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Light scattering by large Saharan dust particles: Comparison of modeling and experimental data for two samples

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ABSTRACT

Light scattering by large mineral-dust particles with small-scale surface roughness is investigated by comparing model simulations with laboratory-measured scattering matrices of two distinct dust samples collected from the Sahara desert. The samples have been chosen on the basis of their large effective radii, and the simulations are based on their measured size distributions. Size parameters larger than about 30 are modeled using a modified ray-optics model RODS (Ray optics with diffuse and specular interactions), while smaller particles are simulated with a *T*-matrix model. RODS allows us to mimic the surface roughness of large dust particles by covering the particle surface by a thin layer of external scatterers with specific single-scattering properties. The Gaussian-random-sphere geometry is used for the shapes of large dust particles. Small particles are modeled as an axial-ratio distribution of spheroids with smooth surfaces. One of the samples consists wholly of large particles and its scattering matrix can be reproduced very well by the RODS model, except for the phase function. The incorporation of wavelength-scale roughness is, however, necessary for good fits. The other sample, consisting of both small and large particles, proves more challenging to match with simulations. The analysis indicates, however, that the difficulties arise at least partially from the small-particle contribution, while RODS results are consistent with the measurements. Further, the results imply that the agreement with measurements would improve if roughness could also be accounted for in the small-particle simulations. Overall, the RODS method seems promising for modeling the optical properties of mineral-dust particles much larger than the wavelength.

1. Introduction

Mineral dust is one of the most abundant aerosols in the Earth's atmosphere. It is known to scatter and absorb solar radiation as well as the thermal infrared radiation emitted by the atmosphere and the surface, affecting the Earth's energy budget. Its presence in the atmosphere also has other complex and often indirect impacts on the Earth-atmosphere system. For example, dust takes part in cloud formation by acting as condensation and freezing nuclei (e.g. [1,2]), thus affecting the water cycle and, indirectly, the distribution of radiative energy. Dust particles fertilize mineral-poor ecosystems such as the Amazonian basin and certain parts of the oceans (e.g. [3,4]). Absorption of solar radiation leads to heating of the atmospheric layers the dust resides in. This also affects hydrostatic stability and, thus, cloud formation.

The direct radiative impact of mineral dust is both considerable and still largely uncertain (e.g. [5,6]). The uncertainty is due not only to the mineral-dust loading

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and distribution within the atmosphere, but also to the poor knowledge of the scattering and absorption properties, hereafter referred to as optical properties, of the dust particles.

Modeling the dust optical properties accurately, however, is very challenging [7]. While wavelength-scale dust particles apparently can be modeled adequately using simple model particles such as a shape distribution of spheroids [8–10], modeling particles larger than the wavelength appears to be quite problematic. One of the key reasons for this difficulty is that the optical properties of large dust particles appear to depend considerably on their surface texture and possible internal structure (e.g. [11,12]), neither of which are well characterized. In addition, there are no exact computational methods that could be applied to the task. Indeed, even the commonly applied ray-optics approximation is unable to account for wavelength-scale texture on the particle surfaces explicitly.

One early attempt at modeling light scattering from dust was made by Nousiainen et al. [11]. These simulations based on traditional geometric optics tended to overemphasize the specular reflection features in the scattering patterns that were absent from laboratory measurements. By making the surface more rough (increasing the variance of surface normals), surface reflections became more diffuse and agreement between the simulations and the measurements improved. Still, even extremely exaggerated roughness did not lead to satisfactory results. It was assumed that the missing factor is the surface roughness in the wavelength scale, which cannot be explicitly accounted for by tilting the surface elements. This small-scale surface roughness was taken into account in the simulations by using an ad hoc parameterization where a user-defined fraction of Fresnellian surface reflections/refractions were treated as Lambertian instead. Such a modification improved the agreement between the simulations and measurements drastically, and essentially proved that the impact of the small-scale roughness needs to be accounted for when modeling optical properties of large dust particles, although the method used was not physically sound. Nousiainen et al. [11] also considered internal Lambertian screens to mimic the impact of possible internal inhomogeneities on scattering. This modification also improved the agreement between the simulations and measurements, but was judged to be of secondary importance compared to the small-scale surface roughness.

One of the uncertainties involved in the study by [11] was that only a part of the size distribution of the sample could be accounted for in the simulations, because a considerable fraction of particles were clearly smaller than that of the ray-optics domain. An attempt was made to account for the small-particle contribution by modeling it using the Mie theory, but this only made the agreement worse. Subsequently, Muñoz et al. [12] measured the scattering matrix in the laboratory for another sample with even larger particles (all in the ray-optics domain) and repeated the modeling exercise with very similar results. Again the Lambertian modifications improved the agreement between the simulations and measurements, but good overall agreements could only be obtained by combining the Lambertian modification with exaggerated large-scale roughness. This provided motivation to consider an improved method of addressing the small-scale effects, using a physically rigorous methodology.

The improved method based on a radiative-transfer framework was introduced by [13]. This model combines Monte Carlo ray tracing with radiative-transfer modeling to make it possible for the light rays to interact with different types of scatterers. It incorporates diffuse, non-specular interactions, in addition to the specular Fresnellian reflection and refraction. Our goal in this work is to reproduce scattering by two distinct mineral dust samples whose scattering matrices have been measured in the laboratory. We assume that surface roughness of the large particles is partially caused by smaller particles aggregated on them, and use three different types of surface scatterers to account for this effect. The only essential factor that is not accounted for in the present treatise is that roughness elements are considered independent scatterers although they are obviously in the near field of the host particle. Sensitivity studies are conducted to see how the remaining free parameters affect scattering, and the results are compared against the laboratory measurements to pinpoint which models result in best fits with the measurements. The paper is organized as follows: in Section 2 we introduce the necessary theoretical concepts and definitions; Section 3 describes the samples of interest as well as their laboratory measurements; the modeling approach is described in details in Section 4; the results are given in Section 5, followed by discussion and conclusions in Section 6.

2. Theoretical aspects

The rate of energy scattered and absorbed by single particles are expressed by scattering and absorption cross sections, $C_{\rm sca}$ and $C_{\rm abs}$. These quantities have dimensions of length squared and relate the power scattered and absorbed to a normal surface area upon which equal power is incident. The total attenuation of incident power is the sum of $C_{\rm sca}$ and $C_{\rm abs}$ and is called the extinction cross section $C_{\rm ext}$. The relative portions of scattered and absorbed power is customarily expressed by the single-scattering albedo $\varpi = C_{\rm sca}/C_{\rm ext}$.

The angular dependence of scattering is specified by the scattering matrix. If the incident and scattered fields are expressed using the normalized Stokes vectors $(I,Q,U,V)^T$ (e.g. [14]), where I is the intensity, Q and U specify the state and the degree of linear polarization, and V is the handedness and the degree of circular polarization, the scattering matrix has a form of a 4×4 Mueller matrix. The scattering matrix relates the properties of incident and scattered light by

$$\begin{pmatrix} I_{\rm s} \\ Q_{\rm s} \\ U_{\rm s} \\ V_{\rm s} \end{pmatrix} = \frac{C_{\rm sca}}{4\pi d^2} \begin{bmatrix} P_{11} & P_{12} & P_{13} & P_{14} \\ P_{21} & P_{22} & P_{23} & P_{24} \\ P_{31} & P_{32} & P_{33} & P_{34} \\ P_{41} & P_{42} & P_{43} & P_{44} \end{bmatrix} \begin{pmatrix} I_{\rm i} \\ Q_{\rm i} \\ U_{\rm i} \\ V_{\rm i} \end{pmatrix}, \tag{1}$$