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Decomposition of atmospheric aerosol phase function by particle size and asphericity from measurements of single particle optical scattering patterns

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ABSTRACT

We demonstrate an experimental approach that provides insight into how particle size and shape affect the scattering phase function of atmospheric aerosol particles. Central to our approach is the design of an apparatus that measures the forward and backward scattering hemispheres (scattering patterns) of individual atmospheric aerosol particles in the coarse mode range. We captured over 30 000 scattering patterns during winter (January 2007) at an urban site in Las Cruces, NM. The size and shape of each particle is discerned from the corresponding scattering pattern. In particular, autocorrelation analysis is used to differentiate between spherical and non-spherical particles, the calculated asphericity factor is used to characterize the morphology of non-spherical particles, and the integrated irradiance is used for particle sizing. We found that the fraction of spherical particles decays exponentially with particle size, decreasing from 11% for particles on the order of 1 μm to less than 1% for particles over 5 μm. The average phase functions of subpopulations of particles, grouped by size and morphology, are determined by averaging their corresponding scattering patterns. The phase functions of spherical and non-spherical atmospheric particles are shown to diverge with increasing size. In addition, the phase function of non-spherical particles is found to vary little as a function of the asphericity factor. Our results support the current remote sensing practice of characterizing atmospheric aerosol particles as a composition of spherical and non-spherical particles with less concern about the diversity of morphology within non-spherical particles. In addition, our results suggest that assuming a constant spherical fraction independent of particle size may not accurately reflect the real morphological distribution of atmospheric aerosol particles.

1. Introduction

Atmospheric aerosol particles impact our Earth's climate by scattering and absorbing radiation as well as by modifying the radiative properties of clouds [1]. Modeling and quantifying this contribution to the Earth's energy balance are needed for climate research. However, the specific contribution of atmospheric aerosols to the Earth's climate is largely unknown and represents a major source of uncertainty in climate models [2]. This uncertainty is driven in part by a lack of knowledge of the global spatio-temporal distribution of atmospheric particles and in part by inadequate modeling of the optical properties (i.e. scattering and absorption) of atmospheric aerosol

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particles [3]. These two sources of uncertainty are connected since remote sensing data collected to determine spatio-temporal distribution of atmospheric particles is inverted using assumed optical properties of atmospheric aerosol particles [4]. Therefore, an accurate description of the optical properties of atmospheric aerosol particles is critical to reducing uncertainties in climate research.

The diverse range of particle morphologies makes characterization of the optical properties of atmospheric aerosol particles a challenging endeavor. A common approach is to model aerosol particles using simple shapes in which the optical properties can be calculated; a distribution of these shapes is then used to model an ensemble of atmospheric particles. Because of simplicity and computational efficiency, homogeneous spheres are often used as model particles [5,6]. However, homogeneous spheres inadequately reproduce the optical properties of atmospheric aerosol particles, particularly large irregular particles like mineral dust [7–12]. A mixture of homogeneous spheroids and spheres has gained substantial traction as a model for aerosol particles ensembles and is currently being used in the inversion of AERONET (AErosol RObotic NETwork) retrievals [13]. This model works surprisingly better than other proposed shapes such as prisms [14], but more validation studies are needed. For example, it is unclear if the spheroidal model is applicable to size distributions with effect radii larger than 1 μm [4]. Furthermore, the fraction of spherical particles is often assumed to be constant for all sizes, which might not be representative of real atmospheric aerosol distributions [15]. Finally, the distribution of aspect ratios of the particles used in the spheroid model is fixed and based on a fairly small selection of measurements, and therefore may not be representative of a wider range of aerosol particles [16,17].

Atmospheric aerosol measurements can play a critical role in answering these and other key questions regarding the validation and improvement of contemporary particle modeling. Historically, atmospheric aerosol measurements involving multi-angle light scattering have proven particularly useful in the evolution of particle models. For example, the measurement of full Mueller matrices of dust collected from the Earth's surface was used to constrain and validate the current spheroidal model [8,13]. More recently, analysis of laboratory experiments combining multi-angle scattering, polarimetry, and IR extinction data has suggested that a single spheroidal shape distribution for all particles sizes is not representative of silicate clay aerosol [17]. Other experimental measurements have made similar contributions to the accuracy of particle modeling.

However, these measurements, while providing critical insight to the field of climate research (see Ref. [18]), have certain limitations since they are performed on particle ensembles; optical properties specific to single particles or even particle subpopulations are partly lost in these ensemble measurements. Recently, scattering measurements of individual atmospheric particles have emerged [19–21,15]. However, these studies focus on measuring particle morphology, specifically particle shape, without measuring optical properties, such as the scattering phase function. Thus in the field of climate research, there appear to be two disparate multi-angle light-scattering techniques, ensemble measurements that explore key optical properties for climate research and single-particle measurements that explore particle morphology needed for aerosol modeling. Neither techniques are currently capable of measuring both optical properties relevant to climate research and morphology information at the single particle level. A technique that did achieve this could decompose the optical properties of atmospheric aerosol by particle subpopulations (e.g. how the scattering phase function varies as a function of particle size and shape), which would greatly aid the validation and evolution of particle modeling thereby facilitating advances in aerosol characterization and climate modeling.

Here, we devise a powerful experimental technique that can obtain the scattering phase function of individual aerosol particles as well as capture optical information related to particle size and morphology. Essential to our approach is the measurement of two-dimensional angular optical scattering (TAOS) patterns from single atmospheric aerosol particles in the coarse mode range [22,23,15,24]. Individually, TAOS patterns provide key information about particle morphology. For example, we demonstrate how autocorrelation analysis of these TAOS patterns can identify optically smooth spherical particles and distinguish them from other particles with overall spherical shape but having rough surface or inhomogeneous distribution of refractive index. And as a collection, TAOS patterns provide ensemble-averaged optical properties. In particular, by grouping TAOS patterns based on particle size and shape, we explore how the scattering phase function depends upon particle morphology. Analysis of over 30 000 TAOS patterns collected from atmospheric aerosol particles in Las Cruces, NM, indicates (1) a decrease in the fraction of optically smooth spherical particles with increasing scattering size, (2) a divergence of the scattering phase function between spherical and non-spherical particles as particle size increases, and (3) little variation in the scattering phase function for non-spherical particles that are further classified using the asphericity factor.

2. Experiment design

The apparatus to capture two-dimensional angular optical scattering (TAOS) patterns is slightly modified from a previous design [15]. The path of the incident laser beam relative to the scattering volume is rotated by 90° so that both the forward and backward scattering hemispheres from single aerosol particles are detected [24]. Furthermore, to optimize the scattering geometry for the analysis performed in this paper, the polarization of the incident laser beam is changed from linearly polarized to circularly polarized. The remainder of the experimental geometry remains very similar to the previous design.

Briefly, aerosol was sampled through a 4-cm diameter, 3-m long, conductive hose inserted through the outside wall of a first floor laboratory (further details in Section 3). A manifold attached to this hose provided aerosol flow for two virtual impactor concentrators, one for single-particle