Geophysical Research Abstracts, Vol. 8, 01997, 2006
SRef-ID: 1607-7962/gra/EGU06-A-01997

Homogeneous nucleation rates of nitric acid dihydrate (NAD) at simulated stratospheric conditions

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Polar stratospheric cloud (PSC) particles play a well-recognized role in the Antarctic and Arctic stratospheric ozone loss because they act as micro-reactors for heterogeneous activation of halogen reservoir species. The main particle types in PSCs are (1) ternary solution droplets (STS) of water, sulphuric acid, and nitric acid, (2) ice particles, and (3) solid hydrates of nitric acid, either nitric acid dihydrate (NAD) or nitric acid trihydrate (NAT).

Despite their importance for the polar ozone loss, knowledge of the formation and composition of nitric acid hydrate particles in the polar stratosphere is still fragmentary. Previous field studies give evidence of both NAD and NAT phases to exist and suggest various pathways to the hydrate formation, including homogeneous nucleation in STS particles, nucleation on pre-existing ice particles, or heterogeneous nucleation on solid aerosol particles. Most modelling studies cannot explain the observed solid hydrate formation and subsequent denitrification processes by particle sedimentation if nucleation rates are applied which have been derived from laboratory studies for the volume or surface nucleation of NAD and NAT in solution particles.

In simulation experiments at the AIDA (Aerosol Interaction and Dynamics in the Atmosphere) facility of Forschungszentrum Karlsruhe we measured NAD nucleation rates in nitric acid solution droplets. The molar composition of the solution droplets and the NAD particles were accurately retrieved from FTIR extinction spectra. Activation energies for the formation of NAD were calculated from the measured nucleation rates. A new parameterisation for the activation energy as function of temperature and the NAD saturation ratio is fitted to our data and compared to various literature data

sets. The experimental data are also compared to the results of a numerical model which includes detailed schemes of thermodynamic and microphysical processes and also considers specific wall boundary conditions of the aerosol chamber experiments.