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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.

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Firstly we thank anonymous referee (#1) greatly for the extensive and useful review of our paper entitled Implementing growth and sedimentation of NAT particles in a global Eulerian model, which has lead to improvements on the original draft. Detailed answers to the points and queries listed in the referees response are listed below.

Specific comments:

a.)Particle sizes produced in the model

The referee agrees that the particle sizes found using both the approaches are larger than those calculated using a Lagrangian model [Carslaw et al., 2002] and compared with in-situ observations [Fahey et al., 2001; Northway et al., 2002]. However, we wish to highlight that the maximum particle sizes found are ˜24m in radius across the range of sensitivity studies used to test both algorithms. The size distributions shown in Fig-

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ure 3 are representative of the maximal particle number concentrations which may be resident in a specific size bin and not, as suggested by the referee, the actual size distributions achieved at the end of the 10-day simulations. We realize that this has not been described clearly enough in the manuscript and have changed this accordingly. A more detailed explanation can be found under point f in this reply. We further agree with the referee that a temporal change in particle size with respect to height during the 10-day run would be useful for the interpretation of the particle growth and evaporation. Therefore, we have added an additional figure (new Figure 5), showing the vertical distribution of the average particle size. The maximum vortex value is shown for each time and altitude. This new figure provides the reader with further insight into the rate at which large NAT particles form and highlights the differences exhibited by both approaches. Additional text has also been added related to the reasons for the simulated differences.

b.) Comparisons of the standard run with Carslaw et al. [2002] and ER-2 particle observations

The referee highlighted correctly that our comparisons with the results of Carslaw et al [2002] were performed at an incorrect altitude. Therefore, we have now revised Figure 4 so that the orthographic cross-sections of the NAT particle field are interpolated onto a 430K surface, instead of using the original surface, whose altitude we agree was too low. This leads to a smaller difference between our results and those of the previous study by by Carslaw et al. [2002], i.e. there are smaller NAT particles resident at = 430K. The reader is referred to section 4.1.1 for more detailed explanation.

c.) Denitrification of the vortex

The referee stated that analyzing the (de)nitrification at two fixed potential temperature levels (475 K and 385 K), as shown in Figure 6, does not given an accurate overview of the extent of the repartitioning of HNO3. We agree on this point with the referee and included different theta levels in Figure 6, which now represents HNO3 changes

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in a 2D theta equivalent latitude domain for all three simulation periods. This amended figure shows the maximum (de)nitrification which occurs for all three 10-day periods, thereby taking into account the lowering of the bottom of the region where NAT may exist. Moreover, it also shows the differences between both approaches more clearly, which have also been discussed in the text. However, the general conclusions drawn from the previous figure concerning the amount of denitrification and the differences between both approaches do not change.

This paper presents the results of the first sensitivity tests conducted to investigate the accuracy and robustness of both NAT algorithms. For this purpose we have restricted ourselves to three 10-day simulation periods performed for a single Arctic Winter. Our choice of the 1999/2000 winter provided us with an excellent winter for which to test our evaluation as it contained both cold and moderate periods. The motivation for our choice was that these periods allowed a direct comparison of the results with those taken from the Lagrangian study of Carslaw et al. [2002] and the ER-2 measurements [Fahey, 2001]. Generic [HNO3] and [H2O] profiles were used in a similar fashion to those used as the Lagrangian model input.

The referee questions whether a comparison with observations of denitrification over the entire winter is valid, due to the limited time periods over which the model is run. Any denitrification occurring prior to these periods, for instance, is disregarded. We agree that a full validation of (de)nitrification calculated by our algorithms can only be done by extending the simulation period and have therefore changed the strongly worded statement that our results suggest that we can fully capture the effects of NAT to that our results are in line with observations for this particular period, which are taken at a fixed point in time.

Our ultimate goal is to conduct a simulation for an entire winter, which requires a model with a full chemistry scheme active, as well as a more sophisticated three-dimensional input field of both [HNO3] and [H2O] in the presence of both Ice and STS particles. We plan to perform such integrations in the near future, as stated in the manuscript (see

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section 5). Regarding the conclusions drawn in this paper, we feel that they are instructive in giving any potential user confidence in the performance of our NAT algorithm and indicate that the number of tracers needed for transporting NAT is relatively low (thus providing an impetus for the inclusion in other higher-scale atmospheric models).

In addition to the results shown here, we have also performed simulations for 10-day periods during other, more moderate, Arctic winters. For conciseness, we excluded these from our original paper. A motivation for doing these simulations was to investigate whether using a prescribed set of particle number concentrations measured during one winter results an exaggerated amount of re-partitioning during other winters and how such simulations compare with a standard equilibrium run for more moderate temperature histories. In view of the suggestions made by the referee regarding other simulation periods we have included additional text in the discussion section summarizing the most interesting findings, i.e. that the extent of denitrification is temperature driven and our approach consistently results in a substantial difference in both the extent and region of repartitioning compared to a standard equilibrium approach.

d.) Introduction

In line with the recommendation of the referee we have added a comment to the introduction addressing the possibility denitrification in the Antarctic has a lesser impact on ozone depletion, compared to the Arctic.

The referee suggests that also during this Arctic winter, much of the vortex was denitrified to levels approaching equilibrium HNO3 values during this winter, limiting the scope for discrepancy between different denitrification algorithms. Although this statement in itself is interesting, and deserves further research, e.g. in a full winter simulation, the meteorological situation in the Arctic vortex is entirely different, with more mixing taking place and more temperature variation, even during a cold winter. Moreover, we were not able to add an example from literature to this introduction that supports this statement.

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e.) Section 2.2.

The results of the box model study were excluded from the paper for brevity, as they were identical to the results shown in Carslaw et al. [2002] and this was simply reiterated to ensure that no errors were made in the implementation of the microphysical model into TM5. The first-order slope approach refers to applying the vertical tracer gradient to calculate the downward mass flux of tracers (particles) by sedimentation. One could simply use the downward flux proportional to the sedimentation rate as calculated in the parameterization. However, this will introduce enhanced diffusion, since in reality downward mass flux depends on the vertical tracer gradient. Therefore the tracer mass gradient has been included in a similar way as in the tracer advection. This explanation has been added in this section.

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

Extensive changes have been made to this section given that the referee found it difficult to follow. In summary, we have modified the text of paragraph 3.2 and 3.3, explaining Figure 3 more thoroughly and also the nature of the model size distribution in the different sensitivity studies. As stated above, the size distributions depicted pertain to the maximum allowable particle number densities in each size bin rather than the actual size distribution achieved at the end of each simulation. Once the temperature falls below TNAT the first size bin fills immediately with particles (as a consequence of the limited radii which may reside in this bin) of radius 0.1 microns, with further growth being governed by the availability of gaseous precursors and the temperature. The NAT mass is allowed to disappear from the size bins, when temperature increases above the NAT formation temperature. For the FixedRad approach, this results in a reduction of the particle number density in all size bins. For the FixedDens method, particles from the higher size bins may move towards the lower size bins.

For the base run, we used the observed large mode value of 2.3e-4 particles cm-3 [Fahey et al., 2001] to constrain the size distribution from 0-24 microns. The referee

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states that this value represents particles larger than 5 microns, so not including the first two size bins mentioned for the base run size distribution. We agree with the referee that the total integrated number density should be used when constraining the complete size distribution. It is important to note that we only take into account the number densities for the large mode particles, also for the first bin, due to the absence of a proper nucleation rate.

In addition, the sensitivity studies yield information about the effect of total number density on denitrification (Figure 9). Doubling the total number density increases denitrification with 34%. However, it is not likely that by applying the observed number densities we have introduced such a strong underestimation and thus the effect on denitrification will probably be smaller. We have to stress that no information about the total integrated number density is available. Moreover, if available, this number could not be used given our selection criteria for the particles in the first bin (as stated above). Considering also the uncertainties in the observations (30% on the observed number densities) we feel that the base run model simulation, together with the sensitivity studies, give a good overview of the uncertainties generated by our methods. Nevertheless, we have stressed this uncertainty in the manuscript stronger, which urges the need for a proper description of the nucleation rate.

Figure 3e and f have been changed the way the referee suggests.

The source of the observed size distribution is Fahey et al. [2001]

The standard run is, in fact, the base run, which has now been changed in the text for consistency

g.) Section 4.1.1.

By the term atmospheric mixing processes we mean to say that both approaches, FixedDens and FixedRad, are affected differently by the transport of NAT, because transport directly affects the radius in FixedDens,and the number density in FixedRad.

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This fundamental difference was one of the most important arguments to test both approaches. The text has been expanded to clarify this point.

h.) Section 4.2.1: Comparison with the equilibrium approach

The differences in the horizontal (longitudinal) distribution of denitrification between Figures 7a and 7c are mainly due to differences in the extent and altitude of the denitrification calculated using the equilibrium and non-equilibrium methods. This point has been mentioned in the text, but we agree with the reviewer that it has not been linked to differences in the horizontal distribution. The instantaneous production of relatively large particles when using the equilibrium approach results in the sedimentation flux of such particles being larger for the first few days. On the other hand, the temperature history of a particular region has no effect when using the equilibrium approach (i.e. the spin-up time for sedimentation of particles to lower levels is less) meaning that repartitioning occurs much faster. This causes differences in both the vertical and horizontal distribution of HNO3 and we have expanded upon this in the text.

We agree with the referee that 7.25 m is bound to overestimate denitrification. However, because of the non-linear dependence of denitrification on particle size, it remains difficult to determine the ideal particle radii which should be used throughout an entire winter period. For this reason we chose a second particle of 4m so as to not exaggerate the differences (Figure 7d). Moreover, we have now updated the text and integrated the comment of the referee into the discussion.

i.)Section 4.2.2

In line with the referee we feel that the prescribed total number density of the resident NAT particles is the achilleas heel of all modelling studies associated with PSC formation and the use of such algorithms to predict the consequences of future changes in climate. (we also refer to the adjustments in section 3.2 and 3.3 concerning the constraints with the total number density and the related uncertainties) For this reason we urge members of the measurement community to suggest a statistically derived aver-

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age value elucidated from the data gathered from many flight tracks already available. We now state this point more succinctly in the text of paragraph 4.2.2 and also in our conclusions.

j.) Section 5: Discussion and conclusions

The 17% change in denitrification between the two approaches, which is quoted in the conclusions, is related to the sensitivity studies shown in Figure 9. However, this value appears to be erroneous when looking at the FixedDens results in Figure 9. Here the change in denitrification is ˜17% when increasing the permissible particle number densities from 0.4-1.0 x 10-4 particles cm-3 thus there is an increase of 250% in the total particle number density, which may be an unrealistic enhancement. This has been changed and clarified further in Section 5. Moreover, as stated above, we agree that uptake of HNO3 by STS should be included during a full winter run, which we have now added to our recommendations for future research.

Typographical errors:

We have amended the majority of the typographical errors listed by the referee in the final version of the paper.

Figures:

Figure 3: This plot shows the maximum permissible number concentrations per size bin across the entire size spectrum chosen for NAT particles. At the start of the model run, particles form in the first size bin only, with subsequent bins been filled upon particle growth. This is now clarified in the description of this Figure in paragraph 3.3.

New Figure 5: This new figure shows the vertical and temporal development of large NAT particles for both approaches. The Maximum grid box average for the entire vortex region are shown.

Figure 6 (Figure 5 before): The text sizes have now been increased to aid legibility.

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Figure 8 (Figure 7 before) This plot refers to latitude (with this information now added to the figure). Total HNO3 (NAT + gas phase) is now shown for all model runs.

Figure 11 (Figure 10 before): The source of the observed size spectrum is Fahey et al. [2001], which is now declared in the figure caption.

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