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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Simulation of denitrification and ozone loss for the Arctic winter 2002/2003" *by* J.-U. Grooßet al.

**J.-U. Grooßet al.**

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We thank the referee for his questions that allow us to clarify some points of the paper. We consider these clarifications in the revised version of the manuscript as indicated in detail below. Further, since the submission to ACPD, all relevant sensitivity studies have now been calculated in high resolution (100 km) to avoid discussion between lower and higher resolution. In these runs we also changed the treatment of the lower boundary which is now an open boundary. This has the advantage to get more realistic ozone depletion near the lower boundary in a descending vortex. In the higher resolution runs the results with respect to the favored nucleation rate did change. We did change the revised manuscript accordingly.

## Detailed answers to the reviewers questions

**a) Model particles:** We did not show the simulated particle size distribution originally as they cannot be determined for a single location where it may be compared with observations. To deduce a smooth size distribution one has to average over the area of a circle of about 1000 km radius. It is also not trivial to derive a particle size distribution. However, it is nonetheless interesting to show a vortex-average particle size distribution. As the paper presents the first description of the sedimentation parameterization we do so in the revised manuscript. The scaling factor is is given by the ratio of particle parcels to air parcels pre volume (≈4.0) times the given NAT density, e.g. now 47 $m^{-3}$ for the reference simulation. The largest number of particle boxes was simulated on 24 December, about 145,000. This is now mentioned in the revised manuscript.

**b) STS treatment:** In our code the STS does compete with the small and large NAT particle mode with respect to uptake of $HNO_3$ from the gas phase. STS and the small particle mode (where present) are simulated in equilibrium. The large NAT particle mode, however, is calculated later in the code and has no direct feedback into the STS (besides that the uptaken $HNO_3$ is missing in the next time step).

As pointed out correctly by the referee, the 'Carslaw code' does not parameterize heterogeneous chemistry and microphysics of the high density mode (STS and NAT) below temperatures of 185 K. In our case, temperatures below 185 K are treated as if they were 185 K. However, we think that this does not have a major effect on the shown results as explained below.

In the considered simulation period the ECMWF analysis showed indeed temperatures below 185 K between December 4 and 11, but always above 180 K. The area covered by temperatures below 185 K at any altitude reaches its maximum on December 5, where it is $3 \cdot 10^6$ $km^2$ or 1.2% of the hemisphere. This maximum



ACPD

4, S3923–S3927, 2004

volume covered by temperatures below 185 K corresponds to 5.3% of the volume covered by temperatures below 195 K.

The consequences for heterogeneous reaction rates and chemistry should thus be small. Even if the heterogeneous reaction rates were larger, not much change in the corresponding chlorine activation is expected, since chlorine activation is likely to be complete for these low temperatures.

In the calculation of the vertical sedimentation velocities of the large particle mode, no temperature threshold is incorporated.

**c) small NAT particle mode:** We agree with the statements made by the referee to some extent. We cannot decide, whether the small particle mode of Fahey (2001) is only the tail end of a STS population. Even if it was only STS particles, those need to be included in the simulation. In the simulated high density particle mode there are STS and/or NAT particles. We present different sensitivity simulations in which the NAT formation within this mode is altered.

A mixed population of STS and NAT particles is certainly possible in the simulation. It does indeed occur in the simulations shown in the paper.

The point made here is that where ever a high particle density mode exists, $HNO_3$ in the gas phase and in the particle phase are in equilibrium. If a large NAT particle enters such a region, e.g. by sedimentation, the 'driving force' for its additional growth is zero. This is the case, even if the amount of $HNO_3$ in this mode is low.

**d) Noisy fields:** Part of the apparent noise in figure 2 was unfortunately caused by a small error in the interpolation algorithm used for plotting the CLaMS results on an isentropic surface. This has now been corrected. However, still the distribution within the polar vortex is simulated to be inhomogeneous. When comparing along the flight tracks, the inhomogeneity is similar in the simulations and observations, even though the absolute values do not agree exactly.



**ACPD**

4, S3923–S3927, 2004

**d) Geophysica comparisons:** The $N_2O$ discrepancy could arise due to uncertainties both in initialization and transport. The simulations shown in the revised manuscript, that are consistently calculated in high resolution, do compare better with the tracer observations. We cannot decide whether the remaining discrepancy is due to transport uncertainties or initialization. This is now mentioned in the text.

Comment on figure 7: For higher nucleation rates we simulate both higher denitrification and higher renitrification. This is the case because the amount of sedimented increases. If one had much higher nucleation rates yielding many small particles this would hinder vertical transport as those particles have small sedimentation velocities. But the model is not yet in this range.

There is also no temperature dependence of the nucleation rate in our code. Of course particles are only nucleated at locations with $T<T_{NAT}$). Up until now, no nucleation mechanism has been identified to our knowledge. Therefore, one must introduce assumptions about the nucleation rate. A globally uniform, temperature independent, volume averaged nucleation rate is of course the easiest approach. If a more realistic nucleation process would be identified, one could of course incorporate it into the model, however, from the current simulations and the comparison with the data, we cannot deduce more information about the nucleation rate. One advantage of this study is the investigation of the sensitivity of the denitrification and renitrification on the nucleation rate. As pointed out in the paper, it is most sensitive in mid December.

**e) Ozone loss comparisons** ozone loss comparison SLIMCAT/CLaMS. With the high resolution CLaMS simulation and the changed lower boundary condition, the agreement between SLIMCAT- and CLaMS did largely improve. During March, the discrepancy between SLIMCAT and CLaMS ozone loss increases slightly. In this study, SLIMCAT did use a simplified denitrification scheme (Davies et al., 2003) that was tested only for a cold winter. We added also a figure to illustrate the



**ACPD**

4, S3923–S3927, 2004

sensitivity effects of denitrification and different $Cl_2O_2$ absorption cross sections on ozone loss. For clarity and better comparability with other studies we also added the evaluation of ozone column loss to the discussion.

To our knowledge no other simulation has been published that focuses on -catalyzed ozone depletion above the altitudes where chlorine-catalyzed ozone loss takes place.

**Figures and typographical errors** have been changed according to the suggestions of referee 1. We thank the referee for pointing them out.

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Interactive comment on Atmos. Chem. Phys. Discuss., 4, 8069, 2004.