Retrievals and Uncertainties Analysis of Aerosol Single Scattering Albedo from MFRSR Measurements

Bangsheng Yin, Qilong Min*

Atmospheric Science Research Center, State University of New York,

Albany, New York

Everette Joseph

Howard University, Washington, DC

*Corresponding author: Qilong Min, qmin@albany.edu

Abstract

Aerosol single scattering albedo (SSA) can be retrieved from the ratio of diffuse horizontal and direct normal fluxes measured from multifilter rotating shadowband radiometer (MFRSR). In this study, the measurement channels at 415 nm and 870 nm are selected for aerosol optical depth (AOD) and Angstrom coefficient retrievals, and the measurements at 415 nm are used for aerosol SSA retrievals with the constraint of retrieved Angstrom coefficient. We extensively assessed various issues impacting on the accuracy of SSA retrieval from measurements to input parameters and

assumptions. For cloud-free days with mean aerosol loading of 0.13~0.60, our sensitivity study indicated that: (1) 1% calibration uncertainty can result in 0.8~3.7% changes in retrieved SSA; (2) without considering the cosine response correction and/or forward scattering correction will result in underestimation of 1.1~3.3% and/or 0.73% in retrieved SSA; (3) an overestimation of 0.1 in asymmetry factor can result in an underestimation of 2.54~3.4% in retrieved SSA; (4) for small aerosol loading (e.g., 0.13), the uncertainty associated with the choice of Rayleigh optical depth value can result in non-negligible change in retrieved SSA (e.g., 0.015); (5) an uncertainty of 0.05 for surface albedo can result in changes of 1.49~5.4% in retrieved SSA. We applied the retrieval algorithm to the MFRSR measurements at the Atmospheric Radiation Measurements (ARM) Southern Great Plains (SGP) site. The retrieved results of AOD, Angstrom coefficient, and SSA are basically consistent with other independent measurements from co-located instruments at the site.

Keywords: MFRSR, single scattering albedo, diffuse-direct ratio, aerosol optical depth, Angstrom coefficient

1. Introduction

Aerosols are recognized as a significant climate forcing, altering the Earth's radiation balance in the global climate system. In recent decades, substantial efforts have been devoted to estimate aerosol climate effects. However, these estimates are subject to large uncertainties [1]. One of the primary causes for the large uncertainties is the lack of the reliable information on the spatial and temporal distribution of aerosol properties. As one of the most important properties of aerosol, the aerosol single-scattering albedo (SSA) plays a key role in the direct radiative effect of aerosols. The variation of SSA can alter the magnitude, even the sign of aerosol direct effect, depending on the surface albedo and asymmetry factor [2, 3]. Therefore accurate information of aerosol SSA is crucial to determine the aerosol effects on radiative flux changes.

In order to derive the SSA and other key properties of aerosols, various measurement systems have been developed, from an in-situ measurement suite of an integrating nephelometer and a particle soot absorbance photometer to radiometers. For different radiometers, the basic principles in retrieving aerosol SSA are similar: under clear-sky conditions aerosol SSA can be derived from diffuse irradiance, or from the diffuse to direct normal irradiance ratio (DDR) [4]. However, both diffuse and DDR are sensitively dependent upon the optical properties of aerosols, surface reflectivity, and

gaseous absorption and scattering in the atmosphere, which can introduce uncertainties into the aerosol SSA retrieval. Therefore, improving retrieval accuracy and making the retrieval algorithm more robust and operational are the key issues in the study of aerosol SSA retrieval.

As widely deployed radiometers, Cimel Sun-photometer (CSPHOT) and multifilter rotating shadowband radiometer (MFRSR) have been utilized to infer SSA. Based on CSPHOT measurements, Dubovik et al [5, 6] developed a retrieval algorithm to derive the aerosol SSA, which has been widely applied by Aerosol Robotic Network (AERONET) sites. In recent decade, based on MFRSR measurements, several aerosol SSA retrieval algorithms have been developed. For example, Kassianov et al. [7, 8] developed a complicated retrieval algorithm to infer the aerosol SSA from MFRSR measurements in two steps: (1) derive the aerosol size distribution from the spectral measurements of the direct irradiance at five channels (415, 500, 610, 665, 870 nm) and (2) retrieve the imaginary refractive index or SSA from the diffuse irradiance with the constraint of the aerosol size distribution in the first step. This algorithm is able to derive both SSA and aerosol size distribution. However, retrieving aerosol size distribution from finite wavelength channels is an ill-posed problem. Errors associated with direct beam measurements for all five channels and the contamination of ozone and water vapor at three channels of 500, 610, and 665 nm introduce significant errors/uncertainties in the retrieved aerosol optical depths, and consequently in the

derived aerosol size distribution and SSA. To analyze the optical properties of different types of aerosols in Central Mediterranean, Meloni et al. [9] developed a retrieval algorithm to retrieve aerosol SSA at two channels (i.e., 415 nm and 870 nm) from MFRSR measurements, with pre-calculated asymmetry factors. To evaluate the retrieval accuracy, Meloni et al. [9] did an extensive sensitivity study for different uncertainty sources, including non-Lambertian surface, cosine response, forward scattering peak, aerosol asymmetry factor, and retrieved aerosol optical depth (AOD).

In this paper, we also use the ratio of the diffuse horizontal to direct normal flux measured by MFRSR to retrieve the aerosol SSA. Since the basic approach of retrieving SSA is well established, our efforts are (1) to deal with uncertainty sources explicitly for a robust and routine retrieval algorithm; (2) to evaluate all major uncertainty sources from measurements to retrieval assumptions and inputs with our retrieval algorithm; and (3) to validate and evaluate our retrieval results against those derived from other independent measurements, e.g., AERONET and Aerosol Observing System (AOS). Specifically, a minimum set of measurements, i.e., two most stable channels of MFRSR (415 and 870 nm), is used in the retrievals to reduce the impacts of these uncertainties for robust retrievals under various conditions. A low order scattering approximation is used to compute irradiance in forward direction [10]. The aerosol asymmetry factor is estimated from the observed aerosol Angstrom coefficient, which provides a better constraint on a key uncertainty source. Other

uncertainties associated with retrieval algorithm assumptions and MFRSR measurements are also considered, including the calibration accuracy, the cosine respond factor, Rayleigh scattering uncertainty, and the surface albedo uncertainty.

At the Atmospheric Radiation Measurements (ARM) Southern Great Plain (SGP) site, besides radiometers like MFRSR and CSPHOT, an AOS is deployed to measure near-surface aerosol optical properties, including TSI 3563 Nephelometer (450, 550, and 700 nm) and Particle/Soot Absorption Photometer (467, 550, 660 nm). The Nephelometer measures aerosol scattering coefficients at each channel, from which near-surface aerosol Angstrom coefficients can be derived. By combining both instrument measurements, aerosol SSA at these three channels can be derived. Therefore, we can evaluate our retrieved aerosol SSA with AOS and CSPHOT co-located measurements.

2. Retrieval Algorithm

The MFRSR is a seven-channel radiometer with six passbands 10 nm FWHM (Full Width Half Maximum) centered near 415, 500, 610, 665, 862, and 940 nm, and an unfiltered silicon pyranometer [11]. It uses an automated shadowbanding technique to measure the total-horizontal, diffuse-horizontal, and direct normal spectral irradiances through a single optical path. It guarantees that the separated spectral irradiance

components share the same passbands and calibration coefficients; hence the Langley regression of the direct normal irradiance taken on clear stable days can be used to extrapolate the instrument's response to the top of the atmosphere, and this calibration can then be applied to both components of irradiance. Transmittances can be subsequently calculated under both clear-sky and cloudy conditions as the ratio of the uncalibrated output to the extrapolated top-of-the-atmosphere value. Also, the ratio of diffuse to direct normal irradiances is independent of absolute calibration. Utilizing the unique capability of MFRSR, the spectral direct normal irradiance can be used to derive optical properties of aerosol and optically thin cloud accurately with a forward scattering correction [10] and the diffuse irradiance can be used to infer cloud optical depths and fractional sky cover [12-14].

Under clear-sky conditions, as the diffuse irradiance is sensitive to both scattering and absorption of aerosols, aerosol SSA can be derived from diffuse irradiance, or from DDR. However, it is advantageous to use the DDR of MFRSR measurements for SSA retrievals, since the DDR is insensitive to the calibration errors. In general, the direct normal irradiance (I^{dirn}) and diffuse-to-direct ratio of solar irradiance (R) can be written as

$$I^{dirn} = I_0 e^{-(\tau_{ray} + \tau_{gas} + \tau_{aer})/\mu_0}$$

$$\frac{I^{dif}}{I^{dirn}} = \frac{I^{dif}(\tau_{ray}, \tau_{gas}, \tau_{aer}, g_{aer}, \alpha_{aer}, A_{ref})}{I^{dirn}(\tau_{ray}, \tau_{gas}, \tau_{aer})} = R(\tau_{ray}, \tau_{gas}, \tau_{aer}, g_{aer}, \alpha_{aer}, A_{srf}) \quad (1)$$

where I_0 is the solar constant at the top of atmosphere, I^{dif} is the hemispheric diffuse

irradiance, and I^{dir} is the direct normal irradiance at the cosine solar zenith angle of μ_0 . τ_{ray} , τ_{gas} and τ_{aer} are the optical depth values of Rayleigh scattering, gaseous absorption, and aerosol extinction, respectively. g_{aer} , α_{aer} and A_{srf} are the aerosol asymmetry, aerosol SSA, and surface albedo, respectively. Figure 1 shows both diffuse transmittance and DDR as a function of AOD and SSA. This indicates that the direct aerosol effect is sensitive to the aerosol SSA, especially under heavy aerosol loading situations.

The retrieval algorithm uses the 415 and 870 nm channels to minimize the interference of gaseous absorption, and separates aerosols from thin clouds based on the temporal and spectral characteristics of direct and diffuse irradiances [14]. As outlined in *Min et al* [10], aerosol optical depth and Angstrom coefficient can be retrieved accurately from simultaneous spectral measurements of direct normal irradiances. The aerosol asymmetry factor can then be estimated from inferred Angstrom coefficient with assumptions of aerosol size distribution and the refractive index. To reduce the impact of the surface albedo on the retrievals, we focus on 415 nm where the surface albedo is small and relatively stable without snow on the ground. Under these constraints and assumptions, we solve the radiative transfer equation iteratively to derive the SSA by matching the measured DDR. To simulate both direct beam and diffuse irradiance measured by MFRSR, we first compute the radiative intensity field using our fast and accurate radiative transfer model [15], which combines the exact radiance of low orders

of scattering with multiple scattering radiance. Then we determine the blocked irradiance by applying the shadowband geometry of MFRSR to the simulated radiance field. To speed up the retrievals, we build a look up table of the simulated direct beam and diffuse irradiances under various solar zenith angles and surface albedos for different aerosol optical depths, asymmetric factors, and SSAs. During the retrieval process, the aerosol SSA can be derived by matching the observed DDR with the simulated DDR in a lookup table. This method can avoid the time-consuming iterative radiative transfer calculation and substantially improve the speed of retrievals.

3. Uncertainties Analysis

The accuracy of retrieved SSA is affected by the uncertainties of many input parameters, which can be categorized into two types: measurement systems and environments. The first type of uncertainty is associated with MFRSR measurements, including MFRSR readout noise, calibration, diffuser cosine response, and forward scattering issue due to the shadowband blocking mechanism. The other type of uncertainty stems from the input parameters and assumptions for radiative transfer calculation, such as the surface albedo, Rayleigh scattering optical depth, gaseous absorption, and aerosol asymmetry factor.

3.1 Uncertainties Associated with MFRSR Measurements

Understanding measurement uncertainties and systematic bias is crucial for any instrument. For all radiometers, radiometric calibration and readout noise are two most critical uncertainties. Although the calibration independent diffuse-to-direct ratio is used to retrieve SSAs, the aerosol optical depth and Angstrom coefficients are derived from the direct beam irradiance. Hence, radiometric calibration is still crucial for SSA retrievals. Due to the uncertainty in the standard transferable lamp, the in-lab calibration accuracy is about 4%. However, MFRSR direct beam measurements allow in-situ Langley calibration under stable clear-sky conditions. For example, the MFRSR deployed at the ARM SGP site has been continuously operated for a decade, and more than 60 Langley events have been obtained each year. The relative solar constant (I_0) at the passband from Langley regressions can be interpolated and extrapolated to any particular day by using a temporal and spectral analysis procedure [16]. To facilitate the analysis, the I_0 values calculated from every specific Langley calibration are defined as Langley values; the smoothed and interpolated or extrapolated I_0 values for every day are defined as Forgan values. The Langley values and Forgan values are calculated by the MFRSR processing package (<ftp://169.226.118.87/pub/tmp/mpp.package.html>).

Figure 2a shows the Forgan values of I_0 at the passbands of 415 and 870 nm from 1997 to 2012. Compared to the 415 nm channel, I_0 at 870 nm is relatively stable.

Specifically, Figure 2b shows the comparison between Langley values and Forgan values in 2008. Based on the I_0 (Forgan values), we can convert the uncalibrated solar irradiance measurements to relatively accurate transmission measurements. The accuracy of the relative solar constant at a non-gaseous absorption passband, based on the Langley regression calibration, is determined by the occurrence frequency of Langley events. At ARM SGP site, it can be within 1% when the occurrence frequency of Langley events is high. In particular, $0.99 * I_0$ at 415 nm is a better estimate in March 2008, assessed from the Langley I_0 during the month. In other channels of MFRSR, such as at 500, 610, and 665 nm, the solar radiation can be absorbed by ozone and water vapor. These gaseous absorptions will introduce additional error to the Langley calibrations, and increase their uncertainties to about 2% or more. Considering gaseous absorption correction issues, we avoid using these three channels for aerosol optical depth retrievals.

Each instrument has its own strengths and weaknesses. MFRSR utilizes the shadowbanding technique to measure the total-horizontal, diffuse-horizontal, and direct normal spectral irradiances through a single diffuser and associated fore-optics. Beyond the standard radiometry calibration and readout noise issues, MFRSR has its own issues due to its shadowbanding technique. As shown in Figure 3a, the surface of MFRSR diffuser is not truly a Lambert surface, its response to the incident radiance varies with the direction. The cosine correction coefficient of the diffuser was first

analyzed by Harrison et al. [11]. For the ARM released MFRSR data, the cosine correction coefficient for the specific instrument is measured by the ARM observation center in lab, and it is attached in the MFRSR data files as instrument parameters. Although direct normal irradiance of MFRSR has been cosine corrected, the diffuse irradiance has not. Based on the cosine correction coefficient of diffuser and simulated radiation intensity, the impact of cosine correction factor on the diffuse irradiance is up to 3%, shown in Figure 3b, varying with solar zenith angle and aerosol loading. Basically, the larger solar zenith angle, the smaller the cosine correction factor is; and the larger the aerosol optical depth, the larger variation range of the cosine correction factor is. Overall, without considering the cosine correction factor of the diffuser, the measured diffuse irradiance at 415 nm and 870 nm can be underestimated by 1 ~ 3%. Such uncertainty of diffuse irradiance will impact on the diffuse-to-direct ratio, and thus on the retrievals of the SSA.

The direct normal irradiance of MFRSR is not directly measured. It is calculated by subtracting the direct beam blocked horizontal irradiance, i.e., the diffuse irradiance, from the total (without blocking) horizontal irradiance, with cosine correction and two side-blocking corrections. Because the blocking angle of the MFRSR shadowband is 7.8 degrees, the shadowband blocks not only the direct beam of the Sun, but also the forward scattering lobe of the solar aureole. Even though a simple two side-blocking at 9 degrees has been used to compensate the forward scattering of the solar aureole,

the large scattering particles with strong forward scattering can result in a large error in measured direct normal irradiance.

Min et al. [10] originally proposed a correction algorithm and applied to the retrievals of optically thin clouds and aerosols from direct beam irradiance. In that algorithm, Min et al. modified the DISORT radiative transfer code [17] by combining the δ -fit method [18] with the Nakajima-Tanaka correction procedure [19] to accurately and rapidly compute the forward scattering radiances with error less than 1%. By adopting Min et al.'s approach and considering the cosine correction for diffuse irradiance, the correction equation for DDR can be written as follows:

$$R_R = \frac{I_R^{dif}}{I_R^{dirn}} = \frac{I^{dif} * f^{CRC} + \Delta^{FS}}{I^{dirn} - \Delta^{FS}} \quad (2)$$

$$I_R^{dirn} = I_0 e^{-(\tau_{ray} + \tau_{gas} + \tau_{aer})/\mu_0} - \Delta^{FS}$$

where the variables I_0 , I^{dif} , I^{dirn} , μ_0 , τ_{ray} , τ_{gas} , and τ_{aer} are the same as Eq. (1); f^{CRC} and Δ^{FS} are the cosine response correction factor and forward scattering correction factor, respectively; and the subscript R indicates that the related variables are recalibrated.

This combined correction is important and necessary for heavy aerosol loading and/or larger size aerosol particles as demonstrated in sensitivity studies discussed in the next section. Eq. (2) provides an accurate way to deal with both forward scattering issue and the cosine response issue of MFRSR.

3.2 Uncertainties associated with prior information (or model inputs)

(a) Rayleigh scattering

Rayleigh scattering is dominant at short wavelengths. In recent decades, various efforts have been devoted to get better estimation of Rayleigh optical depth from ultra violet to visible spectrum [e.g., 20-23]. However, the differences among various approximations of Rayleigh optical depth at short wavelengths are substantial, particularly in ultra violet region. Even at 415 nm, listed in Table 1, the difference is up to 0.0175, indicating non-ignorable uncertainty in retrievals of SSA, particularly when aerosol optical depth is comparable or smaller than Rayleigh optical depth. In Table 1, the latest three versions of values are estimated by Thomasi et al. [22] and Srivastava et al. [23], in which the estimation of Thomasi is the median value and it is also very close to the estimation of Boadhaine et al. [21]. Therefore, in the retrievals, Rayleigh optical depths of 0.3094 and 0.0152 (derived by Thomasi et al. [22]) were used for the passbands at 415 and 870 nm, respectively.

(b) Surface albedo

Surface albedo influences the diffuse irradiance in the shortwave band. The relationship between surface albedo A_{srf} and total downward irradiance I^{tot} can be written as [24]:

$$I^{tot} = I^{tot0} * \frac{1}{1 - A^{srf} * A^{sph}} \quad (3)$$

where I^{tot0} is the total downward irradiance with no surface reflection, and A^{sph} is the spherical albedo determined by the optical properties of the air layers above the surface. Surface spectral albedo over the ARM SGP was measured under both clear-sky and cloudy conditions by using Multi-filter Radiometer (MFR), Analytic Spectral Device (ASD), and Solar Spectral Flux Radiometer (SSFR) [25]. As listed in Table 2 the surface albedos derived from these three sensors are consistent with relatively small differences at 415 and 870 nm. Based on in-situ observation from two towers at the site, however, the surface albedo at 870 nm varies substantially although the surface albedo at 415 nm is relatively constant when snow is absent. Therefore, it is important to evaluate the impact of surface albedo on the retrieval of SSA. As a basic set for the retrievals, we set the values of 0.033 and 0.274 at 415 and 870 nm, respectively, which were measured by the MFR (a similar sensor as the MFRSR).

(c) Asymmetry factor

The phase function of aerosol scattering plays a key role in determining the diffuse irradiance under clear-sky conditions. In general, particle phase function, as well as the asymmetry factor, is sensitive to refractive index, particle shape, and particle size distribution. The latter can be constrained by spectral dependence of aerosol optical depth, i.e., Angstrom coefficients [26, 27]. As shown in Figure 4, the AOS Nephelometer measurements indicate that the asymmetry factor (for submicron particles) is dependent on the Angstrom coefficient (between 450 and 700 nm). However, for a given Angstrom coefficient, the asymmetry factor varies within ± 0.1 . The uncertainty is due to various aerosol size distributions, aerosol shapes, and chemical compositions (or refractive index). The asymmetry factor - Angstrom coefficient relationship also can be simulated by the Mie simulation (shown in Figure 4). In this study, to simplify the radiative transfer calculation, the shape of aerosol

particles is assumed to be spherical. For the non-spherical aerosol, the spherical assumption will introduce some errors in the radiative transfer calculation, and this issue warrants further study in the future. We choose a single-modal lognormal particle number size distribution, with the mode diameter varying within 0.06~0.80 μm with a standard deviation of 1.2 μm . The aerosol refractive index is set as $1.5+0.007i$ at 415 nm, which is chosen based on observations at the SGP site [8]. We derive the aerosol asymmetry factor from Angstrom coefficients through the simulated Asymmetry factor - Angstrom coefficient relationship. Compared to the in-situ surface measurements, the simulated asymmetry factor is relatively larger (0.03-0.05). In this study, the AOS derived aerosol asymmetry factor is measured under dry condition, which could be underestimated compared to the real situation. In the other hand, the real atmospheric aerosols have different properties as assumed in the simulation, inevitably resulting in errors in the simulated asymmetry factor. However, as shown in Figures 5-7 in the following section, our estimated asymmetry factor is within ± 0.02 of AERONET independent estimations.

4. Retrieval Evaluation and Sensitivity Study

4.1 Retrieval Evaluation (Case studies)

The retrieval uncertainties have been discussed systematically. Based on those analyses, we improve the retrieval algorithm by applying calibration and correction to the measurements and by choosing reasonable prior input parameters for radiative transfer model. Here, we apply our retrieval algorithm to the observational data from the ARM SGP site. In this study, the diffuse irradiance and associated DDR is based on ARM original MFRSR measurements with our calibration and corrections in Equation 2; the related AOD and Angstrom coefficients are derived from the calibrated direct normal irradiance [28]. Because this retrieval algorithm is based on the plane-parallel assumption, we select cases with substantially long clear-sky

periods to minimize cloud and associated 3D effects. According to the measurements of CSPHOT, lidar, radar, and Sky Radiation (SKYRAD) collection of radiometers [29] (not shown), March 10 and 11, 2008 (shown as 20080310 and 20080311 in the later) were good for the SSA retrieval study with small aerosol loading; June 6 and 7, 2011 (shown as 20110607 and 20110608 in the later) were good for the SSA retrieval study with heavy aerosol loading. To evaluate our retrievals of AOD and Angstrom coefficients, we compare MFRSR retrievals with the collocated CSPHOT products (level 2.0), which is processed by the standard AERONET processing package [5, 6, 30]. For CSPHOT, although it does not have a 415 nm channel, the AOD at 415 nm can be interpolated from the AODs measured at 380 nm and 440 nm channels by using the Angstrom's empirical relationship between aerosol optical depth and wavelengths [22, 26]. To evaluate our retrievals of SSA, we compare MFRSR retrievals with the collocated AOS measurements and the collocated CSPHOT products (if the qualified retrieved SSA are available).

Figure 5 and 6 show retrieved aerosol optical depth, Angstrom coefficient, asymmetry factor, and aerosol SSA during the previously stated four days. On 20080310 and 20080311, the MFRSR derived aerosol loadings at 415 nm were 0.13 and 0.19, respectively, and the Angstrom coefficients and asymmetry factor were about 1.5 and 0.69, respectively. On 20110607 and 20110608, the aerosol loadings are much higher, with AOD at 415 nm being 0.59 and 0.55, respectively, and the Angstrom coefficients

and asymmetry factor were about 1.65 and 0.67, respectively. As shown in Figure 5(a-b) and 6(a-b), the retrieved aerosol optical depths from MFRSR and CSPHOT agree well, within an uncertainty of 0.01. The above comparisons indicate that whether the aerosol loading is small or big, the retrieval accuracy of AOD from MFRSR is high. The Angstrom coefficients from MFRSR are slightly different from CSPHOT. The AOS measured Angstrom coefficients are also shown in Figure 5(c-d) and 6(c-d). They are computed from extinction coefficients (scatter + absorption) at 450 nm and 700 nm. They are basically consistent with the MFRSR and CSPHOT measurements except on 20080311(after 20:00 GMT) and 20110607. As shown in Figures 5(e-f) and 6(e-f), the aerosol asymmetry factors from MFRSR are slightly different from CSPHOT. Compared to column integrated aerosol asymmetry factors, the aerosol asymmetry factor from AOS sometimes show obvious difference, e.g., it is upto 0.05 or more on 20080310 and 20080311.

To illustrate the potential impact of uncertainty associated with asymmetry factor, we retrieve aerosol SSA in two ways: 1) using the asymmetry factor determined from Angstrom coefficient (baseline) and 2) artificially add 0.1 to the previously determined g ($g+0.1$). Under small aerosol loading situations, the AERONET products (level 2.0) do not provide a valid retrieval of aerosol SSA. Therefore, only the SSAs derived from MFRSR and AOS are compared in Figures 5g and 5h, which indicate that the temporal variations of retrieved aerosol SSA from these two

instruments are consistent with each other. The mean values of MFRSR retrieved SSA (baseline) were 0.967 and 0.958 for 20080310 and 20080311, respectively. However, on 20080311, the difference between the MFRSR SSA (baseline) and AOS SSA increased with time and was substantial in the afternoon. On 20110607 and 20110608, the aerosol loading is much bigger (with $AOD > 0.5$), and valid retrievals of aerosol SSA were provided by AERONET products (level 2.0). As shown in Figures 6g and 6h, the SSA (baseline) derived from MFRSR is consistent with that from AERONET products for both days. However the SSA (baseline) retrieved from MFRSR is 0.03 higher than the SSA derived from AOS measurements on 20110607.

The difference between MFRSR SSA and AOS SSA is probably caused by two factors: one is the retrieval errors from both instruments; the other is the changes in the mixing process in the boundary layer. As shown in bottom plots of Figure 5 and 6, the uncertainty (0.1) of aerosol asymmetry factor can result in substantial difference in the retrieved SSA. Especially for cases on 20080311 and 20110607, the difference between MFRSR SSA ($g+0.1$) and AOS SSA is much smaller. Additionally, the SSA derived from MFRSR is averaged over the entire column of the atmosphere while the AOS measures aerosol SSA at near-surface. Changes in the mixing process in the boundary layer will result in substantial differences of optical/microphysical properties between aerosols aloft in the atmosphere and aerosols near the surface. Furthermore, the observation of polarization lidar also clearly shows that an aerosol

layer exists aloft on 20080311(not shown). Interestingly, when the difference of Angstrom coefficient between MFRSR and AOS is larger, the difference of MFRSR retrieved SSA and AOS measured SSA is also larger. This indicates that the difference between MFRSR SSA and AOS SSA more probably represent the real difference between aerosols in the atmosphere and at the near-surface.

4.2 Sensitivity Study

Since the retrieval accuracy is affected by several factors, it is necessary to perform an extensive sensitivity study to assess the retrieval uncertainty and error associated with those factors. In this study, four cases are separately chosen to analyze the retrieval uncertainties under different aerosol loading. Table 3 lists the results of our sensitivity studies. It is clear that even though we used the calibration independent DDR, the calibration is still an important factor in the SSA retrievals. An increased solar constant (I_0) or reduction of transmittance results in an overestimate of aerosol loading, and consequently an underestimate of aerosol SSA. The calibration uncertainty of 1% results in uncertainties of 0.036 (3.73%), 0.024 (2.54%), 0.007 (0.72%), and 0.008 (0.85%) in retrieved SSA, respectively for aerosol loading of 0.13 (20080310), 0.20 (20080311), 0.47 (20110607), and 0.60 (20110608), which indicates that the uncertainties of retrieved SSA decrease with the aerosol loading. The cosine respond correction factor (f^{CRC}) effectively reduces the diffuse irradiance, resulting in

a reduction of retrieved SSA, i.e., 0.032 (3.32%), 0.024 (2.54%), 0.013 (1.33%), and 0.010 (1.06%) in retrieved SSA, respectively for 20080310, 20080311, 20110607, and 20110608. Similar to the calibration uncertainty, the uncertainties of retrieved SSA resulted from f^{CRC} also decrease with the aerosol loading. Since the Angstrom coefficients were about 1.5 or 1.65, it suggests that aerosol particle sizes were relatively small. This sensitivity study indicates that the effect of forward scattering on the aerosol SSA retrievals, during these four days, is also small, no bigger than 0.007 or 0.73%. However, for a heavy aerosol loading with large aerosol particle size, the impact of the forward scattering is not negligible.

As discussed previously, the asymmetry factor, even constrained with observed Angstrom coefficient, has some uncertainty (± 0.1). In this study, an overestimate of 0.1 in asymmetry factor results in an underestimate of 2.54%~3.4% in retrieved aerosol SSA on these days (shown in Figures 5(g-h) and 6(g-h)). Rayleigh optical depth is another source of uncertainty. If we adopt the common value of Rayleigh optical depth of 0.2930 [20], the changes in retrieved aerosol SSA are 0.015, 0.009, 0.003, and 0.002 for aerosol loading of 0.13 (20080310), 0.20 (20080311), 0.47 (20110607), and 0.60 (20110608), respectively. It is clear that for a small aerosol loading, the uncertainty associated with the choice of Rayleigh optical depth value is not negligible. For the 415 nm passband, the baseline of surface albedo is very small and varies within a narrow range (0.005). However, to illustrate the sensitivity of surface albedo, we intentionally prescribe an uncertainty of 0.05 for surface albedo. As indicated by the sensitivity study, this will result in changes of 0.052 (5.40%), 0.035 (3.70%), 0.011 (1.13%), and 0.014 (1.49%) in retrieved SSA, respectively for 20080310, 20080311, 20110607, and 20110608. As many others use measurements at the wavelengths with a large albedo uncertainty, the uncertainty associated with surface albedo could result in a substantial error or uncertainty in aerosol SSA retrievals, especially for the instances with low aerosol loading.

4.3 Statistical Analysis of Retrieved SSA

To evaluate the performance of the retrieval algorithm, MFRSR measurements for year 2008 at ARM SGP site has been processed to derive aerosol optical properties, including aerosol SSA. Because there is no snow at ARM SGP site, the surface albedo at 415 nm remains 0.033 through the year. As shown in Figure 7a, total AOD at 415 nm shows significantly seasonal variation: increasing from January, reaching the maximum in July and August (days 180-240), and then decreasing from August to December. The aerosol asymmetry factor derived from MFRSR (415 nm), from AOS (450 nm), and from AERONET (440 nm), are shown in Figure 7b: the former two column integrated values are close to each other, most of them are in the range of 0.65~0.75; the AOS derived one has obviously smaller value and bigger variance range. Difference between them is probably caused by two main causes: one is the retrieval error and measurement error, the other is the real difference of aerosol between column integrated aerosols and near surface aerosols. The aerosol SSAs derived from MFRSR (415 nm) and from AOS (450 nm), shown in Figure 7c, exhibit a consistent seasonal variation. Most of daily mean SSAs are larger than 0.85 and the monthly mean SSAs are close to or exceed 0.9, suggesting that aerosols over the ARM SGP site are mainly consists of low absorption aerosols. Based on total AOD and the retrieved SSA, we derive the absorption AOD, as $AOD \cdot (1 - SSA)$, also shown in Figure 7a. Although absorption AOD is much smaller than total AOD, it has a similar seasonal cycle as total AOD does. Figure 7d shows the comparison of MFRSR

retrieved SSA with AOS retrieved SSA. The correlation coefficients between MFRSR and AOS SSAs are 0.47 (or 0.55) for AOD smaller (or bigger) than 0.25, suggesting some coupling of near surface aerosols with the total aerosols in the atmosphere. When AOD is bigger than 0.25, most of the MFRSR retrieved SSA is bigger than AOS measured SSA.

Figures 7e and 7f show the scatter plots of aerosol SSA versus AOD and Angstrom coefficients, respectively. Both retrieved MFRSR SSA and measured AOS SSA increase with aerosol loading (AOD) and Angstrom coefficients, with consistent tendencies between MFRSR SSA and AOS SSA. It suggests that the lower MFRSR retrieved SSA at lower AOD is not due to larger uncertainty but fundamentally a higher percentage of absorption substance at a lower aerosol loading. Although aerosol absorption depth increases with total aerosol optical depth (Figure 7c), a smaller percentage increase of total aerosol is in the absorbing component. Over the SGP site, aerosols are mainly originated from air pollutant that transported from surrounding cities and industrial areas, and partly from the biomass burning and farming activity [31]. For aerosols over this site, the aerosol SSA roughly decreases with the aerosol particle size, as suggested by the Angstrom coefficients.

5. Discussion and Conclusion

In this paper, we developed a retrieval algorithm of aerosol SSA by using both diffuse horizontal and direct normal fluxes simultaneously measured by a MFRSR. We utilized the unique shadowband capability of MFRSR, i.e., both components can be calibrated through Langley regression calibration, and the diffuse-direct ratio is independent of absolute calibration. Furthermore, we carefully selected measurement channels at 415 nm and 870 nm to minimize the interference of gaseous absorption for aerosol optical depth and Angstrom coefficient retrievals. We focused on 415 nm for aerosol SSA retrievals to reduce the effect of the surface albedo, since the surface albedo at 415 nm is low and stable without the snow on the ground. Under those constraints and assumptions, we solved the radiative transfer equation iteratively to derive the SSA. To speed up the retrievals, we built a lookup table of simulated DDR for different atmospheric conditions. Through matching the observed DDR and the simulated DDR, we can derive the SSA quickly without time-consuming iteratively radiative transfer calculating. We applied this retrieval algorithm to the MFRSR measurements at the ARM SGP site. The retrieved results of MFRSR measurements are compared with other independent measurements at the site. The MFRSR derived aerosol optical depth, Angstrom coefficient, and SSA are well consistent with AEROENT measurements. Due to the difference of observed targets (i.e., column integrated aerosols vs. near surface aerosols), the aerosol SSA derived from MFRSR and AOS are not always well consistent with each other.

We assessed various issues impacting on the accuracy of SSA retrieval from measurements to input parameters and assumptions. The uncertainty assessments of surface albedo, cosine response, forward scattering, and asymmetry factor are consistent with the findings of Meloni et al. [9]. Our sensitivity study indicated that: 1) Calibration accuracy is still an important factor in the SSA retrievals, especially for observations with small aerosol loading, e.g., 1% calibration uncertainty can result in 0.8~3.7% changes in retrieved SSA for aerosol loading of 0.13-0.60. 2) The cosine response correction factor, which has been ignored by previous studies, effectively reduces the diffuse irradiance, which can consequently result in an underestimation of 1.1~3.3% in aerosol SSA. 3) Forward scattering correction is important for a heavy aerosol loading and a large aerosol particle size, which could result in an uncertainty of up to 0.73% in retrieved SSA. 4) The uncertainty associated with asymmetry factor, even with the constraints of Angstrom coefficient and/or retrieved aerosol size distribution, is large and is one of the biggest error sources for aerosol SSA retrievals other than the instrument calibration. An overestimation of 0.1 in asymmetry factor can result in an underestimation of 2.54~3.4% in retrieved SSA. 5) The uncertainty associated with the choice of Rayleigh optical depth value is not negligible, particularly for small aerosol loading, which could result in an uncertainty of 0.015 in retrieved SSA. 6) The uncertainty associated with surface albedo could result in a substantial error or uncertainty in aerosol SSA retrievals, e.g., an uncertainty of 0.05 for surface albedo at 415 nm can result in changes of 1.49~5.4% in retrieved SSA. For

wavelengths longer than 500 nm, the surface albedo is high and varies with time, which results in larger uncertainty in the SSA retrievals. In general, for MFRSR measurements at 415 nm, the uncertainty associated with asymmetry factor, calibration, and surface albedo are the three biggest error sources. Importantly, we developed various constraints and corrections in the retrieval algorithm to minimize those uncertainties. Validation and evaluation of our retrieved aerosol SSA with AOS and CSPHOT co-located measurements indicate the robustness of our retrievals for various aerosol conditions.

We applied this retrieval algorithm to year-long MFRSR measurement at the ARM SGP site, and analyzed the statistically characteristics of retrieved aerosol SSA. The year-long SSAs retrieved from MFRSR are correlated with the co-located near surface SSA measured by AOS. For both instruments, the mean values of retrieved SSA increases with AOD and Angstrom coefficient, but the variance of retrieved SSA decreases with AOD and Angstrom coefficient. Based on the retrieved SSA and total AOD, the absorption AOD can be derived directly. At the ARM SGP site, the absorption AOD has a positive correlation with the total AOD, showing a similar seasonal cycle as total AOD, with large values occurred in the summer months.

Cloud screening is the most important issue beyond those discussed here, since our retrieval algorithm is based on the plane-parallel assumption and any cloud

contamination violates this basic assumption. Cloud information can be obtained from various active sensors, such as cloud radar and lidar. However, most of those active sensors without scanning capability only provide 2- dimensional sky condition (vertical and temporal), which are not measuring at the scales of radiative transfer of both horizontal and vertical inhomogeneity. It is possible to distinguish atmospheric inhomogeneity by using the diffuse transmittance ratio between a longer wavelength (870 nm) and a short wavelength (415 nm) measured by a MFRSR. As diffuse transmittances at different wavelengths vary with solar zenith angle systematically, the transmittance ratio at two wavelengths is less dependent on solar zenith angle (or time). Also since there are different spectral dependences of cloud and aerosol optical depth, the diffuse transmittance ratio is strongly sensitive to the cloud amount within the field of view of MFRSR. By analyzing the variability of the diffuse transmittance ratio, a long stable period can be selected for the aerosol SSA retrieval. Notice that this characteristic has been utilized to infer cloud sky fraction from the measurements of MFRSR [13].

Acknowledgments

This work was supported by US DOE's Atmospheric System Research program (Office of Science, OBER) under contract DE-FG02-03ER63531, by the NSF under contract AGS-1138495, and by the NOAA Educational Partnership Program with

Minority Serving Institutions (EPP/MSI) under cooperative agreements

NA17AE1625 and NA17AE1623.

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Table 1. Values of Rayleigh Optical Depth at 415 and 870 nm

Wavelength (nm)	Hansen and Travis, [20]	Boadhaine et al. [21]	Thomasi et al. [22]	Srivastava et al. [23] (quantaum theory)	Srivastava et al. [23] (Classic theory)
415	0.2930	0.3091	0.3094	0.2992	0.3105
870	0.0151	0.0151	0.0152	0.0147	0.0152

Table 2. Surface albedo measurements from different platforms at ARM SGP site.

(This data was measured on March 3, 2000, and plotted by Michalsky et al. [25].)

Wavelength	MFR	ASD	SSFR
415 nm	0.033	0.034	0.038
870 nm	0.274	0.270	0.284

Table 3. The comparison of retrieved aerosol single scattering albedo (SSA) at 415 nm from MFRSR (average values of 10 minutes) under different retrieval algorithm setting with the SSA derived from AOS and AERONET (level 2.0). I_0 , f^{CRC} , Δ^{FS} , g , τ_{ray} , A_{SRF} are relative solar constant at the top of the Atmosphere, sensor surface cosine respond factor calibration, forward scattering correction, aerosol asymmetry factor, Rayleigh scattering optical depth, and surface albedo (at 415 nm), respectively.

Case #		20080310	20080311	20110607	20110608
Time (GMT)		16:36	16:36	12:54	21:36
AOD : MFRSR 415nm		0.13	0.20	0.47	0.60
g: MFRSR 415 nm		0.70	0.68	0.67	0.65
SSA: AOS 450 nm		0.965	0.937	0.954	0.952
SSA: Aeronet 440 nm		N/A	N/A	0.988	0.955
SSA (MFRSR 415 nm)	baseline	0.964	0.946	0.974	0.940
	$1.01 * I_0$	0.928	0.922	0.967	0.932
	f^{CRC} none	0.932	0.922	0.961	0.930
	Δ^{FS} none	0.957	0.940	0.969	0.936
	$g+0.1$	0.931	0.914	0.949	0.911
	$\tau_{ray} -0.016$	0.949	0.937	0.971	0.938
	$A_{SRF} +0.05$	0.912	0.911	0.963	0.926

Figure Captions

Figure 1. (a) Variations of simulated diffuse transmittance with aerosol optical depth (AOD) for different given aerosol single scattering albedos (SSAs). (b) Variations of the ratio of simulated diffuse transmittance under different aerosol SSAs with AOD. (c) Variations of simulated diffuse-direct-ratio (DDR) with AOD for different aerosol SSAs. (d) Variations of simulated DDR with SSA for different aerosol AODs. Where, the other model parameters such as the solar zenith angle, surface albedo, and aerosol asymmetry factor are set as 45° , 0.033, and 0.65, respectively.

Figure 2. (a) The Forgan values of I_0 at the passbands of 415 and 870 nm from 1997 to 2012. (b) The comparison of Langley values and Forgan values for the period of 2008.

Figure 3. (a) Cosine correction coefficient for the diffuser of MFRSR at 415 and 870 nm; (b) the relationship between the cosine respond factor and solar zenith angle for known aerosol optical depths at 415 nm.

Figure 4. The asymmetry factor and Angstrom coefficient: AOS measurements in March, 2008 (scatter points), and Mie calculations (lines).

Figure 5. Comparisons of aerosol optical depth, Angstrom coefficient, asymmetry factor, and single scattering albedo (SSA) inferred from MFRSR, AERONET and AOS on March 10 and 11, 2008, where the baseline or g+0.1 indicate aerosol SSA is retrieved by using the baseline setting or an overestimated (+0.1) asymmetry factor, respectively.

Figure 6. Comparisons of aerosol optical depth, Angstrom coefficient, asymmetry factor, and single scattering albedo (SSA) inferred from MFRSR, AERONET and AOS on June 7 and 8, 2011, where the baseline or g+0.1 indicate aerosol SSA is retrieved by using the baseline setting or an overestimated (+0.1) asymmetry factor, respectively.

Figure 7. Daily mean values of (a) aerosol optical depth AOD (total and absorption component), (b) aerosol asymmetry factor, and (c) aerosol single scattering albedo (SSA) at SGP C1 site in 2008. (d) Comparison of aerosol SSA derived from MFRSR with that from AOS. The scatter plots of aerosol SSA versus (e) the related AOD and (f) Angstrom coefficients.

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