

Interactive comment on “Tropospheric NO₂ columns: a comparison between model and retrieved data from GOME measurements” by A. Lauer et al.

Anonymous Referee #1

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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.

2. Section 1, paragraph 6, last line - although technically correct that prognostic 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

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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.

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 - extinction 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 vertical distribution of absorber, I^* is the intensity calculated by the radiative transfer model without absorber, F is the instrument function, and σ is the cross-section of absorber.

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 -3×10^{14} . In the model, the stratosphere extends to 10 hPa, leaving up to 20×10^{14} in the stratosphere. This stratospheric amount could vary by the required

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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.

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.

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.

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.

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

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