Atmos. Chem. Phys. Discuss., 8, S7670–S7677, 2008
www.atmos-chem-phys-discuss.net/8/S7670/2008/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Spatiotemporal variations of NO_y species in the northern latitudes stratosphere measured with the balloon-borne MIPAS instrument" *by* A. Wiegele et al.

A. Wiegele et al.

Received and published: 30 September 2008

We would like to thank the referee for the valuable comments which we answer in the following (referee comments are inserted *in italics*).

1. The authors state in Section 5.3.1 that model data differences with respect to temporal evolution of NO_2 around sunrise could be caused by a too slow model photolysis. However, it is stated in Section 5.1 that the box model output is written out hourly. If the model results are then interpolated to the measurement times (i.e. every 5 minutes), it is not surprising that NO_2->NO conversion appears to be slower in the model than in the measurements taking into account that the NO_2 chemical lifetime is in the order of minutes.

During the three days of modelling prior to the measurement along the calculated backward trajectories the model uses time steps of ten minutes for calculation and an output interval of one hour. During the period of measurements each time step and output of the model is similar to the temporal resolution of the corresponding measurements (i.e. about 5 minutes). This has been made clear in the text.

An additional aspect is that NO_2 photolysis rates depend strongly on the UV-vis albedo. The authors should specify whether a constant albedo (i.e. 0.4) or a variable albedo in dependence of snow/cloud cover has been applied.
For the box model calculations, a constant albedo of 0.7 has been used. This has now been specified in the text (p. 4704, line 20). Sensitivity runs with the radiative transfer model ART have been performed to characterise the influence of different ozone columns and assumed values of the albedo on the NO_2 J values shortly after sunrise. The albedo chosen has got large effect on the NO_2 J value shortly after sunrise and the sensitivity calculations with ART have pointed that smaller values (0.3 to 0.4) would have been more appropriate for this Arctic box model study. We now discuss this point in an additional section (Sect. 6) in the text.

2. The pronounced disagreement of modeled and observed N_2O_5 profiles discussed in section 5.3.2 and shown in Figure 12 is striking, in particular when taking into account that the box model was initialized by the observed total NO_y and O_3. A downward shift of the N_2O_5 maximum of about 5 km in the model compared to the observations can hardly be explained by erroneous O_3 overhead columns as proposed by the authors. In this case, increased (decreased) N_2O_5 abundances would be expected at all altitudes for lower (higher) O_3 overhead columns. Given that N_2O_5 formation during night depends strongly on temperature it would be useful to compare the ECMWF temperatures used in the trajectory calculations with independent measurements such as provided by satellite observations (i.e. MIPAS-ENVISAT).
As stated in the paper, ozone and total NO_y have been initialised with MIPAS-B data but the partitioning between the individual NO_y species has been deduced from KASIMA

ACPD

8, S7670–S7677, 2008

3-D data. The sensitivity of the modelled N_2O_5 vmr on the ozone initialisation has been investigated in our box model studies. The differences in the resulting N_2O_5 vmr are less than 0.1 ppbv. ECMWF temperatures have been compared to MIPAS-ENVISAT observations in winter 2002/03 by Ridolfi et al. (Geophysical validation of temperature retrieved by the ESA processor from MIPAS/ENVISAT atmospheric limb-emission measurements, Atmos. Chem. Phys., 7, 4459-4487, 2007). Mean differences in the lower stratosphere (below about 30 km) are generally within 1 and 2 K, dependent on altitude. The ECMWF temperatures also agree with the temperatures retrieved from MIPAS-B within 1 to 2 K and do not show a significant bias. We performed model calculations to check the magnitude of the temperature sensitivity of the nighttime formation of N_2O_5 and found that a temperature variation of 2 K alters the N_2O_5 mixing ratio by less than 5% after a built up of 12 or 24 hours in the altitude region between 15 and 31 km. Thus, initialisation problems with respect to the partitioning of the individual NO_y species and particularly the unknown liquid sulphate aerosols surface area densities are assumed to have the most important impact on the discrepancy for N_2O_5 between box model results and observations. We now discuss these issues in Section 5.3.2.

3. One of the conclusion of the paper is that the currently assumed chemistry affecting the N_2O_5, and to a lesser extent to NO_2, is "too slow". Do then the authors suggest that that chemistry should be revised? If so, I think this should be explicitly stated in the manuscript, in the conclusion section. Although, for that end, the authors should make sure that the points mentioned above are not the responsible for the model/measurements discrepancy and they should: a) check if the actual measured temperatures are different from ECMWF and hence if they could affect significantly both the absolute N_2O_5 values and its variation during the measurements period; and b) check the potential parameters affecting the photodissociation rates such as O_3 column above, albedo, and maybe also the cross-section temperature dependence.

We do not suggest that the chemistry should be revised, since a a detailed analysis of the NO_2 decrease due to photodissociation showed that the differences can at least partly be explained by the interpolation of the photolysis rates in the model and as-

ACPD

8, S7670–S7677, 2008

sumptions made for the albedo in the radiative transfer model. Thus, there is no need for a revised chemistry. We discuss these effects in a new section (Sect. 6) which has been added to the manuscript.

Specific comments and Technical corrections

p. 4694. Abstract, l. 3. Please specify which "lower altitudes"
We have changed the sentence to: "At lower altitudes (below about 22 km) ..." although there is of course no hard boundary.

l. 5. I would delete "along the cross section", e.g.., "... reveal the dynamics through the edge ..."
OK.

l. 11-12. I suggest to delete "in terms of quantity" (if it is no indicated the contrary, one would assume it is a "quantitative" agreement, as it is specified later).
"In terms of quantity" is meant in the sense that the absolute volume mixing ratio is met quite well by the model whereas the evolution in time shows some differences. In order to make this point clearer, we have changed "in terms of quantity" to "in terms of volume mixing ratios".

"... slightly too slow..."? Do you mean "slightly slow"?
No, we mean "slightly too slow", compared to the measurements.

Introduction, l. 23. Insert a "," after "$BrONO_2$"
OK

p. 4695 l8. "The reformation of NO_2 after sunset is about as fast as its photolytic dissociation after sunrise." This is valid for the middle and lower stratosphere, only.
We have changed the sentence to: "In the middle and lower stratosphere, the reformation ..."

p. 4696, l.6-7. I suggest to delete "a time period of"
OK

ACPD

8, S7670–S7677, 2008

Interactive Comment

p. 4696 Sec. 2, last par. I would suggest to change the order of the last two sentences. I think last sentence refers to first sentence of the paragraph)
The last sentence refers to the whole paragraph. We have slightly changed the wording to make this clearer.

p. 4696 l24: ... adjusted RELATIVE to the position of the sun
OK

p. 4697 l27. : Suggest to change "mesospheric" -> "thermospheric". (Thermospheric radiative contributions to the measured spectra should be significantly higher than the mesospheric contributions).
We agree to your comment. We have changed the sentence according to your suggestion.

p. 4699, line 7. "that periods" -> "those periods"
OK

P. 4699, last line and first line in the page. I suggest rewriting: In summary, the measurements taken at altitudes between 17 km and 21 km covered the edge of the polar vortex with strong horizontal gradients while weaker gradients could be expected above these altitudes, ...
OK

p. 4701. First par. I think the point made about the small difference in the temperature minimum between outside and inside the vortex, just 1 K (by the way the temperature errors are not mentioned in the manuscript) is not much relevant. The major point, I think, it is that the temperature minimum are located at significantly different altitudes inside/outside the vortex, as mentioned later. I would reduce this paragraph. In any case I would not talk about a "MORE PRONOUNCED temperature minimum" when we are talking about 1 K difference.
We have adapted the text according to the comment. Concerning the temperature minimum, we only discuss the minimum location inside / outside the vortex, and we

ACPD
8, S7670–S7677, 2008

mention the temperature error, which is the order of 1 K (1 sigma, total error), along with the differences to ECMWF temperatures (1 to 2 K).

p. 4701, l3. "Brewer Dobson circulation" -> "meridional circulation"
OK

p. 4702, Sec. 4.3. It would be useful to comment on how the observed NO_y partitioning compares with other observations.
Flights with the second MIPAS balloon instrument are carried out since 1995. Profiles of retrieved NO_y species have been intercompared to independent measurements and chemistry models many times. Some dedicated limb sequences obtained during the March 2003 flight were chosen for the validation of the chemistry instruments aboard Envisat (MIPAS, GOMOS, SCIAMACHY) and the Japanese ILAS-II sensor aboard ADEOS-II. Comprehensive validation and intercomparison activities not only for NO_y related species were performed. All studies exhibit the high quality of data measured by MIPAS-B (see, e.g., Höpfner et al., Validation of MIPAS $ClONO_2$ measurements, Atmos. Chem. Phys., 7, 257-281, 2007; Wang et al., Validation of nitric acid retrieved by the IMK-IAA processor from MIPAS/ENVISAT measurements, Atmos. Chem. Phys., 7, 721-738, 2007; Wetzel et al., Validation of MIPAS-ENVISAT NO_2 operational data, Atmos. Chem. Phys., 7, 3261-3284, 2007; Wetzel et al., Technical Note: Intercomparison of ILAS-II version 2 and 1.4 trace species with MIPAS-B measurements, Atmos. Chem. Phys., 8, 1119-1126, 2008). We have now addressed the comparison with satellite data in the paper.

p. 4702, l24. Please insert a "," after "whereas" and after "VMR peak"
p. 4704, l. 18, climatologic -> climatological
OK

p. 4705 I understand that the total NO_y used for initialization of KASIMA model (to be used later in the box model) has been inferred from the first MIPAS B "azimuth direction" only (e.g., Fig. 6). If so, a possible spatial variability of NO_y over the observed region

ACPD
8, S7670–S7677, 2008

Interactive Comment

would not be taken into account. Could this affect the model-data comparison?
The first azimuth direction was chosen for the NO_y initialisation of the model because this scan was recorded at nighttime where the (difficult to retrieve) species NO can be neglected when calculating the total NO_y budget in the lower stratosphere. We checked the variability of our observed NO_y profiles and found that the standard deviation is less than our inferred NO_y total error which amounts 5-8%. Such small variations should not be relevant for the model-data comparison.

p. 4708, l. 18, Insert a "," after "measurements"
OK

Fig 2: To underlay a PV contour at 450 or 475 K indicating the vortex boundary around 19 km would be helpful in this figure. Also drawing a line separating the night and day sides would also be useful.
We feel that this figure becomes overloaded and maybe misleading, since time and space aspects will be mixed up. Instead we have modified Fig. 3 such that the tangent points for 19.5 and 22.5 km are given in the corresponding plot. Furthermore we have extended the figure caption to: "The sunrise was between 03:40 UTC at the highermost and 04:00 UTC at the lowermost tangent points, during the measurements in the third azimuth direction from the west."

Fig. 3. Are the meridians in the figure the 0° and 30° ones?
No, they are 0° and 45°E, but this figure has now been changed anyhow.

Fig. 6. It would be useful to add different symbols to the different species. Some colors can be confused in some printings.
OK, we have added symbols.

Fig. 11 and 12. I would suggest to show also the modeled/measured differences by difference plots, e.g. in percentage. This gives a better idea about the discrepancies. In order to not increase the number of figures, the left panels of the figures could be removed. I think they do not contain much information.

ACPD
8, S7670–S7677, 2008

We have added the difference plots. However, we want to keep the left panels of the figures in order to show the evolution of the model species during the three days before the measurements.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 4693, 2008.

ACPD

8, S7670–S7677, 2008

Interactive Comment