

Interactive comment on “Nitric acid in the stratosphere based on Odin observations from 2001 to 2007 – Part 2: High-altitude polar enhancements” by Y. J. Orsolini et al.

Anonymous Referee #4

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General Comments: This paper presents a climatology of polar enhancements in nitric acid (HNO₃) in the upper stratosphere and mesosphere from 2001–2007 based on measurements from the Odin Sub-Millimeter Radiometer (SMR). The enhancements result from HNO₃ production after energetic particle precipitation (EPP). The authors show that after Solar Proton Events (SPEs) the atmosphere exhibits two separate HNO₃ enhancements, which they refer to as a two-stage HNO₃ enhancement. The first enhancement is short-lived, and is caused by rapid production of HNO₃ after the SPE. The second enhancement, which requires EPP but not necessarily SPEs, arises from the conversion to HNO₃ of excess NO₂ that is being transported downward from the mesosphere or upper stratosphere to the lower strato-

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sphere. The paper is reasonably well written (it is concise to the point of being hard to understand if one is not already an expert in this area) and presents important results. I recommend that it be published in ACP after consideration of the points below.

Specific Comments: Figures. One major change that is absolutely required before publication is in the presentation of the figures. The original submission (reviewed only for technical errors) had Figures 2 and 3 presented as full-page. In this new submission, however, Figures 2 and 3 are only one-column wide. The labels are completely illegible when printed. This paper should not be published unless Figures 2 and 3 are in a full-page format.

Abstract Line 5. The authors note that both SPEs and precipitating electrons in the auroral zone affect HNO₃. In fact, HNO₃ will be affected not only by *auroral* electron precipitation, but by higher energy electrons that are produced by non-auroral processes and precipitate into the mesosphere and lowermost thermosphere (below the auroral altitude and in a different spatial distribution than the auroral zone). This same omission is made in line 24.

Abstract Line 10 and elsewhere. I think that referring to a two-stage HNO₃ enhancement is misleading. The problem in my opinion is that many, if not most, readers will interpret a two-stage HNO₃ enhancement to mean that the original HNO₃ enhancement (the first stage) somehow goes through a second stage of development. But if I understand the paper correctly, the so-called second stage is unrelated to the HNO₃ produced in the first stage. I believe that the authors are suggesting that the second HNO₃ enhancement results from excess NO_x that was produced at the same time as the first enhancement, but in the upper mesosphere or lower thermosphere. Thus the second HNO₃ increase was not observed until that NO_x had descended to the upper stratosphere; but the second HNO₃ enhancement is in no way a second stage of development of the first enhancement. Therefore, I recommend that the two different HNO₃ enhancements be referred to as two different episodes, or some other similar term that would imply that

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the HNO₃ in the second stage is independent of the HNO₃ in the first stage.

Introduction. I found it difficult to follow the mechanisms that the authors were proposing for their first stage. I believe that they are trying to explain that the first, short-lived, enhancement is caused by one of two mechanisms. Both mechanisms are initiated by the production of excess NO_x and HO_x in the upper stratosphere or lowermost mesosphere by energetic particles (whether protons or energetic electrons). In the first mechanism, the excess NO_x (NO₂ specifically) reacts in the gas phase with the excess HO_x (OH specifically) to produce HNO₃. In the second possible mechanism the excess NO_x is converted to N₂O₅, which then through some kind of heterogeneous reaction (ion chemistry?) is converted to HNO₃. If this is correct, the authors should describe this explicitly, and should describe whether both reactions are equally likely to occur. Also, the heterogeneous chemistry should be clarified. If this is not correct, clearly something needs to be rewritten. Also, why is this first enhancement short-lived? Is HNO₃ a long-lived reservoir in the polar night only in the lower stratosphere? This should be explained in the text.

Page 9594 Line 29. The authors state that the HNO₃ enhancements do not need to follow or coincide with the EPP-induced NO_x enhancements. This does not make sense to me, but I wonder if it was just written backwards. Since both the first and second episodes of HNO₃ enhancements require excess NO_x, I think that the HNO₃ enhancements *must* either coincide with or follow the NO_x enhancements. Perhaps what the authors really meant was that the NO_x enhancements are not always accompanied or followed by HNO₃ enhancements?

Page 9596 Line 5. I would avoid the use of polar cap since it can have a very specific meaning of poleward of 60 degrees geomagnetic latitude. Instead, I would recommend simply stating, ...cross-sections of HNO₃ averaged over equivalent latitudes poleward of 70 degrees N.

Page 9597 Line 10. It looks like two SPEs occurred in December; why only

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remark about one of them? Also, in line 11, do you really mean that the measurement are only indicative of the "second" stage (episode), not a "two-stage" enhancement, which would imply that the first enhancement was also visible?

Page 9597 Line 14. The enhancement of descending HNO₃ seems to occur slightly "before" the SPE, so it is hard to believe it would be attributed to the SPE itself. And if the SPE were responsible for enhancements near 1200 K, why would we not see a "fast" enhancement in HNO₃ at slightly higher altitudes as well, around 1500-2000 K? Therefore, I think the authors should reconsider the implication that the SPE in mid-July 2002 caused any HNO₃ enhancement in the stratosphere. There does seem to be some periodic enhancements in HNO₃ in July and August (note the oscillating red/yellow regions), but this was a winter with substantial wave activity (throughout the winter; not just at the final warming) in the southern hemisphere, which might explain the periodic behavior.

Section 2.2. The southern hemisphere winters of 2005 and 2007 had HNO₃ enhancements that were as large as in 2002. The authors should comment about what is driving this, since there were no SPEs during those winters. For instance, was there significant auroral activity or were there other geomagnetic disturbances? Was the meteorology particularly favorable for descending HNO₃?

Page 9598 Line 8. The authors note that sudden stratospheric warmings might dampen the HNO₃ anomalies. It is important to note that the sudden stratospheric warmings might also play a role in "enhancing" the anomalies. As noted earlier in the paper, the vortex in 2004 and 2006 was very strong, so this gave rise to large amounts of NO_x descending into the stratosphere. The conditions that led to this very strong vortex were probably related to the recovery of the vortex after the warming; so while the immediate effect of a strong warming is to dampen the effects of EPP, a longer term effect might be to strengthen them.

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Page 9598 Line 13. The authors should be more specific when they write that the descending enhancements can only develop if started not too close from the winter-to-summer transition. What month or months? Do you mean that the production must occur in late fall or winter?

Page 9598 Lines 14-19. I don't understand the point of this paragraph, so I would like to see a statement explaining why it's important to note when the descending HNO₃ merges with the main layer. In addition, shouldn't it be mentioned that the reason the abundance in the SH has decreased so much is because of significant denitrification, which doesn't happen in the NH? Are the authors implying that the descending HNO₃ will mitigate the ozone hole chemistry by re-nitrifying the air?

Page 9598 Lines 20. The mixing ratios only increase during part of the descent; then they begin to decrease. So whether there is continued production seems to depend on the time frame.

Summary and Discussion. The authors should quantify how important observing the HNO₃ enhancements is for understanding the contribution of EPP to the stratospheric NO_y budget. Are historical estimates based on solar occultation instruments that do not measure HNO₃ in error because they only include NO_x?

Technical Corrections: Page 9594 Line 16. Change 'has reached'; to 'had reached'. Page 9595 Line 15. Change 'makes also'; to 'also makes'. Page 9596 Line 21. Change 'mixing ratios'; to 'mixing ratio'. Page 9597 Line 6. Remove 'the'; before '1400 K'. Page 9597 Line 11. Should be 'associated with solar flares'; not 'associated to'. Page 9597 Line 8. Change 'damp'; to 'dampen'. Page 9597 Line 8. Change 'in sunlit regions'; to 'to sunlit regions'.

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