



Heterogeneous freezing of droplets with immersed surface modified mineral dust particles

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In the framework of the international measurement campaign FROST II (FReezing Of duST), the heterogeneous freezing of droplets with an immersed surface modified size-segregated mineral dust particles was investigated at LACIS (Leipzig Aerosol Cloud Interaction Simulator, Stratmann et al. 2004).

The following measurements were done: LACIS, CFDC (Continuous Flow thermal gradient Diffusion Chamber, Rogers (1988)) and FINCH (Fast Ice Nucleus Chamber Counter, Bundke et al (2008)) were used to analyze the immersion freezing behavior of the treated Arizona Test Dust (ATD) particles at different temperature regimes. The ability to act as IN (Ice Nucleus) in the deposition nucleation mode was quantified by the PINC (Portable Ice Nucleation Chamber) and the CFDC instrument. AMS (Aerosol Mass Spectrometers, e.g. Schneider et al. (2005)) and ATOFMS (Aerosol Time-Of-Flight Mass Spectrometer) measurements were applied to determine particle composition. The hygroscopic growth and the critical super-saturations needed for droplet activation were determined by means of an H-TDMA (Humidity-Tandem Differential Mobility Analyzer) and CCN counter (Cloud Condensation Nucleus counter, Droplet Measurement Technologies, Roberts and Nenes (2005)).

The 300 nm ATD particles were chemically and physically treated by coating with sulphuric acid (H_2SO_4 , three different coating thicknesses) and ammonium sulphate ($(NH_4)_2SO_4$) or by thermal treatment with a thermodenuder operating at 250°C. The H_2SO_4 coating modified the particles by reacting with particle material, forming soluble sulfates and therefore changing surface properties. AMS showed free H_2SO_4 only for thick H_2SO_4 coatings. In the heated section of the thermodenuder coating materials were evaporated partly and the surface properties of the particles were additionally altered.

Uncoated particles and those coated with thin coatings of H_2SO_4 , showed almost no hygroscopic growth. Particles coated with thicker coatings of H_2SO_4 and of $(NH_4)_2SO_4$ grew noticeably above 95% relative humidity. All investigated particles were found to activate at atmospherically relevant super-saturations.

All kinds of treatment lower the IN-ability, whereas the deposition nucleation was more sensitive to treatments than the immersion freezing mode. Considering the immersion freezing behavior, pure ATD particles and particles coated with thin coatings of H_2SO_4 were more efficient IN, than particles with thick H_2SO_4 or $(NH_4)_2SO_4$ coatings. Thermal treatments of the particles led to further decrease of the IN capability except for particles coated with $(NH_4)_2SO_4$, where the heating did not effect the immersion freezing behavior likely due to their already reduced IN ability.

In order to specify the temperature-dependent immersion freezing, two parameterization based on either stochastic or singular hypothesis were performed. From both theoretical approaches it can be concluded that the treatments lead to particle surface modifications lowering the nucleation efficiency.

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