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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Enhancement of N_2O during the October–November 2003 solar proton events" *by* B. Funke et al.

M. López-Puertas

puertas@iaa.es

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We thank Reviewer #2 for his/her very helpful comments and suggestions that have significantly improved our manuscript. The "Reviewer Comments" are noted first and then we give our "Reply:" to the comment. We are submitting a revised manuscript that includes all the actions noted below.

Specific Comments: Page 4670 near line 25. It would be appropriate to add a reference to the ACE data here [Rinsland, C. P., C. Boone, R. Nassar, K. Walker, P. Bernath, J. C. McConnell, and L. Chiou (2005), Atmospheric Chemistr y Experiment (ACE) Arctic stratospheric measurements of NOx during February and March 2004: Impact of intense solar flares, Geophys. Res. Lett., 32, L16S05, doi:10.1029/2005GL022425].

Reply: We actually thought about including this reference but since it does not treat the NOx enhancement during solar protons events we left it out. Please recall that this paragraph and the Introduction deal with changes of species abundances related to SPEs only.

Page 4673, top: The retrievals are done in a sequential manner, with N2O and CH4 retrieved last. I would like to see a comment about the species separation errors that might be significant for N2O - what is the magnitude of the errors in the retrievals of other constituents, and what errors do they lead to in the N2O retrievals, particularly if a bias is expected? This might be discussed in Glatthor et al., but it is directly relevant to this paper, so a summary would be helpful. Along similar lines, a short statement defining the "total error" (line 11) would be helpful.

Reply: Separation error between the jointly fitted species CH4 and N2O is implicitly taken into account in the noise error. Retrieval errors due to uncertainties in the abundances of pre-fitted species or those for which climatological profiles are used, are less than 1% for both, N2O and CH4. Total error is the square root sum of all systematic and random error components plus model errors (non-LTE). Systematic error sources taken into account are uncertainties in temperature and interfering species abundances, pointing, spectral data, and calibration. Systematic errors are in the order of 10-30%.

Action: A couple of sentences about this point have been included in the revised manuscript.

Page 4673, line 23. Please state why two different versions of NO2 are used (_9 and _11) - do these pertain to different time periods, for instance?

Reply: In effect, the reason for using two different versions for NO2 is because they pertain to different time periods. MIPAS IMK/IAA retrievals are performed for selected episode periods and the data analyzed in this work span over two of those periods. We should mention, however, that the differences between these two versions for NO2 in

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the regions studied are minor. Action: A sentence along this line has been included in the manuscript.

Page 4675, near line 10: The authors assert that the spatial correlation between N2O and NO2 supports their explanation of N2O being produced by the reaction of NO2 and N(4S). Certainly a qualitative comparison of figures 1 and 2 supports this conclusion. I fail to see a strong correlation in Figure 3, though, so I question the value of this figure. I would prefer to see a quantitative analysis of the data in Figures 1 and 2 - even something as simple as a scatter plot, with a correlation coefficient reported, and some discussion of locations or times when the correlation is perhaps not as robust (e.g., 20031031 and 20031111).

Reply: We agree with the reviewer. We have replaced Fig. 3 with a scatter plot. This clearly shows much better the N2O/NO2 correlation after the solar storm and how its slope changes from non-SPEs (all latitudes before SPEs and mid to low latitudes for all conditions) to SPEs conditions. This then shows that N2O during SPEs is produced from NO2 and the source is of a different nature than that for non-SPE conditions.

Page 4675, line 25: Here the authors speculate that there is an indication of aurorally enhanced N2O in Figure 4 on 26 October. I assume they are referring to the region of light blue/green extending above 55 km near the pole, narrowing to around 52-55 km near 60 deg latitude. I agree that this is beyond the focus of the current work, and that it is appropriate to include just a brief speculation about the cause. I question their speculation, however, since the enhancement, if it is an enhancement over normal conditions, occurs at such low altitudes. Auroral precipitation would occur above about 100 km. I recommend that the authors check to see if photochemical lifetimes and descent rates at this time are compatible with descent from ˜100 km to ˜60 km. Another possibility is that it is due to higher energy particle precipitation, which would affect the upper mesosphere directly, if such particles were available before 26 October.

Reply: Yes, we are referring to the region the reviewer mentions. The referee is right,

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such enhancements are not produced by auroral processes. We meant they are produced by energetic electron precipitations (EEPs). MEPED measured large fluxes of high-energy electrons during those days and EEPs can produce, in situ, both N and NO2, required to form N2O. The required NO2 could also be originated from descent of air-masses rich in NO2 that was produced at higher altitudes (in the upper mesosphere/lower thermosphere) a few weeks earlier by auroral electrons (not EEPs). Action: We have changed the manuscript accordingly and make a reference to the manuscript by Funke et al. (2008) that describes the mesospheric N2O enhancements observed by MIPAS during several polar winters.

B. Funke, M. López-Puertas, M. García-Comas, G. P. Stiller, T. von Clarmann, and N. Glatthor, Mesospheric N2O enhancements as observed by MIPAS on Envisat during the polar winters in 2002-2004, ACPD, accepted, 2008.

Page 4675, line 25 (same as above): In addition to the above comment, but related more to the overall conclusions of the paper, it would be interesting to see a plot similar to Figure 4 for NO2, especially for 20031026. The 20031026 panel in Figure 2 suggests that there were no NO2 enhancements on this date at 58 km, calling into question the proposed mechanism for forming N2O, unless the 26 October N2O "enhancements" are actually due to a tropospheric source. Perhaps this is just an issue of color scales, or chemistry that might lead to a poorer correlation between N2O and NO2 at lower altitudes, or retrieval errors. In any event, I think it is important that the authors comment quantitatively about this apparent contradiction.

Reply: NO2 on 20031026 do show a clear enhancement in the same region as N2O (see new Fig. 5a in the revised manuscript). Fig. 2 did not show it so clearly because of the large scale used. Actually we proposed such explanation because we noted this enhancement in NO2 and also the large high energetic electron fluxes measured by MEPED. For completeness, and answering a similar question risen by the other reviewer, we think most of the NO2 enhancement we see on 26 Oct is not produced locally by EEPs (MEPED electron fluxes are not strong enough that day) but it is the

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result of the descent of NO2 produced at higher altitudes by less energetic auroral electrons a few weeks earlier. Action: We have included a new Figure (Fig. 5) similar to Fig. 4 but for NO2. The text has been changed accordingly.

Page 4678, line 25. Here the authors state that below 70 km, nighttime NO2 is a good proxy for NOx. This statement contradicts the model results in Figure 7, which show that at 60 km (for example), NO mixing ratios that are more than half as large as NO2 mixing ratios.

Reply: The reviewer is right. That assumption can only be considered correct below about 50 km. We used that argument only to justify the selection of NO2 to night-time values. We have removed that assertion and mention that we use only night-time data because the production mechanism of N2O is operating at night-time only.

Page 4678, line 28. The authors should quantify "rather good agreement". Even just considering the first few days after the SPE, it looks like the level of agreement varies with altitude and time, and in some cases differences are as much as 40-50%.

Reply. We have quantified now the "good agreement". During the first few days after the SPEs, the model predictions for NO2 are (see Fig. 1 below) generally smaller than the measurements by about 10-30% below about 55 km, and larger by a similar amount in the 55-70 km region.

Action: We have included a few sentences quantifying these differences. We also have removed the old figure 7. This does not add any new evidence for supporting the proposed mechanism (much better represented now with the new Fig. 3 and the addition of new Fig. 5) but could mislead the reader about the quantification of model/measurements NO2 differences. These differences are now discussed on he basis of new Figs. 7, (old Fig. 6).

Page 4679, line 3. The authors state that the model overestimate of NO2 from 55-65 km is consistent with the "slightly larger values" of the predicted N2O enhancements in

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this altitude region. Again, this should be quantified, as should the overestimate in the N2O predictions (which are hard to infer quantitatively from Figure 5). Is there a one-to-one correspondence between errors in simulated NO2 and N2O? If so, this would suggest an over-prediction in N2O of around a factor of 2 near 60 km (based on this difference between the model and observed NO2 in Figure 7), which is not "slight".

Reply: In addition to the differences in NO2, we have also quantified the model/ measurements differences in N2O (see Fig. 2 below). The model generally overestimates the measurements in about 1-2 ppb in the same altitude region where it overestimates NO2, 55-70 km. This overestimation is about 20-30%, similar to that for NO2. Hence the model-measurements differences in both species are similar and take place in the same regions, which further supports the proposed mechanism.

Figures: In a printed version of the paper, the text labels on the figures are small and quite difficult to read. I would appreciate it if the authors made them larger. There is some superfluous information that can probably be removed from the figures themselves and simply incorporated into the caption (e.g., "N2O (NO2) night 58.0 km" in Figures 1 and 2, etc.).

Reply: The reviewer is absolutely right. We were expecting most figures to appear as "two-columns" figures, which would make them larger and more readable. Action: We have re-made all figures increasing all labels, fonts, and removing unnecessary information on the title head. We will also ask the editor to increase the size of the figures in the revised version to make them more readable.

Figure 4 caption: I suggest adding "in panels b-d" after "The enhancement of N2O at high latitudes above 40 km is evident".

Reply: We agree. It has been done.

Figure 6: The caption should state whether or not the model results include the MIPAS averaging kernel (I assume they do).

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Reply: The averaging kernel was not applied originally. We have applied it now. The changes are negligible below around 60 km and slightly increase (˜10%) the modelled values (a priori effect) from 60 to 70 km.

Figures 5-7: There is some information at the top of the panels that can probably be removed in order to make the labels bigger (e.g., IMK...spe..."jtab").

Reply: We agree. It has been done.

Technical Corrections: Page 4672, line 23: Change "being" to "to be". Page 4675, line 11: A comma needs to be added: "...and NO2, which indeed is very...." Page 4675, line 23: Change "fews" to "few". Figure 7 caption. Remove "the" before "November 2003".

Reply: All of them have been included. Thank you very much.

Fig. 1. Model-MIPAS NO2 differences in %. (Fig7b-Fig7a) in the manuscript.

Fig. 2. Model-MIPAS N2O differences in ppbv (Fig6c-Fig.6a).

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 4669, 2008.