Optical Properties of Airborne Dust: The Effect of Particle's Nonsphericity on the Single-Scattering Properties and Downstream Applications

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A major component of atmospheric aerosols is airborne dust that may have significant opacity (particularly over arid and semi-arid areas) and can thus have a substantial impact on atmospheric radiation budget and climate



Asian Dust, MODIS RGB (0.65µm, 0.55µm, 0.47µm) Image



The track of a large dust storm that was followed with the SeaWIFS Satellite observations in 1998



Source: R. B. Husar et al, JGR, 2001

- The important role of mineral dust aerosols in the climate system has been recognized (Haywood & Bougher, 2000).
- Modeling the effect of dust aerosols on the transfer of radiation in the atmosphere contributes to our understanding of the climate system (d'Almeida et al, 1991) and aerosol retrieval algorithms (Mishchenko et al. 1995, Kahn et al. 1997).
- Determination of the sign and magnitude of direct radiative forcing by dust on regional and global scales remains a key unsolved problem (e.g. Sokolik et al., 2001).
- The optical properties of dust particles are fundamental to the assessment of the radiative forcing of dust aerosols. There is a pressing need to reduce the substantial uncertainties in the current knowledge of the optical properties of dust particles.

Scattering Geometry



Stokes vector-Phase matrix/Mueller matrix formulation

The electric field can be resolved into components. $E_{//}$ and E_{\perp} are complex oscillatory functions.

The four component Stokes vector (Stokes, 1852) can be defined.

They are all real numbers and satisfy the relation

 $I^2 = Q^2 + U^2 + V^2$

The phase matrix, P, (for a singlescattering event) or Mueller matrix, M, (for a multiple scattering event, or radiative transfer process) relates the incident and scattered Stokes vectors $\mathbf{E} = \mathbf{E}_{/\!/} \, \mathbf{l} + \mathbf{E}_{\perp} \, \mathbf{r}$

 $I = E_{//} E_{//}^{*} + E_{\perp} E_{\perp}^{*}$ $Q = E_{//} E_{//}^{*} - E_{\perp} E_{\perp}^{*}$ $U = E_{//} E_{\perp}^{*} + E_{\perp} E_{//}^{*}$ $V = i(E_{//} E_{\perp}^{*} - E_{\perp} E_{//}^{*})$

$$\begin{bmatrix} \mathbf{I}^{\mathbf{s}} \\ \mathbf{Q}^{\mathbf{s}} \\ \mathbf{U}^{\mathbf{s}} \\ \mathbf{V}^{\mathbf{s}} \end{bmatrix} = \begin{pmatrix} M_{11} & M_{12} & M_{13} & M_{14} \\ M_{21} & M_{22} & M_{23} & M_{24} \\ M_{31} & M_{32} & M_{33} & M_{34} \\ M_{41} & M_{42} & M_{43} & M_{44} \end{pmatrix} \begin{pmatrix} \mathbf{I}i \\ \mathbf{Q}^{i} \\ \mathbf{U}^{i} \\ \mathbf{V}^{i} \end{pmatrix}$$





Reid et al., 2003

Volten et al., 2005

The Finite-Difference Time Domain (FDTD) Method

Basic principle: Discretizatio of two time-dependent Maxwell's curl equations in terms of the finite-difference technique



Discrete Dipole Approximation (DDA) method Purcell E. M., and C. R. Pennypacker, Scattering and absorption of light by nonspherical dielectric grains, Astrophys. J. 186, 705–714, 1973.





Edward Mills Purcell Nobel Prize in Physics (1952)



Diagram: Z. Zhang

Improved Geometric Optics Method

Yang and Liou (1996)



Nonsphericity Effect of Dust Particles

- "...even moderate nonsphericity results in substantial errors in the retrieved aerosol optical thickness if satellite reflectance measurements are analyzed using Mie theory" (Mishchenko et al. 1995).
- "Multiple, multispectral remote sensing observations, such as those anticipated from the Earth Observing System (EOS) multiangle imaging spectroradiometer (MISR), can distiguish spherical and nonspherical particles over calm ocean for mineral-dust-like particles" (Kahn et al. 1997).
- Demonstrated of a significant impact of the nonsphericity of dust particles on the retrieval of dust microphysical and optical properties (Dubovik et al. 2002, 2006)



Comparison between the phase functions computed for spherical and nonspherical dust particles (Feng, Yang, Kattawar, Hsu, Tsay and Laszlo, 2009). The symbols indicate laboratory measurements (Volten et al. 2001).



Elements of the kernel matrices $K_{11}(\Theta, \lambda, n, k, p, rk)$ and $K_{12}(\Theta, \lambda, n, k, p, rk)$ at different scattering angles for prolate randomly oriented spheroids ($\varepsilon = \sim 2.7$, n = 1.53 + i0.003). The red and blue curves show the results obtained with the *T*-matrix code [*Mishchenko and Travis*, 1994] and the *Yang and Liou* [1996] method, respectively. *Adapted from Dubovik et al.* (2006),



A typical situation where utilizing the spheroid scattering assumption [*Dubovik et al.*, 2002b] resulted in the removal of the false fine mode in the size distribution and false spectral dependence in the real part of refractive index. Size distributions and refractive indices are retrieved assuming sphere and spheroid models from spectral radiance measurements covering the full range of scattering angles. The size distribution retrieved from the aureole only ($\Theta < 40^\circ$, where effects of nonsphericity are minimal) and assuming spherical particles is also shown. *Adapted from Dubovik et al.* (2006).

Simulated solar reflectance at the top of a dusty atmosphere. Spherical and nonspherical (spheroidal) shapes are assumed for dust particles (Yang, Feng, Hong, Kattawar, Wiscombe, Mishchenko, Dubovik, Laszlo, and Sokolik, 2007).



• These results indicate that the equivalent sphere approximation leads to an underestimate of the albedo of a dusty atmosphere. This underestimate has an important implication to the study of the effect of airborne dust on the radiation budget within the atmosphere.



MODIS RGB image on March 2, 2003, showing a dust plume over West Africa. The area indicated by the small red box is used to retrieve dust AOD in the present sensitivity study (Feng, Yang, Kattawar, Hsu, Tsay and Laszlo, 2009).

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Upper panels: the retrieved dust AOD based on the nonspherical (spheroidal) and sphere models.

Lower left panel:

retrieved dust AOD based on the sphere model versus those based on the nonspherical model.

Lower right panel:

the relative differences of the retrieved AOD (Feng, Yang, Kattawar, Hsu, Tsay and Laszlo, 2009). 19

MODIS RGB image (05/27/2008) 100 104 108 112 116 120 124 128 132



Retrieved dust optical depths



Does the particle shape have a large effect on radiative flux simulation?

- Pilinis and Li (1998) compared the model calculated single-scattering properties with observations, and claimed that aerosol forcing could be underestimated by a factor of 3 if particle shape effect was neglected.
- Several studies (Kahnert & Kylling, 2004; Kahnert et al, 2005; Kahnert et al, 2007) investigated the particle shape effect on the simulation of radiance and flux simulation and compared the uncertainties associated with dust nonsphericity with the other important uncertainties.
- From modeling simulations, Kahnert & Kylling (2004) found that the misrepresentation of the aerosol phase function by using the spherical counterpart caused an overestimation of yearly averaged TOA spectral net flux, which was five times larger than the errors due to uncertainties in refractive index.
- Kahnert et al (2005) found that an optimal shape distribution of spheroids could significantly reduce the errors compared with the equiprobable shape distribution.
- The errors due to the spherical particle approximation are found to be the same order as the errors associated with the uncertainties in the refractive indices (Kahnert et al, 2007)

Does the particle shape have a large effect on radiative flux simulation?

• No?

- Despite the apparent differences in the phase function, Mishchenko et al. (1995) argued that the sphere model could be a suitable approximation for nonspherical dust in radiative flux simulation, because the single-scattering albedo and asymmetry factor were similar in the two cases.
- Fu et al (2009) found that the errors in reflectivity and absorptivity resulted from the spherical particle approximation were small and concluded that Mie-theory-based single-scattering properties were suitable for radiative flux calculations.
- The controversy has not been resolved!
- The particle shape effect on broadband radiative flux simulation is not yet determined. 22

Integrated net flux of dust forcing at the top of the atmosphere (TOA) and the surface using the spherical and nonspherical models under various conditions

(Solar spectral band: 0.3μ m- 4.5μ m)

Conditions		Integrated Dust Forcing (W/m ²)			
Surface type	Optical depth	TOA		Surface	
		Nonspherical	Difference	Nonspherical	Difference
Sandy	0.5	-1.1	6.1	-71.0	6.3
Sandy	1.0	-3.5	10.9	-136.6	11.0
Water	0.5	-26.7	8.7	-90.1	9.1
Water	1.0	-48.2	14.7	-170.9	15.4

Difference = the nonspherical results minus the spherical counterparts

New Effort

• Objective

Use <u>manageable and optimal particle</u> morphological sets (or, the geometric parameters associated with particle shapes and compositions) to quantify the effect of the nonsphericity of dust particles on their optical and radiative properties from remote sensing and radiative forcing perspectives.

• Particle shapes

Tri-axial ellipsoid (Bi, Yang, Kattawar, and Kahn, 2008) Non-symmetric hexahedra (Bi, Yang, Kattawar, and Kahn, 2010)









Ellipsoids

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- For size parameters from 0.5 to 30, we used the ADDA code of Yurkin and Hoekstra (2007) to calculate the relevant quantities.
- ▶ For size parameters from 15 to 1000, we use the IGOM with the edge effect included.
- We use a numerical method to calculate the edge effect accurately for ellipsoids.



Randomly oriented ellipsoids: m=1.53+0.008i, λ=0.66 μm (Bi, Yang, Kattawar, and Kahn, 2009)

HEXAHEDRA, PHASE MATRIX ELEMENTS (Bi, Yang, Kattawar, and Kahn, 2010)



Polluted Dust Particles

Some dust particles, after passing through high-density industrial regions or oceans, may be coated with soot or seasalt (Parungo et al. 1995).

(a) (b) Some new efforts (e.g., **Building Blocks Closed Cell Open Cell** Liou, Takano and (C) (d) Yang, 2011) Closed Closed **Ballistic Aggregate Diffusion Limited Aggregate** Open Open 27

Diffusion Limited Aggregate

Observed and Modeled Soot

A Novel Geometric-optics Surface-wave Approach (Liou, Takano and Yang, 2011)



Experimental Setup

(a) The experimental setup



Yong-Le Pan, Kevin B. Aptowicz, Richard K. Chang, Matt Hart, and Jay D. Eversole, *Characterizing and monitoring respiratory aerosols by light scattering,* Optics Letters, Vol. 28, No. 8, p589 (2003).



Experimental and Monte Carlo simulated diffusely backscattered Mueller matrix for a $2.02\mu m$ polystyrene sphere suspension. The approximate size of each image is 1.6 cm^2

B. D. Cameron, M. J. Rakovic, M. Mehrubeoglu, G. Kattawar, S. Rastegar, L. V. Wang, and G. L. Coté, "Measurement and calculation of the two-dimensional backscattering Mueller matrix of a turbid medium," Optics Letters 23, 485-487 (1998).

Mueller Image for Three Particles



Available Laboratory Measurements The Amsterdam Light Scattering Database (http://www.iaa.es/scattering/)

- Samples: feldspar, red clay, quartz, Pinatubo, Sahara...
- Wavelengths: 632.8 nm and 441.6 nm
- Scanning microscope images of the particles
- Phase matrix: scattering angles from 5° to 173°
- Particle size distribution

Aerosol Generator



Experimental setup at the University of Amsterdam. http://www.astro.uva.nl/scatter

Summary

- Nonsphericity effect of dust aerosol particles is quite important from remote sensing perspective.
- The effect of the nonsphericity of dust particles on simulating radiative flux (i.e., radiation budget) is still controversial.
- Efforts have been made to develop dust optical properties using numerical models. Follow-up effort is needed to intercompare the theoretical simulations.
- Laboratory data for the single-scattering properties of dust particles can constrain developing optimal and realistic models (overall shape, surface texture, and inhomogeneity) for these particles.
- Measurements of backscatter are extremely useful for modelers.