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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A 3D-CTM with detailed online PSC-microphysics: analysis of the Antarctic winter 2003 by comparison with satellite observations" *by* F. Daerden et al.

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Interactive comment on referee 1

We thank the referee for the review of the paper. Together with additional simulations, the referee's comments have led to a better understanding of our results. Also, the referee has raised some important issues which where undoubtedly lacking or unclear in the paper.

We will revise the paper taking into account the referee's remarks and suggestions, as will be explained in detail below.

General remarks

- Model-observation deviations above 500K.

  We agree with the referee that above 500K important differences exist between the model results and the observations. We believe that these differences have various origins which we think can be sufficiently interpreted and are not indicative of major model deficiencies. We will comment in detail on the deviations in the various quantities, following the order in which they are presented in the paper.

  1. N_2O

     Above 500K the modeled N_2O actually has the smallest deviations from the MIPAS observations. But in minor point 5 the referee raises concern that the deviations in N_2O may not be due to numerical diffusion across the vortex edge, as explained in the paper, but may rather be due to a poorly performing vertical transport (diabatic descent).

     In this context we will add to the paper the comparison for CH_4 between MIPAS and the model, which is much better than for N_2O (in the high resolution case there is nearly a perfect match until October).

     Taking into account that CH_4 is expected to suffer less from numerical diffusion (because of lower cross-vortex edge gradients), we think this is, although not sufficient, nevertheless an additional indication that the diabatic descent is modeled acceptably well.

     In this context, we would also like to mention Strahan and Polansky [2006], who showed that our high resolution is sufficient to isolate the Antarctic polar vortex, and Manney et al [2005] who showed that the operational, high-resolution forecast products which we use are well suited for detailed studies of stratospheric polar processes.

  2. Extinction (figs. 4 and 5)

     The 575K level is very close to the top of the POAM III extinction profiles, which is situated at 25 km, and results at that level should therefore be handled with more care than in the present version of the paper, in which it is treated in the same way as the other levels. The same holds to a lesser scale for the 525K level.

While PSC retrievals at the top altitudes are not problematic, the main problem lies with the background extinction. The background aerosol extinction is getting close to zero and the retrieval is dealing with very small signals. We therefore think that the estimation of the lognormal parameters of fig. 2 is unsure near the top altitudes.

From fig. 5 it can be seen that at the top levels the model extinction overestimates the POAM extinction considerably already from early May. While originally in our study this was assumed to be caused by temperature biases we are more convinced now that this deviation is due to a poorly initialized background aerosol field (especially after checking out various temperature bias correction schemes, which will be described later in this comment).

A further indication of a problem with the background aerosol distribution at the highest altitudes is that the POAM extinction is much more variable than the model extinction, POAM extinction values can become very low, down to 10^{-6} km^{-1}, while the model extinction is limited by the POAM climatological background of about 10^{-5} km^{-1}.

The uncertainties about the background distribution parameters are the largest at the 575K but due to sedimentation deviations on that level may progress into the 525K level.

Regarding the origin of the lognormal parameters of fig. 2 (see also minor point 4), this is explained already in the text. The observational data mentioned are 3 profiles measured by Terry Deshler. There is however only one of these profiles, dating from May 1995, which exceeds 23 km in altitude. Taking into account that in 1995 the background aerosol conditions were significantly different from 2003, this adds to the uncertainty in our estimate.

Besides the initialization problems, also problems related to the vertical transport of water vapor and of mesospheric NO_y have an impact on the calculated PSC extinction on the highest levels, as will be discussed below.

3. Nitric acid (fig. 6)

   We are grateful to Gabrielle Stiller for her comment on the paper regarding the enhanced HNO_3 concentrations in the polar vortex. We had become aware of the problem independently after submission of the paper. The 3D CTM indeed has no description of NO_x-creating processes in the mesosphere and lower thermosphere (MLT). As a consequence the model does not reproduce the second HNO_3 maximum reported by Stiller et al [2005]. This maximum descends down to the top levels presented in the paper (525K-575K) only from mid-July onwards. But as can be seen from figure 6 the model already underestimates the MIPAS HNO_3 values from early June onwards. This early underestimation is likely due to the problems with the background aerosol and PSC formation at these levels (see previous item), where due to a poor background aerosol initialization the model overestimates the extinction and removes too much nitric acid.

   With the information provided by G. Stiller we now can assume that the deviation between model and observations from mid-July onwards is mainly controlled by the downward transport of the second HNO_3 maximum. Indeed the differences between the model and MIPAS during this period and at the 575K level are $\sim$5ppbv, comparable to the values reported by Stiller et al [2005].

   We also want to add here that the differences between model and MIPAS in the first few weeks of the simulation are likely related to the model spin-up. As mentioned the model simulations were initialized by an assimilated MIPAS field, but for the assimilation a different background aerosol climatology was used from the one we use in the paper. As is known, the background aerosol distribution has an influence on the NO_y field, see e.g. Kühl et al [2002]. We attribute the evolution of HNO_3 during May to the fact that the

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model has to reach a new equilibrium state. To overcome this problem we have initialized the simulations on April 1 rather than on May 1, again from a MIPAS assimilated field for April 1 now. This gives the model one month of time to spin up, and indeed the situation for HNO_3 is much better in these new simulations. We will use these new simulations in the revised paper.

4. Denitrification (figs. 7, 8)

   The deviations at the top levels in the calculated denitrification (fig. 8) are partly related to the problems in HNO_3 described above.

   The referee is correct in minor point 7 regarding the NO_y-N_2O relation used for the calculation of the denitrification. First of all, with the new simulations starting in April, the NO_y-N_2O relation is much more comparable to the MIPAS data. When looking in detail there are deviations from the fit used, and we think it is better to do a fit by ourselves of the NO_y-NO_2 relation and calculate the denitrification according to this new reference fit. The result of our curve fit is:

   NO_y = 24.2943 - 0.0647*N_2O - 3.4059 10^{-5} * N_2O^2

   The linear coefficient of 0.0647 is very comparable to previously reported values for the Antarctic, see e.g. Fonteyn and Larsen [1996] and references therein.

   The old relationship did not fit well the correlation at low N_2O and high potential temperature values. This explains to a large extent the deviations at the highest levels in fig. 8. The new relationship gives a better fit in this region, especially at 525K. The remaining deviations on 575K are a consequence of the deviations in HNO_3 (see above) but could also partly be caused by the fact that this level is near the upper limit of NO_y-N_2O curve fit.

   In the revised paper denitrification data in fig. 8 will be replaced by the one calculated using the new NO_y-N_2O relation.

5. POAM water vapor (fig. 10)

The model has problems to reproduce the POAM water vapor field at the levels above 500K. The origin of this problem can for the largest part be explained by the initial conditions at high altitudes. Although the initial model water vapor field was tuned to the POAM water vapor, as explained in the text, this tuning was done rather conservatively at altitudes above 25 km. Differences of 1.5 ppmv or more exist in the initial (tuned) model field and the POAM data at altitudes between 35 km and 40 km, and these can explain the differences in August at the 525K and 575K levels due to adiabatic descent.

6. Ozone

   The deviations in the ozone fields are principally related to numerical diffusion, as has already been addressed in the text.

- Comparison to the papers by Höpfner et.al.

  These papers appeared at a time when the work for our paper was already at a well developed stage and that is why we - although certainly aware of their appearance - did not extensively compare our results with theirs. Another important reason for this was the fact that the paper by Höpfner et al. (2006b, called H06b hereafter) focused on the importance of mountain waves, while we have not yet introduced mountain wave corrections in our simulations. The main goal of the present paper is the presentation of the coupled system and of some first results, and it would be out of scope to add considerable (yet interesting) complexity to the system at this stage.

  Nevertheless we fully agree that the 1st referee has a valid point that results should be compared to the important paper H06b. As this issue is repeated in major point 1, we will address it in detail below.

Major points

1. The model used in the paper is basically the same model as that used in H06b, but here the Eulerian version is used. There is a switch to turn on or off the homogeneous surface dependent freezing of NAD above T_{ice}. Both cases have been studied in H06b and we recently have done additional simulations with this freezing mechanism turned off.

   The comparison of our results with H06b points to four issues:

   (a) first appearance of NAT in the 2003 Antarctic winter
   (b) influence of temperature corrections
   (c) the evidence (or absence) for the homogeneous surface dependent nucleation of NAT above T_{ice}
   (d) the presence of NAD

   (a) We confirm the results of H06b that with the inclusion of the homogeneous NAT formation above T_{ice}, even with considerably reduced freezing rates (1/100 in our case), NAT forms already during May, too early as derived from the MIPAS observations in H06b (June 11). However this NAT presence is very limited and the first considerable formation of NAT takes place around June 6, as can be seen in figure 5. This is much more comparable with the results of H06b. (We will include some horizontal maps in the revised paper to illustrate this.)

   The contribution of NAT to the total extinction is very small before June 6. This indicates that generally the clouds have a mixed composition, with STS the predominant particle type and only very small amounts of NAT. While in the fast cooling mountain wave processes we expect small NAT particles to form, slow synoptic cooling (as is expected to take place in our simulations) would preferentially generate large NAT particles. H06a and H06b state that MIPAS has difficulties to see NAT particles larger than 3 μm, and in some

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cases to distinguish between ice, STS and large NAT. This is the case on June 5 and June 9, as can be seen in fig. 2 of H06b. Taking also into account that MIPAS was not active from May 25 to June 4, we think our results regarding the first appearance of NAT do not necessarily contradict those obtained in H06b. The difference in date regarding the first apparition of considerable amounts of NAT ($< 3\mu$m), 11 June as seen by MIPAS vs. 6 June in the model, can perhaps be explained by either the size limitation of the method of H06b or by biases in the temperature, which are known to exist (see next point).

(b) We have performed additional simulations applying various temperature bias-corrections. More specifically we have applied the monthly mean bias corrections which H06a derived for the McMurdo station, and which were kindly provided by M. Höpfner, to the entire polar region, and separately we have also applied latitude-resolved monthly mean bias corrections derived from CHAMP radio occultation measurements, which were kindly provided by U. Foelsche [Foelsche et al, 2006].

Our conclusion is that for a 3D Eulerian system, temperature bias corrections need to be more detailed than the monthly averages which are currently available. The latitude resolved data from Foelsche et al [2006] are already a step forward but both the temporal as well as longitudinal resolution need to be increased for them to have any positive effect on the results. Often these corrections have a standard deviation which surpasses the actual value of the correction, and from our simulations we observe that applying these coarse corrections can improve the situation on one location but worsens it on another one. This is possibly due to the nature of the biases in the ECMWF fields (Are they zonally symmetric? Are they constant over one month?), and of course deviations may arise because we apply these bias corrections to ECMWF short-term forecast fields while they were derived from the ECMWF analyses.

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To avoid additional complexity we will not include in our revised paper any results by temperature-corrected simulations.

We want to stress that this problem is intrinsic to our approach, as will also be discussed in our response to referee 2. The difference with studies on trajectories is that trajectories are relatively short in time (10 days) and are all independent to some degree. In a full 3D approach the entire polar - and beyond - region is interacting through the transport, and eventual errors created in the simulations (e.g. the too early creation of an ice cloud due to a bias in the temperature) can propagate throughout the system, eventually over a considerable period of time. The combination of diffusive transport and complex microphysics makes the system very sensitive.

(c) The question of a freezing mechanism for NAT above T_{ice} has been addressed numerous times already in the literature. It has been discussed before in e.g. Svendsen et al. [2004]. It is our opinion that currently we are not able to assess details of the NAT nucleation. There is observational evidence in the Arctic that NAT can form at temperatures above T_{ice} [Larsen et al, 2004; Voigt et al, 2005]. However, we cannot say if this occurs through heterogeneous nucleation, through NAD and (rapid) conversion to NAT, or directly to NAT. We only use the parametrisation of Tabazadeh with the applied corrections because this is presently the only available parametrisation, based on laboratory measurements.

In point 1 we concluded that working with the NAD/NAT homogeneous freezing mechanism with reduced freezing rates does not necessarily contradict the first appearance of NAT as derived from MIPAS. Additionally, and perhaps surprisingly, we have verified that either working with reduced homogeneous NAD/NAT freezing rates (1/100 or lower), or with this mechanism completely turned off, actually does not lead to much different scenarios. See also next point and minor point 3.

(d) The fact that MIPAS does not see NAD, as found by H06b could be due

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either to the absence of NAD formation, or to a very fast transition from NAD to NAT - too quick to be detected.

2. The referee is right that we have the ability to present the various fields and quantities in great detail, but we deliberately opted for a concise presentation of results.

   However as the need for more detailed plots exists we will add some selected relevant horizontal maps to illustrate the horizontal distribution of various quantities, e.g. the formation of the NAT belt in the model.

3. After reducing the theoretical homogeneous surface-dependent NAD nucleation rates by 100, the highest values we obtain in the simulations are typically of order $\sim$10 m^{-2} s^{-1}. This leads to rates which are very comparable to the ones measured by Voigt et al [2005], i.e. $\sim$10^{-6} cm^{-3} h^{-1} for particles of a few μm radius. For the largest particles these rates can become 100 times larger.

4. The intent of this line was to express that the model has the possibility to simulate qualitatively and quantitatively the influence of microphysical parameters (e.g. freezing rates) on global fields such as denitrification and ozone loss. This comes from the direct coupling of a microphysical model to a 3D global model.

   We will leave out this line from the abstract because this issue is not explored further in this paper anymore. Indeed referee 2 encourages us to use the reduced freezing rates without further addressing this discussion anymore.

Minor points

1. This is correct, but we did not take into account e.g. a temperature criterion to select profiles for calculating the vortex averages presented in the paper. This

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means that e.g. in the plot for HNO_3 both the effects of deNO_xification and denitrification are involved, but we think it is not evident to leave out the effect of deNO_xification by a simple profile selection criterion based on temperature. We verified that either applying a profile selection criterion or not does not qualitatively modify the model-observation comparisons.

Regarding the effect of contamination by PSCs we think that this is largely taken care of by the error-weighting in the calculation of the averages, because the retrieval of profiles in the presence of PSCs will increase the retrieved observational error.

2. We will include this information in the paper.

3. Meijer et al [2004] illustrate the clear advantage of using forecast fields regarding atmospheric transport in 6-year simulations. They are less diffusive. But the referee is correct that the advantage over the comparably short period of the simulations presented here (9 months) is unclear. Our intent is to do multi-annual simulations and that is why we used the forecast fields also in this shorter timescale study.

   Regarding the vertical resolution of these levels, there are 10 vertical levels between 10 hPa and 100 hPa, the resolution at 50 hPa is about 10 hPa. In May in the Antarctic, the vertical resolution in altitude is 1.25 km above 21 km, and decreasing below to 1 km at 15 km. At the same time and place, the vertical resolution in potential temperature is around 25 K or less below 550 K.

   Regarding the vertical velocity, and to clarify various transport issues in general, the following lines will be added to the paper.

   The advection scheme is the flux form semi-Lagrangian transport (FFSLT) algorithm of [Lin and Rood, 1996], with monotonicity constraints that allow no undershoots or overshoots in the horizontal directions, and only overshoots in the

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vertical direction. This choice, noted IORD=JORD=3 and KORD=5 in the nomenclature of the FFSLT scheme, is necessary when the vertical resolution around the tropopause is coarse, as is the case for our model. This is also discussed in detail for the GMI model in Rotman et al [2001], a model which is very comparable to our CTM.

The model uses the short-term forecasts of temperature, horizontal winds and surface pressure issued by the ECMWF operational system (cycle 25r3 and 25r4) with a 6-hour time resolution. Manney et al [2005] showed that these operational, high-resolution products are well suited for detailed studies of stratospheric polar processes. These ECMWF products are downloaded on a 1degx1deg horizontal grid, averaged in a mass-conservative way to the coarser grid and linearly interpolated in time to the 15-minute timestep of the CTM.

The vertical velocity is computed by the FFSLT scheme in order to ensure mass conservation with the ECMWF horizontal wind fields, taking into account the time evolution of surface pressure. As most CTM's where the wind fields are produced offline by a system with a different grid, we encounter the mass-wind inconsistency problem described by Jöckel et al [2001].

4. This is explained in the text, section 3.2, one but last paragraph.

5. This point has been addressed in the above under *General remarks*.

6. This issue has been addressed in section 3.1, one but last paragraph.

7. Correct observation. Point discussed above under *General remarks-denitrification*.

8. Yes.

9. We think indeed the difference in latitudinal coverage between POAM and MIPAS is the cause for the difference in the behavior of vortex-averaged water vapor.

While POAM measures along a fixed latitude belt with changing location with respect to the vortex throughout the winter and spring, MIPAS has a dense and global coverage along the entire vortex. The fact that the MIPAS plots do barely show any sign of dehydration is because the averages are taken up to the vortex edge, where there is nearly no dehydration. Taking averages within the vortex core clearly show the effect of dehydration also in the MIPAS data. We will add a plot illustrating this.

10. Will be included in the revised text (http://dmiweb.dmi.dk/pub/PSC/).

Corrections

- See minor point 3.
- This is correct, there is a newer JPL compilation that the one we used.

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