LIGHT SCATTERING BY NONSPHERICAL PARTICLES IN THE ATMOSPHERE: AN OVERVIEW

M. I. Mishchenko, L. D. Travis, and A. Macke

NASA Goddard Institute for Space Studies, 2880 Broadway, New York, NY 10025, USA

ABSTRACT

We briefly review several universal theoretical methods for computing scattering properties of nonspherical particles, discuss the implications of particle nonsphericity for remote sensing of mineral tropospheric aerosols and cirrus cloud particles, and examine the effect of particle shape on the aerosol and cloud radiative forcing of climate.

1. INTRODUCTION

Many laboratory and *in situ* measurements show that light scattering properties of small nonspherical particles can differ significantly from those of volume- or surface-equivalent spheres (e.g., Schuerman, 1980). This factor can have serious implications for remote sensing of nonspherical aerosol and cloud particles in the atmosphere and for computing the cloud and aerosol radiative forcing of the earth's climate. The last decade has been marked by major advances in our knowledge of nonspherical scattering (e.g., Hovenier, 1996), and the aim of this paper is to give a brief overview of recent accomplishments. First, we review several universal theoretical methods for computing nonspherical scattering and compare their advantages and respective ranges of applicability. Then we discuss light scattering properties of realistic size/shape distributions of nonspherical particles and their implications for remotely sensing nonspherical aerosols and cirrus cloud particles using radiometric, polarimetric, and lidar techniques. Finally, we examine the possible effect of particle nonsphericity on the aerosol and cloud radiative forcing of climate.

2. NUMERICAL METHODS

2.1. Separation of Variables Method (SVM) for Spheroids

SVM for homogeneous spheroids was first developed by Oguchi (1973) and Asano and Yamamoto (1975) and then improved by Farafonov (1983) (see also Voshchinnikov and Farafonov, 1993). The method was extended to core-mantle spheroids by Onaka (1980) and Cooray and Ciric (1992). Further development of SVM is underway (Stamnes and Spjelkavik, 1995). Practical applications of SVM are reported by de Haan (1987) and Stammes (1989). SVM solves the problem of light scattering by a spheroidal particle in the respective spheroidal coordinate system and is based on expanding both the incident and the scattered fields in spheroidal vector functions. Despite its name, SVM does not provide a complete separation of variables and, therefore, is not fully analytical. Furthermore, the computation of spheroidal functions is a difficult mathematical and numerical problem. These factors limit the current applicability of SVM to equivalent-sphere size parameters less than about 35. The method seems to be best suited to computing light scattering by spheroids with extreme aspect ratios ($\gtrsim$5). The clear limitation of the method is that it is applicable only to spheroidal particles. The main advantages of SVM are that it is exact (based on directly solving Maxwell's equations) and reasonably fast.

2.2. Discrete-Dipole Approximation (DDA)

This technique was first introduced by Purcell and Pennypacker (1973) and has been significantly improved and widely used during the last decade (see, e.g., Bohren and Singham, 1991; Lumme and Rahola, 1994; Evans and Stephens, 1995; Schneider and Stephens, 1995; and reviews by Draine and Flatau, 1994 and Hoekstra and Sloot, 1996). Lakhtakia (1992) has given a general derivation of DDA from the volume integral equation formulation and discussed its connection with the method of moments. DDA is based on partitioning a particle into a number N of elementary scattering units (dipoles). The electromagnetic response of a dipole to an incident field is assumed to be known. The field exciting a dipole is a superposition of the external field and the fields scattered by all other dipoles. This allows one to write a system of N linear equations for N fields exciting the N dipoles. The numerical solution of this system is used to compute N partial fields scattered by the dipoles and, thus, the total scattered field. The physical idea of DDA is very simple and transparent. However, practical computations show that the number of dipoles N should be very large (tens and hundreds of thousands) even for wavelength-sized scatterers, especially if elements of the scattering matrix need to be computed rather than just the optical cross sections (Okamoto *et al.*, 1995). This factor limits the maximum equivalent-sphere size parameter for DDA to about 15 even on supercomputers and makes DDA computations for randomly oriented and/or polydisperse particles very time-consuming. McClain and Ghoul (1986) and Singham *et al.* (1986) have developed analytical DDA procedures for computing scattering by randomly oriented particles based on re-expanding Cartesian tensor products in terms of spherical tensor products and exploiting analytical properties of Wigner D-functions. Unfortunately, this approach assumes time-consuming numerical matrix inversions and is applicable only to particles smaller than a wavelength. The definite advantage of DDA is that, in principle, it is applicable to any particle shapes. However, this still needs to be verified by comparing DDA computations with exact solutions based on solving Maxwell's equations (see, e.g., Hovenier *et al.*, 1996).

2.3. *T*-Matrix Method (TM)

TM is an exact method based on the assumption that both the incident and the scattered fields can be expanded in spherical vector functions. In this regard TM can be considered a generalization of the standard Mie theory. The T matrix transforms the expansion coefficients of the incident

IRS '96: Current Problems in Atmospheric Radiation
Smith and Stamnes (Eds.) ISBN 0-937194-39-5

field into those of the scattered field and, if known, can be used to compute any scattering characteristic of a nonspherical particle. TM was first introduced by Waterman (1971) for single homogeneous scatterers and was then generalized to multilayered scatterers and arbitrary clusters of nonspherical particles by Peterson and Ström (1973, 1974). In the case of clusters composed of spherical components, the *T*-matrix method is equivalent to the so-called multipolar expansion (or superposition) approach (Borghese *et al.*, 1984; Fuller and Kattawar, 1988; Mackowski, 1991; Tzeng and Fung, 1994; Chew *et al.*, 1994). An extension of TM to the case of internal aggregation (particles with inclusions) was developed by Borghese *et al.* (1994), Mackowski and Jones (1995), and Fuller (1995). TM and its extensive applications were recently reviewed by Mishchenko *et al.* (1996a).

The computation of the *T* matrix for a cluster assumes that the *T* matrices of all components are known and employs the translation addition theorem for spherical vector functions. The *T* matrix for single homogeneous and multilayered scatterers is usually computed using the extended boundary condition method (EBCM). In principle, EBCM can be applied to any particle shapes. However, essentially all available codes assume rotationally symmetric shapes both smooth, e.g., spheroids and so-called Chebyshev particles (Barber and Hill, 1990; Wiscombe and Mugnai, 1986; Mishchenko *et al.*, 1996a), and sharp edged, e.g., finite circular cylinders (Kuik *et al.*, 1994; Mishchenko *et al.*, 1996c). Special procedures have been developed to ensure the numerical stability of TM computations for large size parameters and/or extreme shapes such as spheroids with large aspect ratios (Mishchenko *et al.*, 1996a and references therein).

The main advantages of TM are that it is exact, very fast (can be several times faster than SVM and orders of magnitude faster than DDA), and applicable to particles with size parameters exceeding 80 (Mishchenko *et al.*, 1996a). The latter means that a combination of TM and the geometric optics approximation (GO, see below) can be used to cover the entire size parameter range from 0 to ∞ for rotationally symmetric particles. Moreover, Mishchenko (1991) has developed an analytical *T*-matrix procedure which makes computations for randomly oriented nonspherical particles as fast as those for a particle in a fixed orientation. This procedure even further reinforces the practical superiority of TM over SVM and DDA and makes TM an ideal technique for polydisperse calculations. Recently, Mackowski and Mishchenko (1996) have extended the analytical procedure to randomly oriented clusters of spheres. The high numerical accuracy of TM and its applicability to large size parameters were used to provide benchmark results (Hovenier *et al.*, 1996) and to check the accuracy of GO (Macke *et al.*, 1995).

2.4. Geometric Optics Approximation (GO)

A universal approximate method for computing nonspherical scattering is the so-called geometric optics (or ray-tracing) approximation. GO is based on the assumption that the scattering particle is much larger than the wavelength of the incident light and that the incident plane wave can be represented as a collection of independent parallel rays. The history of each ray impinging on the particle surface is traced using the Snell law and Fresnel's equations. The sampling of all escaping light rays into predefined narrow angular bins supplemented by the computation of diffraction of the incident wave on the particle projection yields a quantitative representation of the particle scattering properties. GO was first applied to randomly oriented finite hexagonal cylinders by Wendling *et al.* (1979) and since then has been widely used in scattering computations for ice cloud particles both homogeneous (e.g., Takano and Liou, 1995; Muinonen *et al.*, 1996; Macke *et al.*, 1996a; and references therein) and containing multiple internal inclusions (Macke *et al.*, 1996b).

The advantages of GO are that it is reasonably fast, thus enabling calculations for particle polydispersions, and can be applied to essentially any shape. However, GO is approximate by definition, and its range of applicability in terms of the smallest equivalent-sphere size parameter x_{min} needs to be checked by comparing GO results with exact numerical solutions of Maxwell's equations. Comparisons of GO and Mie results (Hansen and Travis, 1974) show that GO phase function calculations for spheres are reasonably accurate only for x_{min} exceeding several hundred. On the other hand, analogous comparisons of GO and *T*-matrix phase function results for randomly oriented spheroids and finite circular cylinders (Macke *et al.*, 1995) show that for moderately absorbing nonspherical particles x_{min} can be as small as 60. Unfortunately, decreasing absorption significantly increases x_{min} even for nonspherical particles. Furthermore, accurate GO calculations of polarization and depolarization require much larger x_{min} values than analogous GO calculations of the phase function (Hansen and Travis, 1974; Macke *et al.*, 1995).

Special care should be taken in the case of absorbing particles (imaginary part of the refractive index not equal to zero) since in this case the refracted wave becomes inhomogeneous so that the surface of constant amplitude does not coincide with the surface of constant phase. The Snell law can still be formally used, but needs to be modified as derived by Stratton (1941). The effect of this modification on scattering properties of absorbing ice crystals is discussed by Yang and Liou (1995) and Zhang and Xu (1995).

A modification of GO, the so-called physical optics or Kirchhoff approximation, was developed by Ravey and Mazeron (1982). This method implies the computation of the surface field using ray tracing and the computation of the corresponding far field via the vector Kirchhoff integral. The method was used, with some variations, by Muinonen (1989), Arnott and Marston (1991), and Yang and Liou (1995) and was found to be rather time-consuming. Since this technique is still an approximation, it remains to be demonstrated whether it can be used in computations for significantly smaller size parameters than the original ray-tracing approximation, especially when the full scattering matrix, including polarization and depolarization, is computed.

2.5. Other Methods

Two other exact methods that have become rather popular are the so-called Fredholm integral equation method (Holt, 1982) and the finite difference time domain method (Kunz and Luebbers, 1993). Information on other available methods, both exact and approximate, can be found in Appendix A of Wiscombe and Mugnai (1986) and in two special journal issues edited by Barber *et al.* (1994) and Hovenier (1996). Hovenier and van der Mee (1996) discuss general theoretical methods for checking the physical relevance of calculations for any particles irrespective of the computational technique used.

2.6. Summary

The main characteristics of the SVM, DDA, TM, and GO are summarized in Table 1. (Note that the bold font is used to emphasize definite advantages of the methods, whereas the use of the italic font emphasizes definite disadvantages.) The table suggests that no single method can be used to cover the entire range of size parameters from 0 to ∞. However, this can be done for rotationally symmetric particles by combining TM ($x \lesssim 85$) and GO ($x \gtrsim 60$). Currently, only DDA and GO can be used in scattering computations for arbitrarily shaped particles. Unfortunately, their size parameter ranges do not overlap, and DDA is very difficult to apply to realistic size and shape distributions of randomly oriented particles.

3. Single-Scattering Properties of Natural Nonspherical Particles

The potential ability to compute scattering by a specific nonspherical shape does not necessarily mean the ability to theoretically reproduce scattering properties of natural ensembles of nonspherical particles. For example, mineral tropospheric aerosols occur in such a broad variety of shapes (e.g., Jaggard *et al.*, 1981; Okada *et al.*, 1987) that it is essentially impossible to develop a simple shape classification. The only definite facts about such particles are that they are distinctly nonspherical, that all particles are different so that there are no two identical particles, and that the particle aspect ratio (ratio of the largest to the smallest particle dimensions) varies within some finite range. An additional problem with mineral tropospheric aerosols is that they are randomly oriented and distributed over sizes, the range of equivalent-sphere size parameters for visible light extending from zero to several tens. It appears that the only feasible way of computing scattering properties of such natural particles is to use a statistical approach coupled with TM, i.e., to perform computations for a representative size and shape mixture of relatively simple nonspherical particles (e.g., spheroids with widely varying aspect ratios) in random orientation. The main assumption of the statistical approach is that the ensemble of particles chosen for the purposes of averaging need not be in one-to-one correspondence with the ensemble of particles of interest (Bohren and Singham, 1991).

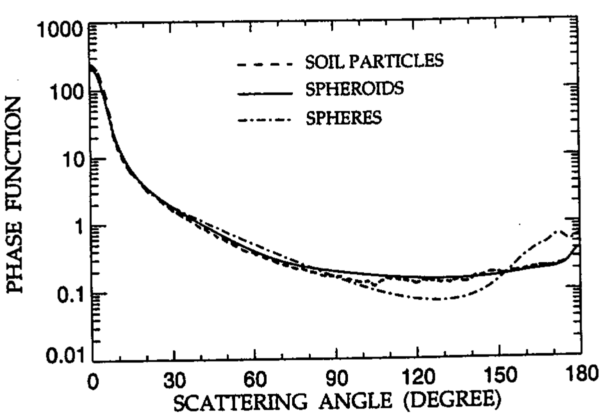

Fig. 1. Comparison of phase functions measured for soil particles and computed for a shape distribution of polydisperse, randomly oriented spheroids and surface-equivalent spheres.

Mishchenko *et al.* (1996d) (see also Hill *et al.*, 1984) have examined the usefulness of the statistical approach by comparing the phase function measured in the visible for micron-sized soil particles (Jaggard *et al.*, 1981) with that computed for a broad shape mixture of polydisperse, randomly oriented prolate and oblate spheroids (Fig. 1). This comparison clearly demonstrates the applicability of the statistical approach to natural dust-like particles. It also shows that phase function differences between mineral aerosols and surface-equivalent spheres are significant, especially at side- and back-scattering angles, and can exceed a factor of 2 (*cf.* Koepke and Hess, 1988; Nakajima *et al.*, 1988; West *et al.*, 1996). Despite the large phase function differences, spherical-nonspherical differences in the optical cross sections, single-scattering albedo, asymmetry parameter, and backscattered fraction are essentially negligible (Fig. 2; *cf.* Mugnai and Wiscombe, 1986; Mishchenko *et al.*, 1996c). On the other hand, Fig. 2 shows that the differences in the extinction-to-backscatter ratio (EBR) can well exceed 100% (*cf.* Waggoner *et al.*, 1972). Extensive *T*-matrix calculations for randomly oriented polydisperse spheroids (Mishchenko *et al.*, 1996a) and cylinders (Mishchenko *et al.*, 1996c) show that spherical-nonspherical differences in all elements of the scattering matrix can be very large. In particular, nonsphericity not only changes the magnitude of linear polarization, but can even reverse its sign.

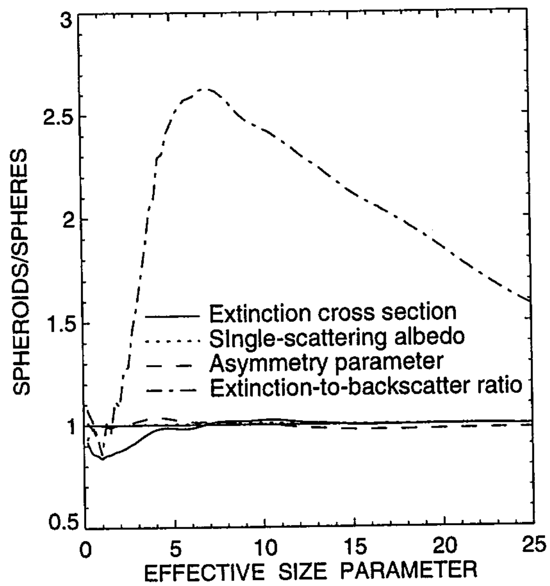

Fig. 2. Ratios of single-scattering characteristics of a shape mixture of polydisperse spheroids to those of surface-equivalent spheres. The refractive index is 1.5 + 0.008*i*.

Another radiatively important type of nonspherical particles in the atmosphere are cirrus ice crystals. In some cases, cirrus clouds exhibit remarkable optical phenomena such as halos, thus indicating that ice crystals have highly regular shapes such as single or aggregated hexagonal columns and plates (Wendling *et al.*, 1979; Takano and Liou, 1989, 1995; Macke, 1993). However, for many if not for most cirrus clouds the halos are not seen even under suitable observation geometries (Minnis *et al.*, 1993b; Sassen *et al.*, 1994), and the ice particle phase function turns out to be rather featureless (e.g., Francis, 1995; Gayet *et al.*, 1995; Posse and von Hoyningen-Huene, 1995; Spinhirne *et al.*, 1996). One way of modeling a featureless phase function is to assume that ice particles lack the perfect hexagonal structure and occur in a wide variety of shapes and to use the statistical approach outlined above. Another way is to model scattering properties of a random ensemble of different shapes using a single but randomly shaped particle and employing GO (Peltoniemi and Lumme, 1989; Muinonen *et al.*, 1996; Macke *et al.*, 1996a). Figure 3 compares phase functions computed, at a wavelength of 0.65 μm, for the ISCCP (International Satellite Cloud Climatology Project) water droplet model (Rossow *et al.*, 1996), regular hexagonal ice columns, and randomly shaped ice particles. The randomly shaped ice particles are modeled as a randomized triadic Koch fractal (Macke *et al.*, 1996a). It is seen that the random-particle phase function is relatively featureless and does not show halos specific of hexagonal crystals and pronounced rainbow and glory features exhibited by spherical water droplets. Interestingly, the phase function computed for the random-fractal particle essentially coincides with that computed by J. Peltoniemi (private communication, 1995) using a different generator of a randomly shaped particle (Peltoniemi and Lumme, 1989). This may suggest that these phase functions indeed capture the scattering character of highly variable and irregular natural ice crystals. It should be noted, however, that the pronounced surface roughness of the theoretically generated randomly shaped particles causes an asymmetry parameter ($g = 0.752$) which is significantly smaller than that for simpler particle shapes such as hexagonal or circular cylinders and spheroids. Therefore, if it turns out that the typical asymmetry parameter value for cirrus particles is about 0.8 or larger and the phase function is still relatively featureless and has no halos, then a possible way to model such phase function is to use the statistical approach (Mishchenko *et al.*, 1996b).

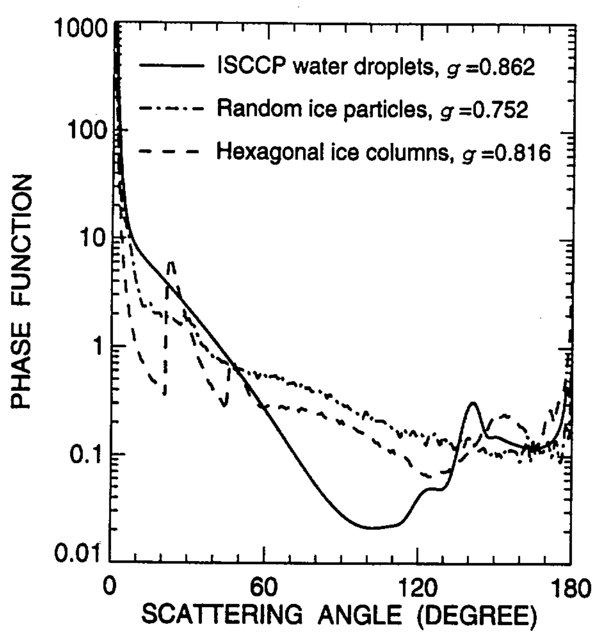

Fig. 3. Phase functions and asymmetry parameters for the ISCCP water droplet model, hexagonal ice crystals, and random ice particles (adapted from Mishchenko *et al.*, 1996b).

Two scattering characteristics which are traditionally considered the most sensitive indicators of particle nonsphericity are the linear (LDR) and circular (CDR) depolarization ratios (Stephens, 1994). Both ratios vanish for perfect spheres, but are nonzero for nonspherical particles. Nonzero LDR and CDR for large ice crystals are usually explained by the depolarizing effect of multiple internal reflections (Liou and Takano, 1994). However, the GO concepts of rays, reflections, and refractions are not applicable to wavelength- and sub-wavelength-sized particles which can also be strong depolarizers (e.g., Asano and Sato, 1980; Mishchenko *et al.*, 1996a,c). Moreover, GO fails to explain the complicated size-parameter and shape dependence of LDR and CDR as demonstrated by exact computations of Mishchenko and Hovenier (1995). The computations also show that although depolarization is an indicator of particle nonsphericity, it can not be considered a universal measure of the degree of nonsphericity. Further theoretical and experimental work in this area is required.

4. REMOTE SENSING AND CLIMATE MODELING

Particle nonsphericity can affect essentially any remote sensing retrievals of aerosols and clouds. For example, large spherical-nonspherical phase function differences (Figs. 1 and 3) can result in an underestimation or an overestimation of the optical thickness if satellite reflectance measurements for cirrus clouds or dust-like tropospheric aerosols are analyzed using Mie theory (Minnis *et al.*, 1993a; Mishchenko *et al.*, 1995, 1996b; Kahn *et al.*, 1996). This factor has already been taken into account in the revised ISCCP algorithm (Rossow *et al.*, 1996; Mishchenko *et al.*, 1996b). EBR is a quantity directly entering the lidar equation (Reagan *et al.*, 1989). Therefore, large spherical-nonspherical differences in EBR (Fig. 2) can make irrelevant lidar retrievals of the optical thickness for nonspherical cloud and aerosol particles based on Mie theory. The necessity of taking into account the shape of ice crystals in polarimetric remote sensing of cirrus clouds has been demonstrated by Coffeen and Hansen (1972) and Masuda and Takashima (1992). The lidar and radar depolarization techniques (Toon *et al.*, 1990; Sassen, 1991; Eberhard, 1992; Vivekanandan *et al.*, 1994; Stefanutti *et al.*, 1995; Tang and Aydin, 1995, and references therein) explicitly use the fact that single-scattering LDR and CDR can have nonzero values only for nonspherical particles.

Despite the strong effect of nonsphericity on aerosol remote sensing retrievals, the effect of particle shape on the direct aerosol forcing of climate is rather weak (Lacis and Mishchenko, 1995). This means that if the optical thickness of nonspherical aerosols is already known, then the aerosol radiative forcing can be computed with high accuracy using the model of surface-equivalent spheres. This result can be explained by small spherical-nonspherical differences in the aerosol single-scattering albedo and asymmetry parameter (Fig. 2). It is important to emphasize, however, that no cancellation of errors occurs if one consistently uses Mie theory in retrieving the aerosol optical thickness and then in computing the aerosol radiative forcing for the retrieved optical thickness value (Mishchenko *et al.*, 1995).

The effect of nonsphericity on the cirrus cloud radiative forcing can be much stronger (e.g., Kinne and Liou, 1989) due to significantly larger spherical-nonspherical differences in the single-scattering characteristics. Figure 4 (adapted from Mishchenko *et al.*, 1996b) shows that spherical albedo of a liquid-water cloud at solar wavelengths can be significantly smaller than that of an optical-thickness equivalent ice cloud composed of hexagonal crystals and much smaller than that of an ice cloud composed of randomly shaped crystals. This result is unambiguously explained by the fact that the asymmetry parameter for the randomly shaped crystals is smaller than that for the hexagonal columns and much smaller than that for the ISCCP water droplets (Fig. 3) and further emphasizes the importance of exact knowledge of the asymmetry parameter for real cirrus cloud particles (Stephens *et al.*, 1990). The potentially strong effect of nonsphericity on the cirrus cloud radiative forcing makes important accurate parameterizations of ice particle scattering properties. The parameterizations by Fu and Liou (1993) and Sun and Shine (1995) are primarily based on the regular hexagonal crystal model, whereas Mitchell *et al.* (1996) use both hexagonal and random-fractal shapes.

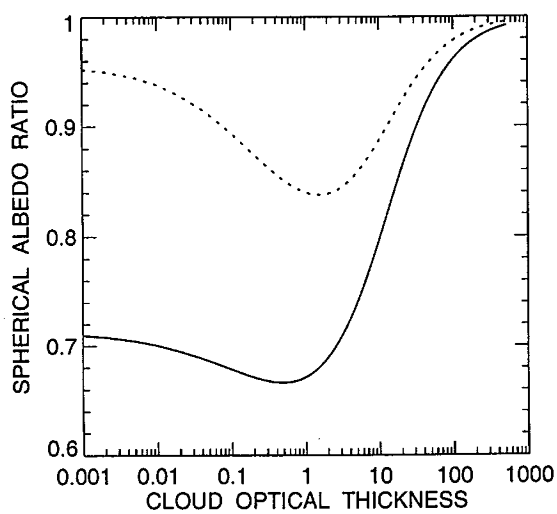

Fig. 4. Spherical albedo of a liquid water cloud relative to that of an optical-thickness-equivalent ice cloud composed of randomly shaped particles (———) and hexagonal columns (·······). The wavelength is 0.65 μm.

ACKNOWLEDGMENTS

We thank J. Lenoble and P. Varanasi for inviting us to give an overview talk on nonspherical scattering at the IRS'96 and J. W. Hovenier for useful comments. This research was funded by the NASA EOS Project managed by Goddard Space Flight Center and by the NASA FIRE III program.

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Table 1. Comparison of different theoretical methods for computing nonspherical scattering.

Method	Accuracy	Shapes	Equivalent-sphere size parameters	Efficiency
SVM	**Exact**	*Spheroids*	$\lesssim 35$	Fast
DDA	Accurate in the limit $N \to \infty$	**Arbitrary**	$\lesssim$ *15*	*Slow*
TM	**Exact**	Rotationally symmetric particles, clusters of spheres	$\lesssim$ ~~85~~ 125	**Very fast, especially for randomly oriented particles**
GO	*Approximate*	**Arbitrary**	$\gtrsim 60$	Moderately fast