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LOW TEMPERATURE PHASES AND TRANSFORMATIONS OF AMMONIUM SULFATE PARTICLES

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Ammoniated sulfate particles are prevalent throughout the global troposphere where they constitute a large number fraction of the ambient aerosol in continental air, and have been observed in the upper troposphere. These hygroscopic particles impact the radiative properties of the atmosphere by scattering incoming solar radiation and by acting as cloud nuclei. These particles may also influence the chemistry of the atmosphere by facilitating heterogeneous reactions such as the hydrolysis of N_2O_5 on their surfaces. Both of these effects are a function of the particles' composition and phase, which are in turn dependent upon the ambient temperature and relative humidity. While models can simulate the equilibrium deliquescence transitions and thermodynamic properties of ammonium sulfate particles reasonably well, the kinetically-controlled efflorescence transitions cannot be presently predicted. Therefore, in order to accurately model the atmospheric effects of these particles, it is necessary to characterize these non-equilibrium phase transitions through laboratory experiments.

We report the application of a single particle trap to investigate the efflorescence transitions of ammonium sulfate particles below 273 K and the formation of the ammonium sulfate tetrahydrate below the eutectic point. The electrodynamic balance allows for the determination of a particle's composition and water activity at the efflorescence point. The efflorescence phase transition, for the homogeneous nucleation of the anhydrous ammonium sulfate salt, has been characterized as a function of temperature from 273 to 220 K. A Mie scattering technique is employed to determine the particle's size, which is used to estimate the rate of nucleation. A comparison between the present experiments and a recent flow tube study, which used much smaller particles, provides for an analysis of the efflorescence transition as a function of particle size. On occasion, heterogeneous nucleation was observed to occur causing efflorescence at lower compositions and higher water activities than could be explained due to particle size differences.

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