Aerosol Properties from Surface Radiation Observations: Anomalous Clear-Sky Absorption May be Due to Small, Highly Absorbing Aerosols

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Introduction

A growing body of evidence indicates that standard models overestimate the diffuse shortwave radiation in clear-sky conditions. The magnitude of the difference (9% to 40% of the diffuse observations, depending on aerosol loading) suggests that the discrepancy is not primarily due to measurement uncertainties. It has been proposed (Kato et al. 1997) that the excess absorption is due to an unidentified gas. But another possibility is the presence of highly absorptive aerosols. In this paper the second possibility is investigated. We base our comparisons on observations of direct and diffuse shortwave radiation obtained at the Southern Great Plain (SGP) site of the Atmospheric Radiation Measurement (ARM). These data are compared to modeling results for a range of aerosol parameters.

Observations

For the comparisons discussed in this paper, two essentially cloud free days (272 or 97/09/29, and 277 or 97/10/04) were selected as they represent extremes in aerosol loading. Day 277 has approximately four times the aerosol optical depth measured on Day 272. The radiation observations used here were acquired by the Baseline Surface Radiation Network (BSRN) at the ARM SGP central facility. observations from a shaded Eppley Precision Spectral Pyranometer (PSP) and an Eppley Normal-Incidence Pyrheliometer (NIP) were used to obtain the diffuse and direct surface shortwave radiation, respectively. The total shortwave irradiance was reconstructed by adding the diffuse irradiance to the product of the direct-normal irradiance and the cosine of the solar zenith angle. The uncertainty on the derivation of the diffuse shortwave component has been estimated to be of the order of 5% to 8% by

Halthore et al. (1998), but could be larger due to improper temperature corrections (Bush et al. 1998). The vertical profile of water vapor was obtained from radiosondes and the Microwave Radiometer (MWR). The Total Ozone Mapping Spectrometer (TOMS) provided noontime estimates of total column ozone. Aerosol optical depth was obtained from observations of the Multi-Filter Rotating Shadowband (MFRSR), which provided optical depth observations at 30-minute intervals within narrow (10 nm) passbands at 414, 499, 609, 665, and 860 nm. The aerosol loading on day 272 is small and shows variation throughout the day (τ= .06 ± .005 @ 0.41 um). Day 277 has a much larger aerosol optical depth, which increases from morning to afternoon by almost 50%. Measurements from tower-mounted up- and down-looking MFRSR provided spectral surface albedo near the central facility. The albedo is about 10% is the visible ramping up to 30% in the NIP.

The Whole Sky Imager (WSI) provides observations of the angular characteristics of the diffuse radiation field at two wavelengths, 450 nm and 650 nm, with a spectral width of 70 nm. These images were compared to our model computations. The uncertainty in the WSI radiometric data is estimated to be about 10% and mostly results from the uncertainty of the calibration lamps and their positioning accuracy (about 1/3 degree).

Model Results and Comparisons

Figure 1 shows the BSRN shortwave observations compared to model predictions made with SBDART, a discrete ordinate radiative transfer (DISORT)-based low-resolution radiative transfer code (Ricchiazzi

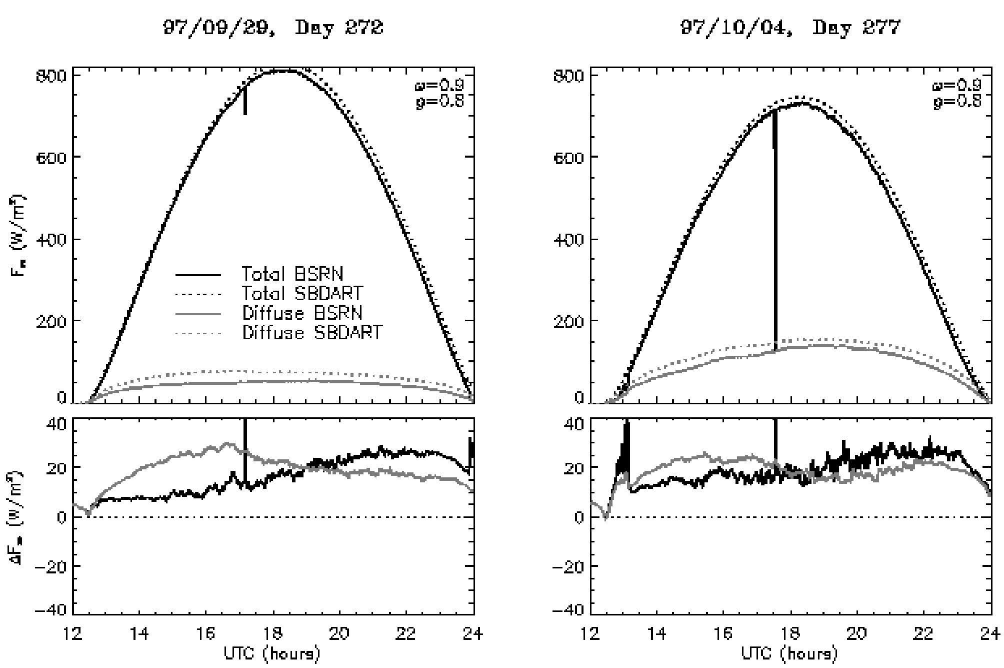

Figure 1. Total shortwave irradiance for days 272 and 277 from the PSP (solid) and as predicted by SBDART with standard aerosol properties (dotted). The lower panels show the difference between SBDART and PSP observations for total (black) and diffuse (gray) shortwave irradiance.

et al. 1998). In this initial SBDART computation, we used a standard rural aerosol model (Shettle and Fenn 1975), with single-scattering albedo, ω, greater than 0.9 and asymmetry factor, g, about 0.8. Whereas the total shortwave is modeled fairly well for day 272 and 277, a large relative discrepancy is obtained for the diffuse radiation field (lower panels). There is a good agreement (15 Wm^{-2} to 20 Wm^{-2}) between model computations (dotted line) and observations (solid line) for the total, which is roughly consistent with the 3% calibration accuracy of the instruments and model errors. SBDART's prediction of direct shortwave irradiance (not shown) is within about 1% of the NIP observations. Such fine agreement is not the case for the diffuse component which is overestimated by SBDART by up to 50% (25 Wm^{-2}). A potential source for this discrepancy may be an incorrect choice of aerosol scattering parameters. To check this possibility we repeated the SBDART simulation with ω=0.5, keeping g=0.8. These input parameters are meant to represent aerosols that absorb much more strongly than the rural, mineral-based aerosols that are thought to represent conditions in the SGP site. With these inputs the predicted diffuse radiation decreased significantly, to a level within a few percent of the observations. A similar improvement can be obtained by fixing ω at 0.6 and lowering g to 0, which is equivalent to assuming very small aerosols particles, with a nearly isotropic scattering phase function. Many other combinations of ω and g, with values in between those extremes, represent possible solutions.

Exploring this issue further, we generated a table of Mie scattering parameters (extinction efficiency, single-scattering albedo, and phase function) for a range of mono- and bi-modal particle distributions. For each day we selected those distributions that displayed a spectral slope of the extinction efficiency that matched those observed by the MFRSR, i.e., approximately 1.4 and 1.8 for days 272 and 277, respectively. The combination of aerosol parameters that produce the required spectral slopes are shown in Figure 2, for the case of a mono-modal radius distribution. These aerosol parameters were used as

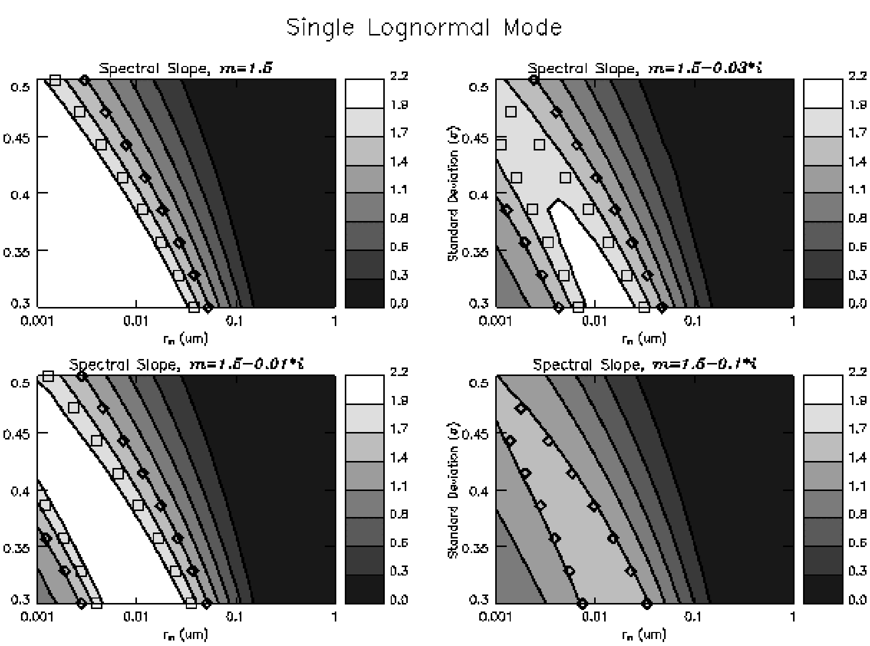

Figure 2. Contour plots of the spectral slope of extinction efficiency as a function of monomodal particle distribution profile parameters, mode radius and standard deviation (i.e., the standard deviation of log 10 [r]).

input for SBDART, thereby generating a corresponding table of radiance profiles. Figure 3a shows a scatter plot of these results. The ratio of predicted to observed diffuse irradiance varies along the

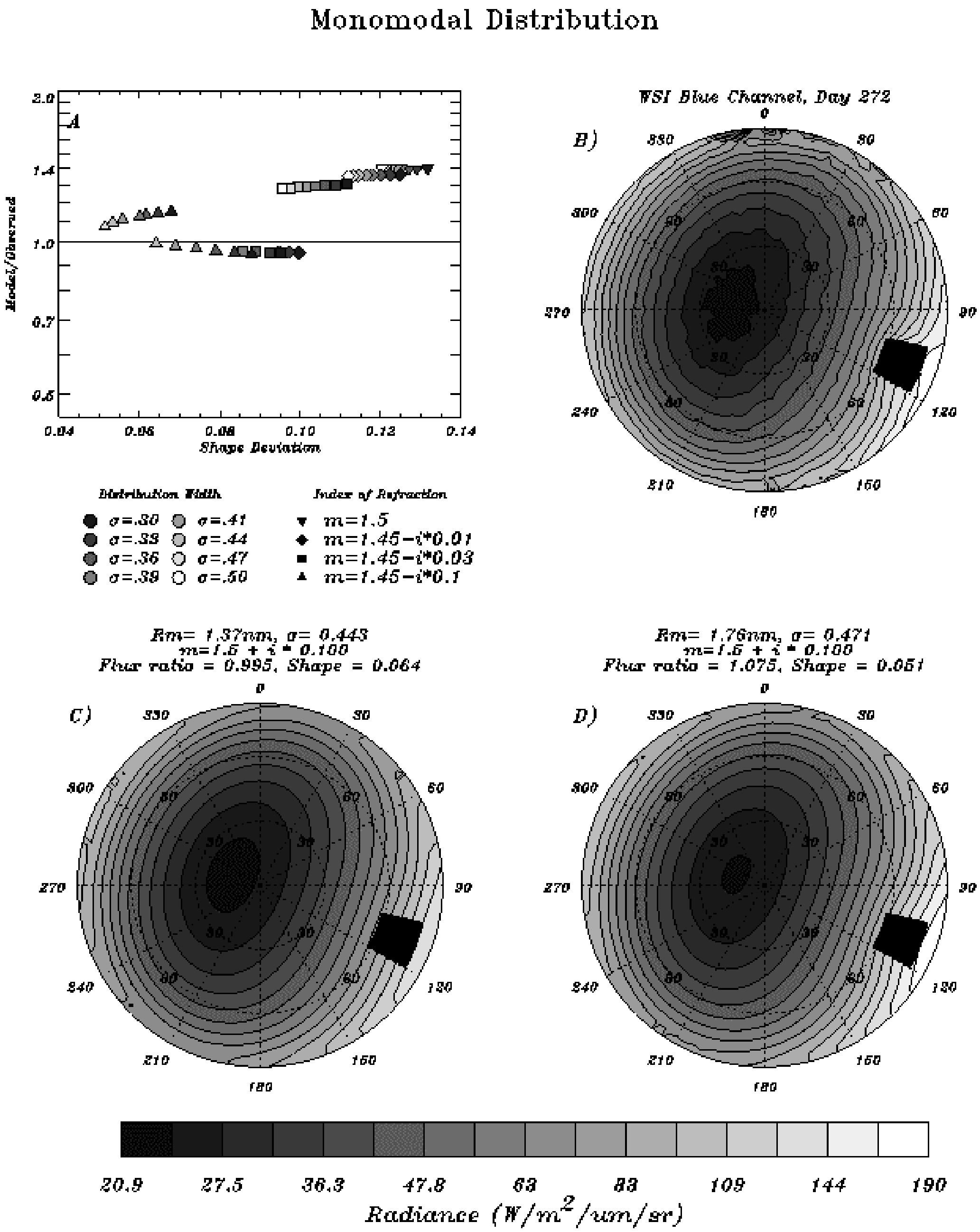

Figure 3. Radiance comparisons, day 272. The mono-modal Mie scattering parameters resulting in spectral slopes close to 1.4 (the diamond symbols in Figure 2) were used in SBDART to generate predictions of sky radiance. a) Scatter plot: of surface irradiance ration (SBDART/WSI) and shape deviation. b) The WSI radiance distribution for the blue channel. c) SBDART radiance for aerosol set that produced the best irradiance match. d) SBDART radiance for aerosol set that produced the best shape.

vertical axis, while the radiance shape deviation (RSD) varies along the horizontal axis. The RSD is defined as the standard deviation of log (Radiance.sbdart/Radiance.wsi) evaluated over all solid angles. This parameter is independent of the overall calibration of the WSI. Figure 3b shows the WSI radiance distribution for the blue channel for the same day and time. The model runs that produced the best irradiance match and best angular radiance distribution are shown in Figures 3c and 3d, respectively.

Conclusion and Discussion

The aerosol retrieval for day 272 yields the imaginary part of the index of refraction of 0.1, and corresponds to a single-scattering albedo of 0.474 (very soot like). For day 277 the imaginary part of the index of refraction was much smaller, about 0.01, and resulted in a single-scattering albedo of 0.865 (similar to mineral dust). These results suggest that the discrepancy found between the modeled and observed diffuse downward shortwave irradiance may be caused by the presence of small particles in the atmosphere, in addition to the aerosols typical of the site. These aerosol particles would be extremely absorbing and possibly similar to large gaseous molecules. The angular distribution observations from the WSI suggest, however, that their scattering properties differ from those of a gas since Rayleigh scattering was not an optimal solution to our minimization problem on the less turbid cloud-free day.

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