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A laboratory study of the nucleation kinetics of nitric acid hydrates under stratospheric conditions

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Measurements of the kinetics of crystallisation of ternary H_2O-H_2SO_4-HNO_3 mixtures to produce nitric acid hydrate phases, as occurs in the lower stratosphere, have been a long-standing challenge for investigators in the laboratory. Understanding polar stratospheric chlorine chemistry and thereby ozone depletion is increasingly limited by descriptions of nucleation processes. Meteoric smoke particles have been considered in the past as heterogeneous nuclei, however recent studies suggest that these particles will largely dissolve, leaving mainly silica and alumina as solid inclusions.

In this study the nucleation kinetics of nitric acid hydrate phases have been measured in microliter droplets at polar stratospheric cloud (PSC) temperatures, using a droplet freezing assay. A clear heterogeneous effect was observed when silica particles were added. A parameterisation based on the number of droplets activated per nuclei surface area (n_s) has been developed and compared to global model data.

Nucleation experiments on identical droplets have been performed in an X-Ray Diffractometer (XRD) to determine the nature of the phase which formed. β-Nitric Acid Trihydrate (NAT) was observed alongside a mixture of Nitric Acid Dihydrate (NAD) phases. It is not possible to determine whether NAT nucleates directly or is formed by a phase transition from NAD (likely requiring the presence of a mediating liquid phase). Regardless, these results demonstrate the possibility of forming NAT on laboratory timescales. In the polar stratosphere, sulfuric acid (present at several weight percent of the liquid under equilibrium conditions) could provide such a liquid phase.

This study therefor provides insight into previous discrepancies between phases formed in the laboratory and those observed in the atmosphere. It also provides a basis for future studies into atmospheric nucleation of solid PSCs.