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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.

Anonymous Referee #1

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General Comments

The authors discuss the integration of an analytic expression for the coupled growth and sedimentation of NAT particles (Carslaw et al, 2002) into the Lagrangian CLaMS chemical transport model. The existing CLaMS microphysics module is retained and specifies the nucleation of NAT particles from STS as a function of the supersaturation. Model simulations of the Arctic winter of 2002/03 are performed and the resulting denitrification is compared with observations from the SOLVE2/VINTERSOL field campaign. The impact of denitrification on ozone loss in this winter is found to be small (˜10% of the total vortex-average loss at 460 K). The authors note that the unusual dynamics of the vortex in 2002/03 produced significant ozone loss at higher altitudes

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in the model due to the NOx cycle.

Brief sensitivity studies of the response of model denitrification to the assumed NAT particle nucleation rate are included and the best fit to observed denitrification is achieved using a NAT particle nucleation rate of 3.4e-6 particles cm-3 h-1. The sensitivity of the denitrification produced by the new microphysical scheme to competition from small NAT particles produced by freezing of the STS aerosol in the existing scheme is also explored.

The inclusion of a microphysical denitrification scheme into CLaMS represents a significant step forward in the ability of the model to accurately simulate denitrification and ozone loss, especially in the Arctic. This is a good study and I recommend it for publication in ACP once the authors respond to the following points:

Specific comments

a) Model particles

No model particle size distributions are shown in this paper.

How large is the scaling factor required to convert actual model particles to particles in the real atmosphere and approximately how many NAT particles would be carried by CLaMS during the coldest period in mid-December?

Approximately how large a sampling region would be required to produce a meaningful NAT particle size distribution at this time?

b) STS treatment

Does STS compete with the large NAT particles for HNO3 or is it only the small NAT mode? A brief sentence stating whether this is the case would be useful in the model description.

The orignial code developed by Carslaw et al (1995) to describe the composition of STS has a cut-off at 185K (or Tice -3K). Have the authors made allowance for this in

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such a cold winter?

c) The small mode in Fahey (2001), Larsen (2004) and Deshler (2003) etc and the need for 2 NAT modes/schemes in CLaMS.

It is clear in the observations made by Deshler et al. (2003) and Larsen et al. (2004) that solid PSCs are more abundant and often form mixed clouds with STS, whether the observations occur in mountain waves or not although the sampling of the vortex by balloon-borne instruments is necessarily sparse.

Whilst there is some discrepancy in the particle number densities (˜10-3 to ˜10-4 cm-3) and sizes (˜1-3 um radius) between the different observations detailed above, is it clearly the case that these particles form a separate discrete mode to large NAT particles observed by Fahey given the different histories of the sampled air in each case?

The small mode shown in Fahey et al (2001) does not necessarily support the notion of a small mode of NAT particles in co-existence with a large NAT mode. It is a product of the analysis of NOy data from the forward-facing inlet. The small mode appears monodisperse (N˜0.002, D˜3.5 um). It is unlikely that such a distribution really exists in the atmosphere as it would be expected to grow at least somewhere during the entire observation period. More likely, the small mode is the tail end of a larger population of particles - probably background sulfate/STS. Solid particles may be present within this mode but there is no way of knowing from ER2 NOy data.

The volume-averaged NAT growth scheme used in CLaMS seems to do a good job of simulating the denitrification, given the uncertainties in NAT nucleation. Is there really a need for the additional scheme to generate a synoptic small NAT mode?

The conclusions to this paper state that the high particle density mode reduces the growth of the large NAT particles. Given that this mode contains only ˜0.2 ppb of condensed HNO3, why is this the case?

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d)Noisy NOy fields

Figs 2 and 5 shows the model calculated denitrification on two separate days during the winter. As noted in the text, the NOy fields are extremely inhomogeneous in both space and time. The Lagrangian/Eulerian approach to the simulation of denitrification used by the Carslaw group produces a degree of smoothing to the simulated NOy. The full Lagrangian transport scheme in CLaMS may be better able to reproduce any fine structure in observed NOy. Does the spatial distribution of the observed NOy from SIOUX demonstrate a similar degree of inhomogeniety to CLaMS?

d) Geophysica comparisons

Is the N2O discrepancy between CLaMS and HAGAR at low N2O attributable solely to transport or is there likely to be a significant contribution from errors in the tracer initialisation?

The agreement between model and observed denitrification is generally very good. Fig 7 - it seems that in order to obtain the best renitrification, the higher nucleation rate causes a deterioration in the simulation of denitrification above 470K.

It is worth noting that the nucleation rates used in this simulation are volume-averaged rates. There is no temperature dependence of this rate in Carslaw et al (2002). Is this also the case in CLaMS?

If so, could the introduction of a temperature dependent rate of NAT nucleation account for the observed higher NAT number densities which are indicated by the ballon-borne observations and improve the comparison with the Geophysica?

e) O3 loss comparisons

CLaMS demonstrates good agreement with observations of O3 from both MIPAS and FOX.

The differences in the denitrification schemes between CLaMS and SLIMCAT are used

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as a possible explanation of the discrepancy between the models' calculated O3 loss. What is the difference between the schemes in the 2 models?

Do other models show similar magnitudes for NOx-induced O3 loss above 600K in February and March?

Figures

Fig 4 - the 2 red lines are difficult to distinguish apart. The colour of 1 of them should be changed. To a lesser extent, this also applies to Fig 3.

Fig 5 - The white contour region is presumably $<$-9 ppbv? What is the vortex edge criterion? The pink circle could be thicker.

Fig 8 - State which nucleation rate is used in the caption.

Typography

P8071 L2 : remove 'the' P8072 L2 : replace 'at reproducing' with 'to reproduce' P8076 L20 : replace 'did barely sample' with 'barely sampled' P8080 L3 : delete one 'show' P8080 L5/6: delete 'a quite'; change 'as' to 'to' P8080 L17/19: re-phrase sentence e.g. 'We conclude that this discrepancy between model and observed renitrification is due to the resolution of the model or uncertainties in the analysed winds and temperatures.' P8082 L23: The sentence needs re-phrasing.

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