Estimation of aerosol single scattering albedo from solar direct spectral radiance and total broadband irradiances measured in China

Fengsheng Zhao¹,² and Zhanqing Li¹

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Aerosol single scattering albedo ($\omega_o$) is a primary factor dictating aerosol radiative effect. Ground-based remote sensing of $\omega_o$ has been employed most widely using spectral sky radiance measurements made from a scanning Sun photometer. Reliable results can be achieved for high aerosol loadings and for solar zenith angle $>$50°. This study presents an alternative method using spectral direct radiance measurements or aerosol optical depths together with total sky radiance to retrieve $\omega_o$. The method does not require sky radiance data that can only be acquired by the expensive scanning Sun photometer. The method is evaluated using extensive measurements by a suite of instruments deployed in northern China under the East Asian Study of Tropospheric Aerosols: An International Regional Experiment (EAST-AIRE) project. The sensitivities of the retrieval to various uncertain factors were first examined by means of radiative transfer simulations. It was found the retrieval is most sensitive to cloud screening, total irradiance and the Angstrom Exponent (AE), but only weakly depends on surface albedo and the fine structure of aerosol size distribution. Using 1 year of rigorously screened clear-sky measurements made at the Xianghe site, the retrieved $\omega_o$ values were found to agree with those retrieved from the Cimel Sun photometer by the AERONET method to within $\sim$0.03 (RMS), and $\sim$0.003 (mean bias). As part of the differences originate from different sky views seen by the Sun photometers and pyranometer under comparison, a further test was conducted by using total sky irradiances simulated with the retrieved aerosol properties from the AERONET. The resulting estimates of $\omega_o$ agree to within 0.01–0.02 (RMS differences) and 0.002–0.003 (mean bias). These values are better measure of the true retrieval uncertainties, as they are free from any data mismatch. The characteristics of $\omega_o$ retrievals were discussed.


1. Introduction

Aerosols have a significant impact on global climate, either directly through scattering and absorption of solar radiation [Coakley et al., 1983; Charlson et al., 1992; Kiehl and Briegleb, 1993; Boucher and Anderson, 1995; Schwartz, 1996] or indirectly by modifying the microphysical properties of clouds [Twomey, 1977; Twomey et al., 1984; Coakley et al., 1987; Albrecht, 1989]. Aerosol absorption of solar radiation may also affect cloud formation and cloud amount [Hansen et al., 1997; Ackerman et al., 2000].

In recent decades, increasing effort has been devoted to the estimation of aerosol climate effects. Direct radiative forcing is estimated to be $-0.4 \text{ W m}^{-2}$ for sulphate aerosols, $-0.2 \text{ W m}^{-2}$ for biomass-burning aerosols, $-0.1 \text{ W m}^{-2}$ for fossil fuel organic carbon, and $+0.2 \text{ W m}^{-2}$ for fossil fuel black carbon aerosols [Intergovernmental Panel on Climate Change, 2001]. These estimates are subject to very large uncertainties. A chief cause for the large uncertainties is the lack of reliable information on the spatial and temporal distribution of aerosol properties. The aerosol single scattering albedo, $\omega_o$, defined as the fraction of the aerosol light scattering over the total extinction, is one of the most important properties of aerosols. The direct radiative effect of aerosols is very sensitive to $\omega_o$. For example, a change in $\omega_o$ from 0.9 to 0.8 can alter the sign of the direct effect, depending on the albedo of the underlying surface, aerosol backscattering fraction etc. [Chylek and Coakley, 1974; Hansen et al., 1997].

Because of technical difficulties, few instruments can measure this key aerosol variable directly. To date, the most widely used means of obtaining aerosol $\omega_o$ on large scales and on a routine basis is to retrieve it using ground-based Sun photometer data from the Aerosol Robotic Network (AERONET) (http://aeronet.gsfc.nasa.gov/) [Holben et al., 1998]. The AERONET network has been successfully developed for long-term observations at over 349 stations.
A pioneering study in retrieving aerosol properties from ground-based observations was conducted by King and Herman [1979] and King [1979] where a statistical algorithm was proposed for inferring the imaginary part of the aerosol index of refraction and surface albedo from the diffuse and direct solar radiation measurements. The algorithm works reasonably well in the case of a clear atmosphere devoid of any cloud cover for a long enough period of time with a large range of change in the solar zenith angle (~40°). A combination of measurements from a Sun photometer, a sky radiometer and a total broadband radiometer were also employed to derive broadband aerosol \( \omega_o \) by Silva et al. [2002]. More recently, Kassianov et al. [2005] described a technique to retrieve aerosol column number density, mean radius and \( \omega_o \) from observations made by the multifilter rotating shadowband radiometer. Compared to the measurements from AERONET, this technique has a higher temporal resolution (~20 s) and its accuracy is independent of the solar zenith angle. The technique can be successfully applied to cases where the aerosols are mainly composed of fine mode particles. If the relative contribution of coarse or giant particles (1 \( \mu \text{m} \) and larger) is significant, their technique fails to retrieve the aerosol properties.

In this study, aerosol \( \omega_o \) is retrieved using measurements of spectral direct solar radiation and broadband downwelling total solar radiation acquired in China for an entire year from 2004 to 2005. In our retrieval algorithm, sky radiation measurements are not needed. The retrieval accuracy is shown to be compatible with those from the AERONET retrievals for cases where both coarse and fine particles are abundant. Also, the accuracy does not deteriorate badly for small solar zenith angles. The proposed method has significant potential implications for studying aerosol effects on regional climate. Aerosol absorption in China has drawn much attention in recent years [e.g., Menon et al., 2002]. However, little quantitative information is available. Under the auspices of the East Asian Study of Tropospheric Aerosols: An International Regional Experiment (EAST-AIRE) project (Z. Li et al., Preface to special section: Overview of the East Asian Study of Tropospheric Aerosols: An International Regional Experiment (EAST-AIRE), submitted to Journal of Geophysical Research, 2007, hereinafter referred to as Li et al., submitted manuscript, 2007), a network of 25 stations spread throughout China provides measurements of direct spectral solar radiation [Xin et al., 2007] that may be used, in combination with total irradiance measurements, to provide nationwide estimates of aerosol \( \omega_o \).

In section 2, sensitivity analyses of \( \omega_o \) to broadband fluxes, size distribution and ground surface albedo are presented. In section 3, the proposed algorithm to retrieve aerosol \( \omega_o \) is discussed. Section 4 describes the observation site from which measurements used in this study were made, the instrumentation deployed and data processing procedures. Application of the algorithm and analysis of the \( \omega_o \) retrievals are given in section 5. A comparison with the AERONET retrievals is presented in section 6. Detailed error analyses are performed in section 7 and a summary of the results and conclusions is given in section 8.
Figure 1b shows the surface broadband downwelling SW fluxes calculated using different exponents in the Junge power law expression. The broadband fluxes increase rather dramatically with increases in the magnitude of the exponent. It is well known that the Angstrom exponent, $\alpha$, is related to the Junge power law exponent through the relation

$$\alpha = p - 3. \tag{3}$$

The value $\alpha$ describes a regression relationship between aerosol optical depths and wavelengths according to the expression

$$\tau(\lambda) = \beta \lambda^{-\alpha} \tag{4}$$

where $\tau$ is the aerosol optical depth at wavelength $\lambda$ and $\beta$ is a constant. It follows from equations (3) and (4) that the larger the value of $p$, the faster the optical depth decreases with increasing $\lambda$. So for the same value of optical thickness at 0.5 $\mu$m, the larger the value of $p$ and the larger the surface broadband downwelling SW flux. Estimation of $\omega_a$ from surface broadband downwelling SW fluxes thus requires information on the aerosol size a priori or simultaneously.

Although the Junge power law distribution has been widely used, many observational studies in recent decades show that the fine structure of the aerosol size distribution may be best described by the bimodal distribution of equation (2). It is of interest to look at the difference between the broadband downwelling SW fluxes calculated using the Junge power law distribution and the bimodal distribution for the same value of $\alpha$. Figure 1c shows the broadband downwelling SW fluxes calculated using the bimodal distribution with the following parameters: $r_{01} = 0.15$ $\mu$m; $r_{02} = 4$ $\mu$m; $\sigma_1 = 2.5$; $\sigma_2 = 4.5$, and $c_1/c_2 = 1$. We first calculated the spectral optical depth using the bimodal distribution then a linear regression technique is used to obtain $\alpha$. The exponent $p$ of the Junge power law distribution is calculated from equation (3) using this $\alpha$. It appears that the detailed structure of the size distribution has little effect on the broadband downwelling SW fluxes, indicating that the Junge power law can be used to approximate the bimodal distribution, provided that the Angstrom exponents derived from the two distributions are the same. This was also noted by Zhao and Nakajima [1997] and Zhao et al. [2002], where they illustrated the validity of using the Junge power law in the satellite remote sensing of aerosols.

Figure 1d shows the broadband downwelling SW fluxes calculated using different values for ground surface albedo. The magnitudes of the ground surface albedo are divided into two groups: one with magnitudes around 0.15 and the other with magnitudes around 0.75. The magnitudes of the broadband downwelling SW fluxes increases with increasing surface albedo because of multiple reflections. The surface albedo can be derived from satellite observations of broadband and narrowband radiances (such as
MODIS, AVHRR, ERBE, CERES, etc.) with an accuracy of about 0.01 ~ 0.03 [e.g., Li and Garand, 1994; Li et al., 2002; Liang, 2003; Liang et al., 2005]. Uncertainties in the surface albedo result in negligible errors in the broadband downwelling SW fluxes, e.g., an uncertainty of 0.05 in the surface albedo results in an error of less than about 4 W m$^{-2}$ in the broadband downwelling SW fluxes.

3. Algorithm for Determination of $\omega_0$

[14] According to the sensitivity analyses described above, for a fixed aerosol optical depth, the broadband downwelling SW fluxes depend mainly on $\omega_0$ and the spectral optical depth from which the Angstrom exponent, or the Junge power law exponent, can be calculated. An algorithm is developed to determine the aerosol $\omega_0$ from the observations of broadband downwelling SW fluxes and aerosol spectral optical depths. The imaginary part of the index of refraction is first estimated by matching measured and calculated broadband downwelling SW fluxes. In the flux calculations, the aerosol optical thickness at 0.5 $\mu$m and $\alpha$ is used. The aerosol size distribution is approximated by the Junge power law with its exponent calculated from the Angstrom exponent. An assumption is that the imaginary part of the index of refraction is independent of wavelength in the SW spectral region. We have performed numerical experiments to investigate the validity of the assumption. The results show that the assumption may cause errors of 0.03–0.04 in the case of hematite mixed with other mineral particles. However, for most aerosol types, such as rural, urban and dust-like aerosols, the results of the numerical experiments indicate that the wavelength independence assumption is reasonable. Especially, for the urban aerosol models in which black carbon absorption is dominating, the retrieved errors are less 0.01. The effect of water vapor on the broadband downwelling SW fluxes is considered in the flux calculations. The column precipitable water amount is determined from both supersites and a nationwide observational network [Xin et al., 2007]. Surface reflection measurements (whole sky cloud views, optical depth, cloud height, liquid water path); (4) aerosol quantities (optical depth, $\omega_0$, size distribution, mass and condensation number); (5) aerosol composition; and (6) precursor gases amounts (ozone, NO, NOx, NOy, CO, SO2).

[16] This study employs a small subset of observations made at Xianghe. The site is under the influence of the East Asian monsoon. From October to April, northwesterly winds prevail, blowing in air from Siberia and the Mongolia Plateau. During the spring season, the strong winds may transport desert dust and pollutants from Beijing to Xianghe, affecting local aerosol properties [Eck et al., 2005]. During the summer season, southeasterly winds are dominant and aerosol properties may be affected by humid oceanic air. The sulfate and carbonaceous particles produced by fossil fuel combustion and biomass burning are important components of the local aerosols. Except for summer, the climate in other seasons is dry. Locally produced windblown dust particles are also an important component of aerosols.

[17] Primary data used in this study are spectral solar direct radiation and broadband downwelling SW fluxes measured by a CIMEL CE-318 Sun photometer and a Kipp & Zonen CM21 pyranometer, although data from several other instruments were employed for selecting cloud-free measurements and for data quality control (described later). The Sun photometer measures the direct solar radiation at 0.340, 0.380, 0.440, 0.500, 0.675, 0.870, 0.940, and 1.020 $\mu$m with a 1.28$^\circ$ full field of view. Details of instrument calibration and data processing are described by Holben et al. [1998]. The errors in the derived aerosol optical depth are less than 0.01 for wavelengths >0.44 $\mu$m and 0.02 for shorter wavelengths. The Kipp & Zonen CM21 pyranometer is used to measure the broadband downwelling SW fluxes in the 0.305–2.80 $\mu$m spectral range. The instrument response time is 5 s and the data sampling time interval is 1 minute. The measurement error of the instrument is estimated to be less than 10 W m$^{-2}$. As a quality control measure, total irradiance was also observed simultaneously with three other instruments, namely a Kipp & Zonen Cimel-11, and Eppley 8–48 black and white pyranometer and normal incidence pyrheliometer. The Eppley instruments were mounted on a solar tracker to measure the direct and diffuse radiation. Discrepancies in total irradiance measurements made by the three independent instruments are usually within 5 W m$^{-2}$ and rarely exceed more than 10 W m$^{-2}$. Larger discrepancies occurred occasionally for which the causes were identified and the data were promptly corrected using independent measurements. The data have been collected continuously from September 2004 till the present.
A critical step of the data processing for this study is separation of cloud-affected data from cloud-free data because the retrieval of aerosol $\omega_0$ is susceptible to cloud contamination. A cloud-screening method proposed by Smirnov et al. [2000] was applied to AERONET data so that clear-sky data were selected in order to retrieve the aerosol optical depths. However, this method only identifies a cloud-free path between the Sun photometer and the solar disk, while the retrieval proposed by this study requires a hemispheric cloud-free sky. A much more rigid cloud screening method was performed that employed time series of radiation data and total sky images of high frequency obtained from a Total Sky Imager (TSI-440A) manufactured by Yankee Environmental Systems [Li et al., 2007]. Only Sun photometer measurements made when the whole sky is clear are employed in this study for two reasons. First, the aerosol optical depths are used together with surface total irradiance to retrieve the aerosol $\omega_0$. One may question the necessity for doing this, given that $\omega_0$ can be retrieved from Sun photometer measurements. It should be borne in mind that retrieving $\omega_0$ requires sky radiance data that can only be obtained by using a complicated and thus expensive Cimel Sun photometers. However, observing the aerosol optical depth can be achieved with a much simpler and cheaper device such as the handheld Sun photometers. The second usage of the subset of Cimel data is to evaluate the retrieval of $\omega_0$ by comparing the two types of retrievals.

5. Results and Discussion

The algorithm described in section 3 was applied to retrieve $\omega_0$ by combining aerosol optical depths at 0.5 $\mu$m, Angstrom exponents ($\alpha_{440-870}$), precipitable water amounts and observed broadband downwelling SW irradiances. According to the sensitivity analyses described in section 2, the retrieval of $\omega_0$ is only feasible for moderate and high aerosol loading. Only data with aerosol optical depths larger than 0.4 (at 0.44 $\mu$m) were selected for the analyses. The same constraint was applied in the AERONET retrievals of aerosol optical properties [Dubovik et al., 2002]. Such heavy aerosol loading is often observed at the Xianghe site [Li et al., 2007].

Data from September 2004 to August 2005 were analyzed with a total of 8513 samples. Figure 2 shows the statistics for the retrieved values of $\omega_0$. The average value is 0.9, with 50.4% of the retrieved values larger than 0.9, 48.3% between 0.80 and 0.9, and 1.3% smaller than 0.8. The average value of $\omega_0$ is nearly the same as those reported for Mexico City and the Maldives [Dubovik et al., 2002]. Aerosols with such values for $\omega_0$ can have a cooling or warming effect on the climate over land surfaces, pending on the magnitude of the surface albedo. The broadband surface albedo around the Xianghe site is about 15%, resulting in a neutral aerosol effect. Matching irradiance data at the top of the atmosphere (TOA) obtained by the Clouds and the Earth's Radiant Energy System instrument with ground-based aerosol data, Li et al. [2007] found that aerosols do not alter the radiation budget at the TOA regardless of aerosol loading, although they substantially reduce the surface radiation budget. This implies that there is strong atmospheric heating due to the aerosols. For the cases with $\omega_0 < 0.8$, these aerosols contain black carbon particles which are a highly absorbing species resulting from either industrial combustion or incomplete biomass.

Figure 2. Statistics associated with the retrieved aerosol single scattering albedo at 0.5 $\mu$m.

Figure 3. Scatterplots between (a) aerosol single scattering albedo at $\lambda = 0.5$ $\mu$m and Angstrom exponent, (b) aerosol single scattering albedo and aerosol optical depth and (c) Angstrom exponent and aerosol optical depth.
Such aerosols have a heating effect on the climate in general.

The magnitude of the Angstrom exponent is determined by the fraction ratio of fine and coarse modes. If the coarse mode is predominant, the Angstrom exponent is small, and vice versa [Eck et al., 2005]. Figure 3a shows the scatterplot of $\omega_o$, as a function of the Angstrom exponent. Although the data shown in Figure 3a are rather dispersed, a general trend can be seen: $\omega_o$ tends to decrease with increasing Angstrom exponent (increase in the fraction of fine-model particles). Fine-mode particles primarily originate from fossil fuel combustion and biomass burning, as well as by the oxidation of gaseous compounds such as sulfur dioxide, nitrogen oxides, and volatile organic compounds. Among these constituents, the black carbon particles from incomplete fossil fuel combustion and biomass burning are the strongest light absorbers. Therefore an increase in the concentration of fine mode particles tends to reduce $\omega_o$. In the Xianghe area under high aerosol loading, the coarse mode particles mainly consist of mineral dust. An increase in the dust particle concentration tends to increase the $\omega_o$ of the polydispersion. In previous studies, the values of $\omega_o$ reported from model simulations [e.g., Shettle and Fenn, 1979; World Meteorological Organization, 1983; Koepke et al., 1997], in situ measurements [e.g., Patterson et al., 1977; Haywood et al., 2001] and column-averaged retrievals [Kaufman et al., 2001; Tanré et al., 2001; Dubovik et al., 2002] vary considerably, rendering a necessity to measure in different parts of the world.

Figure 3b shows that $\omega_o$ increases with increasing aerosol optical depth, indicating that for higher aerosol loading, fossil fuel combustion and biomass burning are not the predominant factors causing the increase in the concentration of aerosols. Heavy aerosol episodes appear to be caused, more likely, by blowing mineral dust. This is partially confirmed by the relationship between aerosol optical depth and the Angstrom exponent as shown in Figure 3c.

Figure 4a shows the daily averaged values of $\omega_o$ at $\lambda = 0.5\mu m$. The dramatic day-to-day variation shown in Figure 4a may be related to meteorological conditions, including variations in air parcel trajectory, humidity, temperature inversions, and dust particles driven by high winds. The seasonal variation shown in Figure 4b indicates that $\omega_o$ has a minimum in spring and a maximum in summer. As described before, in the spring season, northwesterly winds prevail, driving the air pollution from Beijing to the Xianghe site (downwind). Eck et al. [2005] studied the spatial relationship of aerosol properties. They illustrated that in spring, aerosol properties between Beijing and Xianghe are highly correlated, suggesting that the aerosol properties observed in Xianghe are significantly affected by the pollutants from Beijing during this season. The summer maximum in the magnitude of $\omega_o$ indicates that the emission of black carbon during this season is minimum, consistent with the trend in the seasonal variation of black carbon emission in China [Streets et al., 2003]. In addition, the relatively humid air from the southeast and rainy weather (about three quarters of the annual precipitation at Xianghe occurs during the summer season) may be another important factor for the summer minimum in the magnitude of $\omega_o$.

### 6. Comparison With AERONET Retrievals

To assess the validity of our retrievals, the values of $\omega_o$ retrieved by our algorithm are compared with those from AERONET. The two retrievals occur on different time-scales: 1 minute for the retrievals from the proposed algorithm and about 1 hour for the AERONET $\omega_o$. To match the times for comparison purposes, the values of $\omega_o$ from the proposed algorithm were averaged over a period of 30 min before and after the times corresponding to the AERONET retrievals. Figures 5a–5d show comparisons of $\omega_o$ at 0.44, 0.67, 0.87, and 1.02 $\mu m$, respectively. Both products are limited to optical depths larger than 0.4 at 0.44 $\mu m$ and solar zenith angles larger than 50°. The standard deviations and mean differences are 0.0218, 0.024, 0.0369 and 0.0028, 0.0028, 0.0054, 0.0108 at 0.44, 0.67, 0.87 and 1.02 $\mu m$, respectively. Since the contribution of the scattered radiation at 0.44 and 0.67 $\mu m$ to the SW flux is much larger than that at 0.87 and 1.02 $\mu m$, the accuracies of the retrievals are higher at visible wavelengths than in the near infrared. The same is true for the AERONET retrievals [Dubovik et al., 2000]. It is thus not surprising that the difference between the two retrievals is smaller in the visible than in the infrared. Considering that the accuracy of the AERONET $\omega_o$ retrievals is on the order
of 0.03, the consistency between our retrievals and AERONET products is satisfactory.

7. Error Analyses

[25] Despite the good agreement shown in Figure 5, attempts are made to gain further insights into the discrepancies in terms of input data uncertainties and assumptions made in the retrieval algorithm.

7.1. Effects of Instrument Measurement Errors

[26] The instruments used in the present study are the CIMEL CE-318 Sun photometer and the Kipp & Zonen CM21 pyranometer. The CIMEL CE-318 Sun photometer is well calibrated by the AERONET group at least once a year. The errors in the aerosol optical depth derived by this instrument are less than 0.01 ~ 0.02. Such errors can induce negligible uncertainties in the calculation of broadband downwelling SW fluxes. For example, for a solar zenith angle of 50°, an error of 0.01 in the aerosol optical depth at 0.5 μm can cause an error of 1.7 W m⁻² in the SW fluxes, assuming an Angstrom exponent equal to 1.2.

[27] The effects of the CM21 pyranometer measurement errors on the retrieved ω₀ are shown in Table 1. In this table, the instrument measurement error is fixed at 8 W m⁻² (the errors of CM21 measurements are estimated to be less than 10 W m⁻²). As previously mentioned, the errors in the retrieved ω₀ increase with decreasing aerosol optical depth. For aerosol optical depths >0.4, the retrieval errors are less than 0.03. Such errors are comparable to those retrieved from the observations of Sun/sky radiation under high aerosol loading conditions [Dubovik et al., 2002].

7.2. Errors Due to Assumptions

[28] In the present algorithm, the Junge power law is used to describe the aerosol size distribution in an actual atmosphere. It is also assumed that the real part of the index of

Table 1. Retrieval Error δω₀ Arising From CM21 Measurement Errors

<table>
<thead>
<tr>
<th>τ(0.5 μm)</th>
<th>δω₀(0.5 μm)</th>
</tr>
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<tbody>
<tr>
<td>0.1</td>
<td>0.103</td>
</tr>
<tr>
<td>0.4</td>
<td>0.028</td>
</tr>
<tr>
<td>0.8</td>
<td>0.016</td>
</tr>
<tr>
<td>1.2</td>
<td>0.012</td>
</tr>
</tbody>
</table>

*In the calculation of the retrieval error δω₀, the CM21 measurement error is set at 8 W m⁻². The solar zenith angle is 50° and the Angstrom exponent is 1.2. The “true” aerosol single scattering albedo is 0.919.

Figure 5. Comparison between the aerosol single scattering albedo retrieved from the proposed algorithm and AERONET products at wavelengths of (a) 0.44 μm, (b) 0.67 μm, (c) 0.87 μm and (d) 1.02 μm. To match the times for comparison purposes, the values of the single scattering albedo retrieved by the proposed algorithm were averaged over a period of 30 min before and after the times corresponding to the AERONET retrievals.
refraction of aerosols is independent of wavelength and has a fixed value of 1.5. To investigate the uncertainties incurred by invoking these assumptions, many experiments were conducted. First, surface broadband downwelling SW fluxes were simulated using the AERONET retrievals of aerosol size distribution and index of refraction at the Xianghe site. The simulated broadband downwelling SW fluxes, in conjunction with the AERONET aerosol optical depth and Angstrom exponent products, were then employed to retrieve \( \omega_o \) using the proposed algorithm. Figure 6 presents the comparison of \( \omega_o \) retrieved from our algorithm using the simulated surface total irradiances with those retrieved by the AERONET using sky radiances. In this case, the AERONET products are considered as “true” values. The standard deviations and mean errors are 0.013, 0.013, 0.016, 0.022 and 0.0024, 0.0042, 0.0102 at 0.44, 0.67, 0.87 and 1.02 \( \mu m \), respectively. Such errors are not significant, especially for the visible spectral region. Dubovik et al. [2000] found that under error-free conditions, the retrieval accuracy due to computation errors can reach 0.01.

The agreement shown in Figure 6 is a better indication of the true retrieval accuracy of the retrieval algorithm because of the different fields of view seen by the Sun photometer and the pyranometer. Note that the AERONET retrievals of aerosol properties are based on direct solar radiances and sky radiances measurements over a small portion of the sky, while our retrieval is sensitive to both direct radiation and diffuse radiation from the entire sky dome. Any discrepancy would transform into a retrieval disagreement. For example, if a portion of the sky outside the Sun photometer viewing domain is covered by an undetected residual cloud, the two retrievals would differ considerably, simply because they did not see the same targets. Likewise, inhomogeneous aerosol distribution in the sky also incurs a disagreement. As such, the retrievals shown in Figure 6 represent the true uncertainties induced by the retrieval algorithm per se, as the differences caused by a major input variable, namely, the surface irradiance, is removed.

[29] Another retrieval error suffered by most aerosol retrieval algorithms is incurred by the assumption that the aerosol particles are spherical. The validity of such an assumption was discussed by many investigators [e.g., Koepke and Hess, 1988; Mishchenko et al., 1995, 1997].
It was found that differences in the scattering phase functions between spherical and nonspherical particles can be large, but differences in the optical cross sections, $\omega_o$ and asymmetry parameter are much smaller (less than a few percent under most circumstances). Use of broadband irradiances instead of radiance in our algorithm lowers the sensitivity of the retrieval to particle shape. Therefore the spherical particle assumption may not induce significant errors in the $\omega_o$ retrieved by the proposed algorithm.

8. Summary and Conclusions

[31] In this study, we attempt to estimate the aerosol $\omega_o$ from combined measurements of spectral direct solar radiation and broadband downwelling SW fluxes. Tests were performed to examine the sensitivity of the broadband fluxes to aerosol $\omega_o$, size distribution and the ground surface albedo. An algorithm was developed according to the sensitivity analyses. In our algorithm, we first determine the imaginary part of the index of refraction by comparing the measured broadband downwelling SW fluxes with those calculated using the optical thickness at 0.5 $\mu$m and the Angstrom exponent. The aerosol size distribution follows the Junge power law and the real part of the index of refraction is fixed at 1.5. The aerosol $\omega_o$ is calculated by using the retrieved index of refraction and the Junge power law size distribution.

[32] The algorithm was applied to measurements made in Xianghe, China from September 2004 to August 2005 under the auspices of the EAST-AIRE project. The cloud screening was carefully performed by using the movie of the sky conditions obtained from the Total Sky Imager (TSI-440A). The average value of the retrieved $\omega_o$ at 0.5 $\mu$m is 0.9, with 50.4% of the retrieved values larger than 0.9, 48.3% between 0.80 and 0.9, and 1.3% smaller than 0.8. It varies considerably from day to day. In general, the seasonal variation of $\omega_o$ is much less pronounced than the day-to-day variation with a minimum in the spring and a maximum in the summer. The $\omega_o$ decreases as the Angstrom exponent increases, implying that the finer aerosol particles have stronger absorption than the coarser aerosol particles. This indicates that emissions from fossil fuel combustion and biomass burning are major contributors to the aerosol absorption. The majority of the heavy aerosol episodes have large $\omega_o$, presumably due to an increasing proportion of mineral dust.

[33] The $\omega_o$ retrieved from our method are compared with the AERONET product. The two retrievals agree fairly well with each other. The standard deviations and mean differences are 0.0218, 0.024, 0.03, 0.0369 and 0.0028, 0.0028, 0.0054, 0.0108 at 0.44, 0.67, 0.87 and 1.02 $\mu$m, respectively. A significant fraction of the discrepancies originate from differences in the fields of view of the pyranometer and Sun photometer, as indicated by the differences in surface irradiances measured by the pyranometer and those computed from the AERONET-based retrievals of aerosol parameters. If the same surface irradiances were used, the standard deviations and mean errors would be lowered to 0.013, 0.013, 0.016, 0.022 and 0.0024, 0.0032, 0.0042, 0.0102 at 0.44, 0.67, 0.87 and 1.02 $\mu$m, respectively. Such a level of accuracy is compatible with the accuracies of the AERONET retrievals based on sky irradiances. The retrieval errors caused by instrument measurement errors appear to be insignificant for aerosol optical depths at 0.44 $\mu$m larger than 0.4.

[34] The findings of this study have a practical application for studying aerosol radiative forcing both at the surface and at the TOA. It provides a more economical alternative to deriving both aerosol properties and aerosol radiative forcing over large areas. Aerosol optical depths measured by a handheld Sun photometer (or placed on a solar tracker) can be combined with direct radiation measurements from a pyranometer to derive aerosol $\omega_o$. For example, in the EAST-AIRE project, 25 handheld Sun photometers have been deployed to obtain the spatial distribution of aerosol loading across China [Xin et al., 2007]. Some of the stations were equipped with pyranometers. Following this approach, we can derive $\omega_o$ across the region which is invaluable to address regional aerosol forcing issues.

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Z. Li and F. Zhao, Earth System Science Interdisciplinary Center, University of Maryland, College Park, MD 20740, USA. (zli@atmos.umd.edu)