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Coating effect on light scattering by irregularly shaped particles

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ABSTRACT

In this article, we study the impact of water coating on light scattering by irregularly shaped agglomerated debris particles. We compute light-scattering properties of three types of irregular particles, which simulate atmospheric particles before and after coating. We consider two types of the Junge size distributions over sub-micron range of r_c. The coating is represented as deposition of same volume of material on original particles. We found that only the degree of linear polarization is sensitive to coating type, especially maximum value of the polarization that is typically acquired at side scattering. Behavior of the degree of linear polarization maximum appears in accordance with the Umov effect. The linear depolarization ratio at backscattering direction δ_l is insensitive to coating material, but it is dependent from particles size.

1. Introduction

LiDAR (Light Detecting and Ranging) is a useful tool for retrieving data about vertical aerosol composition of the atmosphere. By using various inverse algorithms, we can convert collected optical data, i.e. extinction and backscatter coefficients, linear depolarization ratio into microphysical properties, such as refractive index, particle size distribution, shape, concentration, volume, effective radius, etc. [1–3]. Usually, retrieval algorithms assume that aerosol particles have a spherical shape. In most cases, this approach is very suitable to describing the results, however, in case of dust particles, this approach fails. Dubovik et al. [4,5] shows that particleâs non-sphericity makes a significant impact on reconstruction of dust microphysical and optical properties. Dubovik's [4,5] approach provides better results for dust particles, but it is still unclear what shape of parameterization should be used for spheroidal model particles [6]. Additionally, it is possible to reconstruct linear depolarization ratio at 532 nm up to the value of 30%, that is typical values for Saharan dust [7,8]. But one of the main drawbacks of the spheroidal model is that such model provides different shape and size distributions for different wave lengths [4]. In [9] we see that using agglomerated debris particles results in a fair approximation of the degree of linear polarization in red and blue light simultaneously. Zubko et al. [9] also reported that the agglomerated debris particles do a reliable job reproducing experimental results, even though they do not necessarily match morphology of feldspar particles. Zubko et al. [9] suggested that highly irregular morphology of particles resulted in good fit to experimental data at different wavelengths. When retrieving aerosol microphysical properties, one can simulate real shape of aerosol particles and then model the light-scattering characteristics. It is significant that as it was shown in [10], the true irregular morphology has negligible impact on particle light-scattering properties, and further improvement of particle shapes may not be critical for obtaining acceptable fits to measurements when particle samples are highly irregular. Therefore, one can use the agglomerated debris particles to simulate dust aerosol, and this will be a reliable approximation of *in situ* and laboratory optical measurements. We have previously demonstrated that dust particles can interact with high humidity values in coastal area [11–14]. There is a number of processes that affect particles size (absorption, adsorption, coagulation, etc.). As was mentioned by Hocking [15], coagulation does not occur for particles sizes less than 18 µm. In gas absorption, the molecules are taken up by the bulk material, but not by the surface (as in case of adsorption). Adsorption characterizes by adhesion of atoms, ions or molecules from gas, liquid or dissolved solid to a surface. Adsorption is a surface based process. In case of adsorption, there is a defined boundary between coating material and the coated one, In absorption such kind of boundary is absent. In ab- and adsorption, particles change its shape and size. One can suppose, that particles become more spherical, so its optical properties appear similar to the ones produced by spheres. Actually this is strictly valid for final stage of humidification, when volume of coating material greater than volume of original parti-

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cle. In this case effect of local heterogeneity occurring during coating becomes inconsequential. If any of these processes takes place for certain distribution, there should be a shift to larger radii. In this paper we address following questions: 1) what happens with light-scattering response to certain aerosol distribution after particles being coated with different material, 2) what optical characteristics are the most sensitive to particle transformation and 3) is it possible to identify this transformation by LIDARs or/and sun photometers.

2. Theoretical aspects in light-scattering modelling

We compute a 4x4 scattering matrix **S** that in the literature also is referred to as Mueller matrix [16], which relates the Stokes vectors of incident and scattered radiation as follows:

$$\begin{pmatrix} I_s \\ Q_s \\ U_s \\ V_s \end{pmatrix} = \frac{1}{k^2 D^2} \mathbf{S} \begin{pmatrix} I_i \\ Q_i \\ U_i \\ V_i \end{pmatrix}, \qquad (1)$$

where subscripts i, s refer to incident and scattered radiation respectively; Stokes parameter I characterizes the intensity, Q,U - the linear polarization and V - the circular polarization of beam of light; $k = 2\pi/\lambda$ - wave number related to the wavelength λ, D is the distance from the scatterer. All information about the target, which can be reconstructed from the scattered radiation, is determined by the scattering matrix. Its elements are dependent of wavelength, shape, size, complex refractive index and orientation of the scatterer. Elements of matrix **S** are also functions of scattering angle θ and azimuth angle ϕ. In case of randomly oriented scatterers the latter dependence gets vanished. Scattering angle θ is the angle between propagation directions of scattered and incident waves.

In case of randomly oriented mirror symmetric particles, or with the same number of particles and their mirror-symmetry particles, we have only six independent elements of matrix **S**.

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

Even though these conditions do not strictly valid, scattering matrix elements for complex particles (eg. dust particles) will conform with the form of (2). This means that in our case we could consider only effects of these six non-zero matrix elements.

In this paper we consider all nonzero Mueller matrix elements that characterize unpolarized incident radiation, among them are intensity of the scattered light S_{11} and the degree of linear polarization

$$P = -\frac{S_{12}}{S_{11}};$$

We also consider an important characteristic arising from the LiDAR studies, the linear depolarization ratio for p-polarized incident light

$$\Delta_l = \frac{S_{11} - S_{22}}{S_{11} + 2S_{12} + S_{22}}$$

and special case - linear depolarization ratio in backward direction $\delta_l = \Delta_l(180^\circ)$. The δ_l is commonly known as indicator for anisotropy in scatterers, most often due to non-spherical shape [17,18]. For isotropic spheres $S_{22} = S_{11}$ and $S_{33} = S_{44}$, so $\delta_l = 0$.

All these quantities follow scale invariant rule [19, p.147] and, thus, are dependent on refractive index and so-called size parameter, which is ratio of particle radius over wavelength λ.

$$x = \frac{2\pi r}{\lambda}. \qquad (3)$$

2.1. Numerical experiment

As shown in [9], agglomerated debris particles have advantages to fit scattering properties of real particles with highly irregular shape (mineral dust and urban aerosols). We consider 10 different shapes generated with the algorithm for agglomerated debris particles (e.g., [10]). Such particles are often accompanied with small fragments that are removed in the present study. The resulting shapes are shown in Fig. 1.

Particle growth was modeled as a random deposition of the same number of dipoles to original particle (i.e. core) by following algorithm.

1. Find all edge cells (having less than 26 neighbors) and add such cells to a list.
2. Select a random surface cell (a) and attach to it an extra cell that is assigned to a mantle material (e.g., water) (b).
3. Test whether the initial cell (a) has 26 neighbors, and, if so, remove such cell from the list, and if true remove it from the list.
4. Test whether the attached cell (b) has an empty neighbor place (i.e., less than 26 neighbors). If this is the case, then such a cell is added into the list of surface cells.
5. Go to step 2 if the number of attached dipoles does not exceed the necessary number.

We prescribe amount of the deposited material to be equal what is in the initial particle shapes. The resulting particle shapes emerging from this procedure are shown in Fig. 2.

In our calculations, we have to determine the way measuring the size of irregular particle. For spherical case r in Eq. (3) corresponds to radius of a sphere. But for arbitrary shaped particle it is somewhat difficult to find equivalent of radius, that will describe particle shape and volume correctly. An obvious way to use volume equivalent radius - radius of a sphere of same volume, another way - use surface area or radius of circumscribing sphere to compare particles. van de Hulst [20] was shown that geometric cross-section is the primary parameter for the extinction of light by a particle larger than the light wavelength. However, both radii hardly characterize light scattering by micron-sized particles as, in this case, interference effects play a key role in forming light-scattering response. The interference effects are largely dependent on base of interference, or distance between two constituent parts of the target particle. Therefore, we use radius of circumscribing sphere r_c that is much better characterizes the largest interference base appearing in a given particle as compared to equal-volume and equal-square radii.

For this paper we consider three types of particles: (1) original particles with refractive index m_1, (2) particles coated with water having core refractive index m_1 and coating refractive index m_2, and (3) the same as in case (2) but coating layer and core are assigned to the same material with $m = m_1$. In latter two cases volume of particle increased by factor of 2. In the experiment $m_1 = 1.40 - 0.01i$ and $m_2 = 1.33 - 0.00i$.

To calculate optical properties of target particles we use implementation of Discrete Dipole Approximation method, earlier developed in [21]. This method has a great advantage of providing a maximum freedom regarding the shape of particle. The accuracy is limited by real shape of particle and its discretization. The latter is depend on both computational resources and the DDA validity criterion, that as was shown [21] in applications to highly irregular particles is as follows $|m|kd \le 1.0$. In this study $|m|kd$ does not exceed 0.75.

Each particle was mapped into tens of thousands of dipoles ($x < 16$). Each particle shape was averaged by 5000 random orientations (50 incident directions and 50 azimuthal orientations and 1 mirroring). All shapes were averaged with equal weight. The com-