



Effect of mineral dust aerosol aspect ratio on polarized reflectance



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ARTICLE INFO

Article history:

Received 21 May 2014

Received in revised form

9 September 2014

Accepted 11 September 2014

Available online 28 September 2014

Keywords:

Dust aerosol

Aspect ratio

Polarization measurement

Satellite

ABSTRACT

The effects of dust particle aspect ratios on single- and multiple-scattering processes are studied using the spheroidal model in order to obtain a better understanding of the radiance and polarization signals at the top of the atmosphere (TOA) under various dust-aerosol-loading conditions. Specifically, the impact of the particle aspect ratio on the polarization state of the TOA radiation field is demonstrated by comparing the normalized polarized radiances observed by the POLDER (POLARization and Directionality of the Earth's Reflectances) instrument on board the PARASOL (Polarisation et Anisotropie des Reflectances au sommet de l'Atmosphère, couples avec un Satellite d'Observation important un Lidar) satellite with the corresponding theoretical counterparts. Furthermore, presented are the aspect ratio values inferred from multi-angular polarized radiance measurements of Saharan and Asian dust by the POLDER/PARASOL.

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1. Introduction

Atmospheric aerosol affects the radiation budget of the Earth–atmosphere system, both directly by a magnitude comparable to the radiative effect of greenhouse gases [1] and indirectly by acting as cloud nuclei [2]. Due to the significant spatial and temporal variations in its microphysical and chemical properties, aerosol has been recognized as a significant source of the uncertainties that afflict global climate models [3,4]. Mineral dust aerosol, originating from arid and semi-arid regions, contribute a major portion of atmospheric aerosol mass loading [5]. The estimated global annual mineral dust aerosol emission is between 1604 and 1960 Tg [6]. Depending on the particle size and influenced by gravitational settling, the atmospheric lifetime of mineral

dust aerosol may vary from a few hours to several years [7]. After being lifted into the atmosphere, mineral dust aerosol may be transported thousands of miles by large-scale circulation and may affect both regional and global climate [2,6,7]. Quantification of the radiative effects of mineral dust aerosol requires global scale property measurements, which can be achieved only by satellite remote sensing.

Retrieval of the mineral dust aerosol properties from satellite measurements relies on our knowledge of dust particle single-scattering properties, including the single-scattering albedo, extinction coefficient, and scattering phase matrix [8,9]. Unfortunately, in laboratory measurements, the phase matrix can be obtained only at specific wavelengths and in limited angular scattering regions, for example, 3° to 177° [10], because experimentally measuring the forward and backward scattering is difficult to do in the visual region of the spectrum. Therefore, some important parameters, such as the asymmetry factor, cannot precisely be determined. Theoretical simulations of the scattering of light by

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real dust particles are also impractical. From electron microscope imaging and energy-dispersive spectroscopy, dust particles are generally found to be irregular in shape and inhomogeneous in composition [11], which pose a pronounced challenge to the simulation of light scattering. Numerical techniques, such as the discrete-dipole approximation (DDA) [12,13], may be suitable for modeling complicated morphologies; however, in the visible spectral region, these methods are impractical for coarse mode dust particles with relatively large size parameters. The computational burden becomes even more severe when ensemble averaging over the particle size distribution and orientation is necessary for practical applications. The simple spherical mineral dust aerosol model, which has an exact theoretical solution based on the Lorenz–Mie theory, has been extensively used in aerosol property retrieval, but evidence has shown that the spherical shape assumption leads to substantial errors in retrieving aerosol optical thicknesses and in estimating the climate forcing effect of mineral dust aerosols [14–17]. To account for the non-sphericity of dust particles, the spheroidal model has been developed and has significantly improved aerosol property retrieval [18–20]. In the spheroidal model, the aspect ratio, defined as the ratio of the major axis to the minor axis of the spheroid, is introduced in addition to the particle size [21]. Special attention must be called to the fact that, even with modern parallel computation, obtaining the single-scattering properties of particles lacking an axis of symmetry is computationally costly. In practice, relatively simple morphologies are assumed to model the optical properties of dust particles. For example, a mineral dust aerosol database based on the ellipsoid model has been developed, which allows for fast retrieval of the single-scattering properties of randomly oriented poly-disperse dust particles [22].

Although realistic dust particles may have far from spheroidal geometries and, indeed, the limitations of the spheroidal model have been recognized [23–25], the mean aspect ratio provides a good measure to quantify the particle non-sphericity and other complexities, which are essential to the estimation of dust forcing in the atmosphere [26]. In fact, dust particle models, which are completely different in terms of particle shape, are able to approximately reproduce the measured dust particle phase matrices [27,28]. Thus, a simpler spheroid-like model can be used and the detailed shape effect can be included through the use of the aspect ratio. Ground based measurements have also shown that the mean aspect ratio behavior is related to the relative humidity [29]; therefore, including this parameter in global aerosol retrieval is desirable.

The sensitivity of space-borne radiometric observations to dust particle shape has been examined in several studies. Substantial variations in dust aerosol microphysical properties, such as particle size, refractive index, and shape, may pose severe problems in aerosol retrieval from multispectral single-viewing-angle radiance measurements [30]. Observations from multi-angle radiometry, e.g., the Multi-angle Imaging SpectroRadiometer (MISR), have been found to relieve the problem and to distinguish different mineral dust types over dark water [31–33]. However, due to relatively bright surfaces that may diminish the signal from the mineral dust aerosol, the retrieval

of particle shape information over land from space-borne observations has seldom been reported. Light scattered by atmospheric molecules and dust particles is strongly polarized and the angular features are sensitive to the dust microphysical properties; whereas, light scattered by land surfaces is weakly polarized and spectrally gray in the visible spectral range [34]. The combined multi-angular radiance and polarized radiance measurements have been suggested to be able to derive aerosol properties with sufficient accuracy to be used for long-term global climate research, even when the land surface properties are unknown [35].

We present a theoretical study of the sensitivity of multi-angular and polarization measurements by POLDER/PARASOL to the mean aspect ratio of dust aerosols. A vector radiative transfer code based on the adding-doubling principle is developed to simulate the transfer of polarized radiation in the atmosphere. The modeling results are compared with POLDER/PARASOL observations over dust aerosol source regions. Section 2 reports the single-scattering properties of ensemble averaged poly-disperse dust aerosols based on the spheroidal model. Section 3 describes the vector radiative transfer model. Section 4 presents a sensitivity study of polarized signals at the top of the atmosphere (TOA) to dust microphysical properties. Section 5 presents the mean aspect ratio values of Saharan and Asian dust aerosols inferred from the POLDER/PARASOL measurements. The conclusions of this study are found in Section 6.

2. Single-scattering properties of mineral dust aerosols

In order to cover a wide range of wavelengths, particle sizes, and aspect ratio values to derive the single-scattering properties of dust particles without incurring a tremendous computational burden, we employ an existing tri-axial ellipsoidal mineral dust aerosol database [15]. The development of the database combined the intrinsic merits of several light-scattering computational methods, including the Lorenz–Mie theory, the T-matrix method, the Amsterdam DDA (ADDA), and an improved geometric optics method (IGOM), in terms of their complementary capabilities to compute the single-scattering properties over a rather broad parameter space. Furthermore, the database formatted the single-scattering properties of randomly oriented ellipsoidal particles, such as the extinction cross-sections, the single-scattering albedos, and the phase matrices, into a kernel form [36]. Through the pre-calculated kernels stored in the database, the ensemble averaged single-scattering properties of poly-dispersed ellipsoidal particles can be efficiently derived. In this study, we model dust particles as prolate spheroids only. As a test of the sensitivity of the single-scattering properties to the aspect ratio for both fine mode and coarse mode mineral dust aerosols, Fig. 1 illustrates the kernel elements related to the polarized phase function P_{12} at a visible wavelength of 670 nm. The results show that, for the fine mode particles, an increasing aspect ratio leads to a decrease in the polarization at side scattering angles. Note, the radii of the equivalent-volume spheres for the fine mode particles are smaller than the incident wavelength. For the coarse

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