Interactive comment on "Variability of NOx in the polar middle atmosphere from October 2003 to March 2004: vertical transport versus local production by energetic particles" by M. Sinnhuber et al.

Anonymous Referee #1

The authors would like to thank the reviewer for his/her helpful comments. Below are all the comments followed by replies; in "italics" are changes made to the text of the paper.

1) There is a bit of a contradiction in the Abstract, since it says that local production by precipitating electron is unlikely, but then an upper bound (e.g. 6 ppb at 56-70km) is provided. It should say that the production is weak, smaller than the upper bound. Please clarify.

What we wanted to express is that a significant additional source of NOx due to precipitating electrons below 70 km is unlikely; a weak production is of course possible. To clarify this, the order of the statements in the abstract has been changed to

"Correlations of NOx and CO show that the unprecedented high NOx values observed in the Northern Hemisphere lower mesosphere and upper stratosphere in late January and early February are fully consistent with transport from the upper mesosphere / lower thermosphere and subsequent mixing at lower altitudes. In the polar summer Southern Hemisphere, we observed an enhanced variability of NO and NO2 on days with enhanced geomagnetic activity but they seem to indicate enhanced instrument noise rather than a direct increase due to electron precipitation. A direct effect of electron precipitation onto NOx can not be ruled out, but if any, it is lower than 3 ppb in the altitude range 40—56 km and lower than 6 ppb in the altitude range 56—64 km. An additional significant source of NOx due to local production by precipitating electrons below 64 km exceeding several ppb as discussed in previous publications appears unlikely."

2) What the authors mean by "cross-talk" in retrieved NO between different altitudes is a bit unclear. A couple of sentence on the relevant retrieval issues could be helpful to the reader.

An explanation of this was added at the end section 2 (MIPAS data):

"NO mixing ratio increases strongly from the lower mesosphere to the lower thermosphere. Thus observations at mesospheric tangent altitudes has a strong thermospheric NO signal in the line-of-sight which might provide an additional positive offset of up to 1.5 ppb, henceforth called "thermospheric cross-talk".

For the retrieval of NO2 no such information crosstalk happens because the concentration of NO2 decreases strongly from the upper stratosphere to the thermosphere even during night-time. Thus the measured signal is dominated by radiance emitted near the tangent altitude of each limb observation. Therefore, in the winterhemisphere where NOx values are in the order of tens of ppb to thousands of ppb, observations of NOx (NO+NO2) are used, while in the summer hemisphere, where mixing rations might be in the range of below 1 ppb, night-time NO2 and day-time NO are treated separately."

3) Fig 9: units for the electron fluxes on the y-axis should be included.

The electron and proton fluxes in Fig 9 are given in arbitrary units; a comment has been

added to the figure caption: "..., both in arbitrary normalized units". However, the units of the NO mixing ratio have been added to the y-axis for the left-hand panels.

4) Given the results of Fig 10, wouldn't it be more relevant to show also the day-time SH NO anomalies at a level above 60km, where they are higher (rather than 48, 54 and 60 km)?

Instead of 60 km the 62 km level is now shown. Above 60 km, the sensitivity of the NO observation as defined by the averaging kernel criterion decreases in such a way that for observations with low NO amounts, no data are available. Above ~64 km altitude, the anomalies decrease again because of the low instrument sensitivity and the small number of significant measurements above this altitude as seen, e.g., in Fig. 8. An explanation of this has been added at the end of section 4.2, and the vertical range in which an upper limit can be provided has been changed to below ~64 km altitude, both in section 4.2. and in the abstract; also, Fig 10 now only shows results up to 64 km.

"Above ~64 km altitude, no meaningful NO and NO2 data can be obtained for quiescent polar summer conditions when NOx concentrations are very low."

Spelling/English

P2: : : :during "high" winter: : :: unclear

changed to mid-winter

P4: continuosly

corrected

P6: "for the four scenarios shown in Fig 8". Scenario might not be the appropriate word here.

"scenarios" changed to "cases"

Interactive comment on "Variability of NOx in the polar middle atmosphere from October 2003 to March 2004: vertical transport versus local production by energetic particles" by M. Sinnhuber et al.

Anonymous Referee #2

General Comments: The authors investigate the impact on the atmosphere of energetic particle precipitation during the October 2003 to March 2004 time period using MIPAS observations of NOx and CO. They conclude that the NOx enhancements observed in the Arctic upper stratosphere and lower mesosphere during January-February of 2004 were caused by descent of NOx produced in the upper mesosphere and lower thermosphere by low (e.g., auroral) energy electrons, not by solar protons or higher energy electrons. This contradicts previous reports that direct production of NOx below 70 km was responsible for a significant portion of the observed Arctic NOx enhancement. The paper is well written and organized. Improved understanding of the atmospheric

impact of radiation belt electrons, and how this compares with lower energy electrons and solar protons, is an important contribution to the science community. I recommend publication after the following, mostly minor, comments are addressed.

The authors would like to thank the reviewer for his/her helpful comments. Below are all the comments followed by replies; in "" are changes made to the text of the paper.

Specific Comments p6, line 1. For people who wish to repeat any of this work, it would be helpful to cite a reference for the averaging kernels.

The description of the averaging kernel criterion in section 2 (MIPAS data) has been changed to:

"Where daily averages have been used, NO, NO2 and CO data with low sensitivity have been excluded by applying a threshold to the mean value of the averaging kernel (AK) matrix (Rodgers, 2000) diagonal element. The AK diagonal elements represent a measure of the sensitivity of the retrieval at a given profile grid point to the "true" vmr. Values close to zero (here <0.03) indicate that there is no significant sensitivity to the abundance at the corresponding altitude."

p10, line 8. Will transport and mixing really act on CO and NOx in the same manner? Doesn't this depend on vertical and horizontal gradients in the two constituents, which are not the same?

This is correct, and the reason we do not expect (and indeed do not see) a linear correlation in Fig 6.

Figure 6. I suggest adding color scales that denote the dates of the various colors in the two panels.

Done

p10, lines 16-21. On the basis of the CO/NOx relationship, the authors conclude that the high values of NOx below 70 km in late January are most likely due to subsidence of air, rather than direct production. I believe that the rationalization is that since the NOx values in air that had descended to 70 km were already high by 19 January – before the storms – there is no need to invoke a direct production source to explain them. This was not obvious at first reading, though, so if I am correct, a more explicit discussion would be helpful. Particularly important is to point out that even though the high CO / high NOx correlation *by itself* cannot rule out direct production (since the production could have occurred in air that had already descended), the fact that high NOx existed in the descended air prior to the geomagnetic storms is sufficient justification for the conclusion. (Of course, if that is not what is meant, the discussion should be modified accordingly).

There are two arguments here: both NOx and CO increase at the same time, during a relatively short time-interval between January 14 and January 19. The increase of both NOx and CO at the same time suggests downwelling as a source. Additionally, the largest amounts of both NOx and CO are reached on January 19, several days before a large storm on January 22; the large geomagnetic storm on January 22/23 was thus not the

source of the large NOx values, as suggested, e.g., by Renard et al., 2006. The paragraph has been changed to

"The upper branch of the relationship with CO values >10 ppm and NOx values >700 ppb appears only after 14 January, at altitudes above 60 km. Maximal values of more than 12 ppm CO and more than 2000 ppb NOx are reached on 18/19 January. The increase of both NOx and CO at the same time suggests downwelling of air from the upper mesosphere / lower thermosphere as a source. As maximum values of NOx are reached on January 18/19, their source can therefore not be direct production during the strong geomagnetic storm of January 22, as suggested, e.g., by Renard et al., 2006."

p10, last paragraph. If photochemical loss of CO moves the NOx-CO pairs to the left, can't this result in the appearance of points above the secant line? Related to this: What do the vertical "error" bars in Figure 6 denote? Also with regard to Figure 6: The red and orange data (at least; some other points are hidden) have a very peculiar shape – I think the authors should comment on this, if only to ensure that they are not overlooking some important physics that would affect their conclusions.

That is correct – photochemical loss of CO could move the points across the upper secant line as well. The vertical error bars denote the error of the mean of the daily averaged area weighted NOx values. The peculiar shape in the reds and oranges in the right panel of Figure 6 comes from the development of a NOx "knee" around mid-February as seen, e.g., in the upper panel of Figure 3 and discussed in the last paragraph of page 9; in the CO-versus-NOx view, this "knee" than shows up as a local NOx maximum at median CO values. The last paragraph of page 10 has been changed to:

"In the left panel of Fig. 6, the secant to the NOx-CO relationship is marked as a dashed line. The region comprised by this secant and the NOx-CO distribution (grey area) can be filled in by mixing processes. Photochemical loss of NOx would move the NOx-CO pairs down, while photochemical loss of CO would move NOx-CO pairs to the left. Values above this region can be gained by direct local production of NOx, e.g., due to energetic particle precipitation, or by photochemical loss of CO. In the first days after 20 January 2004, the grey mixing region is indeed filled in, while later (mid to late February), the NOx-CO pairs are moved to the left and downward. This indicates that during the timeinterval from 20 January 2004 to 1 March 2004, both mixing processes and photochemical loss of NOx and CO are important. As discussed before, from mid-February on, a "knee" develops in NOx due to the different photochemical loss rates of NOx and CO in the upper stratosphere and lower mesosphere. This is observed as a local NOx maximum at moderately high CO values in the right panel of Figure 6. Values above the mixing secant are not observed, so there is no evidence for a direct local production of NOx below 70 km during this time from these observations."

As suggested by the reviewer, a colorbar for the dates has been added to this Figure, so the red and yellow colors can be easily attributed to mid-February. The error bars are now mentioned in the Figure caption of Figure 6.

p11, lines 19-22. The most significant conclusion from this paper is that the NOx enhancements observed in the lower mesosphere in Jan-Feb 2004 were not caused by direct production of NOx, as suggested previously based on an analysis of GOMOS data. This conclusion relies not only on the evidence from MIPAS, but also on the ability of the authors to show that the previously published interpretation of GOMOS

data was incorrect. The authors suggest here that had the GOMOS analysis taken into account sampling biases with respect to the "moving wave 2 structure", an NO2 enhancement would not have been found. Because contradicting the GOMOS data interpretation is fundamental to their overall point that the enhancements were not caused by direct production (e.g., that the current interpretation of MIPAS data is more correct than the previously published interpretation of GOMOS data), I think the authors should support their speculation more strongly. In particular, they should show that the GOMOS sampling in Jan-Feb 2004 would indeed have led to a bias that would have resulted in a misinterpretation of the data.

We have made a comparison between Mipas footprints and the Gomos data who were used in Clilverd et al., 2009. Night-time (>106°sza) NO2 mixing rations at 60km on January 28/29 and at 52 km altitude on February 08/09 and February 14/15 are also compared; a figure will be included in the paper. Gomos data have been provided by Dr. Seppälä who has been added as co-author. Both instruments are qualitatively in very good agreement, showing the same pattern of high/low NO2 data, but are averaged over different areas of the vortex; even inside the vortex, NOx is not distributed homogeneously. The text on page 11, line 16 ff is adapted in the following way:

"However, the polar vortex as indicated by high values of both NOx and CO is elongated at the beginning of this period indicating a wave 2 pattern, which moves and expands to a more circular form from 8 February to 15 February. Because of the wave 2 pattern, the vortex edge is shifted far to the North on February 08/09, and Gomos observations poleward of 70° are taken partly outside the polar vortex (see Figure). Gomos observations both inside and outside the polar vortex agree very well with neighboring Mipas data points, but significantly higher values are also observed by Mipas further in the vortex core. On February 14/15, Gomos observations poleward of 70° are obtained all inside the polar vortex. They are significantly higher than the neighboring Mipas observations, though well within the range of Mipas data elsewhere in the vortex. The reason for the apparent disagreement between daily averages of NO2 from Mipas and Gomos in this time-period thus appears to be the strongly elongated vortex as well as

an inhomogeneous distribution of NOx inside the vortex. This inhomogeneous distribution is already apparent and observed by both instruments in late January, see Figure."

p12, first paragraph. The authors should be more precise when discussing the problems with analyzing NOx: What is meant by "cross-talk"?

An explanation of this was added at the end section 2 (MIPAS data):

"NO mixing ratio increases strongly from the lower mesosphere to the lower thermosphere. Thus observations at mesospheric tangent altitudes has a strong thermospheric NO signal in the line-of-sight which might provide an additional positive offset of up to 1.5 ppb, henceforth called "thermospheric cross-talk". For the retrieval of NO2 no such information crosstalk happens because the concentration of NO2 decreases strongly from the upper stratosphere to the thermosphere even during night-time. Thus the measured signal is dominated by radiance emitted near the tangent altitude of each limb observation. Therefore, in the winterhemisphere where NOx values are in the order of tens of ppb to thousands of ppb, observations of NOx (NO+NO2) are used, while in the summer hemisphere, where mixing rations might be in the range of below 1 ppb, night-time NO2 and day-time NO are treated separately." p13, lines 1-3 (and perhaps on previous page). This refers to NOx and the lower left panel of Fig. 8, which only shows NO. The text should be clarified.

In the publication mentioned, NOx is used; in the lower left panel of Figure 8, only daytime NO is shown. This has been clarified in the text as follows: "If the complete polar cap is sampled, enhanced NOx values decrease more steadily (exponentially) after the solar proton event, as shown, e.g, in Friederich et al. (2013) for NOx, and in the lower left panel of Fig. 8 for daytime NO."

p14, lines 13-14. The authors refer to the upper panel of Figure 10, but there is only one row of panels in Figure 10. Since Figure 10 pertains to the same situations as Figure 8, it would be convenient if both figures were formatted the same.

The reference to the upper panel was eliminated, also Figure 10 was formatted as suggested.

Finally, I was surprised that several references on the 2003-2004 winter, which I believe are relevant to the current paper, were not cited. I recommend that the authors consider the following papers:

1. Hauchecorne, A., J.-L. Bertaux, F. Dalaudier, J. M. Russell III, M. G. Mlynczak, E. Kyrölä, and D. Fussen (2007), Large increase of NO2 in the north polar mesosphere in January–February 2004: Evidence of a dynamical origin from GOMOS/ENVISAT and SABER/TIMED data, Geophys. Res. Lett., 34, L03810, doi:10.1029/2006GL027628.

2. Pancheva, D., et al. (2008), Planetary waves in coupling the stratosphere and mesosphere during the major stratospheric warming in 2003/2004, J. Geophys. Res., 113, D12105, doi:10.1029/2007JD009011.

3. Randall, C. E., et al. (2005), Stratospheric effects of energetic particle precipitation in 2003–2004, Geophys. Res. Lett., 32, L05802, doi:10.1029/2004GL022003.

4. Semeniuk, K., J. C. McConnell, and C. H. Jackman (2005), Simulation of the October–November 2003 solar proton events in the CMAM GCM: Comparison with observations, Geophys. Res. Lett., 32, L15S02, doi:10.1029/2005GL022392.

5. Seppälä, A., M. A. Clilverd, and C. J. Rodger (2007), NOx enhancements in the middle atmosphere during 2003–2004 polar winter: Relative significance of solar proton events and the aurora as a source, J. Geophys. Res., 112, D23303, doi:10.1029/2006JD008326.

Those references are now included. Thanks for pointing them out.

Minor Technical Corrections

p3, l28. Add "and" before "by Clilverd:" *Changed*

p5, I25. Add "and" before "about CO:" *Changed*

p6, I12. "combining"

Changed

p6, I15. "available" instead of "provided" *Changed*

p6, I22. Remove the comma after "21". *Sentence changed*

p9, I2. "off in late" not "of in late". *Changed*

p9, I5. "on the order" not "in the order" [and anywhere else this appears] *Changed*

p9, I6. "sunlit" (no hyphen) [and anywhere else this appears] *Changed here, and in a few other places*

p9, l8. "lifetime" and "sunrise " (no hyphens) Changed here, and in a few other places

p9, I25. "development" not "developement" *Done*

p10, I10. "observed by the MIPAS NOM data". I'm not sure what exactly was meant here, but it can probably be changed to just "observed by MIPAS". *Deleted NOM*

p10, I19. "seems" rather than "turns" *Done*

p10, l21. Depending on how you handle the above comment, "NOM" should be defined here. *Deleted NOM*

Deleterario

p11, l12. "continuously" *Done*

p11, l20. "stellar" instead of "star" (and no hyphen) *Done*

p11, l21. Remove "a" before "NO2" *Changed to singular*

p12, I14. Add "and" before "NO2". Done

p14, I24. Either "Exceptions are" or "An exception is" *Done*

Figure 4 caption. "rate" not "ratef" in the last line *Done*

Figure 9 caption. "violet" not "violett" Done