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The impact of surface roughness on scattering by realistically shaped wavelength-scale dust particles

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ABSTRACT

The impact of dust particles surface roughness on scattering is investigated using model particles with realistic shapes based on stereogrammetry, up to size parameter 10. The surface roughness is introduced at the particle surfaces by randomly adding or subtracting volume elements in random surface locations with a Monte Carlo method, in such a way that the total number of volume elements comprising the particle is approximately conserved. Assuming an ensemble of randomly oriented particles, roughening seems to decrease the backscattered intensity, reduce the horizontal and vertical polarization, especially in the backscattering hemisphere, and increase diagonal and circular polarization at all scattering angles. The asymmetry parameter and single-scattering albedo are virtually unaffected, while lidar ratio and linear depolarization ratio are increased significantly, especially for large size parameters. These results are qualitatively similar for all particles and the impact of roughness on scattering is clearly related to the amount of roughening.

1. Introduction

Mineral dust particles, originating mainly from deserts and other exposed arid regions, are important constituents of the Earth's atmosphere [1]. They can exert a considerable impact on radiation, influencing the radiative balance and remote sensing of the Earth–atmosphere system, e.g., [2,3]. In addition, they act as freezing nuclei and sometimes also as condensation nuclei, contributing to the global water cycle and indirectly to radiation through cloud formation [4,5]. Furthermore, they can be important fertilization agents in iron-limited ecosystems such as surface ocean waters [6] and contribute to air quality and health.

The radiative impacts of dust depend not only on their concentrations and spatial distributions, but also on the dust particles' single-scattering properties, which in turn depend on their sizes, shapes and compositions [7,8]. Regarding the shape, it has been realized that the dust particles' small-scale surface roughness may also be quite important for their single-scattering properties [9–20]. In general, it appears that the introduction of surface roughness tends to smoothen the angular dependence of the scattered intensity, promote positive linear polarization at side-scattering angles, and alter the size dependence of scattering. However, different studies often differ in the details, suggesting a complex interplay between the roughness elements and the host particle. Clearly, the

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impact of surface roughness on scattering depends on the refractive index, host particle shape, and the particle size relative to the wavelength of radiation [7].

When modelling the single-scattering properties of mineral dust, a shape model for the dust particles needs to be established. Historically, these have often been based on simple mathematically defined geometries (e.g., spheres, spheroids, ellipsoids, Chebyschev shapes), or more complex, phenomenological shape models that attempt to mimic the appearance of real dust particles [21]. Recently, stereogrammetry of electron microscope images was adapted in an attempt to derive the real shapes of a small number of real dust particles [22]. As surface roughness is recognized to be a radiatively important parameter in dust particle modelling, and the impact of roughness appears to depend on the model particles' overall shapes, it appears prudent to investigate the impact of surface roughness on scattering using realistically shaped dust particles. To this end, we will use the dust particle shapes derived by Lindqvist et al. [22] and apply a roughening scheme to manipulate their surface roughness characteristics.

2. Theoretical aspects

In the investigation of the surface roughness on scattering, we will focus on the elements of the scattering matrix **S**, which relate the Stokes vectors for the incident and the scattered wave in a scattering event such that

$$\begin{pmatrix} I_s \\ Q_s \\ U_s \\ V_s \end{pmatrix} = \frac{1}{k^2 d^2} \begin{bmatrix} S_{11} & S_{12} & S_{13} & S_{14} \\ S_{21} & S_{22} & S_{23} & S_{24} \\ S_{31} & S_{32} & S_{33} & S_{34} \\ S_{41} & S_{42} & S_{43} & S_{44} \end{bmatrix} \begin{pmatrix} I_i \\ Q_i \\ U_i \\ V_i \end{pmatrix}, \tag{1}$$

where subscripts ‘i’ and ‘s’ refer to incident and scattered waves, respectively; Stokes parameter I describes the intensity, Q and U the linear polarization, and V the circular polarization of the wave; $k = 2\pi/\lambda$ is the wavenumber related to the wavelength λ, and d the distance from the scatterer. The scattering matrix thus contains all the information about the scattering event that can be carried by the scattered wave.

Assuming that the particles are randomly oriented, and particles are mirror symmetric or particles and their mirror particles are present in equal numbers, the scattering matrix simplifies into six independent elements [23]:

$$\mathbf{S} = \begin{bmatrix} S_{11} & S_{12} & 0 & 0 \\ S_{12} & S_{22} & 0 & 0 \\ 0 & 0 & S_{33} & S_{34} \\ 0 & 0 & -S_{34} & S_{44} \end{bmatrix} \tag{2}$$

Indeed, even when all these conditions are not strictly valid, the scattering matrices seem to closely conform to this form for ensembles of complex particles, such as dust [21]. Therefore, for this application, it is sufficient to consider only the effects on these non-zero matrix elements. The diagonal elements govern how the I, Q, U and V are preserved in the scattering event. The off-diagonal elements govern how the pairs I and Q, and U and V, transform into each other.

We also investigate the effect of roughening on several scalar quantities. The asymmetry parameter g describes how the scattered intensity is divided between forward ($\theta < 90^\circ$) and backward ($\theta > 90^\circ$) hemispheres, where θ is the scattering angle, that is, the angle between the propagation directions of the incident and the scattered radiation. Asymmetry parameter value of unity would indicate that all radiation is scattered into the exact forward direction, whereas g of -1 would mean that all radiation is scattered into the exact backward direction. The definition of g is as follows:

$$g = \frac{2\pi}{k^2 C_{\mathrm{sca}}} \int_0^\pi \sin\theta \cos\theta S_{11}(\theta)\, \mathrm{d}\theta, \tag{3}$$

where C_{sca} is the scattering cross-section, a measure of the scattering power of the particle.

Single-scattering albedo, ϖ, describes the proportion of scattering to total extinction in a single scattering event. Extinction is the sum of absorption and scatterings' cross-sections and is related to total incident energy reduction by the particle. Value of unity for ϖ means that all of the radiative energy is scattered and none is absorbed, whereas a zero value means that all of the radiation is absorbed and none is scattered. Calculating ϖ is straightforward from the scattering and total extinction cross-sections:

$$\varpi = \frac{C_{\mathrm{sca}}}{C_{\mathrm{ext}}}. \tag{4}$$

When lidars are used to retrieve aerosol properties, a quantity called lidar ratio, R, is required. The lidar ratio is the ratio of the extinction to backscattering cross-section, and can be calculated as

$$R = \frac{C_{\mathrm{ext}}}{C_{\mathrm{back}}} = \frac{k^2 C_{\mathrm{ext}}}{S_{11}(180^\circ)}, \tag{5}$$

where $S_{11}(180^\circ)$ refers to the value of S_{11} at $\theta = 180^\circ$.

Linear depolarization ratio, δ_L, is another quantity of interest in aerosol lidar retrievals due to the sensitivity of S_{22} to the shape of the particle. For example, spherical, isotropic particles have $S_{11} = S_{22} \Rightarrow \delta_L = 0$. Linear depolarization ratio is defined as

$$\delta_L = \frac{S_{11}(180^\circ) - S_{22}(180^\circ)}{S_{11}(180^\circ) + S_{22}(180^\circ)}, \tag{6}$$

with $S_{22}(180^\circ)$ referring to the value of S_{22} at the exact backscattering direction, $\theta = 180^\circ$.

3. Modelling approach

3.1. Particle shapes

The shapes used as the basis of the surface roughness analysis are stereogrammetrically retrieved shapes of four single mineral dust particles described by Lindqvist et al. [22]. The particles were selected from the samples collected during the SAMUM campaign over Morocco [24]. They were imaged with the scanning-electron microscope from two different directions, and based on these images, the topographies of the particle surfaces were retrieved using stereogrammetry. Since the undersides of the particles were