Light reflected by an atmosphere containing irregular mineral dust aerosol

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[1] We simulate polarimetric satellite observations of sunlight reflected by a turbid atmosphere over the ocean. We determine the sensitivity of these observations with respect to the optical thickness and the single-scattering albedo of irregular mineral aerosol. Simulated results indicate that both quantities can be retrieved from simultaneous polarization and intensity measurements. Aerosol scattering is modelled using a true measured scattering matrix of irregularly-shaped mineral dust aerosol. We study the suitability of various approximations of the particle shape used for the numerical calculation of scattering matrices. Approximations with spheres or spheroids with a distribution of moderate axis ratios can lead to large errors of the simulated light intensities. A spheroidal approximation including extreme axis ratios is found to be the most appropriate one for simulations of light scattering by irregular mineral aerosol particles, if measured scattering matrices are not available. INDEX TERMS: 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0343 Atmospheric Composition and Structure: Planetary atmospheres (5405, 5407, 5409, 5704, 5705, 5707); 0360 Atmospheric Composition and Structure: Transmission and scattering of radiation; 0669 Electromagnetics: Scattering and diffraction; 3360 Meteorology and Atmospheric Dynamics: Remote sensing. Citation: Veihelmann, B., H. Volten, and W. J. van der Zande (2004), Light reflected by an atmosphere containing irregular mineral dust aerosol, Geophys. Res. Lett., 31, L04113, doi:10.1029/2003GL018229.

1. Introduction

[2] Mineral dust aerosols influence the Earth’s radiation budget by reflection and absorption of solar radiation. Scattering processes increase the photon path length and enhance gaseous absorption. Dust aerosols play a vital role in cloud formation processes and, by virtue of their large surface area, control many heterogeneous reactions in the atmosphere. The dust is mainly raised in dry lakebeds in the Sahara, east Asia and the Saudi Arabian deserts and can be transported over distances of more than thousand kilometers [Husar et al., 1997]. The amount of dust raised and the atmospheric residence time depend strongly on the meteorological conditions and depend amongst others on the size of the particles [Nho et al., 1996]. Satellite observations of reflected sunlight are used to monitor dust aerosols and to estimate their climatic effect. However, the retrieval of the aerosol load as well as its microphysical properties from satellite measurements of reflected sunlight [e.g., Masuda et al., 2002] remains difficult. For a unique solution multi-spectral multi-angle measurements including both the intensity as well as the polarization are necessary [Chowdhary et al., 2001].

[3] We study the sensitivity of simultaneous observations of the intensity and the polarization with respect to the aerosol optical thickness and the single-scattering albedo. Furthermore we specify viewing geometries that are essential for the separation of both quantities. For this purpose we simulate the Earth’s reflectance for a model atmosphere over the ocean loaded with mineral dust aerosol. Aerosol retrievals require an accurate knowledge of the aerosol scattering described by a scattering matrix and the scattering cross section, which depend on the refractive index, the size and the shape of the particles. We model aerosol scattering based on an accurately measured scattering matrix of a mineral dust sample [Volten et al., 2001] that is representative for atmospheric mineral aerosol. The dust sample includes a large variety of highly irregular shapes as we observed on SEM-graphs (not shown).

[4] In many applications scattering matrices are determined numerically. However, for irregularly-shaped mineral aerosol this calculation is so time consuming that it is practically impossible. In practice, a dust aerosol is often approximated with an ensemble of spheres or spheroids with a distribution of sizes and axis ratios. Spherical scatterers can be treated according to the Lorenz-Mie theory. For spheroidal particles the T-matrix method is applicable [Mishchenko et al., 2000], albeit with constraints that are due to computing time and convergence criteria of the numerical code [Mishchenko and Travis, 1998]. We compare the measured scattering matrix with calculations for ensembles of particles with the projected area equivalent size distribution and the refractive index of the dust sample (section 2). Firstly, we use a spherical shape approximation. Secondly, we approximate the aerosol with an ensemble of randomly oriented oblate and prolate spheroids with moderate axis ratios. Finally, we consider a distribution of spheroidal shapes that includes extreme axis ratios up to the convergence limit of the quadruple precision version of the T-matrix code for each size. We estimate the suitability of the shape approximations for aerosol retrievals. For this purpose, we use the reflectance simulations based on the measured scattering matrix as the reference and compare
them with simulations based on calculated scattering matrices (section 3).

2. Scattering Matrices

[5] A light beam is characterized by the Stokes vector \( \mathbf{I} = (I, Q, U, V) \). Its elements represent the total intensity (I) and linearly (Q and U) and circularly (V) polarized components with the units W/m\(^2\) sr. A single-scattering process is described by the matrix multiplication

\[ \mathbf{I}_{\text{scatt}} \approx \mathbf{F} \cdot \mathbf{I}_{\text{in}}. \]

\( \mathbf{I}_{\text{in}} \) and \( \mathbf{I}_{\text{scatt}} \) are the Stokes vectors of the incoming and the scattered light. For an ensemble of randomly-oriented scatterers, where particles and their mirror particles are present in equal numbers, the scattering matrix \( \mathbf{F} \) has the block diagonal form

\[ \mathbf{F}(\Theta) = \begin{pmatrix} F_{11}(\Theta) & F_{12}(\Theta) & 0 & 0 \\ F_{12}(\Theta) & F_{22}(\Theta) & 0 & 0 \\ 0 & 0 & F_{33}(\Theta) & F_{34}(\Theta) \\ 0 & 0 & -F_{34}(\Theta) & F_{44}(\Theta) \end{pmatrix} \]

and depends on the scattering angle \( \Theta \), i.e., the angle between incoming and scattered beam. The scattering angle \( \Theta = 0^\circ \) denotes forward scattering. Volten et al. [2001] have experimentally determined the scattering matrix elements \( F_{ij}(\Theta) \) of a Feldspar dust sample. The refractive index of Feldspar is estimated to be \( m = 1.57 - 0.0005i \). The real part is taken from Huffman [1977], while the imaginary part is adopted from other weakly absorbing silicate minerals (mica) [Sokolik and Toon, 1999]. The size distribution measured is characterized by an effective radius \( r_{\text{eff}} = 1 \) \( \mu \text{m} \) and a standard deviation \( \sigma_{\text{eff}} = 1 \), defined by Hansen and Travis [1974]. The dust sample is representative for weakly absorbing silicate aerosol [see Dubovik et al., 2002]. The matrix elements have been measured at the wavelength 632.8 nm in the range from 5\(^\circ\) to 173\(^\circ\) with intensities relative to the intensity at \( \Theta = 30^\circ \). Since the full range from 0\(^\circ\) to 180\(^\circ\) is required for radiative transfer simulations, we extend the scattering element \( F_{11}(\Theta) \) according to a method suggested by Liu et al. [2003]. In the forward-scattering direction the scattering of small particles with moderate aspect ratios is dominated by Fraunhofer diffraction and is largely the same as for projected area equivalent spheres. Hence, we merge the Lorenz-Mie result from 0\(^\circ\) to 5\(^\circ\) with the measured \( F_{11}(\Theta) \) that is extrapolated to 180\(^\circ\), and scale the measurement until the standard normalization condition

\[ \frac{1}{2} \int_0^\pi F_{11}(\Theta) \sin(\Theta) \, d\Theta = 1 \]

is met. For the relative scattering matrix elements \( F_{ij}(\Theta)/F_{11}(\Theta) \) a cubic spline extrapolation is used for both the forward and the backscattering direction. Forward and backscattering values are determined consistent with the conditions given by Hovenier and Van der Mee [1996] such that they allow the elements \( F_{ij}(\Theta)/F_{11}(\Theta) \) to run smoothly towards 0\(^\circ\) and 180\(^\circ\) and satisfy the Cloude test at all angles.

![Figure 1](image_url)

**Figure 1.** The measured scattering matrix elements \( F_{11}(\Theta), -F_{12}(\Theta)/F_{11}(\Theta), F_{22}(\Theta)/F_{11}(\Theta) \) and \( F_{33}(\Theta)/F_{11}(\Theta) \) of Feldspar dust (green) are compared with calculations based on shape approximations with spheres (solid blue) and spheroids including moderate (dash-dotted red) and extreme (dashed black) axis ratios. The areas (shifted by a factor of 10 in the upper left graph) show that the mismatch of measurements (light green) with Lorenz-Mie calculations for spheres with a projected area equivalent size distribution (light blue) is found as well for other irregular mineral dust samples.

[6] For comparison we consider 3 types of numerically determined scattering matrices. Firstly, the irregularly-shaped aerosol is approximated with an ensemble of spherical particles with the projected area equivalent size distribution of the dust sample. The scattering matrix is then calculated using the Lorenz-Mie theory. Secondly, the irregularly-shaped aerosol is approximated with an ensemble of randomly oriented oblate and prolate spheroids (Bohren and Huffman [1983]). Scattering matrices are determined using the T-matrix method [Mishchenko et al., 2000] for a simple distribution of moderate axis ratios (major/minor \( \leq 2 \) that does not vary with the particle size. Thirdly, we consider a distribution that includes extreme axis ratios up to the convergence limit of the quadruple precision version of the T-matrix code for each size. The maximum axis ratio for both oblate and prolate spheroids where convergence is achieved decreases with the size parameter \( 2\pi r/\lambda \) given for the projected area equivalent radius \( r \) and a wavelength of 632.8 nm (Table 1). Scattering matrices are determined for 31 sizes and up to 20 axis ratios. The ensemble average scattering matrix is obtained by weighting with the scattering cross sections and the normalized size distribution of the dust sample. We assume an equiprobable distribution of axis ratios on a logarithmically equidistant grid.

Table 1. Maximum Axis Ratios, Where Convergence is Achieved

| Axis ratio: | 15  | 9  | 6  | 3.5 | 21.2 |
| Size parameter: | 1 | 510 | 20 | 5080 |
measurement improves when the particle shapes are approximated with spheroids with moderate axis ratios (dash-dotted red). Especially for achieving the bell shape of the element $-F_{12}/F_{11}$ it is essential to incorporate extreme axis ratios (dashed black). The element $F_{23}/F_{11}$ obtained from T-matrix calculations shows a peak around 160° that is not present in the measurements. This has been observed as well by Nousiainen and Vermeulen [2003]. T-matrix calculations of Zakharova and Mishchenko [2001] indicate that discrepancies of the element $F_{23}/F_{11}$ will not vanish by taking into account even more extreme aspect ratios.

[8] The measured scattering matrix, including the elements $F_{34}/F_{11}$ and $F_{44}/F_{11}$ (not shown), is rendered best, when extreme axis ratios are included. We tried various modifications of both the shape as well as the size distribution, but none of them significantly improves the agreement with the scattering matrix measured. The discrepancy may be partly or entirely due to the fundamental difference between scattering by irregular and by spheroidal particles.

[9] We find similar differences between true and calculated scattering matrices for the feldspar sample presented as well as for other aerosol samples with different properties, i.e., a green clay sample ($r_{eff} = 1.5, m = 1.52 - 0.001i$) and a quartz sample ($r_{eff} = 2.4, m = 1.54 - 0i$). We estimate the spheroidal shape approximation including small particles with extreme axis ratios to be of practical use for the simulation of light scattering by atmospheric irregular mineral aerosol.

3. Simulations

[10] We simulate satellite observations of the intensity and the polarization of sunlight reflected by a turbid atmosphere over a rough oceanic water interface at a wavelength of 632.8 nm. The radiative transfer model is based on the doubling-adding method [De Haan et al., 1986] and includes multiple scattering and polarization. We incorporated a module describing the reflection by an oceanic surface [see Mishchenko and Travis, 1997]. The roughness of the water interface is dependent on the near-surface wind speed. We assume a low wind speed of 0.5 m/s in order to avoid a broad sun glint. The atmosphere is sampled by 5 homogeneous plane-parallel layers. Each layer is characterized by the scattering matrix, the single-scattering albedo and the optical thickness of the aerosol. Molecular scattering is described by Rayleigh scattering with a depolarization factor of 0.0310 for air [Hansen and Travis, 1974], while molecular absorption is neglected.

[11] We present simulations of reflected sunlight for a solar zenith angle of 30° and various viewing angles within the principal plane. Figure 2 shows simulations with the viewing angles $-30^\circ$, $+15^\circ$, $+45^\circ$, and $+60^\circ$. The nadir is at 0° and the peak of the sun glint is observed at the viewing angle $+30^\circ$ (not shown). The polarization is represented by the Stokes parameter $Q$ (ordinate) and is plotted against the total intensity $I$ (abscissa). Both parameters are given as radiiances in W/m$^2$sr for an unpolarized incident solar flux of $\pi$(W/m$^2$) measured perpendicular to the incoming direction. The Stokes parameter $U$ is zero due to the symmetry of the scenario.

[12] We study the sensitivity of these satellite observations with respect to the aerosol optical thickness and the single-scattering albedo. Simulations are shown as grids representing the variation of both the aerosol optical thickness [0.1, 0.2, ..., 0.5] and the single-scattering albedo [0.8, 0.84, ..., 1]. An aerosol optical thickness of this order of magnitude is often observed in the lee of deserts [Díaz et al., 2001]. The range of single-scattering albedos is chosen according to an imaginary part of the refractive index ranging from 0 (e.g., for quartz) up to 0.02 (e.g., for volcanic ash [Patterson, 1981]). The single-scattering albedo of the Feldspar sample is close to unity according to its estimated refractive index. For the simulations based on the measured scattering matrix, we vary the single-scattering albedo without modifying the scattering matrix. This is justified by comparisons of scattering matrices of various samples; we find that differences due to the refractive index are significantly smaller than differences due to the particle shapes used. For all calculated scattering matrices we use the refractive indices that correspond to the various values of the single-scattering albedo. Simulations using the measured scattering matrix are depicted green and are compared to simulations based on shape approximations with spheres (blue, bullets) and spheroids including moderate (red, squares) and extreme axis ratios (black, diamonds).

**Figure 2.** Simulations of the intensity $I$ (abscissa) and the polarization $Q$ (ordinate) of reflected sunlight. The grids represent variations of the aerosol optical thickness and the single-scattering albedo. The thick lines indicate a single-scattering albedo of 1; the markers denote an aerosol optical thickness of 0.5. Simulations using the measured scattering matrix (green, marked with asterisks) are compared with simulations based on shape approximations with spheres (blue, bullets) and spheroids including moderate (red, squares) and extreme axis ratios (black, diamonds).
angles close to $-30^\circ$. In this geometry the intensity is overestimated by up to 135% when irregular particles are approximated as spheres and by up to 42% or 23% when using the spheroidal shape approximation including moderate or extreme axis ratios respectively.

[14] In geometries, where the sun glint contributes to the signal, the presence of aerosol reduces the surface reflection that dominates the polarization observed. This leads to an increase of the Stokes parameter Q. When the surface reflection is not too strong, the decrease of the surface reflection is overcompensated by the increase of the aerosol scattering. This is the case for the viewing angles $+15^\circ$ and $+45^\circ$. Here the lines of constant aerosol optical thickness and lines of constant single-scattering albedo are nearly orthogonal. This is not the case for other geometries, where the area spanned by the two variations is degenerated to a narrow stripe (e.g., at viewing angles $-30^\circ$, $-15^\circ$ (not shown), $0^\circ$ (not shown) and $+60^\circ$).

4. Conclusion

[15] Both the polarization as well as the the intensity of reflected sunlight are sensitive to the shape of the particles. This indicates that the retrieval of the amount and the properties of mineral aerosol from satellite observations requires an appropriate description of the particle shape. This is in agreement with the findings of Deuzé (private communications) who use the intensity and the polarization of reflected sunlight observed by POLDER-ADEOS-1 (POLarization and Directionality of the Earth’s Reflectance, [Deuzé et al., 1999]). He found that in cases when mineral dust is present the retrieval of the aerosol optical thickness can be significantly improved by using an average scattering matrix based on scattering matrices measured by Volten et al. [2001] instead of Lorenz-Mie calculations.

[16] Simultaneous observations of the polarization and the intensity are sensitive to the single-scattering albedo as well as the aerosol optical thickness. Such observations in geometries where the sun glint contributes weakly to the observed signal provide independent information about the single-scattering albedo and the aerosol optical thickness, provided that the specular reflection is described accurately.

[17] Based on our sensitivity studies, we conclude that the aerosol optical thickness and the single-scattering albedo retrieved using the observation geometries studied, may comprise large error contributions, if the particle shape is approximated by spheres or spheroids with moderate axis ratios. Polarimetric observations of light scattered by irregular mineral aerosol appear to be best, although not perfectly, represented by simulations using the spheroidal shape approximation including small particles with extreme axis ratios. The presented results indicate that the use of measured scattering data instead of models using simple shape approximations has the potential to improve aerosol retrievals and radiative forcing estimates.

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References


