

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 #1

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General comments:

The paper presents time series of HNO₃ in the polar stratosphere and lower mesosphere measured by Odin/SMR in the period 2001 to 2007. The data as such as well as their representation (vs. equivalent latitudes and potential temperature) already are presented in the companion paper by Urban et al. This paper here focusses on HNO₃ enhancements in the upper stratosphere and mesosphere which are proposed to be linked to high energetic particle activity. The authors propose a so-called two-stage process (I would prefer the wording “two-phase process”) of HNO₃ enhancement after solar proton events with a rapid short-lived increase immediately

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after the SPE and a longer-lasting enhancement, revealing also downward transport in the stratosphere, for some weeks to months after the SPE.

Besides the presentation of the data as such (which is slightly different to that in the Urban et al. companion paper, since altitude-time cross sections besides latitude-time cross-sections are presented additionally), the paper does not make the attempt of a quantitative analysis of atmospheric processes leading to the observed HNO₃ enhancements in the upper stratosphere/mesosphere. As already mentioned, a two-stage process on HNO₃ enhancements initiated by solar proton events is proposed. No mechanisms leading to this two-stage increase are discussed. I could even accept the paper if, as the only analysis of the data, a thorough attempt had been made to prove this two-stage increase and its relation to solar proton events. However, the paper in its current form is not at all stringent in this point: there are winters observed with and without such a two-stage increase after SPEs, with enhancements before the SPE, with after-SPE enhancements from the middle stratosphere to the mesosphere and those in the middle stratosphere only, but the paper does not make any attempt to discuss and explain all these different observations versus the two-stage enhancement hypothesis.

As consequence, I do not think that the paper can go into ACP in its present form. It needs significant improvement in terms of analysis and discussion of the observations. My recommendation to the authors is to focus on their hypothesis of the two-stage enhancement and provide a proper elaboration of this hypothesis on basis of the presented data. In detail, I have the following comments regarding the paper:

- 1) *Does the paper address relevant scientific questions within the scope of ACP?* Yes, the topic as such is appropriate for ACP.
- 2) *Does the paper present novel concepts, ideas, tool, or data?* The paper presents new data, which, however, are already presented in the companion paper by Urban et al..
- 3) *Are substantial conclusions reached?* This is difficult to say; to my opinion, findings

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from earlier work are repeated here, without making a serious attempt to confirm or disprove them on basis of the new Odin data.

4) *Are the scientific methods and assumptions valid and clearly outlined?* As far as I can see, there are no scientific methods; presentation of the data as time series vs. equivalent latitude and potential temperature has been taken from the Urban et al. paper.

5) *Are the results sufficient to support the interpretation and conclusions?* I don't see obvious contradictions, i.e. the new Odin data confirm findings from earlier work which are repeated here; however, the interpretation is too shallow and does not really dig into the data.

6) *Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)?* Regarding retrieval of HNO₃ distributions from Odin data, and the representation of the data in the plots, the authors refer completely to the companion paper by Urban et al. and earlier Odin papers. Besides this, there are no experiments and calculations to be described.

7) *Do the authors give proper credit to related work and clearly indicate their own new/original contribution?* At several places, references to relevant earlier work are missing.

8) *Does the title clearly reflect the contents of the paper?* Yes.

9) *Does the abstract provide a concise and complete summary?* The abstract could be more concise. What is summarized in the abstract, are not really new findings.

10) *Is the overall presentation well structured and clear?* It is not always clear what is repetition of earlier findings, and what is meant as new interpretation of the presented data.

11) *Is the language fluent and precise?* The English is very good, as far as I can judge; one term is somewhat misleading and should be replaced (see below).

12) *Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?* Yes, as far as present in the paper.

13) *Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced,*

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combined, or eliminated? Sections 2.2 and 2.3 are so short that they hardly provide information. If the paper will be rewritten, they must be extended, and a more thorough interpretation must be provided.

14) *Are the number and quality of references appropriate?* As stated under 7), some relevant references are missing. On the other hand, the fourth paragraph of the introduction refers to some findings and cites a number of papers which are not closely related to the topic of the paper.

15) *Is the amount and quality of supplementary material appropriate?* N/A.

Specific comments:

Abstract:

The enhancements as shown in the figures reach up to the mesosphere (appr. to the 1800 K level), not only to the upper stratosphere.

I agree with reviewer # 4, for the same reasons, that the term "two-stage enhancement" might be misleading. I would prefer "two phases of enhancements" or something similar.

The sentence "We highlight the fact that ... energetic particle precipitation." is very un-specific and, thus, appears close to meaningless to me. The authors should be more specific here.

Introduction:

p9593, l15: References to Kawa et al. (1995) (CLAES observations) and Stiller et al. (2005) (MIPAS observations) are missing.

p9593, l20-21: The list of references is rather incomplete; at least add "and references therein".

p9594, l9-13: The topic introduced here is not really closely related to the topic of the paper and could be removed. This would reduce the length of the introduction and the reference list, which both are disproportionately long for such a short paper.

p9594, l27-29: As already pointed out by reviewer # 4, the statement that HNO₃ enhancements do not need to coincide or follow NO_x enhancements indeed is very

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strange, and I agree with reviewer # 4 that it must be meant vice-versa: not all NO_x enhancements lead to HNO₃ enhancements.

Section 2:

p9595, I24: Further references to work with ACE-FTS, HALOE and POAM data would be appropriate here, for example various papers by Randall et al. (see "References" below).

Figs. 2 and 3 must become 2-column figures, otherwise they are too small.

Section 2.1 and 2.2:

The two sub-sections mainly provide a description of the figures, but hardly any interpretation of the observations and no quantitative analysis in terms of atmospheric processes at all. The text is extremely short and I doubt that it might be useful for a non-specialized reader. Further analysis of the data should take into account: a quantitative analysis of the relation of inter-annual HNO₃ variation to 1) varying NO_x enhancements in the different winters (data from other sensors, eg. GOMOS, OSIRIS, etc. may be used), or, if this is not possible, at least dependence on varying ionization of the atmosphere by EEPs and SPEs; and 2) the varying dynamical situation in the polar vortices, e.g. strength of descent, major and minor stratospheric warmings etc.; 3) intercomparison of the NH and SH HNO₃ enhancements; 4) quantitative analysis of the relevance of the time of the year the SPEs occurred (e.g. in terms of solar zenith angle or non-illuminated area of the polar vortex) etc..

Regarding the individual winters, following questions occur to me:

Why is there no enhancement in the NH uppermost stratosphere and mesosphere during and just after the SPE in December 2001, while enhancements occur in this altitude region after other SPEs?

Why don't we see HNO₃ enhancements in late winter 2003/2004 which is known to have encountered extremely high NO_x intrusions (Randall et al., 2005; Seppälä et al., 2007; Hauchecorne et al., 2007; etc.)

The winter 2005-2006 showed severe NO_x intrusion from above (Randall et al., 2006).

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Why is the HNO₃ enhancement so little in this phase of the winter?

The winter 2005-2006 is not mentioned at all. How does it compare (in terms of NO_x amounts, dynamical situation, ionization etc.) to the other NH winter without SPE (2002-2003)?

Why are the HNO₃ enhancements in winter 2006-2007 after the December 2006 SPEs so different to other SPE-related enhancements? Why is there no enhancement above 1500K during and immediately after the SPEs?

The discussion of the SH winters is very poor. We see large differences, most of the winters are SPE-free, but the differences are not analyzed at all. The discussion of winter 2003 simply repeats the findings of Stiller et al. (2005) without involving own conclusions from the new Odin data.

Section 3:

p9597 - p9598, l2: This statement is, as I believe, true, but no attempt has been made within this paper to relate observed HNO₃ enhancements to increased levels of NO_x, in order to prove or disprove it on basis of the new Odin data.

p9598, l4: NO_x is not only produced by auroral electrons but by also by higher energetic electrons.

p9598, l4-l9: Again, this statement is believed to be true from earlier scientific work, but, again, no attempt has been made within this publication to prove or disprove it on basis of the new Odin data.

First para of section 3 provides the state-of-the-art of knowledge on the relevant processes, but does not provide findings or conclusions based on the data presented.

Second para: The statement "While short-lived HNO₃ enhancements could be triggered by EPP in summer, ..." seems not correct to me as it stands. During summer, HNO₃ will immediately be photolyzed; if N₂O₅ should play a role in HNO₃ formation via heterogeneous reactions on hydrated ion clusters, as stated in the introduction, N₂O₅ would not be sufficiently available in polar summer.

p9598, l17-l19: Do the authors state here that the enhanced HNO₃ from the upper

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stratosphere has impact on the denitrification of the lower stratosphere and the heterogeneous processes leading to ozone depletion? If so, the authors should state this more clearly and elaborate which impact they expect/observe.

References:

Hauchecorne A., J.-L. Bertaux, F. Dalaudier, J. M. Russell III, M. G. Mlynczak, E. Kyrölä, 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.

Kawa, S., J. Kumer, A. Douglass, A. Roche, S. Smith, F. Taylor, and D. Allen (1995), Missing Chemistry of Reactive Nitrogen in the Upper Stratospheric Polar Winter, *Geophys. Res. Lett.*, 22(19), 2629-2632.

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

Randall C. E., V. L. Harvey, C. S. Singleton, S. M. Bailey, P. F. Bernath, M. Codrescu, H. Nakajima, J. M. Russell III (2007), Energetic particle precipitation effects on the Southern Hemisphere stratosphere in 1992 - 2005, *J. Geophys. Res.*, 112, D08308, doi:10.1029/2006JD007696.

Randall C. E., V. L. Harvey, C. S. Singleton, P. F. Bernath, C. D. Boone, J. U. Kozyra (2006), Enhanced NO_x in 2006 linked to strong upper stratospheric Arctic vortex, *Geophys. Res. Lett.*, 33, L18811, doi:10.1029/2006GL027160.

Seppälä A., M. A. Clilverd, C. J. Rodger (2007), NO_x 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.

Stiller G. P., et al. (2005), An enhanced HNO₃ second maximum in the Antarctic midwinter upper stratosphere 2003, *J. Geophys. Res.*, 110, D20303, doi:10.1029/2005JD006011.

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