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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 #1**

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Review of 'Implementing growth and sedimentation of NAT particles in a global Eulerian model' submitted to ACPD by M.M.P. van den Broek.

General Comments

In this paper, the authors use the analytic expression for the coupled growth and sedimentation of NAT particles derived by Carslaw et al (2002) in the Eulerian TM5 model. This contrasts with the Lagrangian approach of Carslaw et al where the growth and sedimentation of individual NAT particles were calculated along trajectories and resulting changes in nitric acid concentrations were passed to the Eulerian SLIMCAT CTM for gas-phase advection. As the authors correctly identify, the coupled Lagrangian/Eulerian approach adopted by Carslaw et al is computationally expensive and may not be suitable for performing multiannual simulations in climate models. Obser-

vations by Fahey et al (2001) of large NAT particles at abundances sufficient to cause denitrification of the Arctic stratosphere indicate that existing thermodynamic equilibrium treatments of denitrification by models may be physically inaccurate, requiring the use of more complex kinetic growth algorithms to accurately simulate denitrification caused by the sedimentation of these particles.

Two different approaches are adopted in this paper to overcome the numerical difficulties associated with a full Eulerian description of NAT particle transport : 1) assuming a fixed number density of NAT per size bin ('FixedDens') in which a change in bin mass is represented as a change in particle radius and, 2) assuming a fixed radius in each bin ('FixedRad') in which the change in bin mass is represented by a change in the number density of particles. The benefits of and limitations to each approach are identified and discussed concisely.

The results from three 10-day idealised simulations during the winter of 1999/2000 are compared with those obtained by Carslaw et al (2002) for the same periods. A number of sensitivity studies investigating the effect of prescribing the total number density, the number of size bins and the shape of the assumed size distribution are included. The model was sensitive to the total number density of NAT particles assumed for each simulation but demonstrated lwer sensitivity to other parameters. The particle size distributions produced by the model tend to be larger than those observed by the ER-2 or produced in the equivalent Lagrangian model. Comparisons with an equivalent equilibrium model in which the NAT particle fall velocity is prescribed demonstrate significant differences in the vertical distribution of denitrification.

This scheme offers the potential for the realistic simulation of Arctic denitrification by large NAT particles in an Eulerian framework which would be of value to the atmospheric modelling community in their predictions of future Arctic ozone loss. In my opinion the paper is generally fairly well written and should eventually be published in ACP. I have detailed below a number of points which should be addressed by the authors before publication.



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Specific comments

a) Particle sizes produced in the model.

The model produces particles which are significantly larger than either observations or the equivalent Lagrangian model. Particle diameters of 24 microns are reported in January for both approaches although the 'FixedDens' approximation is worse in this respect. Maximum observed particle diameters were around 20 microns in diameter for the same time period. This discrepency is evident in the sensitivty study (Figure 3c and d) where 30 micron particles are produced and in Figure 4. It should be noted that the particle sizes described in Northway et al. (2002) and Carslaw et al. (2002) relate to a subset of the whole model domain close to the flightpath of the ER-2 on January 20.

It takes approximately 5 days to grow a 20 micron NAT particle assuming nucleation at 25 hPa. During this time such a particle will have sedimented around 6kmassuming a typical non-peturbed distribution of nitric acid and water. In order for a NAT particle to grow from 20 to 24 microns under such conditions requires a further 2 days. Fall velocities of such large particles are $>$2km per day which indicate that temperatures along particle trajectories must be below Tnat for around 8 days and 10 km vertically. The non-linearity of this process means that it is very difficult to grow NAT particles beyond 20 microns, however low the number density.

It would be useful to see how the particle size distributions change with time during the 10 days of each model run in order to understand the final model particle size distributions shown in this paper.

This feature of the 'FixedRad' and 'FixedDens' assumptions within the model may be of less significance if the model is capable of reproducing the magnitude and spatial distribution of denitrification in the course of an Arctic winter.

b) Comparisons of the 'standard run' with the Carslaw et al. (2002) study and ER-2



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particle obervations.

Figure 4 and the discussion thereof in Section 4.1.1 describe the mean particle diameters from the 'FixedDens' model run at 96 hPa. It is important to note that the equivalent diagram in Carslaw et al. (2002) is shown for the 420 - 440 K potential temperature region - the cruise altitude of the ER-2 where the in-situ particle measurements were made. There is a significant altitude discrepancy ($>$2km) between the model field shown in this study and the altitude of observations (˜60 hPa). The mean radius (and number density) of NAT particles is likely to be sensitive to the altitude at which they are sampled, especially at the lower levels. Figure 5 actually shows this sensitivity clearly. I suggest that the appropriate model level(s) corresponding to 420-440K be sampled when comparison is made to observations and previous work.

c) Denitrification

The location of the region of nitrification is governed not only by the radius (and hence fall velocity) of NAT particles but also the altitude of the bottom of the NAT region which may also vary with time. There is clearly less nitrification at 380K in Fig 6b and 6c when compared with Fig 6a. It not evident that this is due to nitrification at a higher altitude due to a reduction in mean particle radius and therefore fall velocity alone. A change in the altitude of the bottom of the NAT region could also contribute.

I think it is misleading to conclude that the denitrification producd by the sedimentation of NAT in this model is realistic because the three 10-day idealised simulations produce denitrification in the range observed by the ASUR instrument and the ER-2 NOy instrument, as stated in the conclusions. Temperatures were conducive to PSC formation for around 6 weeks prior to January 20, 2000 and the model HNO3 field in completely unrepresentative of the real atmospheric HNO3 distribution prior to denitrification.

The authors correctly state that a model run of the entire winter using realistic initial HNO3 profiles should be undertaken as an initial test of the model denitrification scheme. It seems to me that the greatest potential of this Eulerian kinetic denitrification



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scheme lies not in the prediction of NAT particle size distributions but in the computationally efficient simulation of realistic seasonal denitrification by large NAT particles in the polar lower stratosphere.

I would like to see a model simulation of whole 1999/2000 winter in order to assess the ability of this scheme to reproduce observed denitrification.

I suggest that the authors consider simulations of other cold winters in future work where frequent observations of denitrification exist. The Arctic winter of 2002/03 would be a good candidate.

d) Introduction

Denitrification is not necessarily considered to be more important in the Antarctic than the Arctic. It is certainly a more prevelant and intense feature of the Antarctic vortex but it has been suggested that the impact of denitrification on ozone loss may be less due to persistence of low temperatures into springtime and continuing activation of halogen reservoirs on sulphate aerosol e.g. Portmann et al (1996).

The agreement between thermodynamic equlibrium denitrification schemes and observations in 1999/2000 may indeed be fortuitous, which is certainly the case in Sinnhuber et al. (2000). However, the unusually prolonged period of low temperatures during this winter may mean that much of the vortex was denitrified to levels approaching equilibrium HNO3 values which may limit the scope for discrepancy between denitrification schemes whatever their underlying assumptions.

e) Section 2.2 :

Are the results of the box model simulation described here shown in this paper? Please state 'not shown' if appropriate.

The authors should explain a little more about the method used in the model to limit vertical numerical diffusion - the 'first-order slopes' approach.



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f) Sections 3.2 and 3.3 of the paper are difficult to follow.

The text indicates that 'n_bin' is used to set a threshold for the 'base run'. This is a feature of the 'FixedRad' assumption. Is that correct or have I mis-interpreted the description?

Initial discussion of the results of sensitivity studies (shown in Figure 3) are found in Section 3.3 whilst more detail is found in Sections 4.2.2-4.2.4.

I was under the impression that the value of 2.3e-4 cm-3 derived from ER-2 data related to particles having diameters greater than 5 microns - the so-called large mode (Northway et al, 2002). The first two model particle bins range from 0.2 to 4 microns and would constitute the small mode in the ER-2 data. Can the authors explain this apparent discrepancy?

The text states in Section 3.2 that in Figure 3a shows that the particle number density in the largest bin was redistributed to all other bins because these particles were unrealistically large when compared with observations. However, the black line in Figure 3a indicates ˜2e-6 particles cm-3 in the largest bin. Can the authors explain this?

Why is it valid to redistribute particle number density from larger bins to smaller bins in a particle growth model to obtain agreement with observations?

How is the number density of these large particles re-distributed to the remaining bins?

Figures 3e and 3f are difficult to read and interpret - the colours listed in the legend do not accord with the plot in 3f or the text in Section 4.2.4. I suggest these final 2 plots (e and f) be amended to aid readability.

What is the source of the observed size distribution plotted in Figure 3f?

How does the 'standard run' (section 4.1) relate to the base run?

g) Section 4.1.1



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Differences between 'FixedRad' and 'FixedDens' particle diameters are attributed to 'atmospheric mixing processes'. What exactly is being referred to by this term?

h) Section 4.2.1: Comparison with the equilibrium approach

In addition to considerable differences in the vertical distribution of denitrification between equilibrium and kinetic model denitrification schemes, there appears to be a significant difference in the horizontal distribution of denitrification too (especially between Figures 7a and 7c). The authors have not commented on this feature.

Applying a large assumed radius e.g 7.25 microns (and rapid fall velocity) to the calculated equilibrium amount of condensed nitric acid is bound to overestimate denitrification as ER-2 and MASP observations show a significant proportion of the condensed nitric acid is present in a small particle mode with a negligible fall velocity (e.g. Northway et al., 2002).

i) Section 4.2.2

The model is clearly very sensitive to the total number density of NAT particles assumed in these simulations. The uncertainties in the measurements (30%), their timing (after the peak denitrification period in 1999/2000) as well as the mechanism(s) of particle nucleation mean that it may prove difficult to use a single value which is valid throughout the winter or between different winters.

j) Section 5 : Discussion and conclusions

A figure of 17% is quoted for the change in denitrification over a 10-day period when the total particle number density is increased by 40%. To which run is this value related?

k) Uptake of HNO3 by STS should be included in any full winter simulation of denitrification by large NAT particles. Although STS have a higher vapour pressure than NAT, their significantly higher abundance means that local uptake of HNO3 may dramatically reduce the availability of gas-phase HNO3 for NAT growth in the short term when STS are present.



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Typography

'cold temperatures' should be replaced with 'low temperatures' throughout the text.

Page 3091 line 20: 'bi-model' should be 'bi-modal'

Page 3092 line 24: replace 'is' with 'may be'.

Page 3092 line 25: By 'complex atmospheic models' do you mean CCM/GCMs?

Page 3095 line 11: replace 'a Eulerian' with 'an Eulerian'

Page 3100 line 7: Should '47K' be '475K'?

Page 3101 line 22: 'the fraction of total NAT' should be 'the mass fraction of total NAT'.

Page 3102 Equation 3: 'n_tot' should be defined.

Page 3104 line 27: 'As foreseen' should be removed.

Page 3105 line 16: replace 'criterium' with 'criterion'

Page 3105 line 18: Is 80N referring to latitude or equivalent latitude?

Page 3106 line 5: 'Similar differences' should be replaced with 'Similar changes in the vertical distribution of denitrification'.

Page 3107 line 12-28: Don't use 'tracers' and 'size bins' interchangeably, stick with size bins unless you specifically need to refer to tracers.

Page 3110 line 4 : By 'extent' do you mean the magnitude or the spatial distribution of nitric acid?

Page 3110 line 11 : Does the comaprison of 'FixedDens' and 'FixedRad' refer specifically to the denitrification produced by these schemes or is this a more general statement?

Page 3111 line 7: replace 'surplus' with 'additional'.



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Figures

Figure 3: The plot does not indicate which day of the simulation is shown in the size distributions. Nor is the sampling domain indicated - are these size distributions at 96hPa?

Figure 5: Labels a-f are not easily read. The text size needs increasing.

Figure 7: Is this latitude or equivalent latitude? Is the nitric acid mixing ratio shown here the gas-phase value or the sum of gas-phase and condensed nitric acid?

Figure 10: State the source of data for the 'observed' size distribution in the figure caption.

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