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Application of spheroid models to account for aerosol particle nonsphericity in remote sensing of desert dust

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[1] The possibility of using shape mixtures of randomly oriented spheroids for modeling desert dust aerosol light scattering is discussed. For reducing calculation time, look-up tables were simulated for quadrature coefficients employed in the numerical integration of spheroid optical properties over size and shape. The calculations were done for 25 bins of the spheroid axis ratio ranging from ~0.3 (flattened spheroids) to ~3.0 (elongated spheroids) and for 41 narrow size bins covering the size parameter range from ~0.012 to ~625. The look-up tables were arranged into a software package, which allows fast, accurate, and flexible modeling of scattering by randomly oriented spheroids with different size and shape distributions. In order to evaluate spheroid model and explore the possibility of aerosol shape identification, the software tool has been integrated into inversion algorithms for retrieving detailed aerosol properties from laboratory or remote sensing polarimetric measurements of light scattering. The application of this retrieval technique to laboratory measurements by Volten et al. (2001) has shown that spheroids can closely reproduce mineral dust light scattering matrices. The spheroid model was utilized for retrievals of aerosol properties from atmospheric radiation measured by AERONET ground-based Sun/sky-radiometers. It is shown that mixtures of spheroids allow rather accurate fitting of measured spectral and angular dependencies of observed intensity and polarization. Moreover, it is shown that for aerosol mixtures with a significant fraction of coarse-mode particles (radii $\geq \sim 1$ μm), the nonsphericity of aerosol particles can be detected as part of AERONET retrievals. The retrieval results indicate that nonspherical particles with aspect ratios ~1.5 and higher dominate in desert dust plumes, while in the case of background maritime aerosol spherical particles are dominant. Finally, the potential of using AERONET derived spheroid mixtures for modeling the effects of aerosol particle nonsphericity in other remote sensing techniques is discussed. For example, the variability of lidar measurements (extinction to backscattering ratio and signal depolarization ratio) is illustrated and analyzed. Also, some potentially important differences in the sensitivity of angular light scattering to parameters of nonspherical versus spherical aerosols are revealed and discussed.

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1. Introduction

[2] Adequate modeling of light scattering by nonspherical particles is widely recognized as one of the major difficulties in remote sensing of tropospheric aerosols in

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general and desert dust in particular. There are various in situ and laboratory measurements [e.g., *Heintzenberg*, 1998; *Nakajima et al.*, 1989; *West et al.*, 1997; *Volten et al.*, 2001] as well as remote sensing results [*Koepke and Hess*, 1988; *Kaufman*, 1993; *Sasano and Browell*, 1989; *Liu et al.*, 1999; *Dubovik et al.*, 2002a; *Deuzé et al.*, 2000; *Sinyuk et al.*, 2003; *Müller et al.*, 2003; *Wang et al.*, 2003; *Herman et al.*, 2005; *Kalashnikova et al.*, 2005] that reveal significant deviations of light scattering of desert dust aerosols from scattering properties of homogeneous spheres [*Mishchenko et al.*, 2000]. As a result, there have been numerous efforts to account for particle nonsphericity in aerosol retrieval algorithms [e.g., *Kahn et al.*, 1997; *Krotkov et al.*, 1999; *Liu et al.*, 1999; *Dubovik et al.*, 2002b; *Mishchenko et al.*, 2003; *Herman et al.*, 2005; *Kalashnikova et al.*, 2005]. Nevertheless, at present, there is no single widely accepted light scattering model for applications to the retrieval of desert dust properties. The development of such a model appears to be difficult both methodologically and technically. For example, *Kalashnikova and Sokolik* [2002, 2004] proposed the strategy of modeling light scattering by an ensemble of nonspherical particles using some geometrical particle parameters derived from in situ measurements of ambient desert dust. However, both the measurement of the aerosol microphysical properties as well as the modeling of light scattering by a single particle with varying size, shape and composition have significant limitations. The in situ and laboratory measurements have issues with sampling of unperturbed aerosol, isolation of aerosol particles, and evaluation of their physical and optical parameters [e.g., *Bond et al.*, 1999; *Haywood et al.*, 2003; *Reid et al.*, 2003a, 2003b; *Tanré et al.*, 2003]. The modeling of optical properties of single particles with diverse geometrical shapes and morphologies has been addressed by many fruitful studies for a number of years [e.g., *van de Hulst*, 1957; *Bohren and Huffman*, 1983; *Mishchenko et al.*, 2000, 2002]. However, because of the complex nature of the light scattering formalism, the models available for applications are rather limited. Specifically, the exact solutions describing the interaction of the electromagnetic field with a single particle exist only for a few selected geometrical shapes [*Mishchenko et al.*, 2000, 2002]. Existing numerical methods, such as the discrete dipole approximation [e.g., *Draine and Flatau*, 1994] and the finite difference time domain technique [e.g., *Yang et al.*, 2000] theoretically have no obvious limitations, but, in practice, require excessive computer resources. Therefore the ability to simulate the interaction of electromagnetic radiation with individual particles of various shape, size and morphology is probably the most critical factor in driving the advances in modeling of the optical properties of nonspherical aerosols.

[3] Thus the majority of approaches employed in aerosol retrievals to account for particle nonsphericity are based on modeling simulations limited in terms of particle size range, geometrical shapes, and compositions. As a result, comparisons of retrieval algorithms utilizing different assumptions for modeling optical properties of nonspherical aerosols are difficult and, quite often, inconclusive. For example, the benefits and limitations of using even the simplest nonspherical shapes, such as spheroids (ellipsoids of revolution), for modeling light scattering by nonspherical aerosol are not completely clear. Specifically, *Mishchenko et al.* [1997] showed that a mixture of randomly oriented spheroids with different sizes and axis ratios can reproduce the flattening of the phase function at side scattering angles for desert dust – perhaps the main scattering feature associated with nonsphericity of aerosol particles. Therefore light scattering by spheroids has been used extensively in remote sensing applications [e.g., see *Mishchenko et al.*, 2004b]. At the same time, there is no physical reason to expect all dust particles to be perfect spheroids, and, indeed, microphotographs of natural aerosols show a great variety of shapes, often different from spheroids. Therefore there are studies aimed at modeling desert dust scattering for more complex and, arguably, more realistic geometrical shapes [e.g., *Yang et al.*, 2000; *Kalashnikova and Sokolik*, 2002, 2004]. However, such simulations require very long computation times and are not possible for the entire size range of desert dust aerosol. For example, the studies of *Kalashnikova and Sokolik* [2002, 2004] limit the calculations to particles with volume-equivalent radii smaller than 1 and ~2 μm correspondingly, while both in situ and remote sensing observation suggest high concentrations of particles with radii up to 5 μm and greater in desert dust aerosol [e.g., see *Arimoto et al.*, 1997; *Li-Jones and Prospero*, 1998; *Dubovik et al.*, 2002a; *Reid et al.*, 2003a, 2003b; *Haywood et al.*, 2003].

6. Conclusions

6.1. Forward Simulations

[63] The look-up tables of phase matrix, extinction and absorption were calculated for polydisperse randomly oriented spheroids for 25 logarithmically equidistant axis ratios ranging from ~0.3 (flattened spheroids) to ~3.0 (elongated spheroids). The kernels computed in 41 narrow logarithmically equidistant size bins cover the size parameter ($x = 2\pi r/\lambda$) range from ~0.012 to ~625. In order to cover such a large range, two complementary methods were used for computing kernels. For size parameters below $x \sim 30$–60 (depending on refractive index and aspect ratio), the simulation were performed using the *T*-matrix method [*Mishchenko and Travis*, 1994] which provides an exact solution for electromagnetic radiation scattering by spheroids. For size parameters exceeding the *T*-matrix convergence limits, the approximated geometric-optics-integral-equation method of *Yang and Liou* [1996] was used. The kernels cover the following ranges of complex refractive index: $1.33 \leq n \leq 1.6$ and $0.0005 \leq k \leq 0.5$ and allow for simulations of $P_{ii'}(\Theta)$ for entire range of scattering angles Θ with 1° resolution. For effectiveness and flexibility of using these computed kernels, a software package has been designed. This software allows for quick simulation of τ_{ext}, τ_{scat} and $P_{ii'}(\Theta)$ using following parameters of the spheroid mixture as input: n, k, size distribution ($dV(r_k)/d\ln r$) and axis ratio distributions ($dn(\varepsilon_p)/d\ln\varepsilon$). The simulations can be done for any λ and r_k within the kernel range of x with expected average accuracy below 3% for $P_{ii'}(\Theta)$ and below 0.005 for τ_{ext} and τ_{scat}. The kernels and software package with detailed description of its functions can be available from the lead author upon request.