) may cause the large-scale depletion of the atmospheric column.

The α frequency histograms show differing ranges for the four seasons (Fig. 4). The frequency distribution of α values within the range of −0.20 to 0.40 accounts for 15.8 %, 3.9 %, 5.5 %, and 7.1 % of the total readings in spring, summer, autumn, and winter, respectively. The frequency distribution of α values larger than 1.50 accounts for 2.6 %, 8.6 %, 3.4 %, and 1.0 % of the total readings in spring, summer, autumn, and winter, respectively. These results indicate that there are larger particles present in the air in spring and winter, while there are finer particles present in summer and autumn.

The frequency distribution of SSA values between 0.90 and 0.99 accounts for 95.8 %, 96.6 %, 90.3 %,
and 82.8% of the total readings in spring, summer, autumn, and winter, respectively (Fig. 5). The high SSA values in spring could be due to the coarse particles that are present in that season. In summer, vertical air turbulence is strong and greater numbers of particles are transported through uplift into the air. Because of the high water vapor content in the atmosphere during this period, pollutants can easily absorb moisture, grow, and finally enhance aerosol scattering (Yan et al. 2009). Approximately 17% of SSA values are less than 0.90, which may be due to their absorption by aerosols during winter. Li et al. (2013) found that the proportion of black carbon and brown carbon particles in the air can reach between 25% and 38% during pollution events in winter.

3.3 Seasonal characteristics of AOD, $\alpha$, and SSA values in Beijing

Aerosol optical properties in Beijing exhibit significant seasonal variance. Figure 6a shows seasonal variations in AOD values at different wavelengths. The AOD decreases depending on wavelength during every season. Higher and lower AOD values at 500 nm occur in summer and winter with values of 0.56 ± 0.50 and 0.33 ± 0.28, respectively (Table 1). High AOD values at 500 nm also occur in spring with values of 0.43 ± 0.37. This seasonal AOD pattern may be related to weather conditions. Dust events during spring can induce higher AOD values (Che et al. 2009). Average AOD values for the whole winter appear to be lower; this may be due to frequent cold air events. The diffusion of pollutants may be accelerated by strong winds, thus leading to lower AOD values in winter (Giavis et al. 2005).
Figure 6b shows the seasonal distribution of $\alpha$ values. The boxes in Fig. 6b correspond to 50% of the values' distribution (from 25% to 75%), while the squares and lines within the boxes indicate the mean and median values, respectively. The “×” and “-” symbols correspond to 1%/99% and min/max values. $\alpha$ values are higher in summer and lower in spring with values of 1.05 ± 0.36 and 0.82 ± 0.39, respectively (Table 1). $\alpha$ values are lower in spring, indicating that aerosol particle sizes are larger. This most probably is a result of the long-distance transport of dust particles from north and northwest China (Wang et al. 2011). $\alpha$ values in summer are commonly higher than in other seasons, indicating that fine particles predominate during the summer season. Pollutants are mainly industrial and are caused by man-made emissions or are the product of photochemical reactions (Sun et al. 2010).

Average SSA values are commonly higher in spring and summer with a maximum average value of 0.96 ± 0.03 and lower in autumn and winter with a minimum average value of 0.93 ± 0.04 at 500 nm (Table 1).

The result above shows that the aerosol scattering ability is stronger in the spring and summer, while in autumn and winter there is a greater proportion of absorption component in the atmosphere. The value of SSA in spring is higher, which may be caused by the coarse particles from the dust events in spring. The high values of SSA in summer may have an important relation with the generation of secondary aerosol particles. Larger contents of absorption aerosols such as BC (black carbon) aerosol in winter result in lower SSA values.

Figure 6c also shows seasonal average SSA values for wavelengths of 400, 500, 675, 870, and 1020 nm. SSA values vary between 0.89 and 0.96, 0.96 and 0.94, 0.96 and 0.93, and 0.91 and 0.93 during spring, summer, autumn, and winter,
respectively, for these 5 different wavelengths. For two infrared wavelengths (870 nm and 1020 nm), SSA values showed a slightly decreasing trend with increasing wavelengths; however, for visible wavelengths (400 nm and 500 nm), the SSA values showed an increasing trend. In summer, the SSA values showed a significant decreasing trend from 500 to 1020 nm. During the other seasons, the SSA values showed an increasing trend with middle wavelengths (500 nm and 675 nm).

3.4 Seasonal aerosol volume size distributions in Beijing

Aerosol size properties are one of the most important sources of information for both the observation and modeling of radiative forcing (Dusek et al. 2006). Aerosol size distributions vary greatly under different atmospheric conditions such as clear air or heavy pollution episodes. Figures. 7a–d show the seasonally averaged volumes of different aerosol particle size distributions (dV/dlnr) for spring, summer, autumn, and winter in Beijing. These particle size distributions show typical bimodal patterns during every season. In spring, the effective radii of fine and coarse mode particles are about 0.12 and 3.80 µm, respectively, whereas in summer, the effective radii are about 0.18 and 3.80 µm, respectively. Fine mode volumes exhibit effective radii of 0.15 µm in autumn and 0.10 µm in winter, while coarse mode volumes show effective radii of 4.00 µm in both autumn and winter.

Size distributions show distinct differences in their dominant modes for the different seasons. In spring, the volume of coarse aerosol particles relative to the whole is much larger than for other seasons, probably due to the presence of dust particles of a relatively large size (Wang et al. 2010). However, in summer, the volume of fine mode aerosol particles is larger in relation to the total volume of particles as compared to other seasons, indicating that mainly fine particles
contribute to aerosol optical properties in summer. This increase in the volume of fine mode particles may be related to the growth of anthropogenic aerosol particles with strong hygroscopicity on summer days when water vapor content was found to be high (Li et al. 2010). The size distribution for autumn and winter exhibits a mixture of both fine and coarse mode particles.

Table 1. Seasonal AOD and SSA values at 400, 500, 675, 870, and 1020 nm and \( \alpha \) values (500–870 nm) for Beijing for the period from June 2008 to May 2010.

| AOD400 | AOD500 | AOD675 | AOD870 | AOD1020 | SSA400 | SSA500 | SSA675 | SSA870 | SSA1020 | \( \alpha \)500–870nm |
|--------|--------|--------|--------|---------|--------|--------|--------|--------|---------|------------------|------------------|
| Spring | 0.54±0.45 | 0.43±0.37 | 0.32±0.28 | 0.27±0.24 | 0.23±0.19 | 0.95±0.03 | 0.96±0.03 | 0.96±0.04 | 0.94±0.05 | 0.93±0.05 | 0.82±0.39 |
| Summer | 0.71±0.61 | 0.56±0.50 | 0.39±0.38 | 0.31±0.30 | 0.24±0.23 | 0.96±0.03 | 0.96±0.03 | 0.94±0.05 | 0.93±0.06 | 0.91±0.07 | 1.05±0.36 |
| Autumn | 0.47±0.39 | 0.37±0.31 | 0.26±0.22 | 0.20±0.17 | 0.16±0.13 | 0.91±0.05 | 0.95±0.03 | 0.95±0.04 | 0.94±0.05 | 0.92±0.06 | 1.02±0.33 |
| Winter | 0.42±0.36 | 0.33±0.28 | 0.23±0.20 | 0.18±0.16 | 0.16±0.13 | 0.89±0.05 | 0.93±0.04 | 0.94±0.03 | 0.95±0.04 | 0.93±0.05 | 0.97±0.35 |

3.5 Relations between AOD, \( \alpha \), and SSA values in Beijing

A scatter graph of instantaneous AOD and \( \alpha \) values is shown in Figs. 8a–d. We can see from the scatter graph that higher \( \alpha \) values accompany an increase in AOD values for all four seasons, evincing the urban aerosol characteristics typical of Beijing. Kim et al. (2004) demonstrated that the increase in AODs alongside increased \( \alpha \) values is related to the year-long accumulation process of urban aerosols. High AOD values are related to the presence of fine particles,
which play an important role in the aerosol optical properties of Beijing.

Figures 9a and 9b show the scatter graph of SSA and $\alpha$ values at 400 nm and 1020 nm. The frequency for $\alpha > 0.80$ and SSA $< 0.90$ at 400 nm is 53.9 % and the frequency for $\alpha > 0.80$ and SSA $> 0.90$ at 1020 nm is about 69.2 %. This means that the absorption of fine particles occurs with a greater intensity at short wavelengths and becomes more scattered over infrared wavelengths. During dust events, $\alpha$ values are lower while AOD values are higher, and the $(\text{SSA}_{400\text{ nm}} - \text{SSA}_{1020\text{ nm}})$ value is negative. In the past, dust criteria have been determined after conducting multiple sensitivity tests using both the mean and standard deviation. In this study, the dust criteria were set as $\alpha < 0.47$ and SSA$_{400\text{ nm}} - \text{SSA}_{1020\text{ nm}} < 0.018$. Figures 10a–d show the scatter graphs of SSA$_{400\text{ nm}} - \text{SSA}_{1020\text{ nm}}$ and $\alpha$ values for the four different seasons. Under these dust criteria, dust type aerosol accounts for 4.1 %, 5.1 %, 0.5 %, and 1.2 % of total readings in spring, summer, autumn, and winter, respectively. Dust occurrence probability is higher in spring and summer than in autumn and winter. These dust cases could probably be caused by both the natural dust transportation (Zhang et al. 2003) and local fugitive dust emissions in Beijing (Fan et al. 2009). Fugitive or open emissions can dominate the potential environmental impacts of a chemical process. The dust occurrence is significantly increased in spring possibly because of dust events, while in summer, the reason is most likely the strong vertical turbulence which induces more particles to be transported into the air.

The dust occurrence in summer is more close to that in spring. This may be because of an obvious decreasing trend of long term variation of dust events in North China during recent decades (Wang et al. 2010). In addition, the longer and stronger solar irradiation during summer favors the photochemical formation of secondary aerosol particles, which are the major constituents of PM$_{2.5}$ (Wang et al. 2008a).
Fine particles may increase the fugitive emissions in summer to a certain extent. Fan et al. (2009) pointed out that the highest fugitive dust emission of Beijing occurred during summer. Zhang et al. (2012) assessed the major chemical compositions in <10-micron particles using at least an entire year data from various rural and urban sites in 16 areas of the world. The concentration level of mineral aerosols in China, which is partially a result of sand and dust transported from desert areas and contributions from urban fugitive dust/fly ash sources, is found to be almost equivalent to or even higher than the sum of all kinds of aerosols in urban Europe and North America.

All the above reasons could contribute to the higher dust occurrence probability during summer in Beijing than during spring.

4. Summary

Aerosol optical properties over an urban site of Beijing were investigated continuously over two years using sky radiometer measurement data. The results can be summarized as follows.

The AOD values in the urban region of Beijing was found to vary from 0.11 (5th percentile) to 1.14 (95th percentile) with a median of 0.34 at 500 nm. The maximum and minimum $\alpha$ values are $1.05 \pm 0.36$ in summer and $0.82 \pm 0.39$ in spring. SSA values are higher in summer and spring but lower in winter, suggesting more absorption by aerosol particles in winter than in other seasons.

The frequency distribution of AOD values within the range 0.00–0.5 accounts for 69.5 %, 59.2 %, 76.2 %, and 79.0 % of the total readings in spring, summer, autumn, and winter, respectively. The frequency of $\alpha > 1.50$ is greater than 8 % in summer while it is less than 4 % in the other seasons. These results indicate that there are coarser particles in the air in spring and finer particles in summer. The frequency distribution of SSA values within the range 0.90–0.99 accounts for 95.8 %, 96.6 %, 90.3 %, and 82.8 % of the total readings in spring, summer, autumn, and winter, respectively, suggesting

Fig. 8. Seasonal scatter graphs of AOD at 500 nm versus $\alpha$ values (500–870 nm).
Fig. 9. Seasonal scatter graphs of SSA values at 400 (a) and 1020 (b) nm versus α values (500–870 nm).

Fig. 10. Seasonal scatter graphs of SSA values between 400 nm and 1020 nm versus α values (500–870 nm).
that there are more absorbent aerosol particles present in winter than in the other seasons.

The effective radii of fine mode particles is larger in summer than in the other seasons, while the effective radii of coarse mode particles in autumn and winter is larger than in spring and summer. With an increase in AOD values, the $\alpha$ value begins to increase, indicating that fine particles play an important role in the aerosol optical properties of Beijing. Dust type aerosol occurrence accounted for 4.1%, 5.1%, 0.5%, and 1.2% of all measurements data in spring, summer, autumn, and winter, respectively.

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