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Understanding the ice nucleation characteristics of feldspars suspended in solution

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Freezing of liquid droplets and subsequent ice crystal growth affects optical properties of clouds and precipitation. Field measurements show that ice formation in cumulus and stratiform clouds begins at temperatures much warmer than those associated with homogeneous ice nucleation in pure water, which is ascribed to heterogeneous ice nucleation occurring on the foreign surfaces of ice nuclei (IN). Various insoluble particles such as mineral dust, soot, metallic particles, volcanic ash, or primary biological particles have been suggested as IN. Among these the suitability of mineral dusts is best established. The ice nucleation ability of mineral dust particles may be modified when secondary organic or inorganic substances are accumulating on the dust during atmospheric transport. If the coating is completely wetting the mineral dust particles, heterogeneous ice nucleation occurs in immersion mode also below 100 % RH.

A previous study by Zobrist et al. (2008) Arizona test dust, silver iodide, nonadecanol and silicon dioxide suspensions in various solutes showed reduced ice nucleation efficiency (in immersion mode) of the particles. Though it is still quite unclear how surface modifications and coatings influence the ice nucleation activity of the components present in natural dust particles at a microphysical scale. To improve our understanding how solute and mineral dust particle surface interaction, we run freezing experiments using a differential scanning calorimeter (DSC) with microcline, sanidine, plagioclase, kaolinite and quartz particles suspended in pure water and solutions containing ammonia, ammonium bisulfate, ammonium sulfate, ammonium chloride, ammonium nitrate, potassium chloride, potassium sulfate, sodium sulfate and sulfuric acid.

Methodology

Suspensions of mineral dust samples (2 – 5 wt%) are prepared in water with varying solute concentrations (0 – 15 wt%). 20 vol% of this suspension plus 80 vol% of a mixture of 95 wt% mineral oil (Aldrich Chemical) and 5 wt% lanolin (Fluka Chemical) is emulsified with a rotor-stator homogenizer for 40 s at a rotation frequency of 7000 rpm. 4 – 10 mg of this mixture is pipetted in an aluminum pan (closed hermetically), placed in the DSC and subjected to three freezing cycles. The first and the third freezing cycles are executed at a cooling rate of 10 K/min to control the stability of the sample. The second freezing cycle is executed at a 1 K/min cooling rate and is used for evaluation. Freezing temperatures are obtained by evaluating the onset of the freezing signal in the DSC curve and plotted against water activity.

Results

Based on Koop et al. (2000), a general decreasing trend in ice nucleation efficiency of the mineral samples with increasing solute concentrations is expected. Interestingly, feldspars (microcline, sanidine, plagioclase) in very dilute solutions of ammonia and ammonium salts (water activity close to one) show an increase in ice nucleation efficiency of 4 to 6 K compared to that in pure water. Similar trends but less pronounced are observed for kaolinite while quartz shows barely any effect. Therefore, there seem to be specific interactions between the feldspar surface and ammonia and/or ammonium ions which result in an increase in freezing temperatures at low solute concentrations. The surface ion exchange seems to be secondary for this effect since it is also present in ammonia solution. We hypothesize that ammonia adsorbs on the aluminol/silanol groups present on feldspar (viz. aluminosilicate surface) surfaces (Nash and Marshall, 1957; Belchinskaya et al., 2013). Hence allowing one of the N-H bonds to stick outwards from the surface, facing towards the bulk water and providing a favorable template for ice to grow.

The current study gives an insight into the ice nucleation behavior of aluminosilicate minerals when present in conjunction with chemical species, eg. ammonium/sulfates, which is of high atmospheric relevance.

References

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