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Light scattering by a spherical particle with multiple densely packed inclusions*

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This paper calculates light scattering by a spherical water particle containing densely packed inclusions at a visible wavelength 0.55 μm by a combination of ray-tracing and Monte Carlo techniques. While the individual reflection and refraction events at the outer boundary of a sphere particle are considered by a ray-tracing program, the Monte Carlo routine simulates internal scattering processes. The main advantage of this method is that the shape of the particle can be arbitrary, and multiple scattering can be considered in the internal scattering processes. A dense-medium light-scattering theory based on the introduction of the static structure factor is used to calculate the phase function and asymmetry parameters for densely packed inclusions. Numerical results of the single scattering characteristics for a sphere containing multiple densely packed inclusions are given.

Keywords: ray-tracing technique, Monte Carlo method, light scattering, inclusions
PACC: 4225B, 9265D, 4110H

1. Introduction

In recent years, there are many possible increase of the concentration of aerosol particles, for example, volcanic eruptions,[1] industrial combustions,[2] sand and dust storms,[3] may well lead to a large number of particles inside cloud or fog particles. Many of these particles have long been recognized as an important atmospheric pollutant. They play a significant role in the absorption of solar radiation by atmospheric aerosols and possibly also by clouds. So, particles imbedded in atmospheric particles could potentially reduce the cloud albedo, thereby causing a significant indirect forcing of climate. Some researchers[4] even think that it can be used to interpret 'the cloud and atmospheric absorption anomaly'.

At present, however, there are few techniques available for the accurate computation of light scattering by composite particles covering the entire range of sizes and shapes that can occur in natural clouds. Among possible analytical models of the scatterers, multilayered inhomogeneous spherical particles based on Mie theory appear to be most universal. However, some hard particles trapped inside another large particle may not go into solution. Thus, these particles will not change the overall refractive index of the large particle but only as some internal scatterers.

Macke *et al*[5] have studied light scattering by particles with multiple distinct inclusions. He assumed that the inclusions are randomly and sparsely distributed, so the multiple scattering of the inclusions is based on the independently scattering theory. However, when the density of the internal scatterers reaches a limit, the spatial correlation among densely packed inclusions can substantially change their single scattering properties (such as optical cross sections and phase function[6]), thus make questionable the applicability of the independently scattering approximation in calculation of light scattering by multiple inclusions.

In this paper, we try to get rid of this limitation. The scattering phase function changed with the compaction state of scattering particles is taken into account. In our calculations, we use a dense-medium light-scattering theory based on introducing the so-called static structure factor. The main formula and numerical results of this theory are given in the following section. Then, we present the theoretical procedure for the determination of the single scattering properties by combining ray-tracing and Monte Carlo techniques, and give numerical results for the light scattering by a sphere with multiple densely packed inclusions (Fig.1). The results of this paper are summarized in the concluding section.

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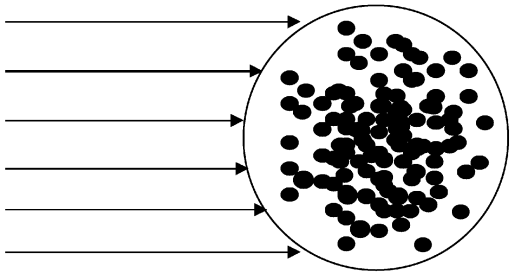


Fig.1. Sketch map of light scattering by a spherical particle with multiple densely packed inclusions.

2. Single scattering characteristics of densely packed inclusions

For sparsely distributed, independently scattering particles, the phase function is given by[7]

$$P(\theta) = \frac{4\pi}{C_{\rm sca}} \frac{{\rm d}C_{\rm sca}}{{\rm d}\Omega}, \tag{1}$$

where θ is the scattering angle and ${\rm d}C_{\rm sca}/{\rm d}\Omega$ is the differential scattering cross section[7] and

$$C_{\rm sca} = \int_{4\pi} {\rm d}\Omega \frac{{\rm d}C_{\rm sca}}{{\rm d}\Omega} \tag{2}$$

is the total scattering cross section.

Densely packed particles are not independent scatterers. Therefore, the concept of the single-scattering phase function cannot be used in the same sense as it is used in computations of light scattering by sparsely distributed particles. However, the concept of the single-scattering phase function can still be used with some modifications to calculate the intensity of light singly scattered by a layer densely packed particles.[6,8] The differential scattering cross section is equal to the product of the independent-scattering differential cross section and so-called static structure factor $S(\theta)$. Thus, instead of Eqs.(1) and (2), for densely packed particles we have

$$P(\theta) = \frac{4\pi}{C_{\rm sca}} \frac{{\rm d}C_{\rm sca}}{{\rm d}\Omega} S(\theta), \tag{3}$$

$$C_{\rm sca} = \int_{4\pi} {\rm d}\Omega \frac{{\rm d}C_{\rm sca}}{{\rm d}\Omega} S(\theta). \tag{4}$$

Note that the theory based on the introduction of the static structure factor has a rather strong physical background and is based on solving Maxwell's equations for calculating light scattering and on statistical mechanics for describing the statistics of mutual positions of densely packed particles.[8,9] So, Eqs.(3) and (4) can be used to calculate the singly scattering phase functions and scattering cross sections of densely packed particles with the consideration of coherent scattering. In addition, Wolf *et al*,[10] Frolen and Maret,[11] and Saulnier *et al*[12] have found that this theory is in good quantitative agreement with results of their laboratory experiments.

The problem of computing the static structure factor for arbitrary particle size distribution and shape is extremely difficult, so we used two usual approximations which have been used by Mishchenko.[6] From the approximation, we assume scattering particles to be nearly spherically shaped and nearly monodisperse (the range of sizes is narrow in comparison with the mean particles size). The structure factor in the Percus–Yevick approximation is given by[13]

$$S(\theta) = \frac{1}{1 - nC(p)}, \tag{5}$$

where

$$p = [4\pi \sin(\theta/2)]/\lambda, \tag{6}$$

n is the number density of scattering particles, λ is wavelength of light, and $C(p)$ is given by

$$\begin{aligned} C(p) =& 24\frac{f}{n}\left[\frac{\alpha+\beta+\delta}{u^2}\cos u - \frac{\alpha+2\beta+4\delta}{u^3}\sin u \right. \\ & - \frac{2(\beta+6\delta)}{u^4} + \frac{2\beta}{u^4} \\ & \left. + \frac{24\delta}{u^5}\sin u + \frac{24\delta}{u^6}(\cos u - 1)\right], \quad p \neq 0, \end{aligned} \tag{7}$$

and

$$C(0) = -24\frac{f}{n}\left(\frac{\alpha}{3} + \frac{\beta}{4} + \frac{\delta}{6}\right), \tag{8}$$

where

$$u = 2pr_0, \tag{9}$$

$$\alpha = \frac{(1+2f)^2}{(1-f)^4}, \tag{10}$$

$$\beta = -6f\frac{(1+f/2)^2}{(1-f)^4}, \tag{11}$$

$$\delta = \alpha f/2, \tag{12}$$

where $f = 4/3\pi n r_0^3$ is the filling factor(i.e., the fraction of a volume occupied by the particles). Note that for sparsely distributed particles (n=0), the structure factor is identically equal to unity.[6]

An interesting question is whether the modified dense-medium phase function given by Eq.(3) can be used to calculate the multiple-scattering contribution to the reflected intensity of densely packed particles. It has been shown in Ref.[6] that it can be used to

compute the full intensity for nonabsorbing or moderately absorbing particles. So the phase function can be used in our Monte Carlo simulations of the light scattering by inclusions.

In our study, the internal scatterers are considered to be ammonium sulfate aerosols ($(NH4)_2SO_4$), which is a naturally occurring aerosol type. The shape of the particles is assumed to be spherical, and the mean radius of the inclusions is assumed to be $0.5\,\mu$m. Mie theory is used to obtain the optical properties of the internal scatterers. It is well known that monodisperse Mie quantities as a function of size parameter contain a high frequency ripple which is usually suppressed in practice because, in nature, there is always a dispersion of grain sizes.[7] Therefore, following Wiscombe and Warren,[14] we reduced the ripple by averaging the phase function over a range of sizes which is small relatively to the mean grain size. The relative refractive indices of $(NH4)_2SO_4$ to outer particle is taken to be 1.15.[5]

Our calculations were performed at a wavelength of λ=0.55 μm, i.e. at the maximum of the solar irradiation. We compared the phase functions of densely packed internal scatterers and sparsely distributed ones in Fig.2. The phase function is shown as a function of the scattering angle θ for five values of the filling factor. It is seen that increasing packing densely results in an angular redistribution of the scattered intensity. The influence of packing density is especially significant at scattering angles $\theta < 50°$. This agrees well with the Mishchenko's[6] conclusion. The other two single scattering parameters, extinction efficiencies ($Q_{\rm ext}$) and asymmetry parameters (g), are listed in Table 1 as a function of filling factor.

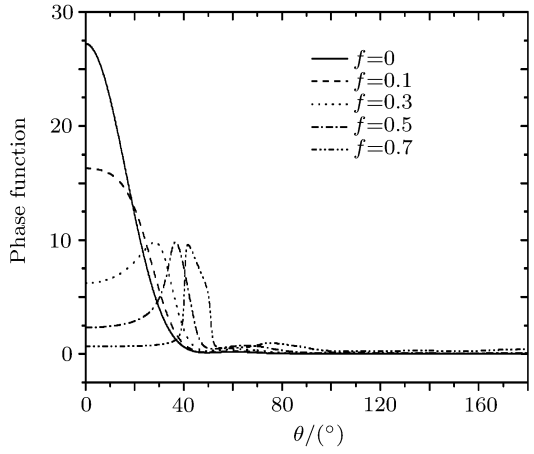


Fig.2. Scattering phase functions of spherical inclusions made of ammonium sulfate.

Table 1. Extinction efficiencies and asymmetry parameters for inclusions as a function of filling factor.

filling factor	$Q_{\rm ext}$	g
0	1.3509	0.9142
0.1	1.0314	0.8930
0.2	0.7726	0.8659
0.3	0.5619	0.8301
0.4	0.3917	0.7815
0.5	0.2571	0.7140

From Table 1 we can see that both extinction efficiencies and asymmetry parameters decrease with the increase of the filling factor. This implies that the scattering approaches to homogeneous scattering.

3. Total scattering simulations with the ray-tracing and Monte Carlo model

The total scattering of light ray by a sphere containing densely packed spherical inclusions is calculated by a combination of ray tracing and Monte Carlo techniques.[5] The ray tracing programs take care of the reflection and refraction events, and the Monte Carlo methods do with the internal scattering processes.

A light ray hitting the surface of the sphere at an incident angle θ_i, will be partially reflected and partially refracted. The reflection angle can be computed with the reflection law, and the refracted angle can be calculated with the Snell's law, i.e. $\sin\theta_{\rm i} = m\sin\theta_{\rm t}$, where $\theta_{\rm i}$ and $\theta_{\rm t}$ are the incident angle and refractive angle, respectively, and m is the relative index of refraction of the particle to the medium. The intensity of the reflected and refracted energy can be obtained by the Fresnel formulas.[15]

After an incident photon is refracted into the sphere, it travels a free path length l given by

$$l = - < l > \log[R(0,1)], \tag{13}$$

where $< l >$ is the mean free path length between two subsequent scattering events and $R(0,1)$ is an equally distributed random number within the interval (0,1). The number density of the inclusion N is described by the mean free path length $< l >$ or by the volume extinction coefficient $\beta_x = 1/ < l >$. For an ensemble of N particles per volume element whose size obeys a standard Gamma distribution, β_x is given by

$$\beta_x = N\int_{r_1}^{r_2} \beta_x(r)n(r){\rm d}r, \tag{14}$$

where $n(r)$ is the normalized particle size distribution function. In this paper, we choose $n(r)$ to be equally distributed.

If the photon has not reached the boundaries of the sphere, its previous direction is changed along the scattering angle and azimuth angle according to

$$\int_0^{\theta} P^{(\mathrm{i})}(\theta)\sin\theta \mathrm{d}\theta = R(0,1)\int_0^{\pi} P^{(\mathrm{i})}(\theta)\sin\theta \mathrm{d}\theta, \quad (15)$$

$$\phi = R(0,2\pi), \quad (16)$$

where $P^{(\mathrm{i})}(\theta)$ denotes the scattering phase function of the internal inclusions. The processes stated in Eqs.(14) and (15) are repeated until the photon reaches the boundaries of the sphere, where it is again subjected to reflection and refraction events. The whole procedure is again repeated for internally reflected component until the photon energy falls below 10^{-5} times the incident energy. However, by setting the minimum free path length l to be 4 times the particle's radius, we ensure the possible 'overlap' phenomenon to be avoided.

The effects of internal inclusions are studied for a water sphere whose radius is equal to 100 μm. Figure 3 presents results of the total phase function for a spherical particle containing densely packed inclusions. Results are shown for the filling factor f=0.001, 0.01, 0.1, 0.5, for comparison, the phase function of pure water sphere is also shown. It can be seen from Fig.3 that dominant features of the phase function are enhanced at scattering angles of 138° for pure water spherical particle, corresponding to the so-called rainbow. With increasing the filling factor f, the phase functions tend to more and more homogeneous except the diffraction region. These results come from the effects of the internal impurities. A photon will suffer much more scattering by the inclusions, so the scattering intensity will be homogeneously distributed in the inner of the water sphere.

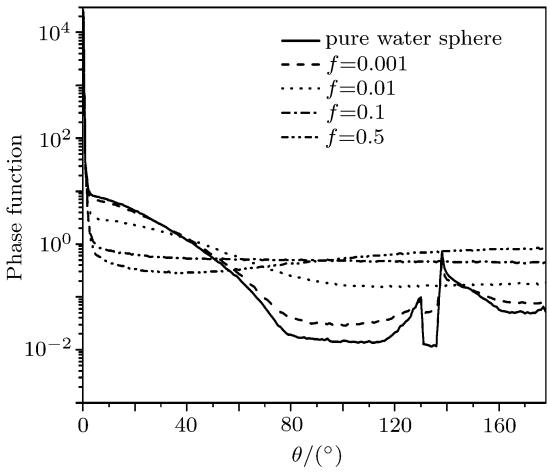


Fig.3. Total scattering phase function of a sphere with internal ammonium sulfate. The filling factor is f=0.001, 0.01, 0.1 and 0.5. The scattering phase function of a pure water sphere without inclusions is given for a comparison.

Table 2 lists the change in the asymmetry parameter g of the spherical particles containing ammonium sulfate as a function of the filling factor. Extinction efficiencies are not shown here since the values approximately equal to 2 when the particle size reaches the limits of geometrical optics. It is obvious that the asymmetry parameter g decreases with the increase of the filling factors, so the overall scattering of the composite particle approaches to homogeneity as the packing density of the inclusions increase to a limit.

Table 2. Asymmetry parameter g of a water spherical particles containing ammonium sulfate as a function of filling factor.

filling factor	g
0	0.8841
0.001	0.8695
0.01	0.7578
0.1	0.5220
0.5	0.4084

4. Conclusion

In this paper, we used a combination of ray-tracing and Monte Carlo techniques to compute the singly scattering phase function for a water spherical particle containing densely packed ammonium sulfate inclusions. A dense-medium light scattering theory based on introducing the static structure factor was used to consider the spatial correlation among densely packed particles, and the singly scattering properties of the inclusions were calculated. A ray tracing program takes care of the individual reflection and refraction events at the outer boundary of the particle. Our calculations show that the effects of the internal impurities on the total scattering phase function are great. The total scattering phase function was smoothed relatively to the pure water sphere. This is because that a photon will suffer much more scattering by the inclusions, the scattering intensity will be homogeneously distributed in the inner of the spherical particle. This may have a noticeable impact of the clouds over scattering and absorption properties.

In this paper, the absorption of the outer particles is nonabsorbing and the shape of the inclusion is spherical, so additional work can be done to consider the non-spherical inclusions and the absorption of the surrounding material.[16]

Acknowledgments

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