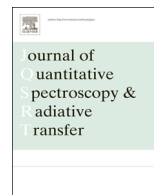


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## Review: Model particles in atmospheric optics

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### ABSTRACT

This review paper provides an overview over model geometries for computing light scattering by small particles. The emphasis is on atmospheric optics, although much of this review will also be relevant to neighbouring fields, in particular to astronomy. Various morphological particle properties are discussed, such as overall nonsphericity, pristine shapes, aggregation, and different forms of inhomogeneity, e.g. porous and compact inhomogeneous morphologies, as well as encapsulated aggregates. Models employed to reproduce the optical properties of complex particles range from strongly simplified to highly realistic and morphologically sophisticated model geometries. Besides reviewing the most recent literature, we discuss the idea behind models of varying degree of complexity with regard to the intended use of the models. Applications range from fundamental studies of light scattering processes to routine applications of particle optics look-up tables in operational modelling systems.

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## 1. Introduction

Light–matter interaction is among the most important processes in physics; it is exploited in numerous experimental techniques and extensively studied in different branches of theoretical physics. An important sub-set of such processes is the interaction of electromagnetic radiation with small particles. Light-scattering processes have numerous applications in atmospheric and planetary sciences, in the studies of solar-system objects [1], proto-planetary disks, exoplanets, the circumstellar environment of red giant stars, as well as the physics and chemistry of the interstellar medium (e.g. [2]). There are also important applications in ocean optics, biomedical optics [3], nano-optics [4], material science [5], process engineering, and combustion diagnostics [6].

In atmospheric physics one frequently encounters ensembles of aerosol particles or hydrometeors that exhibit a large range of different sizes, can have complex, non-spherical shapes, and possess varying, often heterogeneous chemical composition. A theoretical description of the interaction of light with such particle ensembles is a formidable problem. More often than not it can only be solved by introducing simplifying assumptions about the particle shapes.

The use of different model geometries in light scattering computations can sometimes give rise to scientific controversies. To take a concrete example, an ellipsoid model for describing Martian dust particles (e.g. [7]) may be questioned because of its simplicity; ellipsoids have little morphological resemblance to real Martian aerosols. Thus one may question the physical insights that can be gained from such a model. On the other hand, one could equally well criticise that this model is too complex, as a simpler spheroid model may, perhaps, be sufficiently accurate to reproduce the optical properties of Martian

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dust. The point of view we take here is that there is little meaning in discussing such questions decoupled from the question of the intended application of the model. Inversion methods for global remote sensing data sets (e.g. [8,9]) or radiative transfer applications in climate models (e.g. [10]) often necessitate a pragmatic approach based on the use of drastically simplified model geometries. On the other hand, in more fundamental studies aimed at attaining a basic understanding of the relation between morphological and radiative particle properties, one often needs to afford the use of much more elaborate model geometries.

Besides providing an overview of the existing literature, one purpose of this review is to discuss the rationale behind different approaches and approximations to represent complex morphologies in particle optics models, and to discuss their range and limits of applicability with regard to the intended uses of the models. The discussion focuses on atmospheric particles. However, much of this review will also be relevant to other fields, especially to applications of light scattering in astronomy.

Section 2 will first discuss the basic rationale of using particle models of various degree of complexity in light scattering computations. Section 3 will provide a review of modelling approaches for different morphological features. In principle, we could have equally well divided that section into sub-sections for different types of particles, e.g. ice particles and aerosols of different chemical composition (secondary inorganics, black carbon, organics, salt, mineral dust). Alternatively, we could have based the review on the sources of the particles (anthropogenic, marine, wind-blown dust, biogenic, etc). Instead, we organised the discussion according to different morphological characteristics of particles, such as large-scale non-sphericity, small-scale surface roughness, irregularity, aggregation, and inhomogeneity. This is the most logical structuring, as we are mainly interested here in the connection of particle morphology and particle optics, and in how to represent morphologically complex features in optics models.

The main target audience of this review are practitioners. Those who want to obtain more detailed information on electromagnetic scattering theory may start by consulting [11–13]. Reviews of numerical light scattering methods can be found in [14–16]. Available light scattering computer codes are discussed in [17].

## 2. Basic rationale of particle models in electromagnetic scattering

The real world is a messy place. By their very nature, models attempt to focus on the essential aspects of a complex system, while omitting or simplifying less important processes and characteristics of the system under investigation. Atmospheric particles cover a large range of sizes, morphologies, and chemical compositions. Moreover, the position as well as the physical and chemical properties of each particle can change with time. It is usually neither possible nor desirable to compute optical properties by working with exact replicas of each and every particle encountered in a real particle ensemble.

Thus, we are faced with the question how to represent complex particle morphologies in aerosol optics models. The choice of suitable model geometries is constrained by our incomplete knowledge of real-world particles, the computational resources we can afford, and the range of applicability of available numerical methods. Most importantly, the choice of model geometry will be determined by its intended application.

### 2.1. Applications of particle models to environmental modelling and remote sensing

One class of applications comprises large modelling systems designed for routine use (e.g. operational services), such as climate models or inversion schemes for remote sensing data sets. In such applications one rarely employs morphologically detailed model particles. There are several reasons why such large modelling systems almost exclusively employ highly simplified model geometries for aerosol optics modelling. One major limitation is the computational burden involved in creating aerosol optics look-up tables for climate models or remote sensing retrieval methods. The large range of sizes and the high variability in the chemical composition of aerosols require look-up tables that cover a large parameter space. This severely limits the level of detail one can afford in the light-scattering computations.

However, there are also other, equally important reasons that justify the use of highly simplified model particles. To understand these reasons, let us take, as an example, modelling approaches for the direct radiative effect of aerosols in climate models. The number of physical and chemical aerosol properties that are accounted for in climate models is highly limited. For instance, the most detailed modelling systems currently available (known as Earth-system models) contain a chemical transport model (CTM) with an aerosol dynamics module (e.g. [18]) coupled to an atmosphere–ocean circulation model. The CTM delivers the spatial distribution and the temporal evolution of atmospheric aerosols, which are characterised by their mass concentration and number distribution and their size-resolved chemical composition. Based on this information, one can obtain the size-dependent refractive index of the aerosols in each grid cell of the 3-dimensional model domain from which one can compute the aerosol optical properties. This information is fed into the climate model's radiative transfer scheme to compute the radiative forcing effect of aerosols on the atmosphere–ocean system. The salient point is that there are no processes in the model that deliver any information on particle shape (apart from the mixing state of different chemical components). The chemical composition is influenced by the particles' emission sources, chemical transformation processes, and by aerosol dynamic mixing processes, which are accounted for by the CTM. The size distribution results (for secondary particles) from dynamic processes, such as nucleation, condensation, and coagulation (which are described in the aerosol dynamics model), or (for sea salt and wind-blown dust) from emission processes. It is also influenced by dry and wet deposition processes, all of which are accounted for in the model. However, the particle shapes of, e.g., desert dust particles are the result of aeolian erosion and

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