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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.

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Final response (on the referee comment of G. Corlett)

Referee: The explanation of the importance of NO2 in atmospheric chemistry given in the first paragraph of the introduction is a little simplistic. It would be better to refer the reader to a standard textbook such as Finlayson-Pitts and Pitts (The Chemistry of the Upper and Lower Atmosphere).

Response: We think your suggestion to refer the read to a standard textbook is a good idea. Thus we inserted a link to Finlayson-Pitts and Pitts: For more details on the role of NO2 in atmospheric chemistry, the reader is referred to e.g. Finlayson-Pitts and Pitts (1999).

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Referee: The first paragraph of page 413 refers to the direct absorption of radiation by stratospheric ozone and implies a direct link between tropospheric NO2 and stratospheric ozone. This is a rather tenuous link and the authors would be better to consider the link between tropospheric NO2 and ?O3.

Response: The first paragraph refers to direct absorption of radiation by tropospheric NO2, not by stratospheric O3. As our formulation was not that clear, we inserted "by tropospheric NO2" in the related sentence. In our opinion, a link between tropospheric NO2 and tropospheric O3 does already exist: (First paragraph of page 413) As tropospheric O3 is also a significant greenhouse gas, NO2 also contributes indirectly to the radiative forcing.

Referee: The second paragraph of page 413 refers to the differing contributions to the global NOX budget. Again, it would help the reader if a link were given to a better description such as Brasseur et al. (Atmospheric Chemistry Global Change) as the explanation has limitations.

Response: Again, we think this is a good idea and inserted a link to Brasseur et al.: For further details on the contributions to the global NOx budget, see e.g. Brasseur et al. (1999).

Referee: Most of the discussion of the GOME instrument is superfluous whereas important factors such as the spatial and temporal sampling of the instrument are discussed somewhat too briefly. Also, I think it unacceptable that the reader is directed elsewhere to find out the uncertainties of the retrieval method applied here. I would recommend including a table to summarise the assumptions made in the GOME tropospheric NO2 retrievals.

Response: We changed the fifth sentence of section 2.1 from "The atmosphere is scanned by a moving mirror resulting in spatial resolution of 320 km x 40 km (across track x along track)." to "The atmosphere is scanned in spatial resolution of 320 km x 40 km (across track x along track) (forward scan) and 960 km x 40 km (back scan)." In

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addition, we inserted the following sentence: Each individual orbit of ERS-2 takes about 100 minutes. Although the repeating cycle of a orbit is 35 days, nearly global coverage (except for a small gap around the poles) is achieved within three days applying the maximum scan width of 960 km (ESA, 1995). We agree with your recommendation of an additional table on the assumptions made for the derivation of the tropospheric NO2 column amounts from the GOME, because you are absolutely right when saying it is not very nice to direct the reader elsewhere to find out the uncertainties. We couldn't paste the table to this ASCII document, so we only include the title of the new table:

Table 2. Summary of the assumptions for derivation of the tropospheric NO2 column amounts from the GOME measurements (version 1.0 of the IUP/IFE-UB TEM NO2 Dataset) (after Richter and Burrows, 2001). Negative values indicate error sources that tend to lead to an underestimation of the tropospheric NO2.

Referee: Bullet point b) on page 416 should include a reference to justify this assumption.

Response: We absolutely agree. Bullet point b) has been rewritten: at remote locations over the Pacific, the tropospheric NO2 column amounts are nearly constant and negligibly small. This is shown by the results of aircraft measurements (Schultz et al., 1999) and by the GOME measurements themselves (Richter and Burrows, 2001).

Referee: On page 416, the statement describing the model as a "spectral interactively coupled" GCM is somewhat confusing. A brief sentence expanding on this description would be useful.

Response: Ok, our description was somewhat too brief. We rewrote the first sentence and inserted an additional one: ECHAM4.L39(DLR)/CHEM (hereafter referred to as E39/C) is a spectral atmospheric chemistry - general circulation model. The model consists of two parts, the atmosphere general circulation model ECHAM4.L39(DLR) (Land et al., 1999) and the chemistry module CHEM (Steil et al., 1998). ECHAM4.L39(DLR) and CHEM are fully coupled, facilitating the representation of feedback mechanisms

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between changes in concentrations of chemical species and the simulated dynamics. E39/C...

Referee: The authors point out that the CHEM part of the coupled model does not include either PAN or N2O5 hydrolysis. The effect of these omissions on the model output is not adequately discussed where the subject is introduced in the paper or in the discussion of the observed differences. For example, Schultz et al. (JGR, 2000) suggest that PAN could be responsible for up to 27% of the NOX production in the tropical upper troposphere and that N2O5 hydrolysis contributes around 20% the loss of NOX. I feel a more substantial discussion of the errors that these omissions would give on the model output should be included as they disagree with the suggestions and choice references in the paper.

Response: We don't think there is disagreement with the choice references in the paper. However, we decided to insert the following paragraph: In contrast, a study by Schultz et al. (2000) analysing aircraft measurements at 6-12 km altitude over the tropical South Pacific states that the N2O5 hydrolysis is less important (in the free troposphere over the tropical South Pacific) than previously assumed by other (modelling) studies. However this not contradictory to the conclusion of this study (based on the study of Dentener and Crutzen (1993)) on the effect of the N2O5 hydrolysis on the tropospheric NO2 column amounts, as the major fraction of the tropospheric NO2 column amounts is located in the boundary layer and lower troposphere and not in the free troposphere.

Referee: In general, the discussion of the differences between the model results and the instrument data is not very convincing and appears to repeat the usual arguments found when completing such a comparison, although directed this time towards NO2 and its chemistry. A more useful analysis would be to show what effect is observed by changing the NOX emission or running the model over smaller areas at a more appropriate time for overlap with the ERS-2 satellite. A study of this type should be able to answer questions such as: a) How large are the errors on the NOX emission

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inventories? What effect does a small perturbation have on the model output? b) Are the errors produced by a lack of understanding of the basic chemistry? I feel the inclusion of PAN and N2O5 hydrolysis is necessary to answer these questions. These questions are apparent in the paper but there are no answers.

Response: This study is based on two previously existing datasets. It is intended to be a first step in evaluating the model results and cross-checking the retrieval of the satellite measurements. It is not thought to give already convincing answers on now upcoming interesting questions on quantification of single error sources. Of course, the inclusion of the VOC/PAN chemistry and the N2O5 hydrolysis as well as improving the NOx emission datasets is essential for answering the open questions. Nevertheless, in our opinion we have learnt a lot from this first step, enabling us to do the second one at all. This aside, we hope that this study will motivate other scientists to use the now available global climatological NO2 datasets (although not perfect yet) for interesting studies on tropospheric NOx, too.

Technical Corrections

Referee: Page 412 line 8: Should read "earthshine radiance and extraterrestrial irradiance".

Response: Done.

Referee: Page 412 line 10: The sentence beginning "The period of ..." is not grammatically correct and should be rewritten.

Response: We rewrote this sentence: The period of five years of observational data is sufficiently long to facilitate for the first time a comparison based on climatological averages with global coverage, focussing on the geographical distribution of the tropospheric NO2.

Referee: Page 412 line 23: Change "NO2can" to "NO2 can".

Response: We didn't encounter this problem when printing the PDF document.

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Referee: Page 413 line 20: Reference to Leue et al. should be 2001 not 1999.

Response: Done.

Referee: Page 413 line 20 and 21: Change to "In this study, in contrast to recent studies by Leue et al. (2001) and Velders et al. (2001), climatological averages ...".

Response: Done.

Referee: Page 414 line 15: Change "NO2 amou nt" to "NO2 amount".

Response: We didn't encounter this problem when printing the PDF document.

Referee: Page 414 line 23: Change "extra terrestrial to extraterrestrial".

Response: Done.

Referee: Page 416 line 13: Change "below the threshold" to "below a threshold".

Response: Done.

Referee: Page 423 line 8: Change "lightning NOX " to lighning produced NOX ".

Response: Done.

Referee: Page 423 line 18: Change "NOX emi ssions" to "NOX emissions".

Response: We didn't encounter this problem when printing the PDF document.

Referee: Page 424 line 25: change "NOX" to "NOX ".

Response: We used "NOX" intentionally to avoid confusion between "NOx = NO + NO2" (lower case "x") and "NOX = NO + NO2 + NO3 + N2O5 + HNO4" (upper case "X").

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