Derivation of Aerosol Properties From Satellite Measurements of Backscattered Ultraviolet Radiation. Theoretical Basis

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Abstract

We discuss the theoretical basis of a recently described method of detecting UV-absorbing aerosols using satellite-measured radiation in near-ultraviolet (330-400 nm). We show that the interaction between these aerosols with the strong molecular scattering in ultraviolet produces wavelength-dependent variations that can be used to separate aerosol absorption effects from those due to aerosol scattering. This allows positive identification of several different types of aerosols found in the Earth's atmosphere, ranging from non-absorbing sulfates to highly UV-absorbing mineral dust. It is shown that in the near-ultraviolet aerosols of all types can be reliably detected over both land and water, and that absorbing aerosols can be detected over snow/ice-covered surfaces and clouds, and even in a mixed scene containing sub-pixel clouds. However, in order to reliably estimate aerosol column optical depths, the scenes must be cloud-free, and for the absorbing aerosols, it is also necessary to know their altitude. We present a sensitivity analysis of various error sources contributing to the estimation of optical depth, and demonstrate the technique by applying it to the data taken by NASA's Total Ozone Mapping Spectrometer (TOMS) instrument.

1. Introduction

Aerosols affect the radiation budget of the earth-atmosphere system directly by the scattering and absorption of solar and thermal radiation and, as cloud condensation nuclei, they also have an indirect effect by modifying the optical properties and lifetimes of clouds. The net effect on the radiative budget depends upon aerosol composition and physical properties, reflectivity of the underlying surface and altitude of the aerosol layers. Aerosol effects on climate have been the focus of scientific interest for decades. The role of stratospheric aerosol of volcanic origin has traditionally received most of the attention (e.g., El Chichon and Mt. Pinatubo eruptions), because of the large climate modification potential associated with the long lifetime of injected sulfate aerosols. Tropospheric aerosols, on the other hand, have only intermitently been the subject of research in the context of its potential climatic effects. In the last few years there has been a great deal of interest about the importance of tropospheric aerosols of anthropogenic origin from industrial pollution and biomass burning.

The consequent aerosol effect on climate is usually quantified in terms of radiative forcing, i.e., the net flux change at the top of the atmosphere due solely to the direct aerosol radiative effects. Although there are uncertainties in the estimates of aerosol radiative forcing, it is generally agreed that the averaged global direct effects of anthropogenic sulfate aerosols are not negligible and are

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probably similar in magnitude but in opposite direction to anthropogenic greenhouse gas forcing [*Penner et al.*, 1992; *Charlson et al.*, 1991]. Unlike the greenhouse gases, aerosol physical-chemical properties and abundances exhibit large variability in both time and space that result in large uncertainties on the role of aerosols on climate. Because of this, cooling effects from aerosols do not necessarily result in the cancellation of the greenhouse gases' warming effect.

In addition to the well documented cooling effect of aerosols particles by backscattering of solar radiation, some aerosol types also absorb solar radiation reducing their cooling effect. For instance, absorption by both elemental and organic carbon aerosols from biomass burning and industrial pollution may affect the thermodynamics of the atmosphere by changing its thermal structure and perturbing convection and mixing processes in the planetary boundary layer. Silicaceous aerosols, such as desert dust, absorb solar and infrared radiation so that their direct effect on the climate may be significant. The role of anthropogenic sources of mineral aerosols as a climate forcing factor has been examined [*Tegen et. al.*, 1996] and found to be of importance when included in climate change studies. Because of the lack of data on absorbing aerosol physical properties and their spatial and temporal distributions, the heating effects of aerosols particles may not have been adequately included in model calculations. In an effort to improve on the current understanding of the role of aerosols on climate, it has been suggested [*National Research Council, 1993*] that a combination of activities including satellite measurements be undertaken.

The effect of aerosols in the ultraviolet radiation field has traditionally been studied in the context of ozone retrieval from space based measurements of backscattered ultraviolet radiation (buv). The wavelength dependance caused by absorbing and non-absorbing aerosols on the buv radiance was first documented by Dave [1978] in his analysis of aerosol related errors in the ozone retrieval from buy measurements. Additional wavelength and angular dependencies have been discussed that arise from the presence of polar stratospheric clouds [Torres et al 1992] and stratospheric sulfate aerosols [Bhartia et. al., 1993, Torres et. al., 1995] on TOMS (Total Ozone Mapping Spectrometer) ozone measurements. Torres and Bhartia [1995] considered the artifacts introduced by stratospheric aerosols on ozone profile retrieval by the SBUV family of instruments. Torres et al. [1995] used the strong angular dependance of the aerosol effect on the TOMS buy measurements to infer optical depth and particle size of the Mt Pinatubo aerosol layer in the tropics. Several works have been recently published on the detectability by the TOMS instrument of tropospheric absorbing particulate matter such as carbonaceous aerosols [Hsu et al, 1996, Herman et al, 1997] and ash aerosols from volcanic eruptions [Seftor et al, 1997, Krotkov et al, 1997]. These papers discuss the sources and geographic distribution of absorbing aerosols based on an aerosol index from which optical depth can be derived.

In this paper we present the theoretical basis of aerosol detection using satellite measurements of backscattered radiation and the method used to obtain optical depth. First, we describe how the aerosol models that we have used in this study were selected. This is followed by a detailed discussion of the effects of these aerosols on the backscattered radiation in the near-UV (320-400 nm), where the ozone absorption is weak and does not affect the interaction between aerosols and the molecular atmosphere. Next, we discuss alternative methods of detecting these aerosols from the measurements at two UV wavelengths (340 and 380 nm) and present the sensitivity of these methods to various error sources in estimating the aerosol optical depth. Finally, we demonstrate the application of our proposed techniques by applying it to the data taken by the Total Ozone Mapping Spectrometer (TOMS) instrument

2. Aerosol Models

The atmospheric aerosol distribution exhibits large spatial and temporal variability in composition, size and concentration, related to the geographic locations of emission sources, type of emission, diffusive transfer and removal processes. The fluctuations of meteorological parameters (humidity, temperature, wind speed and direction) have a direct effect on the aerosol vertical and horizontal distributions as well as on the processes of diffusion, sedimentation and coagulation of aerosols. A comprehensive discussion of the variability of aerosols characteristics is presented by *d'Almeida et al [1991]*. The optical and microphysical properties of the different types of atmospheric aerosols span a wide range of values. A theoretical analysis of all possible aerosol types is very complicated and impractical. Thus, to carry out a sensitivity analysis it is necessary to simplify the problem by selecting a set microphysical models representative of the most commonly observed aerosol types. A brief discussion on the rationale used in the selection of the aerosol models is presented next.

Non- absorbing aerosols

Suspended sea-salt particles and sulfate aerosols are the most common type of non-absorbing aerosols. Sea salt aerosols are entirely of natural origin whereas the sources of sulfate aerosol include both natural and anthropogenic mechanisms. Two non-absorbing aerosol models covering a wide particle size range are used in this study. A small particle size model, representative of troposheric sulfate aerosols of anthropogenic origin, S1, and a large particle model representative of other non-absorbing aerosol types such as sea-salt aerosols and stratospheric sulfuric acid aerosols, S2.

Carbonaceous Aerosols

Carbonaceous aerosols generated by biomass combustion consist of a mixture of material with varying radiative properties, both absorbing and nonabsorbing. Among these, the group of volatile organic or inorganic components is relatively non-absorbing. Much of the absorption of carbonaceous particles is due to the fraction of elemental or graphitic carbon [*Rosen et. al, 1978*]. Thus, the absorption of smoke aerosol particles depends on the relative amounts of each aerosol constituent. The single microphysical property directly related to the absorptivity of a given material is the imaginary component of its refractive index. The single scattering albedo, a macrophysical measurement of aerosol absorption, is defined as the ratio of the coefficients for scattering and extinction (i.e., the summation of scattering and absorption coefficients). The relation between imaginary refractive index and single scattering albedo is not direct since ω_0 also depends on the absorption cross-section of the particles.

Smoke components are produced from four fire stages : pre-ignition, flaming, smoldering and glowing [*McMahon*, 1983]. The relative amount of elemental carbon and consequently the absorption properties of the resulting smoke vary through the combustion process. Analysis of laboratory fires [*Patterson and McMahon*, 1984] showed significantly larger absorption during flaming combustion than during the smouldering phase of the fire. *Radke et al.* [1991] measured the single scattering albedo of prescribed fires as a function of smoke age at altitudes between 2 to 2.5 km in North America in September and October 1987. The observed single scattering albedo rose from an initial value of 0.7 and stabilized at about 0.9 over an hour after the onset of the fire. *Westphal and Toon* [1991] discussed the dependance of smoke optical properties on the particle size distribution

parameters and examined the time dependance of smoke particle size. Their detailed analysis of direct measurements of size distribution and optical depth in several events of aged forest fire smoke indicates that the size of smoke aerosol particles increases systematically with time reaching geometric mean radii between about 0.10 to 0.14μ in 48 to 90 hours. This range of smoke particle size is consistent with airborne volume density size distributions measurements over Brazilian and African smoke source regions during the TRACE A experiment in September-October 1992 [*Anderson, et. al., 1996*]. The observed size distributions over both areas show a well defined mode diameter at about 0.25 μ characteristic of the accumulation mode aerosols which are a primary combustion product [*Radke et al.,* 1991]. As reported by *Anderson et al.* [1991], the observed aerosol size showed only a slight tendency to increase with smoke age, and no appreciable differences in size distribution were observed between the source areas and the outflow regions. Particle radii derived from sky spectral radiances measured by sun/sky radiometers in Brazil [*Holben et al.,* 1996], also clearly shows the accumulation mode with a well defined maximum volume distribution for radii in the vicinity of 0.1 μ .

It thus appears, that the variability of biomass burning smoke single-scattering albedo over a given area is more likely to be due to the variability of the fraction of elemental carbon in the smoke than to changes in particle size distribution. Based on the documented physical nature of the evolution of biomass burning smoke optical properties, we have selected two carbonaceous aerosol models (C1 and C2). Both models have the same particle size distribution, but differ in the magnitude of the imaginary component of the refractive index. The form of the assumed size distribution is a lognormal function with mode radius 0.12μ and width 1.5. These parameters were found to adequately represent the nucleation mode of the observed size distribution over Brazilian and African sources as depicted in Figures 4, 8 and 9 of *Anderson et al.* [1996]. Model C1, representative of weakly absorbing smoke aerosols away from the source area, has an imaginary refractive index of 0.02. Model C2, representative of fresh, more absorbing, smoke in the vicinity of the source area has an imaginary refractive index of 0.04. These are typical values within the range of recommended values based on experimental work [*Patterson and McMahon, 1984*].

Mineral Aerosols

Mineral or dust aerosol is a common type of atmospheric suspended matter characteristic of the earth's arid and semi-arid regions accounting for about one-third of the land-surface area. Because of the high insolation levels and strong convective processes characteristic of arid regions, fine dust particles are easily lifted to high altitudes and horizontally transported by synoptic-scale atmospheric disturbances to areas thousands of kilometers away from their source regions [*Carlson and Prospero, 1972*]. Measurements of desert dust aerosol composition indicate a mixture of different kinds of materials with calcium, silicon (in the form of quartz), and iron (in the form of hematite), identified as the main elemental components [*Levin and Lindberg, 1979*]. Although desert dust from the same source region exhibits relatively uniform physical and chemical properties over large areas, there is considerable variability in aerosol properties associated with the nature of the crustal material from different source regions [*Gomes and Gillette, 1993*].

The complex refractive index of desert dust is largely determined by the volume fraction of hematite present in the mixture. Empirically-computed aerosol dust refractive index values [*Longtin et al*, 1988] at 400 nm based on a weighted mixing of measured refractive indices of quartz and hematite vary between 1.56 - 0i (for 0% hematite concentration) and 1.66 - .0335i (for 10% hematite

concentration). Real refractive index values of about 1.5 are generally used in the visible based on the measurements of Saharan aerosols by *Patterson et al*, [1977] at 550 and 633 nm. No direct measurements in the near ultraviolet are available. Measured values of the imaginary refractive index at 400 nm at a variety of locations [*Deluisi et. al*, 1976; *Levin et al*, 1980; *Lindberg et al*, 1976; *Lindberg and Laude*, 1974; *Patterson et al*, 1977; *Andronova et. al*, 1993, *Sokolik et al.*, 1993] vary from about 0.005 to about 0.02. While part of the observed variability may result from the use of different measuring techniques [*Sokolik et al.*, 1993], these measurements also reflect the differences in composition of desert aerosols from the different regions of the world where the sampling was made.

The size of desert dust aerosol particles shows significant variability over several orders of magnitude, from Aitken nuclei (aerosol radii less than 0.1μ) to ultra-giant particles with radii larger than 30μ . Since the atmospheric aerosol load over deserts is the direct result of the wind's lifting ability, the airborne particle size distribution is closely related to wind speed. Commonly used dust particle size distributions include the three-model representation for background, wind-carried dust and sandstorm conditions developed by *d'Almeida* (1987), and the wind-speed dependent model of *Longtin et al.* (1988). Model representations of long-range transport mineral aerosols have been developed by *Schütz* [1980] to represent aerosol dust mobilized to maritime environments and by *Shaw* [1979] and *Parungo et al.* [1981] for poleward transport.

We have selected three aerosol lognormal particle size distributions models representative of desert dust to account for the large variability in particle size. A small particle model (D1) representative of the nucleation mode [*d'Almeida*, 1987], and two long range transport models (D2 and D3) representative of larger particles [*Schütz*, 1980]. The complex refractive index for the three models is set to be 1.58 -.02i, considered to be representative of the composition of Saharan dust *Patterson et al*, [1977].

In order to calculate the aerosol scattering phase function making use of Mie theory, it has been assumed that the particles are spherical. The problem of accurately accounting for particle shape in remote sensing is twofold. First, observations are needed to determine the actual shape of the particles. Once, actual shapes are known, the analytical tools to represent distributions of irregularly shaped particles are not always available. For the sulfate aerosol models, the spherical assumption is easily justified since surface tension forces for liquid sulfuric acid particles resulting from the gas-to-particle transformation process tend to produce spherical shapes.

Carbonaceous aerosols, on the other hand, are generally a mixture of spherical, non-spherical and chain aggregates depending on the type of burning matter, combustion phase and the age of the smoke. *Hallett et. al.* [1988], used electron microscopy to determine the shape of forest fire smoke aerosols and found the particles to be mostly spherical. The actual shapes and sizes of combustion aerosols were measured by means of scanning electron microscopy (SEM) during the SCAR-B (Smoke, Clouds and Radiation - Brazil) experiment. As reported by *Martins et. al.* [1996], the SEM analysis showed high variability in particle shape during the flaming phase of the fire and nearly spherical particles in the smoldering phase. They also determined the asymmetry of the particles by measuring the percent change in the integrated light scattering as the particles orient themselves along an applied electric field [*Weiss et al.*, 1992]. The measured asymmetry factor during SCAR-B was always less than 11% [*Martins et. al.*, 1996]. Thus, based on observational evidence, the representation of carbonaceous aerosols as spherical particles seems to be a reasonably good approximation.

For desert dust aerosols, the sphericity assumption is difficult to justify since dry aerosol particles are seldom spheres. Model calculations [*Wiscombe and Mugnai*, 1988; *Koepke and Hess*, 1988; *Mishchenko et. al.*, 1995] show that the phase function of non-spherical particles significantly deviates from that of spherical aerosols. Modeling results of the effects of non-spherical particles on phase function, single scattering albedo and asymmetry parameter are available in the literature [*Mishchenko et. al.*, 1995]. The phase function departure from the spherical model is particle size and scattering angle dependent with the biggest effect at scattering angles between 90° and 140° for particles of effective radii larger than about 0.5μ . The non-sphericity of the particles produces only a small effect in the single scattering albedo and asymmetry parameter. Although the particle shape is important in most of the range of scattering angles relevant to satellite remote sensing, the phase function deviation from sphericity is minimum in the range $150^{\circ}-160^{\circ}$. Depending on the actual particle shape and viewing geometry, space retrievals of aerosol optical depth at visible wavelengths may be underestimated at backscattering geometries and overestimated at side-scattering angular configurations [*Mishchenko et al.*,1995].

Modeling the effect of non-sphericity of aerosol particles in the near uv is required to fully assess the limitations of the spherical approximation of mineral aerosols used in this work. Because of the large multiple scattering contribution to the total backscattered intensity in the near UV the use of the spherical particle approximation to retrieve optical depth from measurements in the 320-400 nm range may produce smaller errors than a similar estimate in the visible region. For near UVmeasurements of absorbing aerosols, the non-spherical particle phase function effect decreases with increasing absorption. Since the desert dust imaginary refractive index in the UV is almost an order of magnitude larger than at 630 nm, the phase function effect and, therefore, the particle shape may also be less important in the UV than in the visible.

These models (S1, S2, C1, C2, D1, D2, and D3) approximate the most common types of aerosols found in the Earth's atmosphere, i.e., non-absorbing sulfate aerosols, organic carbon aerosols and desert dust aerosols. The effect of volcanic ash aerosols is not considered in this analysis since it is only rarely present in the atmosphere. The effects of volcanic ash in the backscattered UV radiation are reported by Krotkov et. al. (1997). The effect of each aerosol model is considered separately, i.e., no aerosol mixtures are considered. The physical properties of the above described aerosol models are summarized in Table 1. In order to have radiatively equivalent models, the optical depth at 380 nm has been set to unity for all six models. The resulting scattering phase functions for all models are depicted in Figure 1 for the range of scattering angles relevant to space measurements. Notice that in spite of the fact that the wavelength dependance of the refractive index has been ignored, the computed optical thickness and single scattering albedo are spectrally dependent due to the incident light-aerosol particle interaction.

3. Effect of aerosols on near ultraviolet radiances

At wavelengths longer than 320 nm, where ozone absorption is weak, the net aerosol effect on the backscattered radiation at the top of the atmosphere is the result of the complex interaction of several competing radiative transfer processes: Rayleigh and Mie multiple scattering, aerosol absorption of both Rayleigh and Mie scattered radiation as well as scattering and absorption of radiation reflected by the underlying surface. Thus, the resulting backscattered radiance may increase or decrease with respect to a purely molecular atmosphere depending on the aerosol physical properties, i.e., complex refractive index, particle shape, particle size distribution and total aerosol optical depth. In addition,

the location in the atmosphere of the aerosol layer and the reflectivity of the underlying surface also affect the resulting impact of aerosols. In the remainder of this section we examine the sensitivity of the bUV (backscattered ultraviolet) radiance at the top of the atmosphere to changes in the physical properties of both absorbing and non-absorbing aerosols.

The sensitivity of the bUV radiance at the top of the atmosphere to changes in aerosol properties, as represented by the assumed array of aerosol models, was examined by means of detailed radiative transfer calculations. The radiative transfer model we have used in this study is a modified version of the Vector Program D (VPD) developed by *Dave* [1972]. It fully accounts for all orders of scattering, including the effects of polarization. Though the effect of the Earth's sphericity is taken into account for the incoming radiation, higher order scattering is assumed to take place in a plane parallel atmosphere. Comparitive analysis of this pseudo-spherical approximation to results using a fully spherical code [*Caudill et. al,* 1997] suggest that errors in the approximation used in the VPD code are small (< 1%) at solar zenith angles less than 80° .

In addition to the aerosol models, input data on surface reflectivity, aerosol profile and viewing geometry are required. Although, the sensitivity analysis to changes in those parameters was examined, a set of baseline values of reflectivity, aerosol altitude and solar zenith angle was selected. The wavelength independent surface reflectivity (5%) is based on the long-term analysis of the 380 nm minimum reflectivity measured by the Nimbus7-TOMS instrument [*Herman and Celarier*, 1997]. The neglect of the spectral dependance agrees with the observational work of *McKenzie and Kotkamp* [1996]. The vertical distribution of mineral and carbonaceous aerosols is represented by a gaussian profile with maximum at 3 km and a width of 1km. This vertical distribution is assumed to be representative of an average profile since lidar measurements of both smoke and dust layers indicate that the aerosol layers typically reside between 2 and 5 km. The baseline values of solar and satellite zenith angles are 40° and 0° respectively, corresponding to a scattering angle of 140° . Calculations at 320, 340, 360, 380 and 400 nm were carried out. A general analysis of the spectral dependance of the aerosol effect is first presented followed by a detailed account of the sensitivity of the 380 nm intensity to changes in aerosol and other related parameters.

The spectral dependance of the aerosol effect, presented as the percent change in backscattered radiance relative to the Rayleigh scattering limit, is shown in Figure 2 for the seven aerosol models. The transition from a radiance increase at all wavelengths for non-absorbing aerosols to a radiance decrease for highly absorbing aerosols is due to the competing effects of aerosol scattering and absorption. Since the aerosols have a strong forward scattering, the non-absorbing aerosols do not significantly alter the background radiation generated by molecular scattering and surface reflection, but add a weakly wavelength-dependent component to the strongly wavelength-dependent molecular scattering. Therefore, for such aerosols (models S1 and S2), the percent change in the radiance from the background is positive and increases with wavelength. By contrast, highly absorbing aerosols (model D3) scatter weakly (particularly at the 140 degree scattering angle assumed for the plot), but they strongly absorb the background radiation. Since this absorption varies slowly with wavelength, there is a net, nearly wavelength-independent, decrease in radiation.

The previous argument also explains the effect of aerosol altitude on the backscattered radiation, as shown in Figure 3. Lack of altitude dependence for models S1 and S2 supports the observation that non-absorbing aerosols interfere little with the background radiation, whereas the increased absorption with height for model D3 reflects the fact that these aerosols strongly absorb the molecular radiation coming from below. That is, the higher the absorbing aerosol altitude the greater is the

fraction of affected molecular radiation. The proportionality of Rayleigh scattering to pressure and the log-linear relationship between atmospheric pressure and altitude also explains why the effect of absorbing aerosols varies non-linearly with height.

Since Figures 2 and 3 illustrate the basis of detecting aerosols from the measurements in near UV, it is worth summarizing their findings. In near-UV, aerosols produce two effects on the backscattered radiation: they produce a wavelength-dependent increase in the background radiation proportional to aerosol scattering, to which is added a wavelength-independent decrease in the radiation proportional to aerosol absorption. The former effect is independent of aerosol height, but the latter is not. It is important to note that the Rayleigh scattering plays an important role in creating the effect; it makes the backscattered radiation highly sensitive to aerosol absorption, and imparts ?color" (i.e. introduces spectral dependence) to the otherwise ?gray" aerosols, thus making them more easily detectable. That is, for gray absorbing aerosols, i.e., aerosols with wavelength invariant optical properties, the wavelength dependence of the backscattered radiance from an atmosphere containing absorbing aerosol particles deviates significantly from that of an aerosol-free atmosphere.

The angular dependance of the aerosol effect is plotted in Figure 4 as a function of both scattering and solar zenith angles. In the single scattering approximation, the percent change in backscattered radiance due to the presence of aerosols would be strictly proportional to the ratio of aerosol- to-Rayleigh scattering phase functions. For small optical depths that are usually associated with scattering angles larger than about 140° (i.e., solar zenith angles smaller than about 40°), the radiance contribution from aerosols is highly correlated with the aerosol scattering phase function. The structure of the aerosol effect in the backscatter direction is related to the glory feature present in the phase function of large particles. At larger solar zenith angles, where the slant optical path through the aerosols approaches unity, the correlation smears out due to the enhancement of the multiple particle scattering.

The effect of the reflectivity of the underlying surface on the bUV radiance when the atmosphere contains aerosols is depicted in Figure 5. As before, the non-absorbing aerosols act to increase the bUV radiance. As the surface reflectivity increases, the Rayleigh backscatter of upward radiance to the surface also increases causing the percentage increase in excess radiance amount over Rayleigh scattering to become smaller. Thus, for extremely high reflectivity (fresh snow), the non-absorbing aerosol effect becomes negligible. The presence of low UV-reflectivity over most land surfaces enables non-absorbing aerosols to be detected in the UV (e.g., from TOMS), but not in the visible wavelengths (e.g., from AVHRR).

The interaction between the reflecting surface and absorbing aerosols is more complex. For low values of surface reflectivity and weak to moderate aerosol absorption, aerosol scattering predominates over absorption so that there is a net increase in the amount of bUV radiance at the top of the atmosphere. At some critical value of the surface reflectivity, there is a near balance between scattering back to the surface and absorption by the aerosol. Such a critical reflectivity value is a function of aerosol single scattering albedo [*Kaufman*, 1987]. For higher values of surface reflectivity, the radiance increase caused by aerosol scattering is overcome by absorption within the aerosol layer and backscattering of the reflected radiance to the surface so that one observes a net reduction of the backscattered radiation. For strongly absorbing aerosols, absorption may dominate over scattering even at low reflectivities.

4. Retrieval of Aerosol Properties from near ultraviolet measurements

Remote-sensing of aerosols, whether from space or the ground, presents formidable challenges. Even for the simpler case of spherical particles, there are five aerosol parameters that affect what one measures from a remote-sensing instrument: Real and imaginary component of the refractive index, size distribution, particle density, and vertical distribution. Additional parameters are needed to characterize non-spherical particles. Measurements made by remote-sensing instruments obtaining 1 or 2 independent pieces of information simultaneously depend on several (5 or more) aerosol parameters. In order to derive the 1 or 2 quantities of interest it is necessary to assume an aerosol model incorporating the other parameters. For space-based remote-sensing there is additional problem of knowing the background reflectivity accurately and of identifying cloud-free scenes. Because of the highly variable background reflectivity for wavelengths longer than 420 nm, measurements in the visible wavelengths have been largely restricted to the dark ocean areas.

As discussed earlier, the near-UV technique can provide at most 2 pieces of information about the aerosols: The spectral dependence of the change in radiances from their background values, that is proportional to aerosol scattering; and the magnitude of this change, that is proportional to aerosol absorption. The strength of the UV technique is that this information can often uniquely identify the type of aerosol present in a scene, thus helping one to choose the correct model for the retrieval of aerosol parameters. We have found two distinctly different ways of using the information contained in the UV radiances. Both have their strengths and weaknesses; the choice of a particular method depends upon the intended application.

3.1 Residue Method

The residue method, more descriptively called the spectral contrast anomaly method, emerged as a by-product of the most recent improvement (Version 7) in the TOMS ozone retrieval algorithm (ref). As originally proposed (ref), the backscattered ultraviolet (bUV) technique uses a pair of wavelengths, one ozone-sensitive the other insensitive, to derive total ozone. To do this succesfully, one is required to separate accurately the spectral contrast (ratio of radiances at the selected wavelengths) caused by ozone absorption from that caused by all other effects, including molecular scattering, surface reflection, aerosols, and clouds. The Lambert-Equivalent Reflectivity (LER) method was proposed by Dave (XXX) to solve this problem. In this method (ref), the spectral variation of bUV radiances is predicted by assuming that the amosphere consists only of molecular scatterers and absorbers (primarily ozone), bounded by a Lambertian surface. With this assumption, one can construct an accurate radiative transfer model (ref) that accounts for polarization, all orders of molecular scattering and Earth's sphericity effects (ref). The reflectivity of the surface is selected to be the value that predicts the measured radiances at one UV wavelength (typically 380 nm).

Although detailed radiative transfer calculations that included the effects of aerosols and cloud scattering (ref), as well as non-lambertian surfaces (ref), had confirmed the theoretical validity of the LER technique for the retrieval of ozone amounts, the method was not directly validated using measurements until the development of the TOMS V7 algorithm. Nimbus-7 TOMS had 4 wavelengths (331, 340, 360, 380) where the ozone absorption is weak, so the validity of the LER method to reproduce the spectral dependence of the measured radiances at these wavelengths could be directly tested. The residue, as defined in equation 1, provides a measure of error in predicting the spectral contrast using the LER method. (It is very nearly equal to the percentage difference

between the measured spectral contrast and that predicted using the LER method, divided by -2.3. The actual definition is in terms of **?**N-values", historically used by the ground-based Dobson network, and adopted by NASA's buv team. 1 unit of N-value is equal to 2.3% change in radiance.)

$$r_{\lambda} = -100 \left[\log(I_{\lambda}/I_{380})_{meas} - \log(I_{\lambda}(\Omega, R_{380})/I_{380}(R_{380}))_{calc} \right] .$$
(1)

An analysis of the residuals revealed that the LER method, though it works well in large number of cases, does not correctly reproduce the measured spectral contrast in several notable instances. These include scenes containing broken or thin clouds, sun-glint from water and UV-absorbing aerosols. All these effects are now well understood. Here we shall concentrate on just the aerosols.

Figure 6 shows the 340 nm residues for the 7 aerosol models that we introduced earlier. Absorbing aerosols produce positive residues that increase with the aerosol optical depth, with the slope being roughly proportional to the single scattering albedo (ω_0) of the aerosols. In addition to ω_0 , the aerosol size distribution can also change the slope, as seen in the slope difference between models S1 and S2. One also notes that the model S2, which also mimics the behavior of thin clouds, produces almost zero residue. These results are consistent with actual observations by Herman et al., 1997, who have shown that UV-absorbing aerosols, particularly smoke from biomass burning and desert dust produce positive residues. Treating the residues as an aerosol index assumed proportional to optical depth, they have been able to track these aerosols with high degree of fidelity, identifying their sources over land and tracking them as they are transported under prevailing winds. Unpublished results show that sulfate aerosols do produce negative residues as predicted by the S1 model, and are located in the region of high industrial activity.

Qualitatively the ability of the residue method to distinguish absorbing aerosols can be understood by noting that the effect of clouds and large sulfate particles (model S2) on bUV radiances gives nearly zero residues, very similar to a simple increase in surface albedo. Other types of non-absorbing small-particle aerosols produce wavelength dependencies that do not match that predicted by the LER model and thus produce residues.

The principal strength of the residue method is that the clouds are automatically eliminated from the data. Only the UV-absorbing aerosols are seen when the positive residues are mapped (Herman et al., 1997). The method works over snow/ice surfaces, or when underlying clouds are present, and even when the scene is mixed, containing absorbing aerosols and clouds. However, weakly absorbing aerosols (models S1, S2, C1 and D1) are harder to detect over the background noise, which is roughly ~0.2 N-units.

The principal limitation of the residue method is that one needs to know which aerosol model to use in order to derive the optical thickness. Selection of a wrong model can give errors of a factor 4 or more. In addition, as shown in Table 1, one needs to accurately know the height of the aerosol layer, particularly for highly absorbing aerosols. From Table 1, one also notes a unique feature of the residue method: It correctly underestimates the optical thickness when the aerosols are obscured by sub-pixel clouds, even though the clouds themselves are much brighter than the aerosols. Most other methods would overestimate the aerosols by a large amount. However, the method overcorrects highly absorbing aerosols, because of non-linearities in the algorithm.

To summarize, the residue method is a useful way to make maps UV-absorbing aerosols over both land and water, without the clouds or sulfate aerosols showing up in the map. However, it cannot be used to make quantitative estimates of optical thickness unless there is external information about the type and altitude of aerosol present at particular geographic locations. The altitude may be inferred by tracking the observed movement of aerosols from one day to next in comparison with parcel model predictions using known wind fields.

3.2 Direct Method

In the direct method the radiances are used to derive two pieces of information about aerosols (e.g., optical depth and single scattering albedo). Figure 7 shows the relationship between the 340/380 nm radiance ratios and the 380 nm radiances for our 7 aerosol models. Within the domain of these 7 models, one can identify the different aerosol types or mixtures (except for differentiating between the C1 and D1 models). As shown in Table 2, the primary sources of error are aerosol height and cloud contamination. Unlike the residue method, the direct method cannot distinguish clouds from aerosols, and therefore overestimates the aerosol amount when sub-pixel clouds are present. However, the direct method is sensitive to all aerosol types, including the weakly absorbing ones, which do not show up in the residue maps.

4. Results

The aerosol detection techniques discussed in this paper have been applied to the measurements of backscattered UV radiation by the Total Ozone Mapping Spectrometer (TOMS). The TOMS sensor measures the backscattered ultraviolet (buv) radiance at six wavelengths (313, 318, 331, 340, 360 and 380 nm). The N7-TOMS instrument on board the Nimbus-7 spacecraft measured bUV radiances during the 1979-1993 period. A second instrument, on the Russian Meteor-3 satellite, collected data from 1991 to 1994. Two more TOMS sensors, on the Earth Probe and ADEOS platforms started routine operation on August 1 and November 1 of 1996, respectively. Results of the application of the residue method to map the global distribution of absorbing aerosols have been adequately covered elsewhere [*Herman et. al.*, 1997, *Seftor et. al.*, 1997, *Hsu et. al.*, 1996].

For the retrieval of aerosol parameters using the TOMS data, we will take advantage of the capabilities of the residue method for detecting clouds, by eliminating all scenes for which the residues are within the range 0 ± 0.5 N-units. (Along with clouds, this process also removes large particle sulfate aerosols and weakly-absorbing aerosols, but may not completely eliminate mixed scenes containing strongly-absorbing aerosols and sub-pixel clouds). In addition, we also eliminate scenes over water within the sun-glint viewing geometry, and scenes with reflectivities greater than 0.2, to eliminate bright clouds (at large solar zenith angles bright clouds can produce large negative residues) and snow/ice covered surfaces. For the remaining scenes we apply the direct method to compute the optical depth by assuming, for this example, that all aerosols are at 2.9 km altitude. The surface reflectivities used in the calculation are obtained from the available climatology of minimum 380 nm TOMS reflectivity [*Herman and Celarier*, 1997].

Figure 8 provides a one day map of the 380nm aerosol optical depth for August 23 1985 over both land and water surfaces where most of the cloud contamination has been screened out by processing only those scenes with reflectivities less than 0.2. The geographical distribution of optical depth clearly shows large aerosol concentrations over the smoke source regions of Brazil and Central Africa. The large values over the Saharan desert and off the western shore of northern Africa are associated with the dust outbreaks known to take place in this area a this time of the year. Figure 9 provides a coded map of the aerosol model that was selected by the direct method. A detailed comparison of TOMS derived aerosol optical depth with ground based measurements will be published separately.

5. Summary and Conclusions

We have presented the basis of a new method for aerosol detection using the strong wavelength dependance of backscattered radiance in the near ultraviolet. By means of an inversion procedure using radiative transfer calculations and realistic aerosol models, it has been shown that the 340-380 nm spectral contrast can be used to derive aerosol optical thickness and identify the aerosol type provided that the aerosol height and surface reflectivity are known or prescribed. Because the UV-reflectivity of most land types and water is very low, the technique presented here can be applied over both water and land areas. This is a major advantage over visible methods that are mostly restricted to aerosol retrieval over dark water surfaces.

The approach discussed here has been applied to measurements of bUV radiance by the TOMS instrument. The results indicate that the TOMS measured 340 nm - 380 nm backscattered spectral contrast and 380 nm radiance can be inverted to provide a global picture of aerosol optical depth and aerosol type for nearly cloud free pixels. As with most methods of aerosol retrieval from space, a way to identify cloud contaminated pixels is required. Due to the absence of an spectrally based internal technique to screen for cloud contamination, a reflectivity threshold is used above which no retrieval is done. The method can be applied to cloud free scenes (low reflectivity) or to areas where the aerosol layer is above a bright surface (clouds, ice or snow) provided that the reflectivity of the underlying surface is known.

Because of the low spatial resolution of the TOMS footprint (50 km nadir and 100 km average for N7-TOMS and ADEOS-TOMS, and 25 km nadir and 50 km average for Earth-Probe TOMS), the aerosol retrieval method is best applied to large scale events such as continental scale biomass burning, desert dust outbreaks and ash clouds of volcanic origin [*Krotkov et. al,* 1997] that are likely to have cloud free pixels. Validation of the TOMS aerosol retrievals have been discussed in terms of distribution and frequency of occurrence [*Herman et al.,* 1997; *Hsu et al,* 1996] by comparison with ground based sun photometer data and AVHRR satellite data. Quantitative comparisons with sun photometer optical depths will be published separately.

The availability of data on aerosol optical depth and single scattering albedo is of paramount importance in order to account for the scattering and absorption effects of atmospheric particulates on the earth's radiation budget and climate. In spite of the difficulties inherent with satellite based sensing, spaceborne measurements remain the most convenient method to characterize aerosol particles and determine their time and space distribution on a global basis. Currently available satellite data sets on aerosol properties do not provide a full description of the atmospheric aerosol load. The AVHRR aerosol data set provides information on optical depth only over the water surfaces of the earth because of the difficulties associated with the variable high reflectivity of land surfaces in the visible region of the spectrum. The SAM and SAGE family of sensors were specifically designed to retrieve information on stratospheric aerosols and do not function well in the lower troposphere. In addition, because of the nature of the solar occultation observing technique, both time and space coverage are very sparse.

The long-term TOMS data record on backscattered ultraviolet radiation makes an important contribution to the global observational aerosol data base required in climate studies. The currently available Aerosol Index (AI) product [*Herman et al.*, 1997] is a useful tool to map the temporal and spatial variability of absorbing aerosols of natural and anthropogenic origin. For a given aerosol event, the AI is approximately proportional to the optical depth. Although the AI can not be directly interpreted as a measure of any microphysical aerosol property, it is an efficient way to identify the sources and monitor the transport of different kinds of absorbing aerosol particles. Due to the time and space variability of aerosol properties, the inversion of the TOMS spectral contrast cannot be done using just one aerosol model. For that reason, the conversion of spectral contrast to optical depth and single scattering albedo is limited to specific cases in which information on aerosol particle size and vertical distribution is available.

As indicated in the error analysis section, the main shortcomings in the application of this method to infer aerosol properties are the presence of sub-pixel clouds and the uncertainty on the height of the aerosol layer. For the TOMS instruments, neither of these obstacles can be overcome without the use of external information. Given the large TOMS footprint, sub-pixel cloud contamination is by far the biggest problem when converting the AI to optical depth. The use of a reflectivity threshold helps to eliminate the obvious cloud contamination cases, but optically thin clouds may not be screened out with this technique. Moreover, a reflectivity based screening method may result in the rejection of aerosol-containing scenes with reflectivities larger than the threshold, even though the high reflectivity value may be produced by aerosol scattering effects. As a consequence of any cloud screening method, the spatial coverage of a daily product is significantly reduced. The problem of aerosol layer altitude may be addressed with the use of a climatological data base compiled from airborne lidar measurements. Several field experiments (TRACE-A, ABLE, SCAR-B) have collected a large amount aerosol backscatter profiles over the main biomass burning regions of the world. Parcel trajectory modeling of observed aerosol motions can determine the plume height relative to known wind-shear regions. (Elaborate on the possible used of Raman scattering to help in the detection of aerosol height using GOME data).

The choice of wavelengths for aerosol sensing from space is driven by the need to avoid the ultraviolet region of the spectrum (less than 330 nm) where ozone absorption is the main attenuation process of the incoming solar radiation. The use of wavelengths longer than about 450 nm reduces the need of accurately accounting for the multiple Rayleigh scattering component of the upwelling radiation and simplifies the inversion process. However, as a result of interest in the study of the ozone layer during the last three decades, radiative transfer models in the ultraviolet that include molecular multiple scattering [*Herman and Browning*, 1965; *Dave*, 1968] as well as particle scattering [*Dave and Gazdag*, 1970] have achieved a high degree of accuracy. The need to include multiple scattering effects is no longer a constraint in inversion algorithms.

In this work it was shown that the strong wavelength dependance of the molecular scattering process in the near ultraviolet is a useful tool to detect and characterize aerosols. The detection capability for absorbing aerosols is enhanced in the near ultraviolet since the absorbing suspended matter acts not only on the Mie scattered radiation but also on the Rayleigh component. As a result, the strong molecular scattering in the ultraviolet becomes an advantage since it effectively acts to increase the spectral contrast in the presence of absorbing aerosols. In addition, the low reflectivity of most surface types allows aerosol detection and inversion into optical depth over both water and land surfaces. Because of these unique advantages, it is recommended that in the planning of future aerosol sensing experiments, the choice of wavelengths be expanded to include the 330-390 nm

range to aid in the detection and characterization of absorbing aerosols.

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Model	Parameters $r_0(\mu) \sigma$		Reference	Refractive Index	ω ₀ 340 380	τ 340 380	
S 1	0.07	2.03	Shaw (1979)	1.4300i	1.00 1.00	1.03 1.00	
S 2	0.4	1.43	Torres et al. (1995)	1.4300i	1.00 1.00	0.98 1.00	
C1	0.14	1.45	Anderson et al (1996)	1.5502i	0.92 0.93	1.04 1.00	
C2	0.14	1.45	Anderson et al(1996)	1.5504i	0.84 0.85	1.04 1.00	
D1	0.07	1.95	d'Almeida (1987)	1.5702i	0.90 0.91	1.06 1.00	
D2	0.25	2.20	Schütz (1980)	1.5702i	0.72 0.74	0.99 1.00	
D3	0.50	2.20	Schütz (1980)	1.5702i	0.63 0.65	1.00 1.00	

 Table 1. Aerosol models used in the sensitivity analysis.

	S 1	S2	C1	C2	D1	D2	D3
R (± 1%)			±5%	±1%	±2%	±0%	±0%
Z (±1 km)			-10 15	-25 55	-10 15	-30 65	-40 70
Cld (10%)			-10%	-10%	-10%	-20%	-20%

Table 2. Error analysis for the retrieval of optical depth using the residue method

	S 1	S2	C1	C2	D1	D2	D3
R (± 1%)	±0.10	±0.10	±0.10	±0.10	±0.10	±0.05	±0.02
Z (±1 km)	-2 2	-2 2	-5% 9	-18 % 45	-5 10	-25 60	-35 65
Cld (10%)	20%	20%	20%	25%	20%	30%	40%

Table 3. Error analysis for the retrieval of optical depth using the direct method

	S1	S2	C1	C2	D1	D2	D3
R (± 1%)	±0.003	±0.003	±0.004	±0.006	±0.002	±0.018	±0.030
Z (±1 km)			±0.012	±0.015	±0.010	±0.025	±0.040
Cld (10%)			.015	.02	.010	0.10	0.15

Table 4. Error analysis for the retrieval of single scattering albedo using the direct method

Figure Captions

Figure 1. Scattering phase functions (at 380 nm) for the aerosol models in Table 1. The phase functions are plotted only for the range of scattering angles relevant to satellite remote sensing.

Figure 2. Spectral dependance of the change in backscattered radiance, with respect to a molecular atmosphere, for the seven aerosol models used in this study (see Table 1). The results in this figure are for solar zenith angle 40° and nadir view. The reflectivity of the surface is 0.05 and the aerosol layer altitude is 2.9 km. The 380 nm optical depth is 1.0.

Figure 3. Effect of aerosol layer altitude on the 380 nm backscattered radiance for the array of aerosol models used in this analysis. The results shown correspond to computations at three altitudes: 1.4, 2.9 and 5.8 km. Other conditions are: solar zenith angle 40°, nadir view, surface reflectivity 0.05 and aerosol optical depth (380 nm) 1.0

Figure 4. Angular dependance of the backscattered 380 nm radiance. Other conditions are: nadir view, surface reflectivity 0.05, aerosol height 2.9 km and aerosol optical depth (380 nm) 1.0.

Figure 5. Sensitivity of the backscattered 380 nm radiance to surface reflectivity. Other conditions are: solar zenith angle 40°, nadir view, aerosol layer height 2.9 km and aerosol optical depth (380 nm) 1.0.

Figure 6. Relationship between the Aerosol Index and 380 nm optical depth for the seven aerosol models. Other conditions are: solar zenith angle 40°, nadir view, surface reflectivity 0.05 and aerosol layer height 2.9 km.

Figure 7. Ratio of the backscattered radiance at 340 and 380 nm as a function of 380 nm radiance for the aerosol models assumed in this work. Other conditions as in Figure 6.

Figure 8. Map of retrieved 380 nm optical depth on August 23 1985.

Figure 9. Map of the distribution of aerosol types selected for the optical depth retrieval.