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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.

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

Major points

1. page 4701, line 18ff, subsidence of about 1 km: Seen in the figure is an altitude shift of the N_2O contour by 1km between the first and the last observed profile. It would correspond to subsidence only if (i) the first profile is well outside the vortex and (ii) if no mixing across the vortex edge would have taken place. The observed N_2O contour should be interpreted as a lower limit for the subsidence in the polar vortex.
OK, we have included the following sentence in line 20: "It should be noted, however,

that the actual subsidence may be larger than the values derived from these measurements because the measurements do not cover the whole range from well outside to well inside the vortex at all altitudes and mixing across the vortex edge may also have an influence on the N_2O concentration."

2. page 4701, line 25ff, mesospheric intrusions: Müller et al., (JGR, 2007) clarified, that the air with the lowest N_2O mixing ratios is rather unmixed air originating from the upper stratosphere. The mesospheric origin of the air masses is visible higher up (24-27 km).
We have changed this sentence making reference to Müller et al., (JGR, 2007)

3. page 4705, line 7ff: Is the normalisation factor close to 1 or significantly different? If it would deviate from 1, non-linearities may become important.
The normalisation factor is in the order of 0.8 to 1.5. However, sensitivity studies performed with different initialisations (not shown) do not exhibit any non-linearity.

The discrepancy between observation and model NO_2 and N_2O_5 is only interpreted qualitatively. The statement that the "model chemistry is too slow" (abstract, l. 14 and p. 4709, l. 4) is very vague and should be quantified and explained better. Most important for the NO_2 decomposition at sunrise is the O_3 mixing ratio and the NO_2 photolysis rate. Model O_3 could be compared with the MIPAS-B observations and NO_2 photolysis is also rather constant with altitude (see e.g. Stolarski, 1995, Scientific Assessment of the Atmospheric Effects of Stratospheric Aircraft, NASA Ref. Publ. 1381, 1995, or Becker et al., J. Atm. Chem., 37, 217-229, 2000.). Thus in principle, the NO_2 decomposition at sunrise should be easy to model and reasons for discrepancy should be provided in a study like this. In the rather un-complex model like the used model, sensitivity studies with respect to the uncertainty of the relevant kinetic parameters would be a good way to investigate this discrepancy.
The time for the NO_2 photolysis has been be quantified and the term "too slow" in the conclusions has been replaced by "about three times slower".
Furthermore, in order to better explain the differences between measurements and

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box model results, the paper has been expanded by a section (Section 6: Photolysis rates of NO_2) dealing with the NO_2 J values which can be deduced from the MIPAS-B measurements around sunrise and those used in the box model runs. For the latter, the impact of ozone and the albedo has been studied.

Minor points

1. page 4695, line 1: The words "fast" and "slow" in a scientific publication are only meaningful if compared to a certain value.
This sentence has been improved by indicating the time scales to the photolytic reactions: "Photolytic reactions of NO_2 and NO_3 are very fast (in the order of minutes), while photolysis of N_2O_5 is slower (in the order of several hours), and photolysis of HNO_3 and $ClONO_2$ is almost negligible in the lower stratosphere at high altitudes in winter."

2. To me it was confusing to read that the MIPAS-B flight was on March 21, 2003 that is one day after also a MIPAS-B flight are published (e.g. Engel et al., 2006). It seems that these are different data. Please confirm that the given date is not a typo and mention the other flight. It would be interesting to see how the two flights compare.
In fact, both datasets are from the same flight on 20th/21th March (see p. 4699, line 13). The balloon was launched at 18:30 UT on March 20th, and the flight lasted until 9:00 UT on March 21th. In this publication we focus on the data acquired in the morning of March 21th in the specified latitude - longitude section (see Fig. 2).

3. p. 4702, l. 24ff: It is not clear how "vertical NOy redistribution" can be seen from this plot.
The HNO_3 VMR is reduced by about 1 ppbv inside the vortex compared to outside along with a broadening of the HNO_3 peak area. A plausible explanation for that is some redistribution of the HNO_3 vertical profile by de-/renitrification earlier the winter. The signal is not that clear any more since after the the major renitrification that took place in early January (see Grooß et al.,2005), dynamics (inside-vortex mixing, mix-

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ing across the vortex edge, inhomogeneous vertical movements) and photochemistry might have washed out any more pronounced structures. We have now mentioned in the paper that the signals of de-/renitrification are very weak at this time of the year.

p. 4704, l. 16ff: Not much is said how the photolysis rates are interpolated to the altitude and zenith angle of the trajectories. This detail may be important as during sunrise the photolysis rates change quickly over orders of magnitude and a not sophisticated interpolation may cause errors especially near sunrise and sunset.
This is a very good comment. The photolysis rates are calculated with the radiative transfer model ART (E.P. Röth, Description of the anisotropic radiation transfer model ART to determine Photodissociation coefficients, Berichte des Forschungszentrums Jülich, JUEL-3960, Germany, 2002). Therein, the solar zenith angle dependence of the J values of each substance at a distinct altitude is given by the following parameterisation: $f = f_0 e^{b*[1-sec(c*\chi)]}$. This parameterisation has been used in the box model studies with different f_0, b, and c for each altitude. However, an exact calculation of the photolysis rate for distinct zolar zenith angles has shown that the interpolation gives too high values around sunrise. We now discuss the effect of interpolation in the newly added Section 6, where we compare the NO_2 J values deduced from the measurements with the ones used in the model.

5. Figure 1: Also important are the thermal decomposition of N_2O_5 and HO_2NO_2. The main product of $ClONO_2$ photolysis is NO_3, not NO_2.
We have modified the figure accordingly. The thermal decomposition of N_2O_5 and HO_2NO_2 is now addressed by a neutral body (M). The arrow for the $ClONO_2$ photolysis points to NO_3.

6. Figure 3: It would be elucidating to see the 7 tangent point locations of the observations at the nearest corresponding altitude over-plotted, not only the location of Kiruna. With that the reader would get a better impression of which data are inside or outside the vortex.
This figure has been modified accordingly.

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7. Figures 4/5 and 7-10: The horizontal gradient in the figures is difficult to read from the color scale. It would be better to complement these figures by a time series (quantity vs. time) for a chosen interesting altitude, e.g. 20 km. Would it be possible to add vortex edge after Nash et al. similarly as the sunrise line or PV or equivalent latitude?
We have provided two additional figures showing the temperature or vmr, respectively, as a function of latitude for a selected altitude (supplementing Figs. 4/5 and 7/8) and vmr as a function of time (supplementing Figs. 9 and 10). Concerning the vortex edge, it is hard to show it in these figures because the spatial distribution of the measurements is highly non-linear. Adding the vortex edge at selected points might be more confusing than helpful. However, we now show the vortex edge according to Nash in Fig. 3. This should also give the reader a better idea about the situation of the vortex with respect to our measurement locations.

8. The above argument also holds for figures 11 and 12. It is very difficult to judge over agreement and disagreement in a quantitative way from these figures, since differences may be hidden in the color contrast or may appear exaggerated depending on the choice of the color scale.
We have added difference plots.

Technical Corrections *1. Abstract line 2: change to "spatio-temporal" or "spatial and temporal"*
OK, we have changed it to "spatio-temporal"

2. p. 4704, l. 17/24: At this paragraph, it is not yet clear what trajectories or trajectory levels are, since this is explained in the following section.
We have exchanged the two sections such that the trajectories are described before the model is discussed.

3. p. 4704, l. 8 (and other places of the paper): change "sunlit" to "sunlight"
In the context used we really mean the ajdective "sunlit" and not the noun "sunlight".

4. p. 4705, l. 18: change "with" to "from"

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5. p. 4706, l. 2: change to ". . . ending at the tangent points. . . "
6. p. 4706, l. 19: change to ". . . to include the box model results for the simulated period."
All these changes have been done.

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

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