Geophysical Research Abstracts
Vol. 14, EGU2012-2132, 2012
EGU General Assembly 2012

Contrasting the Evaporation and Condensation of Water from Glassy and Amorphous Aerosol Particles

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The partitioning of water between the condensed and gas phases in atmospheric aerosol is usually assumed to occur instantaneously and to be regulated by solution thermodynamics. However, the persistence of high viscosity, glassy and amorphous aerosol to low relative humidity without crystallisation occurring is now widely recognised, suggesting that the timescale for water transport to or from the particle during condensation or evaporation may be significant. A kinetic limitation on water transport could have important implications for understanding hygroscopic growth measurements made on ambient particles, the ability of particles to act as ice nuclei or cloud condensation nuclei, the kinetics of chemical aging/heterogeneous chemistry, and the rate or condensation/evaporation of semi-volatile organic components.

In this study we will report on measurements of the timescale of water transport to and from glassy aerosol and ultra-high viscosity solution droplets using aerosol optical tweezers to investigate the time-response of single particles to changes in relative humidity. As a benchmark system, mixed component aerosol particles containing sucrose and sodium chloride have been used; varying the mole fractions of the two solutes allows a wide range of solution viscosities to be studied. We will show that coarse particles can take many thousands of seconds to equilibrate in size and that the timescale correlates with the estimated bulk viscosity of the particle. We will also confirm that significant inhomogeneities in particle composition can be established during evaporation or condensation. Using the experimental data to benchmark a model for equilibration time, predictions can be made of the timescale for the equilibration of accumulation mode particles during water condensation or evaporation and these predictions will be described and their significance explored. Finally, the coalescence dynamics of highly viscous aerosol particles will be reported, reporting for the first time the timescale required for coalesced particles to relax to an equilibrium morphology and size.