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Decomposition of Atmospheric Aerosol Phase Function by Particle Size and Morphology via Single Particle Scattering Measurements

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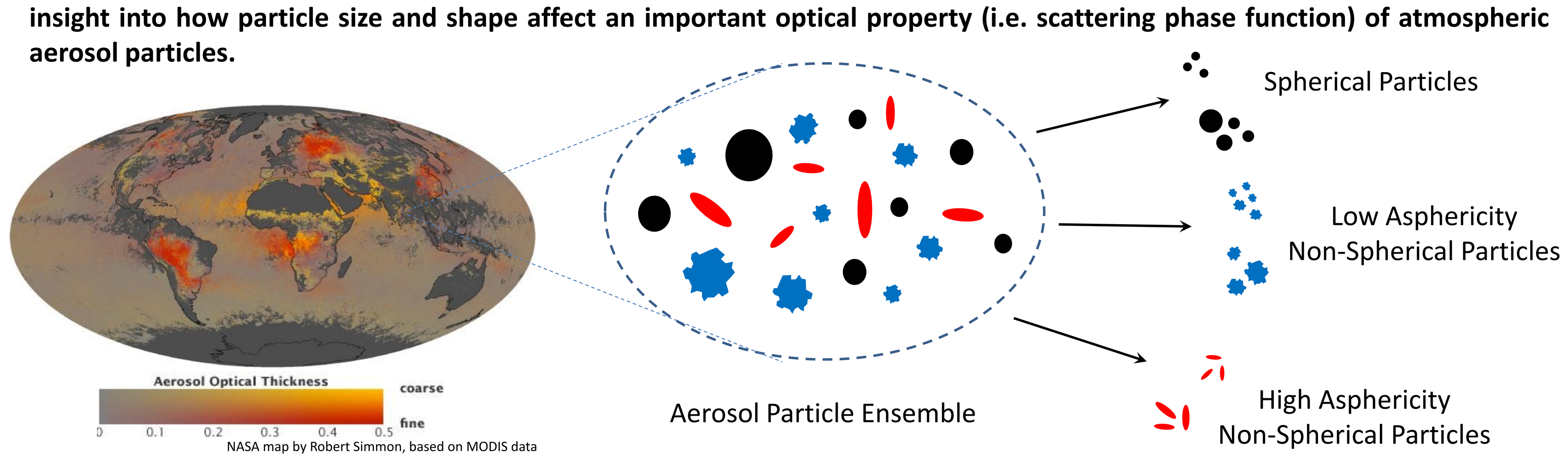
Motivation

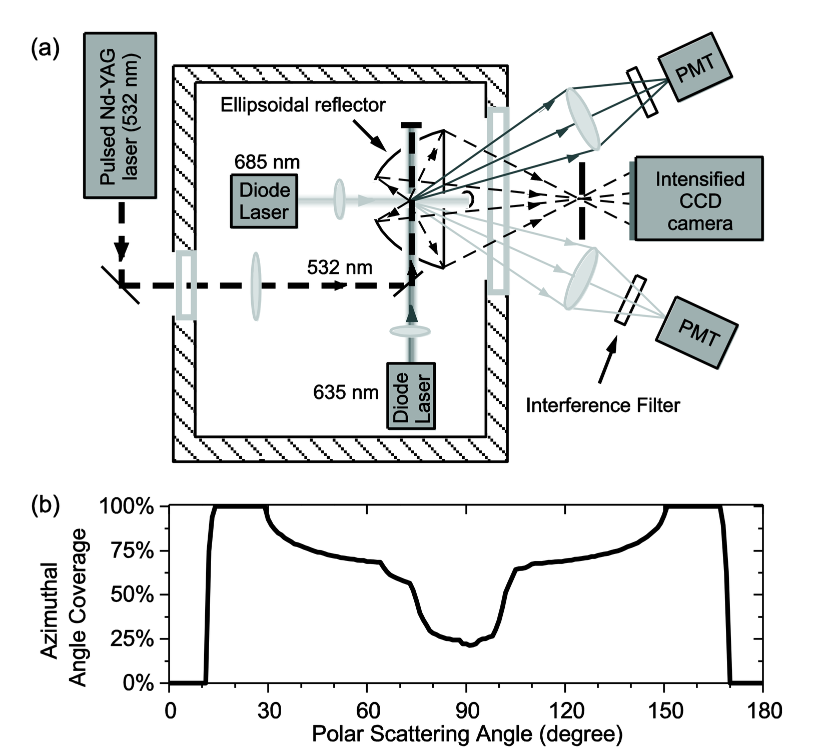
Atmospheric aerosol particles impact our Earth's climate by scattering and absorbing radiation as well as by modifying the radiative properties of clouds. Modeling and quantifying this contribution to the Earth's energy balance is needed for climate research. However, the specific contribution of atmospheric aerosols to the Earth's climate is largely unknown and represents a major source of uncertainty in climate models. 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 particles. 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. Therefore, an accurate description of the optical properties of atmospheric aerosol particles is critical to reducing uncertainties in climate research. **In this work, we demonstrate an experimental approach that provides insight into how particle size and shape affect an important optical property (i.e. scattering phase function) of atmospheric aerosol particles.**



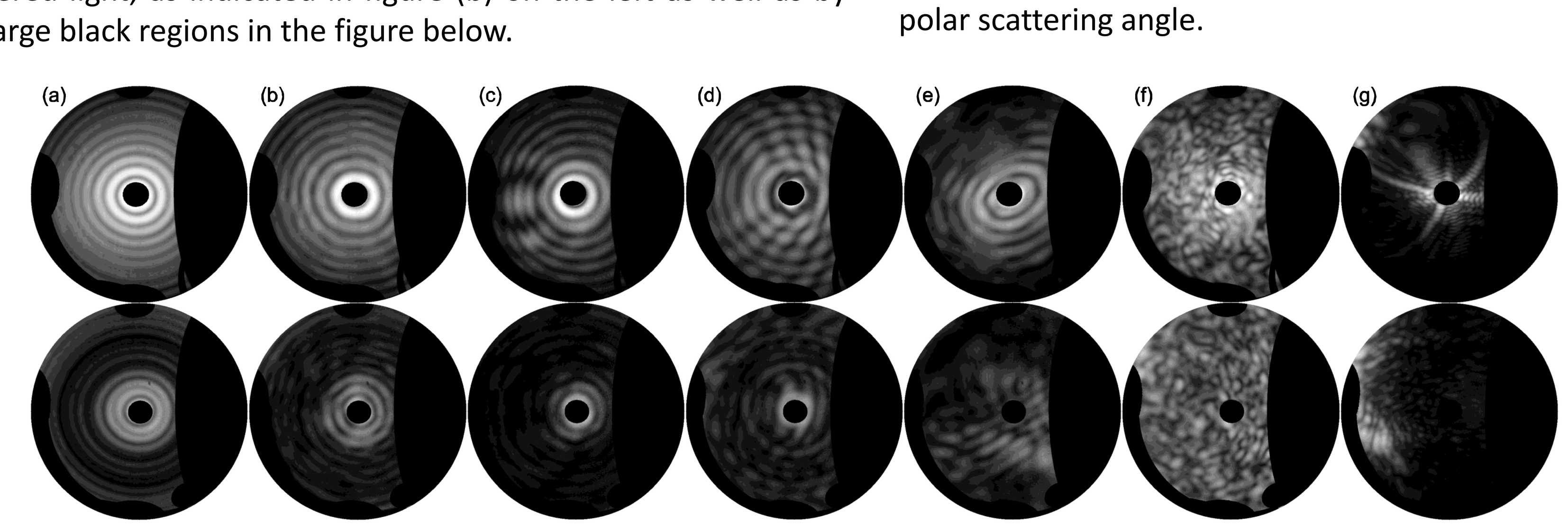
The morphology of atmospheric aerosol particles can be quite varied. This diverse morphology needs to be accounted for in optical modeling. **How does light scattering depend upon particle morphology?**

Apparatus

The apparatus to capture two-dimensional angular optical scattering (TAOS) patterns is shown on the right. The path of the incident laser beam relative to the scattering volume is such that **both the forward and backward scattering hemispheres from single aerosol particles are detected**, as shown in the figure below. Aerosol was sampled through a 4-cm diameter, 3-meter long, conductive hose inserted through the outside wall of a first floor laboratory. Aerosol particles were concentrated by a concentrator (Dycor, XMX) with inlet flow of about 10 liters per second and minority outlet flow of about 1 liter per minute. A conically shaped aerodynamic sheath nozzle focused the minor flow inside the airtight box into a laminar jet around 300 μm in diameter. The aerosol nozzle tip was positioned 0.5 cm above the first focal point of the elliptical reflector. To reduce errors associated with misaligned particles, only particles at or near the first focal point of the elliptical reflector are illuminated. This is achieved with a cross-beam trigger system that restricts the scattering volume to be significantly smaller than the illuminating beam. The experimental geometry does not detect all the scattered light, as indicated in figure (b) on the left as well as by the large black regions in the figure below.



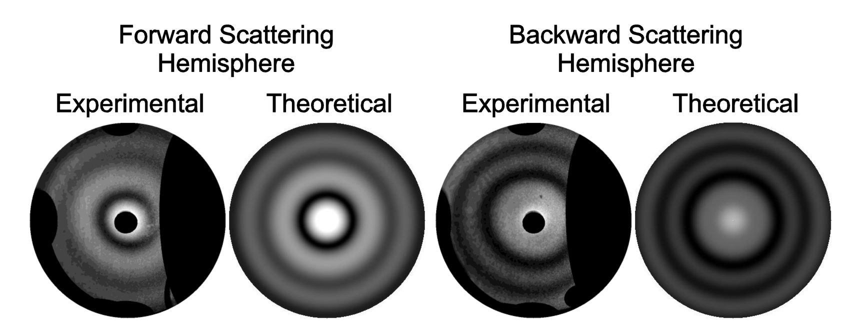
(a) TAOS instrument to simultaneously capture forward and backward scattering hemispheres.
(b) Detected range of azimuthal angles as a function of polar scattering angle.



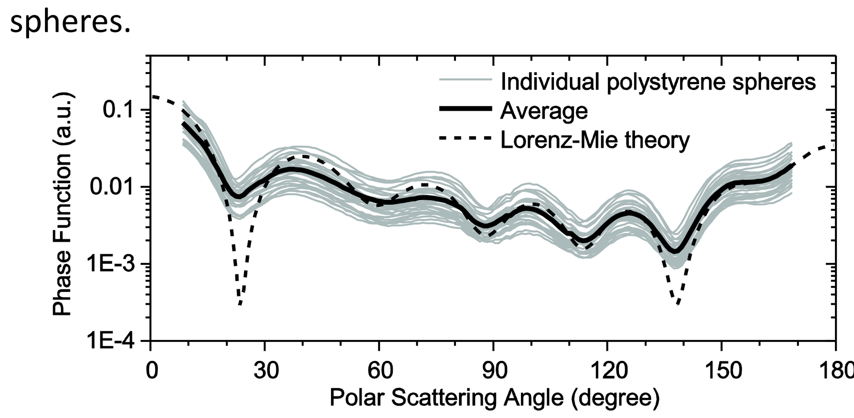
Forward (top row) and backward (bottom row) scattering hemispheres from unknown atmospheric aerosol particles. Intensities are on a logarithmic scale and the backward scattering intensities are scaled by a factor of two as compared with the forward scattering intensities.

Phase Function

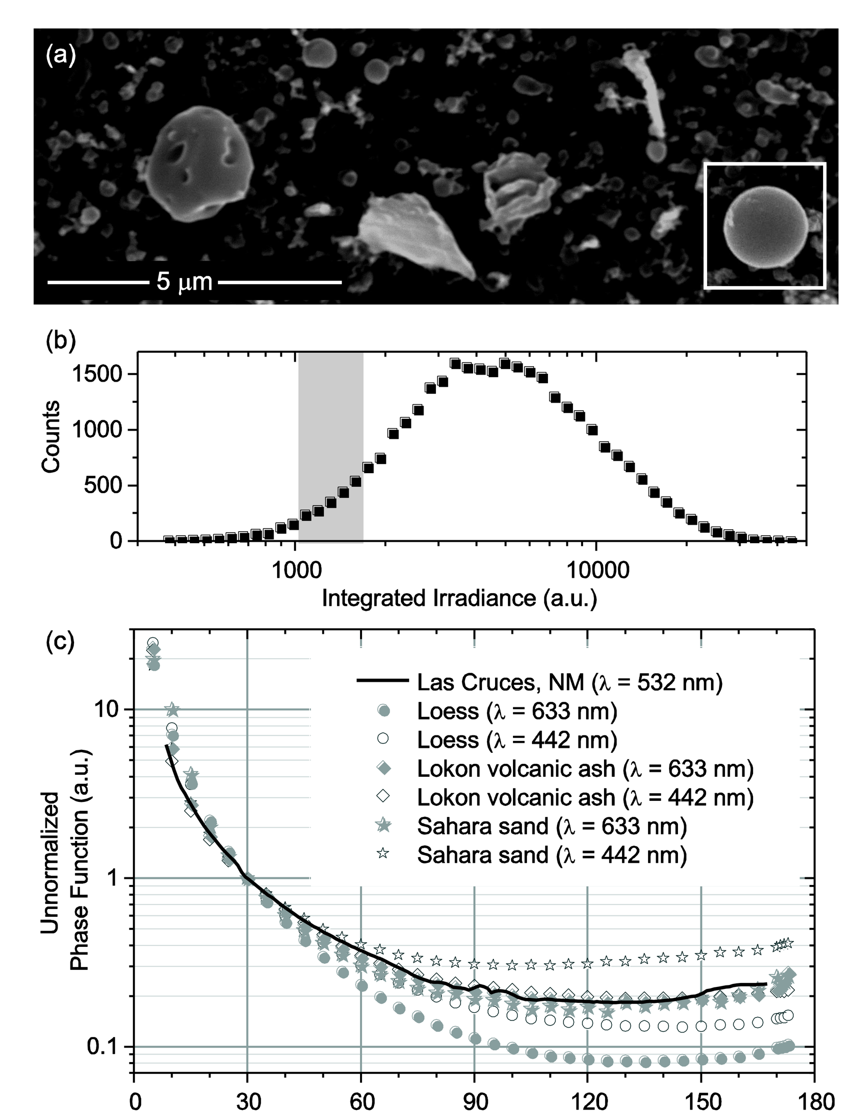
To test the system, measurements were performed on 1 μm polystyrene latex (PSL) spheres. Shown below are the forward and backward scattering hemispheres from experiment and calculated from Lorenz-Mie theory. Black regions in experimental image represent inaccessible angles for our experimental geometry.



Of primary interest for this work is the phase function of aerosol particles. Thus the phase functions were obtained from 30 different polystyrene spheres (gray solid lines) and compared with Lorenz-Mie theory (dashed line). Black solid line indicates the average scattering intensity for all 30 spheres.

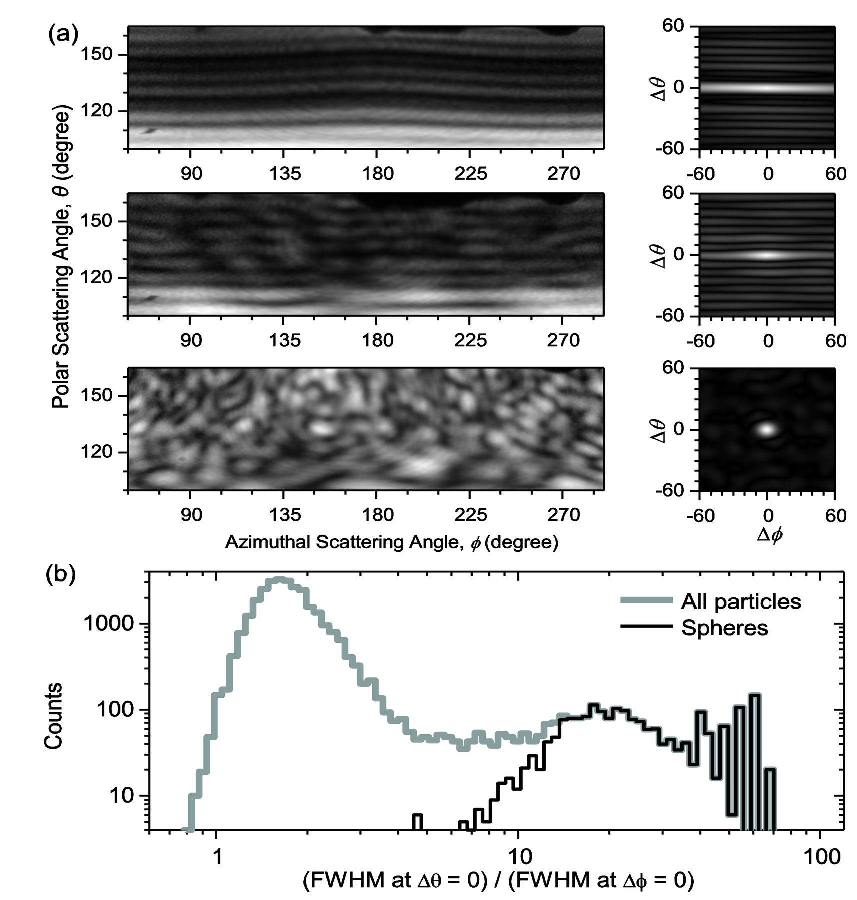


Measurements were conducted on the campus of New Mexico State University, Las Cruces, New Mexico during January 2007. The scattering patterns of 31,407 ambient aerosol particles were analyzed for this work. Scanning electron microscope images of representative particles are shown in (a). To get a sense of the size distribution, a histogram of integrated scattering intensities of all atmospheric particles is shown in (b). Grayed region represents scattering from PSL particles. Finally (c) displays the average phase function of all the particles. Also plotted for comparison are the scattering phase functions from the Amsterdam Light Scattering Database.

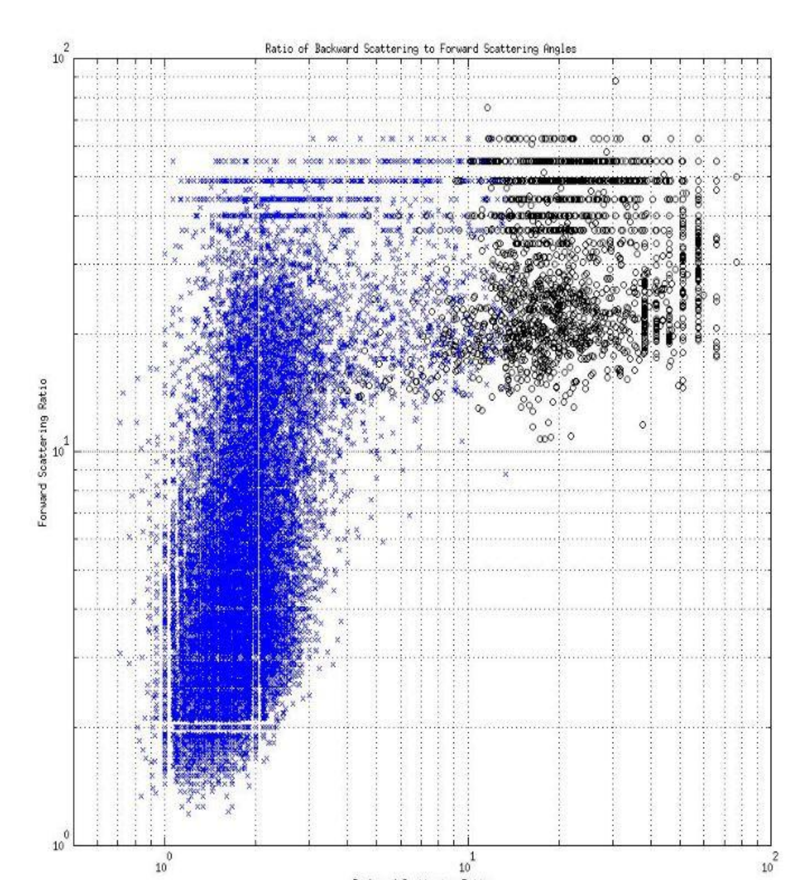


Morphology Analysis

We have also explored using the autocorrelation function of the patterns to discern particle shape. Below shows the scattering irradiance (left) and autocorrelation (right) of three scattering patterns. The size of the central peak of the autocorrelation function strongly correlates with sphericity. See figure (b) below where FWHM stands for full width half maximum of the central peak.

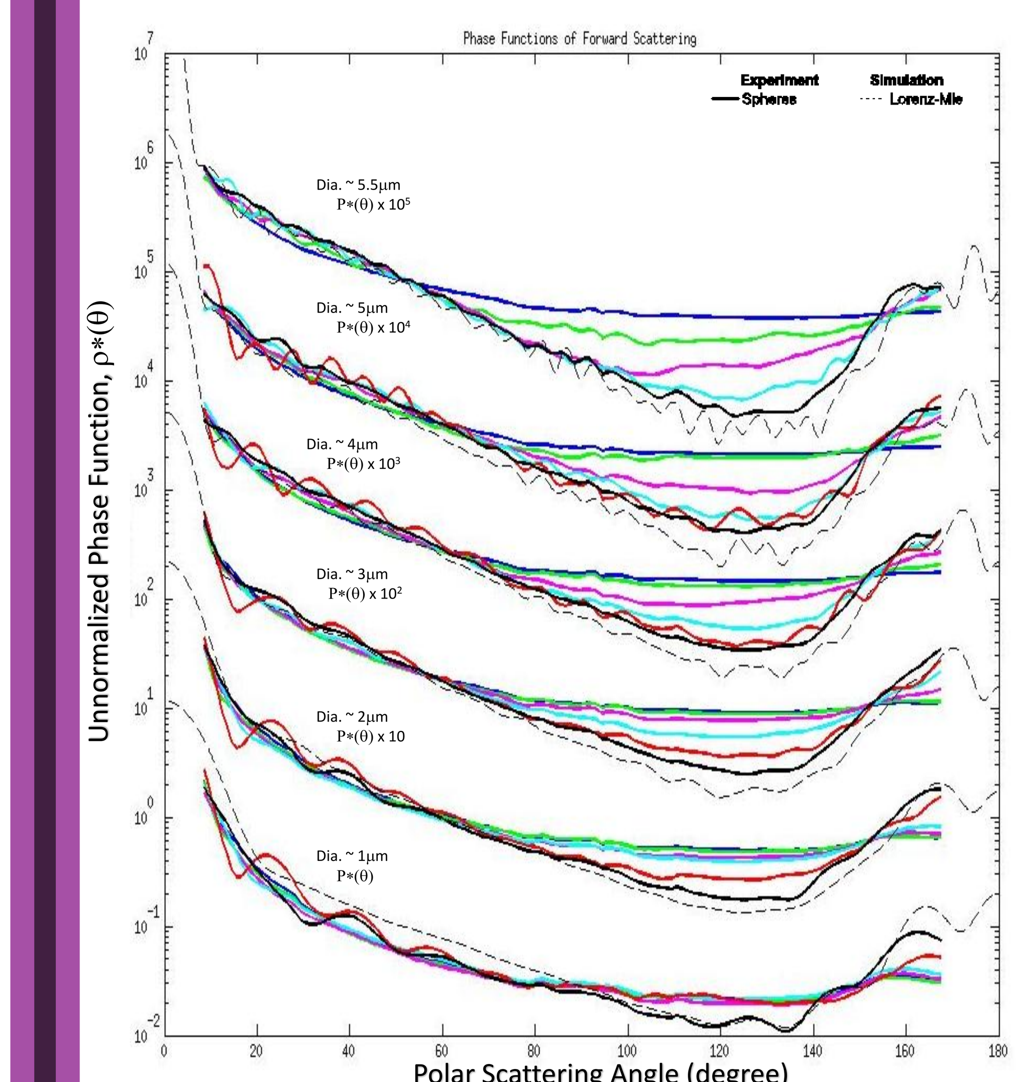


Using the central peaks of the autocorrelation functions of the individual TAOS patterns, we compared the results of the FWHM ratios for the forward scattering patterns with the FWHM ratios of the backward scattering patterns, in order to look for groupings of data. This helped lead to further classification. For example, a high forward ratio and a low backward ratio would be expected to be a sphere with an inhomogeneous distribution of refractive index, or a sphere with a perturbed surface. The figure below shows the distribution of spheres (black circles) and non-spheres (blue x's), in the backward vs. forward scattering ratios.



Phase Function Decomposition

The central result of this work is the determination of the aerosol phase function for different subpopulations of particles. In particular, we binned the particles by size and by Full-Width Half Maximum of the Autocorrelation function, and then calculated the average phase function for each bin. As a check, we also computed the phase function from Lorenz-Mie theory based on the size distribution. The figure below shows decomposition of phase functions based on the data from the forward scattering ratios.



This shows a gradual evolution of phase functions as particles evolve from spheres to non-spheres. The varied colors represent the phase functions of particles as they evolve from a spherical to non-spherical classification.

Summary

For the first time, an image analysis technique has been devised to clearly show the gradual evolution of the scattering phase function of atmospheric aerosol particles from those that are spherical particles, to those that are significantly non-spherical. In particular, the two-dimensional autocorrelation function of the forwards and backwards scattering hemispheres is able to quantify the ring-like structure that occurs in the scattering patterns of spheres. As this ring-like structure diminishes, the phase function evolves from being one associated with spherical particles, to having a shape that is traditionally associated with non-spherical particles. Prior research demonstrated a binary relationship between spheres and non-spheres, without a continuous progression of phase functions. Understanding how the scattering phase function evolves from spherical particles to a non-spherical particles will aid in the accurate interpretation of data collected using these models.