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Full Length Research Paper

Simple technique to evaluate effects of nonsphericity and orientation on particle size distribution retrieval

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This paper reports the formulation of an algorithm based on the anomalous diffraction theory to infer the particle size distribution (PSD) from theoretical perspective, taking the effects of particle nonsphericity and orientation into account. Preliminary analysis of input parameters sensitivity of the analytical retrieval formulas of the different PSDs, it is found that there exist simple scale transformation relations between the retrieved and the true PSD for spheroids and triangular prisms if inappropriate shape and orientation assumption are made, but scale transformation relation is invalid for cubes, because the particle modes occur in retrieval procedure.

Key words: Particle size distribution, extinction spectrum, inversion, atmospheric particles.

INTRODUCTION

Atmospheric particles (e.g. aerosols, raindrops, cloud droplets and ice crystals) play a large role in the energy balance of the earth-atmosphere system by scattering, absorbing and emitting solar and planetary radiation. Meanwhile, the coupling between atmospheric particles and water cycles is of increasing importance. Compared with the radiative properties of long-lived and globally well-mixed greenhouse gases, those of atmospheric particles are size-distribution-dependent and with high temporal and spatial variability.

There has been consequently significant interest in determining the particle size distribution (PSD) over the past few decades (Phillips, 1962; Ramachandran and Leith, 1992; Wang et al., 2006). These results are all based on the assumption of spherical morphology, but atmospheric particles generally have nonspherical geometries. For example, raindrops display spheroidal shapes; the shapes and sizes of ice crystals in clouds are governed by the temperature and supersaturation of the environment in which it grows, and their habits usually show hexagonal prism structure (Liou and Takano, 1994); aerosols show a great variety of shapes (Nakajima et al., 1989). On the other hand, sufficient theoretical computations and experiments (Matthias et al., 1998; Krotkov et al., 1999; Kalashnikova and Sokolik, 2002; Gobbi, 2002) show that the scattering properties of nonspherical dust particles and ice crystals in cirrus cloud are significantly different from those of equivalent spheres, and some retrieval biases cannot be explained by the spherical particles model (Wenzel et al., 1996). If the satellite reflectance measurements over the ocean are to be analyzed using the conventional Mie theory (1908) for moderate dust particles, the caused relative errors in the retrieved optical thickness and size distribution of aerosols will be found to be over 100% (Mishchenko et al., 1995), which exceeds the minimum accuracy necessary for the long-term monitoring of global climate forcings and feedbacks. For these reasons, the nonspherical effects on retrieval have recently become a subject of active research (Liu et al., 1999; Wang et al., 2003, Mishchenko et al., 2003; Kocifaj and Horvath, 2005; Dubovik et al., 2002; Zhao et al., 2003; Wang et al., 2003; Dubovik et al., 2006; Feng et al., 2009). Most of them belonging to the so-called direct inversion method in which the optical properties of particles must be numerically calculated by complex light scattering algorithms, such as discrete dipole approximation (DDA) method (Draine and Flatau, 1994), the finite-difference time domain (FDTD) method

(Yang et al., 2000) and T-matrix method (Mishchenko and Travis, 1994). Unfortunately, incorporating nonspherical scattering into the particle size distribution retrieval is extremely expensive in both time and resources for these complicated computational schemes. Hence, they do not yield understanding commensurate with its complexity.

The anomalous diffraction theory (ADT) is alternate approximation to Mie theory for light scattering by spherical particles in homogeneous medium (van de Hulst, 1957), and has been extensively used for calculating extinctions by particles with complex shapes (Greeberg and Meltzer, 1960; Napper, 1967; Gross and Latimer, 1970; Chylek and Klett, 1991a, b; Streekstra et al., 1994; Fournier and Evans, 1996; Liu et al., 1998; Sun and Fu, 2001).The biggest advantage is that of its simplicity. ADT presumes that the index of refraction is close to unity and that the size parameter is large when compared with the wavelength, which implies that the refraction and the reflection are negligible as the ray passes through the particle, thus the presence of a particle will only produce a change in the complex phase front of an incident plane wave over its geometrical shadow area. Some analytical and numerical inversion schemes based on the ADT have been presented already (Shifrin and Perelman, 1963; Box and McKellar, 1978; Shifrin et al., 1981; Fymat, 1978; Smith, 1982; Klett, 1984; Wang and Hallett, 1996; Franssens et al., 2000; Sun et al., 2008; Zhao, 2010). However, there is still lack of an intuitive view of influences of nonsphericity and orientation on PSD retrieval.

The present paper is devoted to this issue from a theoretical viewpoint. Subsequently, on the basis of ADT, we develop analytical algorithms to retrieve spheroid, triangular prism and cube size distribution from extinction spectrum in the ideal case of nonabsorption. The effects of nonsphericity and orientation for different models are compared and discussed later in the work. Finally, the main conclusions drawn from this study are given.

DEVELOPMENT OF RETRIEVAL ALGORITHM FOR DIFFERENT SHAPE MODELS

Spheroidal model

Although, particles in the atmosphere usually have quite complicated morphologies, spheroid model is reasonable approximation and has been widely employed in relevant remote sensing studies. The convenience of this assumption is that the spheroids can be used to model particle shape over a wide rang from flat oblate disks to spheres to elongated needles by simply varying the aspect ratio. Suppose the condition of single scattering is satisfied, the extinction coefficient of polydispersions of particles with fixed aspect ratio and orientation can be expressed by:

$$\tau(\kappa) = \int_0^\infty \chi\pi a^2 Q_{ext}(\kappa a) n(a)\, da\,, \tag{1}$$

Here, the inverse problem consists in retrieving the unknown PSD, $n(a)$, from a set of $\tau_{ad}(\kappa)$ values, $Q_{ext}(\kappa a)$ is the extinction efficiency of a spheroidal particle, which has relatively simple formula in the frame of ADT (Greeberg and Meltzer, 1960).

$$Q_{ext}(\kappa a) = 2 - 4\frac{\sin(\kappa a)}{\kappa a} + 4\frac{1-\cos(\kappa a)}{(\kappa a)^2}\,, \tag{2}$$

with

$$\kappa = \frac{4\pi(m-1)}{\lambda\sqrt{\cos^2\theta + \mu^2\sin^2\theta}}\,,$$
$$\chi = \mu^{-2}\sqrt{\cos^2\theta + \mu^2\sin^2\theta}\,, \tag{3}$$

where λ is the wavelength, m is the refractive index of the particle relative to the medium (for simplicity, it is assumed that m is real), $\mu = a/b$ is the aspect ratio (for prolates $\mu > 1$ and for oblates $\mu < 1$), a and b correspond to the lengths of the semiaxis of rotation and other axes of the spheroid, θ is the angle between the symmetry axis and the direction of incident radiation.

Inserting Equation 2 into Equation 1 yields:

$$\tau(\kappa) - \tau(\infty) = \int_0^\infty [-4\frac{\sin(\kappa a)}{\kappa a} + 4\frac{1-\cos(\kappa a)}{(\kappa a)^2}][\chi\pi a^2 n(a)]da\,, \tag{4}$$

where $\tau(\infty) = \int_0^\infty 2\chi\pi a^2 n(a)\, da$.

From Equation 4, it is shown that:

$$-\frac{d}{4\pi\chi\kappa d\kappa}\{\kappa^2[\tau(\kappa) - \tau(\infty)]\} = \int_0^\infty [a^2 n(a)]\cos(\kappa a)\, da \tag{5}$$

Using the inverse Fourier cosine transform, the retrieved PSD can be expressed by:

$$n(a) = -\frac{1}{2\pi^2 a^2 \chi}\int_0^{+\infty}\left[\frac{d}{\kappa d\kappa}\{\kappa^2[\tau(\kappa) - \tau(\infty)\}\right]\cos(\kappa a)\, d\kappa. \tag{6}$$

Triangular prismatic model

Using the ADT to a right triangular prism of length l and side a oriented, such that the incident radiation is parallel to the bases, we have (Chylek and Klett, 1991a).

$$Q_{ext}(\kappa a) = 2[1 - \frac{\sin(\kappa a)}{\kappa a}], \tag{7}$$

with

$$\kappa = \frac{\sqrt{3}\pi(m-1)}{\lambda \sin(2\pi/3-\theta)}, \tag{8}$$

where θ denotes angle between the direction of incident radiation and one of the side face. The extinction coefficient $\tau(\kappa)$ for triangular prismatic particles with PSD $n(a)$ is given by:

$$\tau(\kappa) = \int_0^\infty \chi a l Q_{ext}(\kappa a) n(a) da \tag{9}$$

where $\chi = \sin(2\pi/3-\theta)$. Then, we get the result as:

$$\tau(\kappa) - \tau(\infty) = -\chi l \int_0^\infty a \frac{\sin(\kappa a)}{\kappa a} n(a) da, \tag{10}$$

where $\tau(\infty) = \int_0^\infty 2\chi a l n(a) da$. From the Equation 10, it follows that:

$$-\frac{1}{\chi l}\{\kappa[\tau(\kappa) - \tau(\infty)]\} = \int_0^\infty n(a)\sin(\kappa a) da. \tag{11}$$

Using the inverse Fourier sine transform, the PSD can be expressed by:

$$n(a) = -\frac{2}{\pi \chi l}\int_0^{+\infty} \kappa[\tau(\kappa) - \tau(\infty)]\sin(\kappa a) d\kappa \cdot \tag{12}$$

Cubic model

The analytical formula of the extinction for a cube, oriented at an arbitrary angle to the incident beam has not yet been achieved. Following Napper (1967), we only consider three special orientations of the cube cases.

Case 1: Incidence normal to a cube face

If the incident light is normal to one face of the cube with a side length a, then, the extinction efficiency can simply be expressed by:

$$Q_{ext,1}(\kappa_1 a) = 2[1 - \cos(\kappa_1 a)], \tag{13}$$

where

$$\kappa_1 = \frac{2\pi(m-1)}{\lambda}, \tag{14}$$

Then, the extinction spectrum $\tau_1(\kappa_1)$ for cubic particles with PSD $n_1(a)$ is given by:

$$\tau_1(\kappa_1) = \int_0^\infty a^2 Q_{ext,1}(\kappa_1 a) n_1(a) da. \tag{15}$$

As a result of the Riemann–Lebesgue lemma, $\lim_{\kappa_1 \to \infty} \int_0^\infty \cos(\kappa_1 a) n(a) da = 0$ provided that $n_1(a)$ is absolutely integrable. In that case, $\tau_1(\infty) = \int_0^\infty 2a^2 n_1(a) da$, so,

$$-\frac{1}{2}[\tau_1(\kappa) - \tau_1(\infty)] = \int_0^\infty a^2 \cos(\kappa_1 a) n_1(a) da. \tag{16}$$

The PSD can be inverted through a Fourier cosine transform with:

$$n_1(a) = -\frac{1}{\pi a^2}\int_0^{+\infty} [\tau_1(\kappa_1) - \tau_1(\infty)]\cos(\kappa_1 a) d\kappa_1. \tag{17}$$

Case 2: Edge incidence

For this orientation, the acute angle between the incident beam and each cube face is $\pi/4$. Thus, we have:

$$Q_{ext,2}(\kappa_2 a) = 2 - 2\frac{\sin(\kappa_2 a)}{\kappa_2 a}, \tag{18}$$

where

$$\kappa_2 = \frac{2\sqrt{2}\pi(m-1)}{\lambda}, \tag{19}$$

The extinction coefficient of polydispersions of particles is given by:

$$\tau_2(\kappa_2) = \int_0^\infty \sqrt{2} a^2 Q_{ext,2}(\kappa_2 a) n_2(a) da. \tag{20}$$

Consequently, $\tau_2(\infty) = \int_0^\infty 2\sqrt{2} a^2 n_2(a) da$. Multiplying Equation 20 by κ_2 and taking the derivative with respect to κ_2, we can obtain:

$$-\frac{1}{2\sqrt{2}}\frac{d}{d\kappa_2}\{\kappa_2[\tau_2(\kappa_2)-\tau(\infty)]\}=\int_0^\infty a^2\cos(\kappa_2 a)n_2(a)da\,, \quad (21)$$

Using the inverse Fourier cosine transform, the PSD can be written as:

$$n_2(a)=-\frac{1}{\sqrt{2}\pi a^2}\int_0^{+\infty}\frac{d}{d\kappa_2}\{\kappa_2[\tau_2(\kappa_2)-\tau_2(\infty)]\}\cos(\kappa_2 a)d\kappa_2 \quad (22)$$

Case 3: Corner incidence

When the incident beam impinges upon a corner and the direction of propagation lies along the body diagonal of the cube, the extinction efficiency of cube can be formulated by:

$$Q_{ext,3}(\kappa_3 a)=2-\frac{4[1-\cos(\kappa_3 a)]}{(\kappa_3 a)^2}, \quad (23)$$

with

$$\kappa_3=\frac{2\sqrt{3}\pi(m-1)}{\lambda}. \quad (24)$$

The extinction spectrum $\tau_3(\kappa_3)$ is given by:

$$\tau_3(\kappa_3)=\int_0^\infty \sqrt{3}a^2 Q_{ext,3}(\kappa_3 a)n_3(a)da\,. \quad (25)$$

Therefore,

$$-\frac{1}{4\sqrt{3}}\frac{d^2}{d\kappa_3^2}\{\kappa_3^2[\tau_3(\kappa_3)-\tau_3(\infty)]\}=\int_0^\infty a^2\cos(\kappa_3 a)n_3(a)da \quad (26)$$

where we have used $\tau_3(\infty)=\int_0^\infty 2\sqrt{3}a^2 n_3(a)da$. The inversion formula now is given by:

$$n_3(a)=-\frac{1}{2\sqrt{3}\pi a^2}\int_0^{+\infty}\frac{d^2}{d\kappa_3^2}\{\kappa_3^2[\tau_3(\kappa_3)-\tau_3(\infty)]\}\cos(\kappa_3 a)d\kappa_3 \quad (27)$$

EFFECTS OF NONSPHERICITY AND ORIENTATION

Development of retrieval algorithm for different shape models illustrates how to reconstruct the size distribution of nonspherical particle from the extinction data. Of course, the shape and orientation are assumed to be accurately known prior to the size distribution retrieval. However, uncertainties and perturbations may exist. Therefore, it is important to evaluate what happens if correct parameters are not employed.

Spheroidal model

Suppose extinction spectrum is accurately known everywhere, μ_0, θ_0 and $n_0(a)$ are true, μ and θ are used in retrieval procedure and $n(a)$ is the retrieved PSD for spheroidal model. Letting:

$$\gamma=\frac{\sqrt{\cos^2\theta_0+\mu_0^2\sin^2\theta_0}}{\sqrt{\cos^2\theta+\mu^2\sin^2\theta}}, \quad (28)$$

From Equation 6, we can deduce a conclusion:

$$n(a)=\frac{\mu^2}{\mu_0^2}\gamma^4 n_0(\gamma a), \quad (29)$$

This leads immediately to the following relationships about the p th moment:

$$\int_0^\infty a^p n(a)da=\frac{\mu^2}{\mu_0^2}\gamma^{3-p}\int_0^\infty a^p n_0(a)da, \quad (30)$$

the total particle concentration:

$$\int_0^\infty n(a)da=\frac{\mu^2}{\mu_0^2}\gamma^3\int_0^\infty n_0(a)da, \quad (31)$$

and the average length of the semiaxis of rotation of spheroids:

$$\frac{\int_0^\infty a\cdot n(a)da}{\int_0^\infty n(a)da}=\frac{1}{\gamma}\frac{\int_0^\infty a\cdot n_0(a)da}{\int_0^\infty n_0(a)da}. \quad (32)$$

It is of interest to note that the effects of nonsphericity and orientation cannot be separated.

Triangular prismatic model

Similarly, we may assume that the extinction spectrum is accurately known everywhere; also, θ_0 and $n_0(a)$ are true, and θ is used in retrieval procedure and $n(a)$ is the retrieved PSD for triangular prismatic model. Letting:

$$\gamma = \frac{\sin(2\pi/3-\theta_0)}{\sin(2\pi/3-\theta)}. \tag{33}$$

From Equation 12, it now follows that:

$$n(a) = \gamma^3 n_0(\gamma a), \tag{34}$$

and

$$\int_0^\infty a^p n(a)da = \gamma^{2-p}\int_0^\infty a^p n_0(a)da, \tag{35}$$

$$\int_0^\infty n(a)da = \gamma^2 \int_0^\infty n_0(a)da, \tag{36}$$

$$\frac{\int_0^\infty a\cdot n(a)da}{\int_0^\infty n(a)da} = \frac{1}{\gamma}\frac{\int_0^\infty a\cdot n_0(a)da}{\int_0^\infty n_0(a)da}, \tag{37}$$

These results are very similar to those for spheroids.

Cubic model

If we use same extinction data in Equations 17, 22 and 27 for different orientation case, it is easy to find that:

$$n_2(a) = 2n_1(\sqrt{2}a) - \frac{1}{\pi a^2}\int_0^{+\infty}\frac{\kappa_1 d\tau(\kappa_1)}{d\kappa_1}\cos(\sqrt{2}\kappa_1 a)d\kappa_1, \tag{38}$$

$$n_3(a) = 3n_0(\sqrt{3}a) - \frac{1}{\pi a^2}\int_1^{+\infty}\left\{\frac{\kappa_1^2}{2}\frac{d^2\tau(\kappa_1)}{d\kappa_1^2} + \frac{2\kappa_1 d\tau(\kappa_1)}{d\kappa_1}\right\}\cos(\sqrt{3}\kappa_1 a)d\kappa_1. \tag{39}$$

Clearly, the spurious particle modes occur for both case 2 and case 3 as compared to case 1 in retrieval procedure.

Conclusions

The impact of nonspherical particles and their orientation on the retrievals of the PSD is important to consider. The difficulty in evaluating the impact of nonspherical particles on the PSD retrieval is that there is no one complete light scattering method that can be applied to study such sensitivities, this being principally due to particle shape and size limitations. Our approach is to use the ADT applied to oriented spheroids, triangular prisms and cubes to solve for the extinction efficiency and this solution can be incorporated analytically into the solution for the PSD. The major advantage of this approach lies in its simplicity. Through preliminary analysis of input parameters sensitivity of the analytical retrieval formulas of the different PSDs, it is found that the effects of nonsphericity and orientation cannot be separated. There exist some scale transformation relations between the retrieved and the true PSD for spheroids and triangular prisms if inappropriate inputs are made, but scale transformation relation is invalid for cubes in view of the fact that that the particle modes occur in retrieval procedure.

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