Aerosol single scattering albedo retrieved from measurements of surface UV irradiance and a radiative transfer model

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[1] Aerosol single scattering albedo (ω), the ratio of aerosol scattering coefficient to total aerosol extinction coefficient, at UV wavelengths is an important aerosol radiative parameter in determining surface UV irradiance. Surface measurements of total and diffuse UV irradiance in the summer and fall of 1999 at the seven narrowband wavelength channels of an UV multifilter rotating shadowband radiometer (UVMFR-SR) at Black Mountain, N. C., were coupled with a tropospheric ultraviolet radiative transfer model to produce values of ω . Its value ranged from 0.65 to 0.91 at 300 nm, 0.71 to 0.96 at 305.5 nm, 0.73 to 0.97 at 311.4 nm, 0.74 to 0.91 at 317.6 nm, 0.76 to 0.96 at 325.4 nm, 0.77 to 0.97 at 332.4 nm, and 0.80 to 0.99 at 368 nm. Error in this procedure decreases with increasing aerosol optical depth (AOD), from ± 0.63 at AOD = 0.05 to ± 0.04 at AOD = 1.0 averaged over the seven wavelengths. The current values of ω have a slightly wider variation than values reported from a previous study at the same site. The lower values of ω could indicate that, over the site, preferential absorption of UV radiation by black carbon aerosols could be occurring. More values of ω in the UV spectrum will allow for better estimation of this parameter for UV radiative transfer modeling and will lessen error in INDEX TERMS: 0305 Atmospheric Composition and estimation of surface UV irradiances. Structure: Aerosols and particles (0345, 4801); 0360 Atmospheric Composition and Structure: Transmission and scattering of radiation; 0394 Atmospheric Composition and Structure: Instruments and techniques; KEYWORDS: aerosol scattering, ultraviolet radiation, radiative transfer modeling, aerosol single scattering albedo, aerosol optical depth, black carbon aerosol

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1. Introduction

[2] Ultraviolet (UV) radiation has an important role in both animal and plant life. Recent knowledge of stratospheric ozone depletion has led to concerns about increased levels of biologically harmful UV-B (280–320 nm) radiation reaching the Earth's surface, relative to more beneficial UV-A (320–400 nm) radiation. Increased UV-B surface irradiance can lead to damage to both terrestrial and oceanic organisms, and increases in the incidences of cataracts and skin cancer in humans [Madronich et al., 1998]. Clouds and atmospheric aerosols have been found to attenuate UV radiation and are believed to mask the increase of UV irradiance due to stratospheric

ozone depletion [Frederick et al., 1993; Meleti and Cappellani, 2000].

[3] The effect of tropospheric aerosols on UV radiation varies widely in time and space due to their short residence time (2–7 days), variability in size, shape, and chemical composition, and dependence on relative humidity [Reuder and Schwander, 1999]. For this reason, an extensive ground-based network of UV radiation monitoring instruments is required to obtain a spatial and temporal variation of UV irradiance at the surface. Such networks do exist; however, it is difficult to obtain ground-based measurements over the oceans, and radiative transfer modeling offers a much cheaper alternative to quantifying UV surface irradiance. These models are only as accurate as their inputs. More information on aerosol radiative properties is required if they are to be effectively parameterized for use in radiative transfer models.

[4] Reuder and Schwander [1999] conducted a sensitivity study using the radiative transfer model STAR to determine which aerosol properties are most decisive in determining the short-term variability of aerosol effect on UV radiation. It was found that more than 80% of the aerosol effect on UV radiation due to increasing turbidity is determined by aerosol optical depth (AOD) and single scattering albedo (ω) . The latter is the ratio of the aerosol scattering coef-

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ficient to the total aerosol extinction coefficient (scattering plus absorption). Many studies have obtained values for ω in the visible wavelengths for specific locations and times [Anderson et al., 1999; Devaux et al., 1998; Dubovik et al., 1998, 2002; Eck et al., 1998; Ogren and Sheridan, 1996; Waggoner et al., 1981; Yu et al., 2000], as these wavelengths are important to climate forcing [Schwartz et al., 1995]. However, less research has been done to determine values of ω in the UV wavelengths [Wenny et al., 1998; Kylling et al., 1998]. Madronich [1993a] and Lacis and Mishchenko [1995] state that in the UV spectrum ω varies between 0.5 and 1.0, depending on particle composition. Dust and soot aerosols tend to have lower values (0.5 to 0.7), while sulfate aerosols have values closer to 1.0. Single scattering albedo in the UV wavelengths can be expected to vary as the aerosol effect on UV radiation; scattering and absorption properties of aerosols have strong dependence on wavelength, relative humidity, size, shape, and chemical composition [Reuder and Schwander, 1999]. Thus, investigating its variation in these wavelengths will allow for better parameterization of its value.

[5] The objective of this study is to provide values of ω in the UV spectrum that can be considered representative for clear-sky summer days in the southeastern United States. Values of ω were retrieved with an inversion procedure using measurements from an ultraviolet multifilter rotating shadowband radiometer (UVMFR-SR) coupled with a radiative transfer model. Single scattering albedo was determined for the seven operational wavelengths of the UVMFR-SR. Back trajectory analysis was then used to determine if values of ω could be correlated with air mass classifications as determined at the site [Saxena and Yeh, 1989; Deininger and Saxena, 1997; Ulman and Saxena, 1997]. Such information is required to assess the impact of aerosols in these wavelengths [Schwander et al., 1997; Kylling et al., 1998]. Better estimates of this parameter can serve as input into radiative transfer modeling studies of surface UV trends. Furthermore, knowledge of the regional characteristics of ω will lead to the reduction of errors in satellite estimation of surface UV irradiance [Krotkov et al., 1998].

2. Methodology

2.1. Instrumentation and Research Site

[6] The UVMFR-SR was deployed near the town of Black Mountain, NC (35.66°N, 82.35°W, 753 m above mean sea level) atop a 10-m tower. More detail of the setup of the research site is available from Schafer et al. [1996] and Wenny et al. [1998]. The database used for this study was collected between July and December 1999. The UVMFR-SR works under the same principles as the original visible wavelength version described by Harrison et al. [1994] and is detailed by Bigelow et al. [1998]. This instrument measures both total and diffuse irradiance for seven specified wavelengths (300, 305.5, 311.4, 317.6, 325.4, 332.4, and 368 nm) with a 2-nm nominal full width at half maximum (FWHM) bandwidth. Measurements of total and diffuse irradiance were recorded every 20 s, and stored as 2-m averages along with a computed direct irradiance. The cosine response and irradiance calibration of the UVMFR-SR were conducted by the manufacturer,

Yankee Environmental Systems™, in spring 1999 before deployment at the field site.

2.2. Retrieval of ω

[7] The ω retrieval procedure employed the data collected by the UVMFR-SR and the tropospheric ultraviolet radiative transfer model TUV4.1 [Madronich, 1993b]. The TUV4.1 uses a discrete ordinate method of determining radiative transfer through the atmosphere, and was run in the 8-stream mode [Stamnes et al., 1988]. This model has been used in other UV radiation studies conducted at the site [Wenny et al., 1998, 2001]. The original code, which outputs various spectral irradiances and radiances in the UV and visible spectrums, was modified to output diffuse-todirect ratio (DDR) at the wavelengths of the UVMFR-SR. The model inputs relevant to the present study are the time of solar noon, aerosol optical depth (AOD), total ozone column (TOC), asymmetry parameter (g), ground albedo (ga), and ω . The initial output of DDR at the morning solar zenith angle of 45° from TUV4.1 was compared with instantaneous measurements taken by the UVMFR-SR. TUV4.1 output of direct horizontal irradiance was divided by the cosine of the solar zenith angle to give direct normal irradiance so as to match the direct normal irradiance measurements of the UVMFR-SR. The DDR was derived from the UVMFR-SR measurements as the ratio between the recorded diffuse and direct irradiances. The TUV4.1 was iterated by varying ω until the DDR output matched that of the UVMFR-SR, ultimately yielding ω for each wavelength for each morning at a solar zenith angle of 45°. Figure 1 outlines the inversion process of ω retrieval in a flowchart. It is assumed that the value of ω retrieved does not vary with altitude. TUV4.1 does allow for varying ω or g values with altitude, but there is little data on how these radiative parameters vary with height in the UV wavelengths. Ogren and Sheridan [1996] found that ω and g varied little with altitude in the visible wavelengths. Because of the inherent difficulties clouds bring to radiation studies, their influence is neglected here. Overcast sky conditions have been shown to attenuate up to 70% of clear-sky (280-320 nm) irradiance at a solar zenith angle of 50° [Schafer et al., 1996]. However, the three-dimensional distribution of clouds over a region is difficult to obtain and, under the right circumstances, can increase UV irradiance reaching the surface [Schafer et al., 1996; Weihs et al., 2000]. To ensure cloudless conditions at the 45° solar zenith angle, each day's diffuse irradiance was plotted against time of day. If the top of the characteristic "bell curve" of the irradiance plot was seen as uninterrupted for some time before and after this solar zenith angle in the morning, that day was deemed usable for ω retrieval.

[8] The inputs used in TUV4.1 were obtained or assumed as follows. AOD was calculated from the irradiance measurements at the seven wavelengths of the UVMFR-SR's by Wenny et al. [2001] for 74 clear sky days in the observation period of 1999. A detailed explanation of AOD retrieval can be found there. Briefly, Langley plot analysis was used on site to obtain an average value of V_o (the voltage signal the UVMFR-SR would record at the top of the atmosphere) for the morning time, as prescribed by Harrison and Michalsky [1994]. This method assumes that the atmospheric conditions above the research site are stable for the measurement

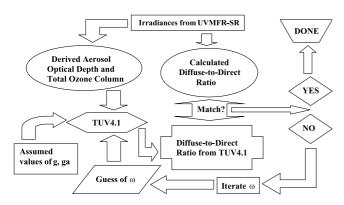


Figure 1. Diagram outlining the inversion process of single scattering albedo (ω) retrieval.

period, a requirement not optimally met at Black Mountain, N. C. This leads to uncertainties in the V_o values, and hence uncertainties in the AOD retrieval, discussed in section 3.2. Given these V_o values for each wavelength channel, the following formula can be used to obtain total optical depth (TOD):

$$TOD = -(\cos Z) * ln(V_{\lambda}/V_{\lambda o})$$

where V_{λ} is the UVMFR-SR voltage signal at wavelength λ , $V_{\lambda o}$ is the extraterrestrial voltage signal at wavelength λ , and Z is the solar zenith angle. From TOD, Rayleigh and ozone optical depths are subtracted, leaving AOD. Absorption by other atmospheric gases is considered negligible for the wavelength channels of the UVMFR-SR. Rayleigh optical depth is assumed constant for the site's elevation, and ozone optical depth is calculated from total ozone column, ozone absorption coefficients, and the filter function as measured by Yankee Environmental Systems . This process was conducted for each 2-min measurement. The model input of AOD is at 340 nm. Angstrom's formula was used to extrapolate AOD to this wavelength from the

AOD retrieved from the four longer wavelengths of the UVMFR-SR. The AOD inputs were morning averages using clear-sky measurements for $Z \leq 60^{\circ}$. TUV4.1 assumes that the model input of AOD at 340 nm varies inversely with the first power of wavelength [Madronich, 1993b], and that it varies with altitude according to the Elterman [1968] profile.

- [9] Total ozone column (TOC), was derived from measurements of the UVMFR-SR, and its retrieval is detailed by *Wenny et al.* [2001]. The technique used to retrieve TOC will not be described here, as TOC will be shown in the error analysis section to be an insignificant input, and could have been assigned a constant value of 300 Dobson Units, with no change in retrieved ω values.
- [10] Asymmetry parameter, g, was assumed a value of 0.70, for all wavelengths and altitudes. This was the average of nine values from a previous UV-B radiation study conducted at the same site [Wenny et al., 1998]. These values were calculated at 312 nm in the same months of the year as the current study. Also, Madronich [1993b] states that g typically falls between 0.6 and 0.8 for the UV wavelengths, of which the median is also 0.70.
- [11] Ground albedo, ga, was assumed a value of 0.04, for all wavelengths and altitudes. This value was originally taken from *Schwander et al.* [1997]. Above grassy meadows such as those surrounding the research site, the ground albedo in UV wavelengths has typically been found to be between 0.01 and 0.03 [*Blumthaler and Ambach*, 1988; *Diffey et al.*, 1995; *Feister and Grewe*, 1995; *McKenzie et al.*, 1996]. A more recent study by *Kylling et al.* [2000] gives a value of 0.08 for the same ground cover.

2.3. Correlation With Air Mass Type

[12] Figure 2 shows the demarcation of geographical sectors of different air mass types influencing the research site. Saxena and Yeh [1989] determined the air mass sectors based on the United States Environmental Protection Agency's source inventory data on SO_x and NO_x emissions [Environmental Protection Agency, 1993]. Deininger and Saxena [1997] and Ulman and Saxena [1997] conducted

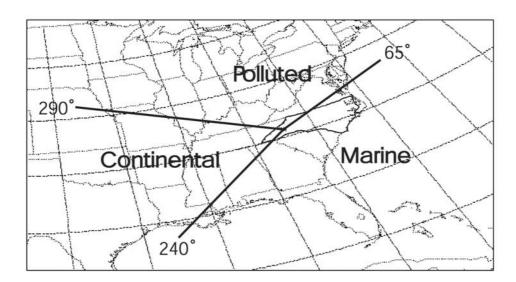


Figure 2. Map of the United States illustrating the geographical location and classification of the different air mass types influencing the research site in Black Mountain, North Carolina.

Table 1. Single Scattering Albedo (ω) Values Retrieved at the Seven Wavelengths of the UV-MFRSR^a

Date	AOD at 340 nm	SZA	At 300.0 nm	At 305.5 nm	At 311.4 nm	At 317.6 nm	At 325.4 nm	At 332.4 nm	At 368.0 nm	Air Mass
7/15/1999	0.827	45.01	0.65	0.71	0.73	0.74	0.76	0.77	0.80	C_p
8/10/1999	0.491	44.88	0.84	0.88	0.90	0.90	0.91	0.93	0.99	C
8/11/1999	0.298	45.02	0.83	0.91	0.92	0.91	0.91	0.92	0.92	C
8/16/1999	0.562	44.97	0.87	0.92	0.93	0.94	0.95	0.96	0.99	M
8/17/1999	0.772	45.12	0.73	0.81	0.83	0.83	0.84	0.85	0.85	C
8/18/1999	0.514	44.89	0.88	0.96	0.97	0.96	0.96	0.97	0.96	HP
8/26/1999	0.509	45.07	0.82	0.86	0.89	0.89	0.91	0.92	0.94	C
9/7/1999	0.653	44.87	0.91	0.90	0.91	0.91	0.91	0.91	0.89	C
9/8/1999	0.324	45.09	0.84	0.88	0.87	0.86	0.86	0.87	0.85	HP
9/13/1999	0.384	44.88	0.89	0.91	0.91	0.89	0.88	0.88	0.81	M
10/5/1999	0.421	45.08	0.78	0.77	0.77	0.78	0.79	0.80	0.83	C
AVERAGE	0.523	44.99	0.82	0.86	0.88	0.87	0.88	0.89	0.89	

^aAsymmetry parameter = 0.70, ground albedo = 0.04. Displayed to the left are the values of aerosol optical depth (AOD) at 340 nm and the actual solar zenith angle (SZA). Displayed to the right are the air mass classifications as determined by the HY-SPLIT 48-hour back trajectories (HP = highly polluted, C = continental, M = marine).

broad-based studies to validate the demarcation of the marine, continental, and highly polluted sectors. The marine sector was found to be dominated by salt aerosols, the continental sector by a combination of silicate and soot aerosols, and the polluted sector is dominated by soot and sulfate aerosols [Deininger and Saxena, 1997]. The pH was found to also be correlated with the sector classifications [Ulman and Saxena, 1997]. The 48-hour back trajectories were computed from the site using the HYbrid Single-Particle Lagrangian Integrated Trajectory (HY-SPLIT) Model for each value of ω to see which sector the air over the site came from [Draxler and Hess, 1998]. Back trajectories were computed as if the air parcel reached an altitude of 1 km above the site. If the computed trajectory went over more than one sector, that trajectory was classified by which sector it longest remained over.

3. Results and Discussion

3.1. Values/Air Mass Correlation

[13] Single scatter albedo (ω) values were obtained for the seven wavelengths of the UV-MFRSR for 11 days from July 15 to October 5 of 1999. The value of ω ranged from 0.65 to 0.91 at 300 nm, 0.71 to 0.96 at 305.5 nm, 0.73 to 0.97 at 311.4 nm, 0.74 to 0.91 at 317.6 nm, 0.76 to 0.96 at 325.4 nm, 0.77 to 0.97 at 332.4 nm, and 0.80 to 0.99 at 368 nm. These results are displayed in Table 1. The error in each day's single scattering albedo is dependent on the aerosol optical depth of that particular day, and is presented in section 3.2.

[14] As can be seen in Table 1, there is no evidence of correlation between air mass type and ω at these wavelengths. This can be attributed to the widely varying aerosol content of air masses coming from each sector. This is especially true for air masses originating in the polluted sector, where efficient scatterers (sulfate) and absorbers (soot) abound. An extensive knowledge of the source inventories within the three sectors is required if a range of expected ω values is to be known.

3.2. Sensitivity/Error Analysis

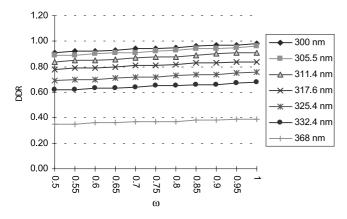
[15] It is necessary to perform sensitivity studies so that error in this ω retrieval technique can be determined. Tests

were conducted at a solar zenith angle of 45° to see which input played the greatest role in determining DDR. One parameter was varied holding the others constant. The other stock inputs, when not being tested, were g = 0.70, ga = 0.04, ω = 0.85 and TOC = 300 DU. Two AOD scenarios at AOD = 0.05 (relatively clean) and 1.0 (relatively turbid) were run to test the sensitivity of DDR to the other input parameters at variable aerosol optical depth. An example of the results of these tests can be seen in Figure 3, which shows the sensitivity of DDR to ω for the two aerosol optical depths. From these tests the order of importance for parameters for determining DDR, based on the rate of increase of DDR with respect to the variable in question, is: AOD, ω , g, ga, and TOC. It was found that DDR has little dependence on realistic values of TOC. In clear-sky periods, variations of tropospheric ozone do little to influence UV radiation, including its scattering, when compared to stratospheric ozone [Barnard et al., 2003]. Figure 3 shows that the sensitivity of DDR to ω increased from AOD = 0.05 to AOD = 1.00, and this was also found to occur for g and ga. A further test was conducted to see how much a change in DDR of 0.02 would affect the retrieval of ω as AOD is varied. Figure 4 shows these results. It can be seen that, as AOD decreases, the change in ω increases. Hence, error analysis in ω retrieval was conducted at several aerosol optical depths, from 0.05 to 1 in 0.05 increment to see how AOD affects the error.

[16] At each AOD, the uncertainty in DDR values due to uncertainty in g, ga, and AOD was determined. This was done by summing together the greatest possible error in DDR due to these three parameters. This error in DDR was combined with the uncertainty in the instrument's DDR measurements through the root mean square error (RMSE) formula. After the total error in DDR was found, how much this error in TUV4.1 output of DDR affected the ω values retrieved could be determined (P. Bloomfield, personal communication, 2001). The value of ω was allowed to vary from a value of 0.86, and the solar zenith angle was 45°. Table 2 displays the assumed values of the model parameters and the uncertainty assigned to these parameters and the instrument measurements.

[17] Uncertainty in AOD was taken from Wenny et al. [2001]. Fractional uncertainties were determined for the V_o values retrieved at each wavelength at Black Mountain.

^bThis air mass was conventionally from the marine sector, but originated in western North Carolina and northern Georgia, and was therefore classified as a continental air mass.



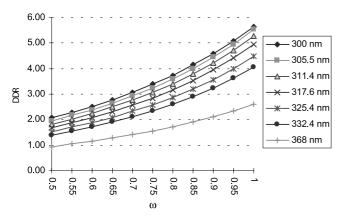


Figure 3. Sensitivity of diffuse-to-direct ratio (DDR) to single scattering albedo (ω) in TUV4.1 (top) at AOD = 0.05 and (bottom) at AOD = 1.00.

These uncertainties translate to uncertainties in AOD at each wavelength, displayed in Table 2. Another source of error for AOD retrieval in the UV wavelengths is that AOD is not the dominating attenuator in the UV wavelengths, and is subject to changes in the other attenuators. Ozone absorp-

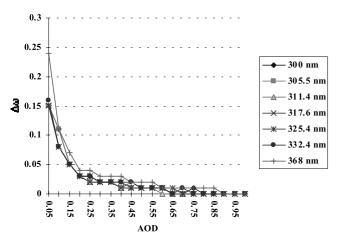


Figure 4. Change in single scattering albedo value $(\Delta\omega)$ with an increase of 0.02 in diffuse-to-direct ratio (DDR) at various aerosol optical depths (AOD).

Table 2. Assumed Values and Related Assumed Uncertainties of Model Parameters for Use in Error Analysis

	Assumed Value	Error (±)
Uncertainty in aerosol		
optical depth due to V _o		
at 300.0 nm		0.103
at 305.5 nm		0.078
at 311.4 nm		0.071
at 317.6 nm		0.076
at 325.4 nm		0.042
at 332.4 nm		0.038
at 368.0 nm		0.037
Asymmetry parameter	0.7	0.05
Ground albedo	0.04	0.02
Diffuse-to-direct ratio		0.02
Single scattering albedo	0.86	

tion is the most important attenuator in the two shortest wavelengths of the UVMFR-SR, and Rayleigh scattering is the most important attenuator in the five longest wavelengths. It should be noted, however, that the uncertainty in ozone optical depth at 311, 317, and 325 nm is larger than the uncertainty in Rayleigh optical depth at these wavelengths. AOD accounts for only 7% of the total optical depth at 300 nm, increasing to 34% at 368 nm [Wenny et al., 2001], and is therefore subject to the uncertainties in the ozone and Rayleigh factors. The error related to this phenomenon has not yet been quantified, but is expected to give greater uncertainty to AOD measurements as wavelength decreases.

[18] The uncertainty assigned to asymmetry parameter, ± 0.05 , is one standard deviation of the values of g from Wenny et al. [1998] at 312 nm. The uncertainty for ground albedo, ± 0.02 , was chosen so as to encompass most of the values found in the literature for open grassland in the UV spectrum. Uncertainty in DDR was estimated to be ± 0.02 . This helps to accounts for blockage of diffuse light by the shadowband of the UVMFR-SR, as well as possible intensification of diffuse irradiance by broken clouds far from the solar disk.

[19] Figure 5 shows the results of the error analysis. Displayed is the estimated uncertainty in ω as it varies with AOD and wavelength. For the analysis conducted here, uncertainty generally decreases with increasing wavelength, due to larger fractional uncertainty values for AOD retrieval in the shorter wavelengths. It does not always do so, likely due to truncations throughout the error analysis. Additionally, the unquantified increasing uncertainty in AOD retrieval with decreasing wavelength in the UV spectrum is expected to also increase uncertainty in ω retrieval as wavelength decreases. Uncertainty in this technique seems to decay exponentially with increasing AOD, from an average error of ± 0.63 for AOD = 0.05 to an average error of ± 0.04 for AOD = 1.0. Error in ω retrieval does not always decrease with increasing AOD, again likely due to truncations throughout the error analysis. For AOD < 0.3, the estimated average error is ± 0.12 . This was deemed to be the threshold for a reasonable ω retrieval, so results from days with AOD < 0.3 were excluded.

3.3. Comparison With Previous Work

[20] Wenny et al. [1998] determined ω for 9 days in 1995 at 312 nm at the same site as the current study for the lowest

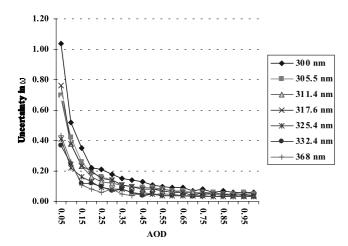


Figure 5. Estimated uncertainty in single scattering albedo (ω) as functions of wavelength and aerosol optical depth (AOD).

kilometer of atmosphere. They used an iterative modeling procedure, involving a UV-B Radiative Transfer Model, very similar to the one used in this study, along with a Mie code. Single scatter albedo, asymmetry parameter, and the refractive index were the result of this procedure. Values of ω ranged from 0.75–0.93, with no discernible dependence on air mass type. This range of values is slightly more limited than the values presented in this study (0.73–0.97 at 311.4 nm). They found a dependence on the relative humidity, as days with relative humidities above 80% yield the higher single scattering albedo values.

[21] Kylling et al. [1998] matched spectral UV irradiance from two spectroradiometers with a radiative transfer model that uses a discrete ordinate algorithm (similar to TUV4.1). This was conducted on two different Greek islands for data from June 1996. To get the best correlation between model and measurement, an ω value for a specific day was taken to be the value that gave the best agreement between the two at noon. The value of ω was assumed to be independent of wavelength. Both instruments measure spectral irradiance at wavelengths covering the wavelengths measured by the UVMFR-SR (Bentham DM 150, 290–500 nm; Brewer MK III, 287.5–366 nm). The value of ω ranged from 0.83 to 0.99 in this study, and falls within the range of the values retrieved in the current study.

[22] One possible cause of the wider range of values of single scattering albedo in this study, as compared to previous work, is the high temporal and spatial variability of tropospheric aerosols. The vast differences in sources, transformation and removal processes, and lifetimes all contribute to this variability [Kiehl and Rodhe, 1995]. Both the quantified and unquantified error in ω retrieval due to error in retrieved aerosol optical depth also contribute to the wide variation in single scattering albedo values. The lower values of ω could indicate that, over the site, preferential absorption of UV radiation by black carbon aerosols could be occurring. Concurrent measurements of black carbon concentration would aid in observation of this phenomenon.

[23] It should be noted that TOMS absorbing aerosol retrievals do not corroborate the present finding of days with dominant UV aerosol absorbing properties at the

research site. None of the days of the present study with successful ω retrievals exhibited a positive Aerosol Index, which indicates the existence of absorbing aerosols [Hsu et al., 1996]. It is possible that the absorbing aerosols lie only in the boundary layer. Yu et al. [2000] found that at Black Mountain, 70% of the total aerosol content lies in the lowest 1 km of the atmosphere. Boundary layer aerosols are not readily seen by TOMS [Herman et al., 1997], and this might explain the disagreement. Nevertheless, due to the lack of corroborating TOMS retrievals and that fact that the database is small (11 retrievals), the values of ω should not be said to be fully representative of the values for the region. A larger database of UVMFR-SR surface UV measurements at the site would aid greatly in the representativeness of these values.

4. Conclusions

[24] A procedure was devised to retrieve single scattering albedo from UVMFR-SR measurements coupled with the radiative transfer model TUV4.1. Values of ω were determined for the seven wavelengths of the UVMFR-SR for 11 days in 1999. The values retrieved vary from 0.65 at 300.0 nm to 0.99 at 368.0 nm. There was no discernible dependence of ω on the air mass classifications used. The range of values is larger than previous studies have found, and this can be attributed to wide variability in aerosol size, composition, and distribution in the air masses influencing the site, as well as error in both the instrument and model. Uncertainties in estimations of asymmetry parameter and ground albedo also contribute, but errors in AOD inputs are a main contributor to error. Some of the errors in AOD measurements have been quantified, but uncertainty in AOD is expected to be higher than what was used here, due to uncertainty in the more dominant ozone and Rayleigh optical depths. Quantification of this error is needed. Error and sensitivity analysis was conducted to determine the uncertainty in the retrieval of single scattering albedo. Error in this procedure was found to decrease with increasing aerosol optical depth, suggesting that this process works best for more turbid atmospheres (AOD > 0.3). The error was determined to be ± 0.63 for AOD = 0.05 and ± 0.04 for AOD = 1.0 averaged over the seven wavelengths of the UVMFR-SR. Long-term UVMFR-SR measurements would allow for observation of trends in UV aerosol radiative properties. The values of ω found here can be used for better estimation of the parameter in these wavelengths for clear-sky summer days in the southeastern United States. This will lead to further development of UV radiative transfer models and lessen error in estimation of surface UV irradiances for the region.

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violet and Visible) Radiation Model information is available from UCAR Atmospheric Chemistry Division at http://www.acd.ucar.edu/TUV/.

References

- Anderson, T. L., D. S. Covert, J. D. Wheeler, J. M. Harris, K. D. Perry, B. E. Trost, D. J. Jaffe, and J. A. Ogren, Aerosol backscatter fraction and single scattering albedo: Measured values and uncertainties at a coastal station in the Pacific Northwest, *J. Geophys. Res.*, 104, 26,793–26,807, 1999.
- Barnard, W. F., V. K. Saxena, B. N. Wenny, and J. J. DeLuisi, Daily surface UV exposure and its relationship to surface pollutant measurements, *J. Air Waste Manage. Assoc.*, 237–245, 2003.
- Bigelow, D. S., J. R. Slusser, A. F. Beaubien, and J. H. Gibson, The USDA Ultraviolet Radiation Monitoring Program, *Bull. Am. Meteorol. Soc.*, 79, 601–615, 1998.
- Blumthaler, M., and W. Ambach, Solar UVB-albedo of various surfaces, *Photochem. Photobiol.*, 48, 85–88, 1988.
- Deininger, C. K., and V. K. Saxena, A validation of back trajectories of air masses by principal component analysis of ion concentrations in cloud water, Atmos. Environ., 23, 295–300, 1997.
- Devaux, C., A. Vermeulen, J. L. Deuzé, P. Dubuisson, M. Herman, R. Santer, and M. Verbrugghe, Retrieval of aerosol single-scattering albedo from ground-based measurements: Application to observational data, J. Geophys. Res., 103, 8753–8761, 1998.
- Diffey, B. L., A. T. Green, M. J. Loftus, G. J. Johnson, and P. S. Lee, A portable instrument for measuring ground reflectance in the ultraviolet, *Photochem. Photobiol.*, 61, 68–70, 1995.
- Draxler, R. R., and G. D. Hess, An overview of the Hysplit_4 modeling system for trajectories, dispersion, and deposition, *Aust. Met. Mag.*, 47, 295–308, 1998.
- Dubovik, O., B. N. Holben, Y. J. Kaufman, M. Yamasoe, A. Smirnov, D. Tanré, and I. Slutsker, Single-scattering albedo of smoke retrieved from the sky radiance and solar transmittance measured from ground, *J. Geophys. Res.*, 103, 31,901–31,923, 1998.
- Dubovik, O., B. Holben, T. F. Eck, A. Smirnov, Y. J. Kaufman, M. D. King, D. Tanré, and I. Slutsker, Variability of absorption and optical properties of key aerosol types observed in worldwide locations, *J. Atmos. Sci.*, 59, 590–608, 2002.
- Eck, T. F., B. N. Holben, I. Slutsker, and A. Setzer, Measurements of irradiance attenuation and estimation of aerosol single scattering albedo for biomass burning aerosols in Amazonia, *J. Geophys. Res.*, 103, 31.865–31.878, 1998.
- Elterman, L., UV, visible, and IR attenuation for altitudes to 50 km, *AFCRL Rep. AFCRL-68-0153*, 49 pp., Air Force Cambridge Res. Lab., Hanscom Air Force Base, Mass., 1968.
- Environmental Protection Agency, *Regional Emissions Inventories* (1987–1991), vol. 2, *Emission Summaries*, Off. of Air Qual. Plann. and Stand., Research Triangle Park, N. C., 1993.
- Feister, U., and R. Grewe, Spectral albedo measurements in the UV and visible region over different types of surfaces, *Photochem. Photobiol.*, 62, 736–744, 1995.
- Frederick, J. E., E. K. Koob, A. D. Alberts, and E. C. Weatherhead, Empirical studies of tropospheric transmission in the ultraviolet: Broadband measurements, *J. Appl. Meteorol.*, 32, 1883–1892, 1993.
- Harrison, L., and J. Michalsky, Objective algorithms for the retrieval of optical depths from ground-based measurements, Appl. Opt., 33, 5126– 5132, 1994.
- Harrison, L., J. Michalsky, and J. Berndt, Automated multifilter rotating shadow-band radiometer: An instrument for optical depth and radiation measurements, Appl. Opt., 33, 5118-5125, 1994.
- Herman, J. R., P. K. Bhartia, O. Torres, C. Hsu, C. Seftor, and E. Celarier, Global distribution of UV-absorbing aerosols from Nimbus 7/TOMS data, *J. Geophys. Res.*, 102, 16,911–16,922, 1997.
- Hsu, N. C., J. R. Herman, P. K. Bhartia, C. J. Seftor, O. Torres, A. M. Thompson, J. F. Gleason, T. F. Eck, and B. N. Holben, Detection of biomass burning smoke from TOMS measurements, *Geophys. Res. Lett.*, 23, 745–748, 1996.
- Kiehl, J. T., and H. Rodhe, Modeling geographical and seasonal forcing due to aerosols, in *Aerosol Forcing of Climate*, edited by R. J. Charlson and J. Heintzenberg, pp. 281–295, John Wiley, New York, 1995.
- Krotkov, N. A., P. K. Bhartia, J. R. Herman, V. Fioletov, and J. Kerr, Satellite estimation of spectral surface UV irradiance in the presence of tropospheric aerosols: 1. Cloud-free case, *J. Geophys. Res.*, 103, 8779– 8793, 1998.
- Kylling, A., A. F. Bais, M. Blumthaler, J. Schreder, C. S. Zerefos, and E. Kosmidis, Effect of aerosols on solar UV irradiances during the Photochemical Activity and Solar Ultraviolet Radiation campaign, *J. Geophys.* Res., 103, 26,051–26,060, 1998.

- Kylling, A., T. Persen, B. Mayer, and T. Svenøe, Determination of an effective spectral surface albedo from ground-based global and direct UV irradiance measurements, *J. Geophys. Res.*, 105, 4949–4959, 2000.
- Lacis, A. A., and M. I. Mishchenko, Climate forcing, cloud sensitivity, and climate response: A radiative modeling perspective on atmospheric aerosols, in *Aerosol Forcing of Climate*, edited by R. J. Charlson and J. Heintzenberg, pp. 11–40, John Wiley, New York, 1995.
- Madronich, S., The atmosphere and UV-B radiation at ground level, in *Environmental UV Photobiology*, edited by A. R. Young, pp. 1–39, Plenum, New York, 1993a.
- Madronich, S., UV radiation in the natural and perturbed atmosphere, in *Environmental Effects of Ultraviolet (UV) Radiation*, pp. 17–69, Lewis, Boca Raton, Fla., 1993b.
- Madronich, S., R. L. McKenzie, L. O. Björn, and M. M. Caldwell, Changes in biologically active ultraviolet radiation reaching the Earth's surface, J. Photochem. Photobiol., 46, 5–19, 1998.
- McKenzie, R. L., M. Kotkamp, and W. Ireland, Upwelling UV spectral irradiances and surface albedo measurements at Lauder, New Zealand, *Geophys. Res. Lett.*, 23, 1757–1760, 1996.
- Meleti, C., and F. Cappellani, Measurements of aerosol optical depth at Ispra: Analysis of the correlation with UV-B, UV-A, and total solar irradiance, J. Geophys. Res., 105, 4971–4978, 2000.
- Ogren, J. A., and P. J. Sheridan, Vertical and horizontal variability of aerosol single scattering albedo and hemispheric backscatter fraction over the United States, paper presented at 14th Conference on Nucleation and Atmospheric Aerosols, Int. Assoc. of Meteorol. and Atmos. Sci., Helsinki, 1996.
- Reuder, J., and H. Schwander, Aerosol effects on UV radiation in nonurban regions, *J. Geophys. Res.*, 104, 4065–4077, 1999.
- Saxena, V. K., and J. R. Yeh, Temporal variability in cloud water acidity: Physico-chemical characteristics of atmospheric aerosols and wind field, *J. Aerosol Sci.*, 19, 1207–1210, 1989.
- Schafer, J. S., V. K. Saxena, B. N. Wenny, W. Barnard, and J. J. DeLuisi, Observed influence of clouds on ultraviolet-B radiation, *Geophys. Res. Lett.*, 23, 2625–2628, 1996.
- Schwander, H. P. Koepke, and A. Ruggaber, Uncertainties in modeled UV irradiances due to limited accuracy and availability of input data, J. Geophys. Res., 102, 9419–9429, 1997.
- Schwart², S. E., et al., Group report: Connections between aerosol properties and forcing and climate, in *Aerosol Forcing of Climate*, edited by R. J. Charlson and J. Heintzenberg, pp. 251–277, John Wiley, New York, 1995
- Stamnes, K., S. C. Tray, W. Wiscombe, and K. Jayaweera, Numerically stable algorithm for discrete-ordinate method radiative transfer in multiple scattering and emitting layered media, *Appl. Opt.*, 27, 2502–2509, 1988
- Ulman, J. C., and V. K. Saxena, Impact of air mass histories on the chemical climate of Mount Mitchell, North Carolina, J. Geophys. Res., 102, 25,451–25,465, 1997.
- Waggoner, A. P., R. E. Weiss, N. C. Ahlquist, D. S. Covert, S. Will, and R. J. Charlson, Optical characteristics of atmospheric aerosols, *Atmos. Environ.*, 15, 1891–1909, 1981.
- Weihs, P., A. R. Webb, S. J. Hutchinson, and G. W. Middleton, Measurements of the diffuse UV sky radiance during broken cloud conditions, J. Geophys. Res., 105, 4937–4944, 2000.
- Wenny, B. N., J. S. Schafer, J. J. DeLuisi, V. K. Saxena, W. F. Barnard, I. V. Petropavlovskikh, and A. J. Vergamini, A study of regional aerosol radiative properties and effects on ultraviolet-B radiation, *J. Geophys. Res.*, 103, 17,083–17,097, 1998.
- Wenny, B. N., V. K. Saxena, and J. E. Frederick, Aerosol optical depth measurements and their impact on surface levels of ultraviolet-B radiation, *J. Geophys. Res.*, 106, 17,311–17,319, 2001.
- Yu, S., V. K. Saxena, B. N. Wenny, J. J. DeLuisi, G. K. Yue, and I. V. Petropavlovskikh, A study or the aerosol radiative properties needed to compute direct aerosol forcing in the southeastern United States, *J. Geophys. Res.*, 105, 24,739–24,749, 2000.
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