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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Implementing growth and sedimentation of NAT particles in a global Eulerian model" *by* M. M. P. van den Broek et al.

Anonymous Referee #2

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In their paper, van den Broek et al. disscuss a non-equilibrium approach to implement the growth and sedimentation of NAT particles in a state-of-the-art 3D chemistry transport model. They use two different methods to transport NAT particles size dependent as tracers in the Eulerian CTM; in one approach, the number density of particles per size bin is fixed (`fixed dens`) and in the other the radii of the size bins are fixed (`fixed rad`). They compare their results in terms of mean particle sizes with a Lagrangian model and observations of the 1999/2000 winter, with equilibrium calculations and they perform sensitivity studies of the simulated denitrification.

The attempt to implement a simple non-equilibrium parameterization of NAT particle growth and sedimentation in a global CTM is a new approach and one of the fundamental missing issues in PSC science. A successfull parameterization of denitrification in global CTMs can help to improve the understanding of polar ozone depletion and the

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prediction of ozone loss. Therefore this paper presents an important effort to deal with this question. The paper is generally well written and should eventually be published in ACP. However, I will present two major topics, which should be addressed by the authors before publication.

1) Denitrification

To my understanding, one of the major purposes of the implementation of NAT particles in a CTM is to simulate de- or renitrification in a CTM and to improve the HNO3 concentrations in the CTM.

Although the total NAT particle number density in the model is fixed to the observations by Fahey et al on 20th January 2000 over the Arctic (2.3*10-4 cm-3), the extent of the denitrification in the model (here simply defined as percentage change in HNO3 (against equivalent latitude) per simulation period of 10 days) depends strongly on the choosen model approach (nbin or r fixed). In the cold period, there are differences by up to 17% in the ten day simulation period, which could increase during a whole winter simulation (Fig.6). Furthermore in the cold period around 20 January, the (`fixed dens`) approach results in significantly more denitrification whilst in the warm period around 26 February the (`fixed rad`) approach leads to larger denitrification.

Therefore I strongly recommend to compare those results with observations. There are plenty HNO3 measurements during the Arctic winter, (e.g. Fahey et al, Science, 2001, Northway et al, jgr, 2002, Popp et al., grl, 2001, Kleinböhl et al, jgr, 2003, or satellite observations). This could lead to a conclusion, which approach `fixed rad` or `fixed dens` is favored.

2) Dependence on total NAT number density

There is a strong dependence of the simulated 10 days denitrification on the total NAT number density Nbin (Fig.8) in both approaches. Changing the NAT number density from 1*10-4 to 4*10-4 cm-3 results in a 17% difference in the 10 day denitrification. I

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encourage the authors to find a total NAT number density, which can reproduce the measured denitrification a) during the 10 days simulation period and b) during a whole winter run. Given the variability in the meteorological conditions in different winters, does this total NAT number density vary from year to year? How can predictions in a cooling stratospheric climate then be made?

In the atmosphere, the NAT number density strongly varies, depending on temperatures, timescales below TNAT and NAT supersaturations. Could you detail more explicitely, how particle formation is dealt with in the model, I assume, there is no NAT nucleation rate?

Minor comments:

3) constant initial HNO3 and water profile

The authors chose a constant initial HNO3 and water profile. How does the use of more realistic HNO3 and H2O profiles change the simulations? Maybe also uptake of HNO3 in ternary solution particles (STS) should be considered, as this strongly changes the available HNO3 gas phase at low temperatures.

Technical comments:

P 3090 L13 (`Fixed dens`) per size bin and...

The model results concerning denitrification should be stated more explicitely in the abstract.

P 3090 L23 Definition of denitrification: for exapmle: irreversible removal of HNO3 from the lower stratosphere (Fahey et al., Science, 2001)

P 3091 L20 bi-modal

P 3094 L8 Is a time step of 900 or 1800 sec appropriate for the NAT growth and sedimentation calculations?

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P 3097 L4 Is there an effect, that sedimentation is calculated after particle growth? Could you comment on the nummerical diffusion in a winter run?

P 3108 L25 $<$8%

P 3109 L19 agree (not really favorably)

P 3109 L25 The comparison with the measured range between 20 and 60% denitrification is rather crude and not sufficient. This should be done in more detail.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 3089, 2004.