# Optical characteristics of composite ellipsoidal solid-phase aerosols with variable carbon content

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**Abstract**

The optical behavior of three-component carbonaceous ellipsoidal aerosols is analyzed. Compared to compact ellipsoids, the fragmented particles show enhanced backscatter and linear polarization. Employment of EMTs may lead to overestimation of both the asymmetry parameter and efficiency factor for absorption, but bulk $Q_{ext}$ is only slightly influenced. Typically the compact particles, whose absorbing component has the largest air-carbon interface, absorb less efficiently than particles with random fragmentation.

## 1 Introduction

Fine submicron-sized aerosols can occur in both the solid phase and the liquid phase. Such particles are too small to settle rapidly or to be washed out by rain, but they are still sufficiently large to avoid coagulation processes. Thus their survival time in the atmosphere is quite long. Solid-phase aerosols are almost typically non-spherical and are rarely homogeneous. While the prevailing constituents of aerosol particles can be constantly identified in some territories, the internal structure of such particles is a notoriously unknown quantity and depends on many factors participating in processes of particle formation. The optical response of the particles (having known size, shape, orientation and composition) to the incident electromagnetic radiation is still uncertain due to the variety of possible internal mixing of individual materials.

Basically, it is impossible to describe the realistic shapes of ambient aerosols. Instead, it is more convenient to characterize the prevailing morphology by means of aspect ratio $\varepsilon$ that relates the largest and smallest characteristic sizes of arbitrarily shaped particle. One of the easiest ways to simulate non-spherical particles having different aspect ratios is to employ mathematically well-defined geometries, like ellipsoids. Then, the ratio of the major semiaxis $a$ to the minor semiaxis $b$ (i.e. the aspect ratio) may vary from $a/b \approx 1$ (for nearly spherical particles) to $a/b >> 1$ (for needle-like particles) or $a/b << 1$ (for disk-like particles) [1].

We present results of numerical simulations of the optical behavior of carbonaceous non-spherical particles with random as well as non-random internal mixing of individual materials. The numerical study is based on the discrete dipole approximation (DDA). At present, many excellent numerical tools are available to calculate optical properties of non-spherical particles, yet there is still a lack of methods applicable for composite particles. Therefore a set of effective medium theories (EMTs) was developed to overcome computational difficulties (typical e.g. for DDA). However, the correctness of EMTs is questionable and their usage may result in errors.

We perform numerical light-scattering simulations for ellipsoidal aerosol particles composed of ammonium sulfate, organic matter and black carbon. Organic matter typically influences the atmospheric radiation through both scattering and absorption. Elemental carbon includes strongly light-absorbing material and is thought to yield large positive radiative forcing. Ammonium sulfates represent a quite important aerosol constituent in urban atmospheres.

## 2 Computational model

Generally speaking, EMTs approximate the optical properties of the inhomogeneous scatterer by those of a homogeneous particle of the same shape. The advantage of an EMT is that a homogeneous particle can be calculated much more easily and rapidly than a heterogeneous particle. One of the easiest, but physically not-well-justified EMTs is volume (mass) weighted mixing for which the resulting refractive index $m$ of a particle is given as follows: $m = \sum_j m_j f_j$ , where $m_j$ is the refractive index of *j*-species and $f_j$ is its volume fraction. To render principal differences between the EMT-based approach and regular (DDA-based) calculation, we consider the volume weighted mixing rule.

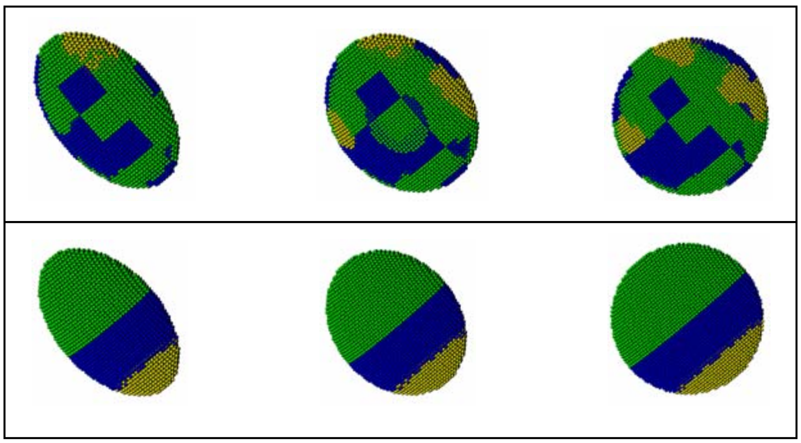

Figure 1: Applied particle models for the low-carbon case. Top shows particles with materials randomly distributed. Bottom shows particles with materials in clumps.

The basic particle model we incorporated into numerical scheme coincides with typical urban aerosols composed of ammonium sulfates (volume content ~ 50%, refractive index 1.52-0.0*i*), organic matter (volume content ~ 40%, refractive index 1.46-0.016*i*), and black carbon with approximately 10% volume content and refractive index 1.75-0.3*i* [2]. However, under some conditions the carbon can occur as an abundant component with volume fraction 20-30%. These situations are studied as "high carbon content case" (where organic matter is varied between 20-40% of particle volume, and ammonium sulfate ranges from approximately 40% to 50 % of particle volume). To check the simultaneous effect of asphericity and material configuration on particle optical properties, we model the aspect ratios of rotationally symmetric particles to be close to 1.4 - 1.6 as this range is well applicable to tropospheric particles [3].

Two basic approaches are employed to simulate the internal mixing of materials: a) the individual pieces of different constituents are distributed randomly in the particle body and b) the individual constituents are not scattered over the particle volume but are rather clumped together into compact blocks (Fig. 1). For both models we calculate the optical characteristics (phase function, polarization and efficiency factors for scattering, absorption and extinction, and asymmetry parameter) using the EMT approximation that assumes the effective refractive index is the volume-averaged refractive index of the components [4]. We analyze particles with *i*) prolate, *ii*) moderate, and *iii*) oblate forms. The classification is based on the mutual relation of particle sizes in X, Y, and Z, which are in our case given as follows: *i*) 1.6/1.0/1.0, *ii*) 1.6/1.3/1.1, *iii*) 1.6/1.6/1.0. All computations are made for incident monochromatic radiation with wavelength $\lambda = 0.525$ μm.

## 3 Numerical results and discussion

When computing the optical properties of composite randomly oriented particles we especially paid attention to modeling the phase function, polarization and efficiency factors for scattering $Q_{sca}$, absorption $Q_{abs}$, extinction $Q_{ext}$, and asymmetry parameter $g$. The computer model of the particle is expanded or contracted to produce scattering calculations for particles of different size. The precision of the DDA scattering calculations approximately depends on parameter $|m|kd$, where $d$ is the inter-dipole separation, $k$ is the wave number ($k = 2\pi/\lambda$) and $m$ is the complex refractive index of the particle. To guarantee that scattering calculations are sufficiently accurate, the parameter given above should be less than 1. We increased the number density of dipoles always when necessary to satisfy this requirement.

The computational results show slight differences between optical properties of particles built from compact homogeneous blocks and particles composed of randomly mixed pieces of material constituents (Fig. 2). As expected the most affected quantity is $Q_{abs}$: particles with characteristic random material configuration absorb more efficiently than particles built from compact blocks (refer to DDA calculations presented in Fig. 2). We discussed this in [5] for very small particles. As known from macrophysical studies the light transmitted into a material generally is increased if the media on either side of the interface have similar refractive indices. The air-carbon interface

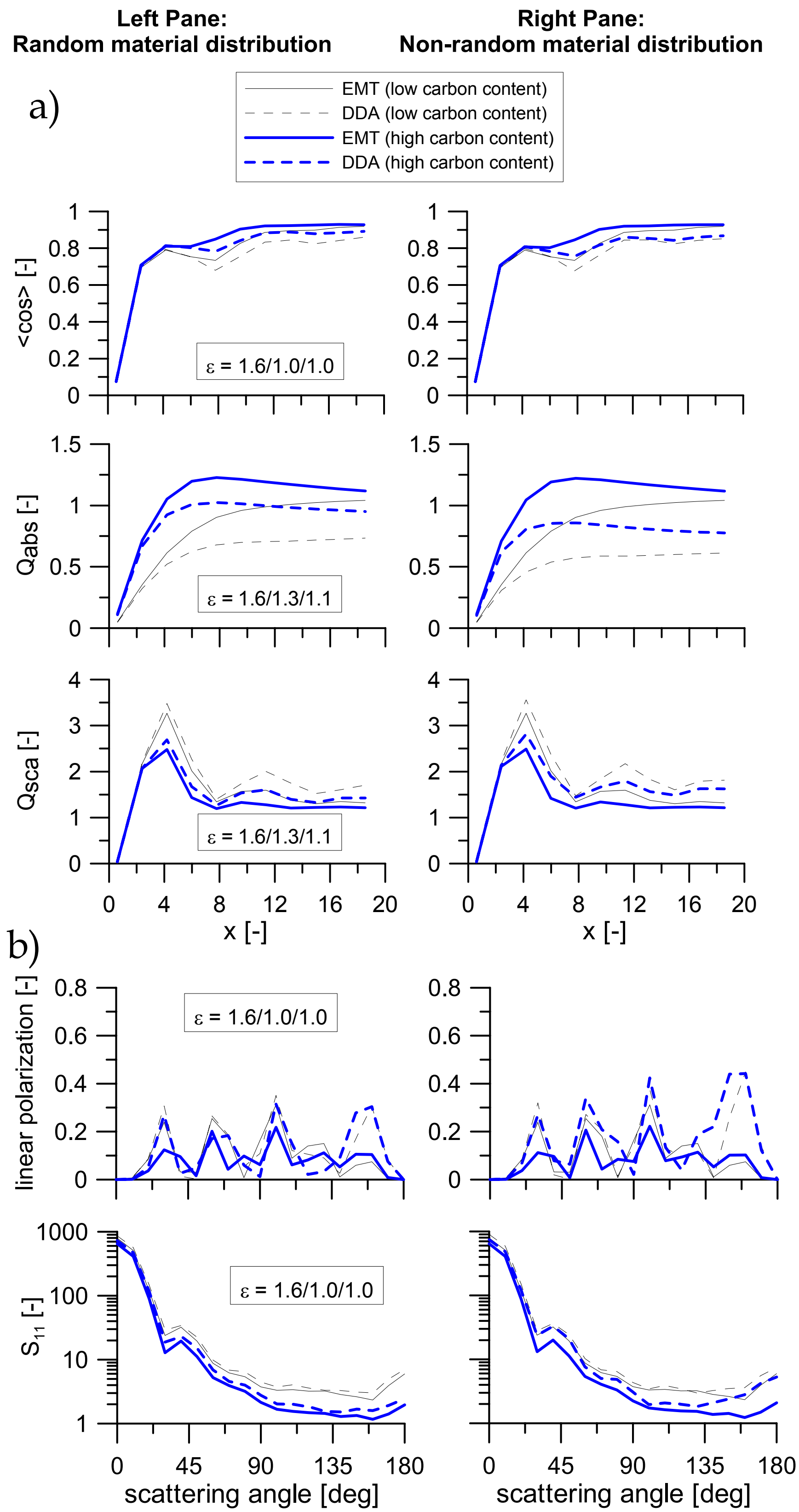


Figure 2: Optical properties of three-component carbonaceous ellipsoidal particles: a) Bulk properties and b) scattering response of x = 6 particles.

tends to reflect more light than the ammonium sulphate-carbon (or organic matter-carbon) interface. Therefore three-segment compact ellipsoids, whose absorbing component has the largest air-carbon interface absorb less efficiently than randomly fragmented ellipsoid.

Compared with DDA results, the volume-weighted EMT results overestimate both the asymmetry parameter and absorption efficiency, and usage of the EMT results in a quite evident reduction of scattering efficiency. Altogether these effects tend to cancel, leaving the extinction efficiency $Q_{ext}$ the most accurate quantity reproduced using EMTs. Discrepancies in the behavior of linear polarization and phase function is observed at large scattering angles for particles whose size is comparable to the wavelength of incident radiation: 1) the fragmented ellipsoids show a bit larger linear polarization than the compact ellipsoid, and also 2) the fragmented ellipsoids with high-carbon content scatter more in the backward direction than the compact ellipsoid. This can be due to reduced absorption or additional internal interactions.

### Acknowledgments

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