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The crystallization kinetics of metastable HNO_3 Hydrates

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Nitric acid hydrates are major constituents of solid PSC- and Cirrus cloud particles. The knowledge of hydrate phases is indispensable for the understanding of radiation balances and ozone chemistry in the lower stratosphere and upper troposphere. We have developed a special preparation technique, which allows us to grow the hydrate phases from amorphous samples of well-known stoichiometry in the course of an annealing program. Preferentially the thermodynamically metastable hydrates (α-NAD, β-NAD, α-NAT, NAP) crystallize and then transform into the stable phases (NAM, β-NAT and ice) at higher temperatures. X-ray diffraction (XRD), infrared and Raman spectroscopy analyze the partially unknown hydrates. An attempt is made to elucidate the mechanisms of their nucleation and their crystal growth under isothermal conditions. Therefore, characteristic XRD reflexes of every phase are chosen and their intensity changes are monitored in time dependence. For the analysis the kinetic model of Avrami is applied. It becomes obvious in samples where ice is the dominating phase that this ice matrix has a strong influence on the nucleation and the phase change of the embedded nitric acid hydrates. Thus, the phase change from α- into β-NAT, proceeding in 20 minutes, requires 175 K at the stoichiometric composition (x=0.25) but needs more than 190 K at a composition of x=0.06. Due to the Avrami model this phase change is unequal at different concentrations, which might also have an impact on the morphology of the crystals. In the same way also the phase change from α- into β-NAD and the nucleation of α-NAD and α-NAT from the amorphous solids have been investigated.