Improving Calculation Accuracies of Accumulation-Mode Fractions Based on Spectral of Aerosol Optical Depths

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Abstract. Anthropogenic aerosols are released into the atmosphere, which cause scattering and absorption of incoming solar radiation, thus exerting a direct radiative forcing on the climate system. Anthropogenic Aerosol Optical Depth (AOD) calculations are important in the research of climate changes. Accumulation-Mode Fractions (AMFs) as an anthropogenic aerosol parameter, which are the fractions of AODs between the particulates with diameters smaller than 1μm and total particulates, could be calculated by AOD spectral deconvolution algorithm, and then the anthropogenic AODs are obtained using AMFs. In this study, we present a parameterization method coupled with an AOD spectral deconvolution algorithm to calculate AMFs in Beijing over 2011. All of data are derived from AErosol RObotic NETwork (AERONET) website. The parameterization method is used to improve the accuracies of AMFs compared with constant truncation radius method. We find a good correlation using parameterization method with the square relation coefficient of 0.96, and mean deviation of AMFs is 0.028. The parameterization method could also effectively solve AMF underestimate in winter. It is suggested that the variations of Angstrom indexes in coarse mode have significant impacts on AMF inversions.

Keywords: Accumulation-mode fraction, Angstrom index, coarse-mode

1. Introduction
Accumulation-mode fraction (AMF) is an important parameter to distinguish the anthropogenic and natural aerosol optical depth (AOD)\(^{[1,6]}\). It is defined as a fraction of accumulation-mode AOD in the total AOD.

Tanre et al.\(^{[7]}\) proposed a method to calculate look-up table for bimodal normal distribution of aerosol, retrieved the aerosol optical thickness and accumulation-mode fraction using MODIS observations over ocean and land. However, there are significant errors when aerosol size distributions and complex refractive indexes are assumed. O’Neill et al.\(^{[8,9,10]}\) estimated AMF using aerosol optical depth spectral deconvolution algorithm (SDA), which only needed the input of multi-wavelength
AOD. But constant truncation radius in the inversion is not used to calculate AMFs in SDA. To obtain AMFs, it is calculated from aerosol volume distributions using Mie scattering code. More researchers do further study on fractions of accumulation-mode volumes. It is reported that Angstrom indexes are sensitive to fraction of accumulation-mode volume radii less than 0.6μm, calculated using 7 wavelengths aerosol optical thickness from AERONET website\(^{[11]}\). Gobbi et al.\(^{[12]}\) presented the graphical method to build the space of fitting polynomial parameters of multi-wavelength AOD. The relationship between fraction of accumulation-mode volume and aerosol size distribution is obtained in the coordinate system of graphical method.

In this paper, we focus on the impacts of coarse-mode aerosol index variations on AMF in spectral deconvolution algorithm. We present a parameterization of coarse-mode Angstrom index instead of the constant coarse-mode aerosol index in SDA to improve the AMF accuracies with AODs (Lev2.0) from AERONET website over 2011.

2. Data and Method

2.1. Data
We apply the inversion results from sun-sky radiometer to calculate AMFs and Angstrom indexes. The data includes multi-wavelength aerosol optical depths, aerosol volume distributions retrieved from AODs, complex refractive indexes and accumulation-mode fractions at Beijing site (116.38ºE, 39.98ºN) over 2011 from AERONET website (http://aeronet.gsfc.nasa.gov/). In our study, we choose 3 common wavelengths (440nm, 675nm and 870nm) as trial data from two types of instrument at Beijing site. Lev2.0 AOD, Lev2.0 inversion results and Lev1.5 AMF (denoted as AMF\(_{AERO}\), hereafter) are used to validate the AMF accuracies.

2.2. AMF derived from spectral of aerosol optical depth
We apply aerosol optical depth spectral deconvolution algorithm (SDA) to estimate AMF. The basic theory used in the algorithm is the total AODs are equal to the sum of fine-mode and coarse-mode AODs as follows:

\[
\tau_a = \tau_f + \tau_c ,
\]

where, \(\tau_a\) is aerosol optical depth; \(\tau_f\) is accumulation-mode aerosol optical depth; and \(\tau_c\) is coarse-mode aerosol optical depth. The derivation of multi-wavelength aerosol optical depth is Angstrom index (\(\alpha\)):

\[
\alpha = - \frac{d \ln \tau_a}{d \ln \lambda} = \frac{\alpha_f \tau_f + \alpha_c \tau_c}{\tau_a} ,
\]

where, \(\lambda\) is wavelength; \(\alpha_f\) is accumulation-mode Angstrom index; and \(\alpha_c\) is coarse-mode Angstrom index. If \(AMF = \frac{\tau_f}{\tau_a}\) then

\[
\alpha = \alpha_f AMF + \alpha_c(1 - AMF) ,
\]

The equation (3) about \(\alpha\) is transformed into that for AMF like:

\[
AMF = (\frac{\alpha_c - \alpha_f}{\alpha_f - \alpha_c}) ,
\]

where, AMF is accumulation-mode fraction.

In the SDA, \(\alpha_c\) is a statistical mean of coarse-mode Angstrom index, in other words, that is a constant (\(\alpha_c = -0.15\)). So derivation of coarse-mode Angstrom index equals to zero (\(\alpha'_c = 0.0\)). However, \(\alpha_c\) is not a constant in any situation, but varies with AOD, size distribution and complex refractive index. So, estimation errors of AMF from SDA derived from the coarse-mode Angstrom index.

2.3. Parameterization of coarse-mode Angstrom index
We calculated the variations of coarse-mode Angstrom index (Figure. 1) using aerosol volume distribution from AERONET website in 2011. Figure 1 shows that Angstrom indexes can be divided into 3 parts according to the range of \(\alpha_c\). The coarse-mode Angstrom indexes vary greatly in the range...
of -0.631 to -0.086 when the values of $\alpha$ are between 1.0 and 1.5, with the mean value of -0.358 (Table 1). However, mean $\alpha_c$ is -0.198 if $\alpha$ less than 1.0, in which case coarse-mode aerosol is dominant. When $\alpha$ is more than 1.5, the values of $\alpha_c$ are equal to -0.15, as large as that in SDA. But the case in this range is rare over 2011.

![Figure 1](image.png)

Figure 1. The variations of coarse-mode Angstrom index ($\alpha_c$). Scattering points are values of $\alpha_c$ calculated by Mie theory; dash line is $\alpha_c = -0.15$; solid line is the averaged $\alpha_c$ in the actual situation.

| $\alpha_c$ | $\alpha < 1.0$ | $1.0 \leq \alpha \leq 1.5$ | $\alpha > 1.5$ |
|------------|----------------|-----------------------------|----------------|
| maximum    | -0.072         | -0.086                      | ---            |
| minimum    | -0.323         | -0.631                      | ---            |
| mean       | -0.198         | -0.358                      | -0.15          |

To build the parameterization according to the relationship between $\alpha_c$ and $\alpha$ as follows:

\[
\begin{align*}
\alpha_c &= -0.198 \quad (\alpha < 1.0) \\
\alpha_c &= -0.358 \quad (1.0 \leq \alpha \leq 1.5) \\
\alpha_c &= -0.150 \quad (\alpha > 1.5)
\end{align*}
\]

(5)

Here, $\alpha_c$ is coarse-mode Angstrom index, and $\alpha$ is Angstrom index of the all particles.

2.4. Experiment design

In order to examine the impacts of $\alpha_c$ on AMFs and improve the accuracies of estimated AMFs, we preform two simulations. The first simulation is the control test with truncation radius $R=0.5\mu m$ (denoted as RCUT, hereafter). AMFs are calculated by Mie code using the inversion results of volume distributions as inputs. The second simulation estimates the AMFs at 500nm using SDA coupled with coarse-mode Angstrom index parameterization method (denoted as PARA, hereafter). In this study, AMFs from control test and contrast test are denoted as AMF$_{RCUT}$ and AMF$_{PARA}$, respectively.

3. Results

3.1. Aerosol optical thickness and Angstrom index

To analyse the impacts of coarse-mode Angstrom indexes on AMFs, we need to verify the control test with observations first. Aerosol optical depth at 440nm, Angstrom index calculated by Mie theory and that derived from AERONET are compared in figure 2a. The errors of AOD are less than 0.1 except two points whose AODs are more than 3.5. The mean values of AODs from AERONET and RCUT are 1.08 and 1.12, respectively, with the mean error less than 0.04 over 2011.
There is a good consistency between the trends of Angstrom indexes (440nm-870nm) from RCUT and that from AERONET website (Figure 2b). The maximum of Angstrom indexes from RCUT is 1.64, closed to 1.65 from AERONET. The errors are less than 0.13, with the mean error of 0.04.

3.2. Estimated accumulation-mode fraction

AMFs represent the extinction contribution of accumulation-mode aerosols whose radii are less than 0.5μm. So, AMFs estimated with a constant truncation radius from RCUT are more accurate. Figure 3 presents the results compared among the three tests. The correlation between AMF from PARA and that from RCUT (Figure 3a) is better than that between AMF from AERONET website and that from RCUT (Figure 3b). The square correlation coefficient in the Figure 3a is 0.96, better than 0.87 from the correlation between AMF_{AERO} and AMF_{RCUT} in Figure 3b. And the slope and intercept of the linear
fitting equation is 0.84 (standard error, 0.010) and 0.16 (standard error, 0.013), respectively. In contrast with the slope and intercept in Figure 3a, that between $\text{AMF}_{\text{AERO}}$ and $\text{AMF}_{\text{RCUT}}$ (Figure 3b) are 0.73 (standard error 0.021) and 0.29 (standard error 0.015), respectively. It is suggested that AMFs from PARA test can better describe the extinction contribution of accumulation-mode aerosols.

3.3. Error analysis of AMF

Figure 4 presents AMFs and errors from PARA test and AERONET. The trends of AMFs from two cases show excellent agreement, and AMFs are smaller in spring than in other seasons (Figure 4a). $\text{AMF}_{\text{PARA}}$ is almost larger than $\text{AMF}_{\text{AERO}}$ from January to May. The errors of AMFs are reduced in summer, smaller than 0.05 from June to August. And the errors are increased more in spring, especially during sand-dust and gray haze events (Figure 4b). The errors from PARA are smaller than that from AERONET, with the mean errors of 0.028 and 0.072 (Table 2), respectively.

The increases of the accumulation-mode aerosol extinctions are caused by winter heating (Julian Day over 1-75) leading to increasing soot concentrations in Beijing over 2011. In order to correct the underestimates of accumulation-mode aerosol extinctions, we involve the parameterization method of coarse-mode Angstrom index $es$. Table 2 presents maximum error of accumulation-mode fractions, which is corrected from 0.192 to 0.115.

![Figure 4](image)

**Figure 4.** (a) Comparison between AMF from PARA test and that from AERONET website; (b) the errors of $\text{AMF}_{\text{PARA}}$ and $\text{AMF}_{\text{AERO}}$ related to control run.

**Table 2.** The errors of $\text{AMF}_{\text{AERO}}$ and $\text{AMF}_{\text{PARA}}$ relative to the control test RCUT.

| Test    | AERONET | PARA  |
|---------|---------|-------|
| Maximum | 0.192   | 0.115 |
| Minimum | 0.000   | 0.000 |
| Mean    | 0.072   | 0.028 |

4. Conclusions

In this study, we couple the parameterization method of coarse-mode Angstrom indexes with aerosol optical depth spectral deconvolution algorithm to improve the accuracies of AMFs using sun-sky
radiometer observations in Beijing over 2011. Coarse-mode Angstrom indexes are derived from statistical calculation. It is indicated that errors of AMFs are reduced and get closer to the results from control test when involving the parameterization of \( \alpha_c \). Compared with AMFs from AERONET website, correlation coefficient \( R^2 \) increases from 0.87 up to 0.96. And using this method could effectively correct underestimates of AMFs in winter.

The errors of anthropogenic AODs associated with AMFs larger than 0.83 are \( \pm 0.05 \) in Brilouin’s research\(^[3]\). The errors of AMFs in our study are reduced from 0.072 to 0.028. It could effectively improve on accuracies of anthropogenic AODs.

It should be noted that Angstrom indexes in the spectral deconvolution algorithm have large impacts on AMF calculation. We discussed the impacts of \( \alpha_c \) on improving the accuracies of AMFs, and the overall effects of accumulation-mode Angstrom indexes will be the subject of forthcoming work.

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