Improving our Understanding of the Optical Properties of Atmospheric Aerosol

Supervisors:
Prof Jonathan Reid (School of Chemistry, University of Bristol) – Main supervisor
Prof Andrew Orr-Ewing (School of Chemistry, University of Bristol)
Host institution: University of Bristol

Project description: As identified in the recent report by the *Intergovernmental Panel on Climate Change (2013)*, aerosols continue to contribute the largest uncertainty to the total estimate of radiative forcing and, thus, diminish our ability to predict future climate change. Aerosols play both a direct role, scattering and absorbing sunlight, and an indirect role, influencing cloud albedo and lifetime. New techniques are crucial to better quantify aerosol processes and properties if the uncertainty in their role is to be reduced. We will exploit a newly developed technique to measure the optical properties of individual aerosol particles and to test and validate commonly used optical models. Further, we will explore the processes that transform aerosol optical properties including the condensation/evaporation of water.

We have recently demonstrated that individual particles in the size regime dominating atmospheric optics (>500 nm and <1.5 μm diameter) can be captured and held indefinitely in an optical trap, and the light extinction, angular scattering and asymmetry ratio measured.[1] This provides the *first ever approach* for measuring the extinction efficiency of an aerosol particle directly, a critical quantify to determine and validate models of light scattering. Uniquely, changes in optical properties of the particle can then be measured over timescales of hours (eg. with change in environmental conditions such as relative humidity, RH). Extinction measurements are made by cavity ringdown spectroscopy, an ultrasensitive technique for determining optical cross-sections.

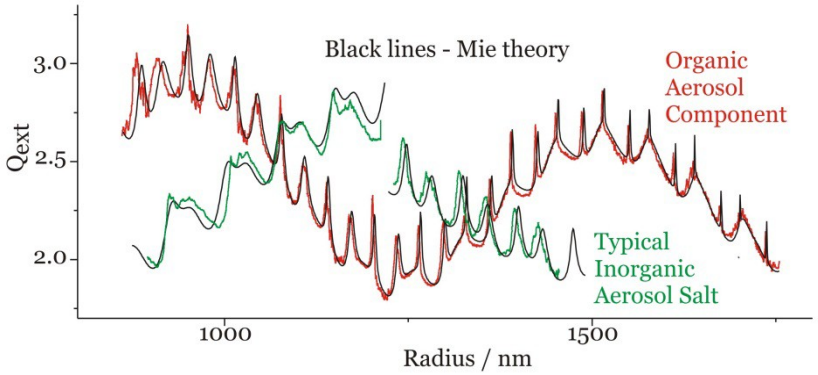

Figure: Examples of light extinction measurements made on inorganic salt solution particles (green, change in optical cross-section with RH change from 49 to 87 %) and organic aerosol (evaporation of organic component over hours leading to change in optical extinction). Mie theory predictions for these simple single and binary aerosol cases are shown by the black lines.

The project will focus on three aims. (1) To assess models for treating the optical properties of mixed component aerosol and mineral dust. There is considerable ambiguity as to the description of the optical properties of complex aerosol systems (eg. mixing rule predictions for refractive index, the formation of core-shell structures, morphology). By examining some benchmark systems, we will establish the limits to the uncertainty in light extinction by complex aerosol arising from uncertainties in composition and particle morphology. (2) To assess models of hygroscopic optical growth for ambient particles. A limited number of studies have explored how the optical cross-section of aerosol depends on RH. We will explore both systems that show equilibrium hygroscopic growth and changes in optical cross-section, or kinetically arrested

states of secondary organic aerosol that are now commonly considered to exist in ultraviscous or glassy states.[2] (3) Heterogeneous chemistry can lead to the formation of brown carbon and absorbing aerosol. We will directly monitor the changes in light extinction by an individual particle as it is chemically aged, with the aim of better constraining the growth in ambient absorbing secondary organic aerosol.

[1] J.S. Walker, A.E. Carruthers, A.J. Orr-Ewing and J.P. Reid, Measurements of Light Extinction by Single Aerosol Particles, J. Phys. Chem. Lett. 4 (2013) 1748-1752.

[2] D.L. Bones, J.P. Reid, D.M. Lienhard, and U.K. Krieger, Comparing the mechanism of water condensation and evaporation in glassy aerosol, PNAS 109 (29) (2012) 11613-11618.