



Phase state is a limiting factor in hygroscopic growth of secondary organic aerosol

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Secondary organic aerosol (SOA) particles formed from oxidation products of volatile organic compounds (VOC) form a significant fraction of the total atmospheric particulate matter affecting climate both directly and indirectly. The dependence of hygroscopicity on particle composition is often represented with the single parameter κ , commonly used in global models to describe the hygroscopic properties of atmospheric aerosol particles. The physical phase state of SOA particles affects the partitioning of organic vapors and also may affect the uptake of water vapor and particle activation into cloud droplets. Thus, hygroscopic behaviour of SOA particles is affected by composition (i.e. oxidation state and molecular size) but also by phase of particles.

In this study the following three distinct studies were performed: (1) particle bounced fraction (BF) measurements, which are qualitatively related to particle phase, as a function of relative humidity using an Aerosol Bounce Instrument (ABI). We assume that the particles with $BF > 0$ are solid or semisolid, and that particles with $BF = 0$ behave mechanically as liquids (2) water uptake measured in the sub-saturated region using hygroscopicity tandem differential mobility analyzer (HTDMA) by measuring the ratio of wet to dry particle diameter following exposure to water vapor at a controlled RH (3) cloud droplet formation in the supersaturated region using a cloud condensation nuclei counter (CCNc). Particle composition and oxidation state was measured with a compact time of flight aerosol mass spectrometer (c-ToF-AMS).

In this study we show that at sub-saturation conditions water uptake by SOA particles is restricted due to the kinetic limitations. Diffusion and solubility limitations inhibit water uptake until the humidity is high enough for dissolution to occur. Our studies show that this "threshold" humidity is dependent on particle composition, oxidation state, and average molecular size. Our laboratory results explain several observations both in laboratory and in the atmosphere that have reported discrepancies between SOA particle hygroscopicity measurements in subsaturated and in supersaturated conditions. The results reported here provide new information about one of the most central aerosol processes in the atmosphere.