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# Scattering and Radiative Properties of Internal Versus External Mixtures of Different Aerosol Types

Michael I. Mishchenko and Larry D. Travis

NASA Goddard Institute for Space Studies  
2880 Broadway, New York, NY 10025  
E-mail: crmim@giss.nasa.gov and pldldt@giss.nasa.gov

## ABSTRACT

An advanced theoretical technique is used to compute the scattering of light by internal aerosol mixtures in the form of polydisperse, randomly oriented two-particle aggregates. These computations are compared with those for composition-equivalent external mixtures, and the effect of aggregation on the scattering and radiative properties of aerosols is discussed.

## INTRODUCTION

Different kinds of aerosols (e.g., dust, carbonaceous, and sulfate particles) can be suspended in air both in the form of external mixtures (different aerosol particles are widely separated) and internal mixtures (two or more aerosol particles are in physical contact and form an aggregate). Since aerosol particles forming an aggregate are in the near-field zone of each other, the scattering and radiative properties of internal aerosol mixtures can differ from those of composition-equivalent external mixtures. These potential differences may influence the results of remote sensing studies of aerosols and calculations of the direct aerosol forcing of climate and, therefore, must be accurately evaluated using theoretical calculations and/or laboratory measurements (Chylek *et al.*, 1995; Fuller, 1995).

In this paper we calculate the optical cross sections and the elements of the Stokes scattering matrix of internal two-component mixtures of carbonaceous, dust, and sulfate aerosols and compare them with those of equivalent external mixtures computed with the standard Mie theory. This comparison is used to derive conclusions about the effect of aggregation on the scattering and radiative properties of multi-component aerosols.

## COMPUTATIONS

The single scattering of light by a small-volume element consisting of randomly oriented particles is completely specified by the cross sections for scattering,  $C_{sca}$ , and extinction,  $C_{ext}$ , and the elements of the normalized scattering matrix  $F(\Theta)$ , where  $\Theta$  is the scattering angle (van de Hulst, 1957). In the standard  $\{I, Q, U, V\}$  representation of polarization, the scattering matrix has the well-known block-diagonal form

$$F(\Theta) = \begin{bmatrix} F_{11}(\Theta) & F_{21}(\Theta) & 0 & 0 \\ F_{21}(\Theta) & F_{22}(\Theta) & 0 & 0 \\ 0 & 0 & F_{33}(\Theta) & F_{34}(\Theta) \\ 0 & 0 & -F_{34}(\Theta) & F_{44}(\Theta) \end{bmatrix}, \quad (1)$$

so that only eight elements of  $F$  are nonzero and only six of them are independent. The (1, 1) element

of the scattering matrix is called the phase function and satisfies the normalization condition

$$\frac{1}{4\pi} \int_{4\pi} d\Omega F_{11}(\Theta) = 1. \quad (2)$$

Additional important quantities are the absorption cross section,  $C_{\text{abs}} = C_{\text{ext}} - C_{\text{sca}}$ , the single-scattering albedo,  $w = C_{\text{sca}}/C_{\text{ext}}$ , and the asymmetry parameter of the phase function defined as

$$g = \frac{1}{2} \int_{-1}^1 d(\cos\Theta) \cos\Theta F_{11}(\Theta). \quad (3)$$

Our computations employ a highly efficient exact technique developed recently by Mishchenko and Mackowski (1994) and based on the T-matrix solution of Maxwell's equations. The method rigorously computes the scattering of light by randomly oriented two-sphere clusters with sizes comparable to and larger than the wavelength. The efficiency of the method is the result of combining the power of the superposition approach in treating light scattering by composite particles (Fuller, 1991; Mackowski, 1994) and the analyticity of the T-matrix formulation in application to randomly oriented nonspherical scatterers (Mishchenko, 1991). The main idea of the method is to employ the superposition approach to calculate the T-matrix of a bisphere in the natural coordinate system with the z-axis connecting the centers of the component spheres and then to use this T-matrix in an analytical procedure to directly compute the optical cross sections and the elements of the scattering matrix for randomly oriented bispheres. We have found that the analytical averaging over orientations makes our method much faster than that based on the standard numerical averaging and, thus, suitable for computations for realistic size distributions.

The computations reported here were performed for two polydisperse aerosol mixtures at a visible wavelength of  $\lambda = 0.628 \mu\text{m}$  and assuming the power law size distribution of Hansen and Travis (1974). Mixture #1 was composed of dust-like particles with an effective radius (as defined by Hansen and Travis, 1974) of  $r_{\text{eff}} = 1 \mu\text{m}$  and an equal number of sulfate aerosols with  $r_{\text{eff}} = 0.6 \mu\text{m}$ . Mixture #2 consisted of equal numbers of sulfate particles with  $r_{\text{eff}} = 0.6 \mu\text{m}$  and soot aerosols with  $r_{\text{eff}} = 0.3 \mu\text{m}$ . We used the following refractive indices:  $1.53 + 0.008i$  for dust,  $1.44$  for sulfates, and  $1.75 + 0.435i$  for soot (cf. d'Almeida *et al.*, 1990). The effective variance for all aerosol types was  $v_{\text{eff}} = 0.2$ . In computations for internally mixed aerosols (polydisperse two-sphere aggregates), we assumed that the ratio of the radii of the particles forming an aggregate was constant and equal to  $1/0.6$  for the dust-sulfate mixture and  $0.6/0.3$  for the sulfate-soot mixture. The results of the computations are shown in Figs. 1 and 2 and Tables 1 and 2 and are discussed in the next section.

## DISCUSSION AND CONCLUSIONS

Figures 1 and 2 show that the differences in the phase function for internal versus external mixing are relatively small and, on average, do not exceed 10%. The only exception is the exact forward-scattering direction, where the interference effects result in a significant enhancement of the aggregate phase functions (Mishchenko *et al.*, 1995). The differences in the other elements of the scattering matrix, including the degree of linear polarization  $-F_{12}/F_{11}$ , are also surprisingly small, especially for mixture #2. The only clear indications of the fact that two-sphere aggregates are nonspherical particles is the deviation of the ratio  $F_{22}(\Theta)/F_{11}(\Theta)$  from 1 and the deviation of the ratios  $F_{33}(180^\circ)/F_{11}(180^\circ)$  and  $F_{44}(180^\circ)/F_{44}(180^\circ)$  from  $-1$  (cf. van de Hulst, 1957; Mishchenko *et al.*, 1995).

Tables 1 and 2 demonstrate that the differences in the integral scattering characteristics ( $C_{\text{ext}}$ ,  $C_{\text{sca}}$ ,  $C_{\text{abs}}$ ,  $w$ , and  $g$ ) between internal and composition-equivalent external mixtures are even smaller, particularly for mixture #2. The differences are especially small for the single-scattering albedo and the asymmetry parameter of the phase function. The latter fact combined with the small phase function and

polarization differences suggest that aggregation can have a relatively weak effect on remote sensing retrievals of the aerosol physical characteristics and aerosol direct radiative forcing. This seems to be especially true when one of the aerosol components is the strongly absorbing soot. More extensive computations are required, however, to further substantiate this conclusion.

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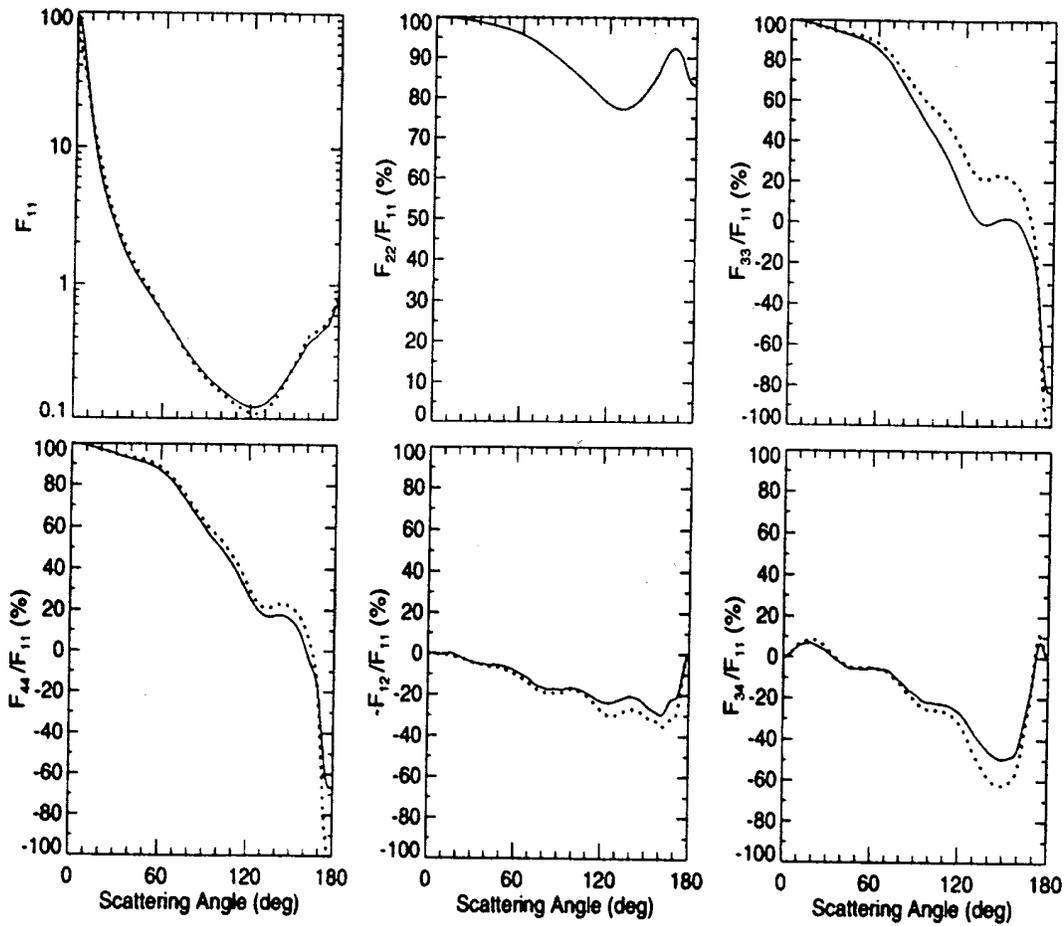


Figure 1. Scattering matrix elements for mixture #1 with internally (solid curves) and externally (dotted curves) mixed components.

Table 1. Integral scattering characteristics for mixture #1.

	Internal Mixing	External Mixing
$C_{\text{ext}} (\mu\text{m}^2)$	5.785	6.408
$C_{\text{sca}} (\mu\text{m}^2)$	5.191	5.821
$C_{\text{abs}} (\mu\text{m}^2)$	0.594	0.587
$w$	0.897	0.908
$g$	0.719	0.717

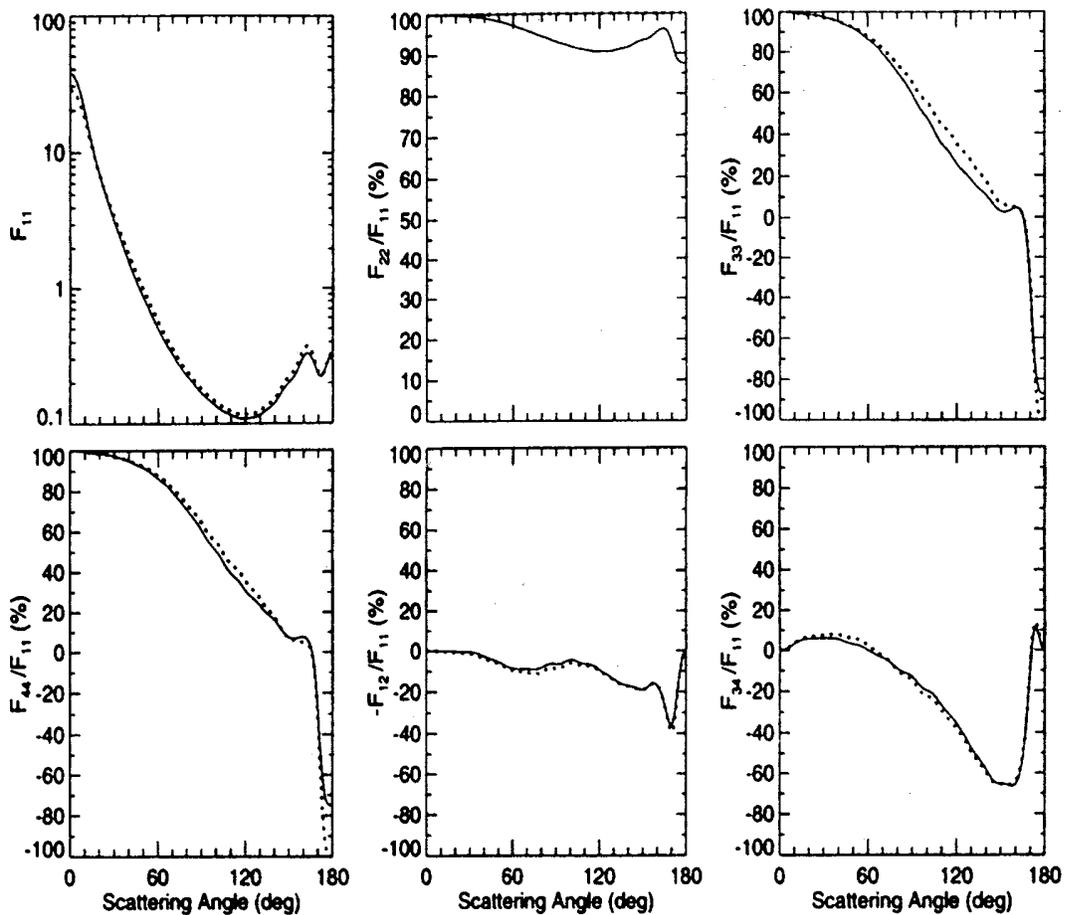


Figure 2. Scattering matrix elements for mixture #2 with internally (solid curves) and externally (dotted curves) mixed components.

Table 2. Integral scattering characteristics for mixture #2.

	Internal Mixing	External Mixing
$C_{\text{ext}} (\mu\text{m}^2)$	2.140	2.295
$C_{\text{sca}} (\mu\text{m}^2)$	1.901	2.070
$C_{\text{abs}} (\mu\text{m}^2)$	0.239	0.225
$w$	0.891	0.902
$g$	0.749	0.726