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NAT nucleation and denitrification in the polar stratosphere

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Nitric acid trihydrate (NAT) particles in the polar stratosphere are known to influence the chemistry of ozone depletion. NAT particles, along with other liquid and crystalline particles, provide heterogeneous surfaces for chlorine activation. More importantly, they can take up significant amounts of HNO_3 from the gas phase and transport HNO_3 downward by sedimentation. This can lead to denitrification, in the Arctic typically at altitudes above about 20 km, and a re-nitrification below, at the level where the NAT particles evaporate.

The nucleation rate of NAT particles is a critical parameter for the simulation of this process. Very low NAT nucleation rates around $2{\cdot}10^{-9}\mathrm{cm}^{-3}\mathrm{s}^{-1}$ have been deduced for low NAT supersaturations from observations. In previous studies, vertical HNO_3 transport has been successfully simulated by Lagrangian 3-D simulations using a constant NAT nucleation rate of around $2{\cdot}10^{-9}\mathrm{cm}^{-3}\mathrm{s}^{-1}$, for the Arctic winters in the years 2003 and 2005. However, for winter 2009/2010, this approach does not generate satisfying results.

Here, saturation dependent NAT nucleation rates were derived from Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO), observations under the assumption that NAT nucleates heterogeneously on dust particles that are characterized by active sites with a certain occurrence probability distribution depending on the contact angle. Simulations with the Zurich Optical and Microphysical box Model (ZOMM) along back-trajectories starting from points where PSCs were observed by CALIPSO allow the parametrisation of heterogeneous nucleation rates for NAT and ice on dust, and the reproduction of the different PSC classes observed.

We present simulations by the Chemical Lagrangian Model of the Stratosphere (CLaMS) of the winter 2009/2010 applying this new parametrisation of heterogeneous NAT nucleation rates. The CLaMS simulation is initialized using a combination of MLS, MIPAS-ENVISAT and ACE-FTS data. The simulation shows good agreement of chemical trace species with observations, especially H_2O and HNO_3, which are important for the reliable simulation of HNO_3 supersaturations over NAT. This is shown by comparisons with in-situ CFH frost point hygrometer sondes and satellite based observations of H_2O and HNO_3 by MIPAS-ENVISAT, MLS, and ACE-FTS.

Comparisons of the size distribution of the simulated NAT particles with the observations by a Forward Scattering Spectrometer Probe (FSSP) aboard the high-flying research aircraft Geophysica operated in the winter 2009/2010 indicate good agreement, even though the largest observed particles are not reproduced by the simulation. The simulation also reproduces the HNO_3 redistribution in the Arctic winter 2009/2010 with denitrification and the re-nitrification peaks as observed by the ACE-FTS satellite instrument and in-situ observations by the SIOUX instrument operated aboard Geophysica.