Atmos. Chem. Phys. Discuss., 1, S215–S220, 2001
www.atmos-chem-phys.org/acpd/1/S215/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Tropospheric NO_2 columns: a comparison between model and retrieved data from GOME measurements" *by* A. Lauer et al.

A. Lauer et al.

Received and published: 5 March 2002

Final response (on the referee comment of Anonymous Referee #1)

Referee: 1. Section 1, paragraph 1 - curiously, the authors forget to mention the important role of NO2 in tropospheric chemistry via its production of NO3 at night, NO3 being the main oxidiser in polluted areas at night.

Response: We included the following sentence and reference about NO3 and its role as important oxidant in the troposphere into the introduction: In addition, NO2 can react with O3 to form the nitrate radical (NO3), which is a strong oxidant and plays an important role, particularly in NOx polluted areas at night (Wayne, 1991).

Referee: 2. Section 1, paragraph 6, last line - although technically correct that prognos-

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tic simulations cannot be performed using a chemical model with prescribed dynamics (a CTM), the idea that prognostic simulations are important implies dynamical feedback due to changes in NO2. This could happen via its greenhouse effect, except that this is surely negligible. But it might happen via the greenhouse effect of changes in ozone caused by changes in NO2. Such changes in ozone are possible, but the greatest greenhouse effect from ozone is in the upper troposphere and lower stratosphere (UTLS). This is just where changes in NO2 have minimal effect on ozone: NO2 creates ozone in the lower and middle troposphere, it removes ozone in the middle and upper stratosphere, and in the UTLS the sign of the effect changes, making any effects small. Hence simulations of the chemical and UV-radiative effects could be made with a CTM, using annually repeating transport, and such simulations would be useful.

Response: Ok, from this point of view you are absolutely right. We therefore decided to remove the statement "which cannot be provided by Chemical Transport Models." completely from the related sentence.

Referee: 3. Section 2.1, paragraph 3, line 11 - The apparent slant column of a gas when observing sunlight scattered from the nadir to space (or from the zenith to ground) is not the integrated concentration along the light path through the atmosphere. There is no single light path through the atmosphere, instead there is a continuum of light paths through the atmosphere. The apparent slant column can be thought of as a weighted average of the integrated concentration along each of the light paths, the weight being the intensity of scattering at the altitude of the scattering point multiplied by (1 - extinc-tion along the path). Scattering and extinction must be calculated by a radiative transfer model. In fact, such intensity-weighted calculations are now known to be incorrect (e.g. Sarkissian et al. 1995). Instead, the apparent slant column is the normalised integral over wavelength of

-ln(I / I*) F / sigma

where I is the intensity calculated by a radiative transfer model, given the actual verti-

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cal distribution of absorber, I* is the intensity calculated by the radiative transfer model without absorber, F is the instrument function, and sigma is the cross-section of absorber.

Response: We fully agree with the reviewer comments, and the airmass factors used in this study have been calculated using the radiative transfer model GOMETRAN that does not make use of the intensity weighting approximation. However, for a qualitative description as that in the relevant paragraph, a more detailed discussion as given by the reviewer does not seem appropriate to us, in particular with respect to the intensity weighting approximation, that for the typical GOME scenarios introduces errors of the order of 1-3%. We tried to account for the reviewers comment by changing the relevant sentence to:

The slant column of a gas is derived from the differential absorption of the gas in question and in first approximation is the integrated concentration along the light path through the atmosphere.

Referee: 4. Section 3, following point 4 - there are some alternative arguments that can be made. For example: "In model data, Ref Sector (TEM) values often go slightly negative - a natural consequence of subtracting a reference sector with some variability - whereas the Thermal Tropopause values must always be positive. The most negative Ref Sector values are about -3e14. In the model, the stratosphere extends to 10 hPa, leaving up to 20e14 in the stratosphere. This stratospheric amount could vary by the required 3e14". The continental outflow cited in the text is unlikely to be giving rise to variability over Antarctica, as seen in the Reference Sector (TEM) model data.

Response: We don't think that the model top of 10 hPa has great influence on the results of TEM although it is correct that not all stratospheric NO2 can be captured with a model top of 10 hPa. But as long as all tropospheric NO2 is included in the calculation of the column amounts, the error of the missing stratospheric NO2 should cancel out when calculating the differences of the modelled total column amounts and

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the modelled stratospheric column amounts.

Referee: 5. Figure 3 - in January in GOME data (upper right-hand plot of Figure 3), the NO2 over Antarctica varies by 30e14, whereas the model variation is about 6e14. This is not discussed in the text. The minimum is over the Antarctic Peninsula, which is very cloudy so that less boundary-layer NO2 will be observed by GOME. The maximum is over the Antarctic Plateau. We now know that significant NO2 is expelled from nitrate in snow by sunlight (e.g. Jones et al 1999), a reaction which is not yet included in the model. The data in Figure 3 could be the first indication that it has a significant regional effect in Antarctica.

Response: Although this observation is correct, the results of the GOME measurements for high latitudes must be interpreted carefully, as some of the assumptions made for the retrieval of the tropospheric NO2 column amounts from the GOME measurements are not valid for this region. In particular, the assumption of longitudinal homogeneity of the stratospheric NO2 is not always fulfilled. To be more precise, we inserted the following sentence: Although obvious from figure 3, the much lower variation of NO2 in the model data compared to the GOME dataset over Antarctica in January is not discussed, because some of the assumptions made for the tropospheric NO2 retrieval from the GOME measurements (see table 2 and 4.2) are not valid for high latitudes. Thus the results for Antarctica are difficult to compare and should only be treated carefully.

Referee: 6. Section 4.1, last paragraph but 2 - the text speculates that the missing reaction of N2O5 on tropospheric aerosols in the model could account for some discrepancies with the measurements. This speculation is well founded, and the estimates of the size of the error are in excellent agreement with the observed differences. The authors should say so.

Response: We decided to insert the following sentence: Particularly the missing N2O5 hydrolysis should account for some of the overestimation of NO2 by the model. The

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estimated size of the error (as stated by Dentener and Crutzen (1993)) is in good agreement with the observed differences.

Referee: 7. Section 4.2, paragraph 4 - errors arising from undetected clouds could be estimated by altering the threshold value in the detection scheme to 20% and to 0%. This would be particularly useful over Northern Europe, where the differences between model and measurements in Figure 4 are rather large.

Response: We agree that varying the cloud fraction and comparing the resulting tropospheric NO2 columns is a useful experiment, and in fact, such an analysis has been performed on a subset of the data (see Richter & Burrows, 2001). While the general outcome of the experiment is in good agreement with the expected change in NO2 column if only the shielding effect of clouds is taken into account, there are two problems with this approach: First, the number of useable measurements is strongly reduced in winter below 10% cloud cover, and even if several years of data are used is hardly of statistical significance. Secondly, there is evidence for substantial amounts of NO2 above clouds in Europe in winter, making interpretation of the results difficult. To clarify the reasons for the selected threshold value, we included the following paragraph in the paper:

This threshold value has been chosen to balance between meeting the needs of the 'clear sky' assumption in the AMF calculations and the number of available measurements. Because of the large dimension of the GOME pixels, the number of usable measurements would drop dramatically when using a lower threshold value.

Referee: 8. Section 4.3, last paragraph - the text suggests that measurements in late morning, when lightning activity is small, may be responsible for less NO2 in the measurements than in the 24-hour average of the model. This sounds plausible. However, Figure 4e shows that the measurements have more NO2 than the model in Australia - the difference has the opposite sign. In Australia, about a third of the area has major thunderstorm activity (tropical north coast and North-East area, east coast as far

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south as Sidney). Furthermore, thunderstorm activity there is a maximum in summer (January) where measurements and model agree well.

Response: Australia doesn't seem to be special with respect to lightning produced NOx. The seasonal variation of the modelled tropospheric NO2 column amounts is determined by the lightning produced NOx emissions, which are only present in summer. Here, the model results and the measurements are in good agreement and the values of the NO2 column amounts from GOME are lower than the modelled ones - as expected. The higher NO2 column amounts from GOME in winter could be a result of an underestimation of the other NOx sources in the model in winter, where the emissions are very low. For clarity, we inserted the following:

In contrast to the other regions, the model shows noticeable less NO2 than the measurements during the winter months, when lightning activity is at its minimum. As lightning produced NOx is dominating the seasonal variation of the NOx emissions in the model (and therefore the seasonal variation of the tropospheric NO2 column amounts), lightning produced NOx seems to be well represented by E39/C as there is good agreement between model and measurements during summer when the lightning activity is high. However, the other NOx sources seem to be too weak, particularly in winter. This could explain why the modelled NO2 column amounts have lower values than the measured ones during the Australian winter.

Interactive comment on Atmos. Chem. Phys. Discuss., 1, 411, 2001.