Geophysical Research Abstracts
Vol. 13, EGU2011-8720-1, 2011
EGU General Assembly 2011

Direct comparison of the thermodynamic properties of sodium chloride and ammonium sulphate aerosol at relative humidities approaching saturation

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When compared with greenhouse gases the impact of aerosols on the radiative balance of the Earth's atmosphere is poorly understood. Although estimates suggest that the net global cooling due to aerosols is likely to be similar in magnitude to the warming caused by anthropogenic carbon dioxide emissions, aerosols remain the dominant uncertainty in radiative forcing estimates. A greater understanding of the thermodynamic, kinetic and optical properties of aerosol particles is required for this uncertainty to be reduced. One important thermodynamic property of aerosols is hygroscopicity, which describes how an aerosol particle changes size with changes in relative humidity (RH) through the uptake or loss of water to the atmosphere. Hygroscopicity governs the optical cross section and also the cloud-forming potential of a particle, both critical factors in the effect of aerosols on climate. Laboratory studies of individual aerosol particles increase our understanding of the thermodynamic behaviour of atmospheric aerosol.

Over recent years optical tweezing has emerged as a powerful tool with which to probe and compare multiple aerosol particles of different composition held within the same environment. In such experiments a laser is used to trap particles for an extended period of time within a trapping cell using optical forces. Trapped particles, normally 1 to 5 μm in radius, are sized with nanometre accuracy from the unique fingerprint of size provided by the stimulated Raman spectrum and comparison with Mie theory calculations. The RH within the trapping cell can be varied from dry conditions extending up to RHs approaching saturation (100% RH) and the response in size of the different trapped particles compared.

In this work we compare directly the hygroscopicity of single component sodium chloride and ammonium sulphate aerosol particles at RHs approaching saturation [1]. This allows us to assess the accuracy with which the solution thermodynamics and wet particle size can be estimated for these systems under the conditions of supersaturation that lead to the activation of cloud condensation nucleii. The evolving size of the aqueous sodium chloride particle is used to accurately infer the RH. Measurements of the equilibrium size of ammonium sulphate aerosol are shown to agree with thermodynamic predictions to within an uncertainty of $\pm$0.2% between 84% and 96% RH. This suggests that estimates of the critical supersaturation, the RH required to activate an aerosol particle into a cloud droplet, are accurate to better than $\pm$0.002% RH for ammonium sulphate aerosol, an important cloud-forming aerosol. It can be concluded that the critical supersaturation estimated from models such as the Aerosol Inorganics Model (AIM) and the Aerosol Diameter Dependent Equilibrium Model (ADDEM) can be assumed to be accurate to within an uncertainty of better than 1% for typical supersaturations of 0.2% and above. This supports the conclusion of recent ensemble measurements comparing the critical supersaturations of ammonium sulphate and sodium chloride aerosol [2].

[1] Walker et al., J. Phys. Chem. A 2010, 114 (48), 12682-12691
[2] Rose et al., Atmos. Chem. Phys. 2008, 8 (5), 1153-1179