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Increased ice nucleation efficiency of mineral dusts in dilute ammonium sulfate solutions

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Mineral dust has lifetimes of several days in the atmosphere and can be transported over long distances. During transport, mineral dust particles may acquire a coating when they come in contact with reactive gases and semivolatile species or when they undergo cloud processing (e.g. Tang et al., 2016). Coatings can influence the ice nucleation (IN) efficiency of mineral dust in different ways. Water-soluble coatings reduce the critical supersaturation required for cloud droplet activation. Coatings may chemically change the mineral surfaces. Moreover, coatings open up a pathway for immersion freezing below water saturation in concentrated solutions. In many cases, the freezing temperature of mineral dust in solutions can be predicted by the water-activity based IN theory (Zobrist et al., 2008).

Here we show examples where this approach to predict freezing temperatures in solutions failed because of specific interactions between the solutes and the ice-nucleating surface. Our focus was on microcline, a K-feldspar known to be highly IN active (e.g. Atkinson et al., 2013). We performed immersion freezing experiments of emulsified droplets with a differential scanning calorimeter. At low concentrations of NH_4^+ -containing salts, namely $(\text{NH}_4)_2\text{SO}_4$, NH_4HSO_4 , NH_4NO_3 , and NH_4Cl , the ice freezing temperature was increased by up to almost 5 K above the value in pure water, whereas at high concentrations the ice freezing temperature was decreased below the predictions from the water activity-based IN theory for all investigated solutes ($(\text{NH}_4)_2\text{SO}_4$, NH_4HSO_4 , NH_4NO_3 , NH_4Cl , Na_2SO_4 , H_2SO_4 , K_2SO_4 and KCl). An increase in IN efficiency was also present in the case of dilute NH_3 solutions. An increase of the IN efficiency in very dilute NH_3 and NH_4^+ -containing solutions followed by a decrease with increasing concentration was also observed for sanidine (a K-feldspar) and andesine (a Na/Ca-feldspar), as well as for mica and kaolinite. This indicates the presence of specific chemical interactions between solutes and the feldspar surface which are not captured by the water activity-based IN theory. We hypothesize that the hydrogen bonding of NH_3 molecules with surface -OH groups could be the reason for the enhanced freezing temperatures in dilute ammonia and ammonium containing solutions as they could form an ice-like overlayer providing hydrogen bonding groups for ice to nucleate on top of it. This enhanced IN efficiency might be of relevance for freezing in condensation mode when ammonium sulfate coatings on mineral dust particles dilute during cloud droplet activation.

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Significance statement

Mineral dust particles may acquire coatings during long-range transport in the atmosphere. We show here that in the case of ammonium sulfate coatings different types of mineral dusts may have an increased ice nucleation efficiency in condensation mode when the coating dilutes during cloud droplet activation.

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