

**Comment on “A nitric acid dataset from IASI for polar stratospheric denitrification studies”
by Ronsmans et al.**

This manuscript analyzes 10 years of IASI HNO₃ total column measurements in conjunction with ERA-Interim temperatures to characterize the onset of PSC formation in the Antarctic lower stratospheric vortex. The high-density horizontal sampling afforded by IASI is valuable, and the approach of using the minimum in the second derivative of the HNO₃ total column with respect to time to identify the onset of HNO₃ uptake into PSCs is interesting and potentially useful. However, several aspects of the analysis and/or its description in the manuscript are flawed. Although referee comments raising serious issues with this manuscript have been posted, there are a number of points that we would like to add and/or further elaborate.

Respectfully,
Gloria Manney and Michelle Santee

General comment: Throughout this manuscript, starting with its title, the term “denitrification” is taken to be synonymous with the uptake of gas-phase HNO₃ through the formation of PSCs. Although not without precedent, this approach is contrary to common practice and may lead to confusion. Condensation of HNO₃ in PSCs is usually referred to as “sequestration”, while the term “denitrification” is usually reserved for the permanent removal of HNO₃ from the lower stratosphere through the sedimentation of PSCs. In the absence of analysis of direct PSC measurements (e.g., from an instrument such as CALIOP), the occurrence of true denitrification can only be inferred from space-borne measurements of gaseous HNO₃ when abundances do not rebound as PSCs dissipate at the end of winter, suggesting permanent removal. Thus the “drop temperature” derived in this study is indicative only of the onset of PSC formation, not the onset of denitrification, as is stated in numerous places in the paper.

Abstract

- L2: It is misleading (particularly for those who read only the abstract of the paper) to characterize the IASI HNO₃ total columns as having “good vertical sensitivity”. Indeed, this optimistic assessment is directly contradicted in Section 2, where IASI is stated to have “low vertical sensitivity ... with only one independent piece of information” (L76).

Introduction

- L48-49: It should be made more clear that this is by no means an exhaustive list of spaceborne instruments that have measured stratospheric HNO₃.

Section 2

- The information provided about the IASI HNO₃ retrieval, data quality, and data screening is insufficient. This information is critical to assessing the robustness of the reported results, and readers should not be forced to refer to previous papers to find it.
- In later sections (e.g., L186, L225), errors in IASI retrievals arising from issues with emissivity above ice shelves are invoked to account for some dubious results, but no

mention of these poor-quality retrievals is made in the “Data” section, nor is it explained why quality-control measures fail to properly filter out these suspect data points.

- L78: 10 km can hardly be characterized as the “mid-stratosphere”.
- L84: “normal” has a specific statistical meaning and is not the appropriate word here.
- L85-86: The validity of the analysis approach depends on the 50 hPa pressure surface and the 530 K isentropic surface being in very close proximity during Antarctic winter. This implicit assumption should be explicitly justified in the paper.
- L89-91: It is highly problematic to use a single theta level to distinguish inside from outside vortex regions for column measurements. This approach implicitly (and erroneously) assumes that the vortex does not tilt, shrink, or expand with height over the altitude range considered. A better approach would have been to check PV over a range of levels and discard measurements classified as outside the vortex at any one of those levels. A similar comment can be made concerning the use of a single pressure level for temperature. Again, it might have been better to use a range of T over the ~10–30 km layer where IASI has most sensitivity. Some attempt is made to justify the latter choice (using 195 K at 50 hPa) in Section 3 (L141-142) and Section 4 (L168-169), but the arguments are not convincing, as the authors themselves appear to recognize when they state (L188-189) “hence, the use of temperature at a single pressure level might be restrictive to some extent”.

Section 3

- The definition of the three “regimes” in the T/HNO₃ relationship seems arbitrary and not well justified. For example, R1 is defined to begin in April, but Fig. 1a shows that HNO₃ values start to increase rapidly and temperatures start to decrease rapidly in March (or even February, as noted in L117), not April. Only R2 encompasses a steep change in HNO₃, but that regime also includes a lengthy period during which HNO₃ remains nearly constant. It might have been better to break R2 into an “onset of PSC formation” phase and a “denitrification plateau” phase. Moreover, as defined in the paper, R2 extends through, not to, September as stated in L108. These problems are evident in the discussion in this section, as in some cases the behavior ascribed to one regime actually occurs in another.
- L102 and Fig. 1 caption: The red line in Fig. 1a is horizontal, not vertical, and Fig. 1b contains no such line – it is on Fig. 1c. Neither red line is defined in the caption.
- L102 and Fig. 1: 2011 was a particularly cold and long-lasting Antarctic winter, and thus it is arguably not representative. Some explanation for why that year was selected for highlighting in Fig. 1b is needed.
- L105-106: The contribution of confined descent inside the developing vortex bringing air rich in HNO₃ from above into the domain where IASI is most sensitive has been ignored here – isn’t descent also a factor leading to the observed high HNO₃ total column values in early austral autumn?
- L115-116: In addition to a lack of citations of earlier papers on renitrification of the lowermost stratosphere (LMS), this sentence is not a very clear expression of the fact that IASI is not sensitive to the LMS and hence renitrification has little impact on the observed evolution of total column HNO₃.

- L119-121: Why is 2010 highlighted in Fig. 1a (green line)? Other recent Antarctic winters were also disturbed with some minor SSW activity, e.g., 2012 and 2013. Did those episodes not affect the HNO₃ distribution? Also, why does the green line show T at 20 hPa, when the other curves show T at 50 hPa? More explanation for why the authors chose to show this particular level for this particular year is needed.
- Fig. 1c: In general this plot is not well explained or well motivated. By showing the position in temperature / HNO₃ space of the bin with the maximum number of observations, important information about the range of those values on a given day is omitted. The ranges in Fig. 1b suggest that the values at a given time may span most of the HNO₃ axis in Fig. 1c, rendering the curves shown less meaningful. In addition, it is stated (L127) that this figure highlights the interannual variability in total HNO₃, but interannual variability is also clearly seen in panel (a), which is much easier to interpret. The discussion relates the picture in Fig. 1c to the three regimes, but since they are not marked on this panel, it cannot easily be examined without reference to Fig. 1a. It is therefore not obvious what additional value this figure brings to the paper.
- L125: HNO₃ columns are said to slowly increase as the T decreases over “February to May, i.e., R3 to R1”. However, R3 is defined to start in October, and actually the slow increase in total HNO₃ starts before February, arguably even as early as December.
- L126: In the discussion of strong and rapid HNO₃ depletion, “June (R1-R2)” should be “June-August (R2)”.

Section 4

- Fig. 2 and its caption: More should be said about the agreement (or lack thereof) between the dashed and solid HNO₃ and the grey and red T lines when they both exist. Some readers may question why the PV approach is used, given the gaps in those curves. Also, perhaps this is just an optical illusion, but the solid blue line appears to be thicker in some years (2011, 2014, 2016, 2017) than in the others. If that is the case, then that also needs to be explained. In the caption, the level to which the stated PV value pertains (presumably 530 K) should be specified.
- L155: It is not appropriate to characterize the total HNO₃ depletion in the inner vortex as being the “coldest”.
- L160: The wording in this sentence is garbled.
- L162-163: 23 is more than “a few” days.
- L174-179 and Fig. 3 caption: The description of the figure is confusing. It is stated in both in L174-175 and the caption that the vertical red dashed line indicates, at 90S, the 10-year average of the drop temperatures (191.1 K) calculated from the HNO₃ second derivative time series in the area delimited by the $-10 \times 10^{-6} \text{ K} \cdot \text{m}^2 \cdot \text{kg}^{-1} \cdot \text{s}^{-1}$ PV contour. It’s not clear how a vertical line on a time series plot can represent a temperature value. Perhaps the authors meant to say the average date on which T dropped below the 195 K threshold at 90S? Moreover, the discussion above indicated that the value of 191.1 K was the average for the inner vortex (defined by either PV or EqL), not specifically at the South Pole (90S). In addition, the scale for the PV contour should be 10^{-5} , not 10^{-6} . Then in L176-177, it is stated that the “delay of 4-23 days between the maximum in total

HNO₃ and the start of the depletion is also visible” – but how is a range of values (which arises from different years) visible in a climatological plot?

- Fig. 4: Very little discussion is devoted to this figure; it is merely noted (L177-178) that it shows the reproducibility of the IASI measurements of HNO₃ depletion from year to year. Since Fig. 1 already makes this point, the added value of Fig. 4 is not clear.
- Fig. 5: How relevant is the PV contour averaged over the May to October period, when the dates of the onset of HNO₃ depletion are May to June (or possibly July)? Why include August, September, and October in this average?
- L181: “the drop 50 hPa temperatures” should be “the 50 hPa drop temperatures”.
- L183: Technically, the isocontour represents -10 , not ≤ -10 .
- L184-185: First, how does the range of dates corresponding to the onset of HNO₃ depletion reported here – mid-May to early July – relate to that reported (L163) in connection with Fig. 2, which was 17 May to 10 June? Does the difference in these estimates arise because the former is based on averages in $1^\circ \times 1^\circ$ bins, whereas the latter is based on a vortex average within the PV contour? July seems rather late for the onset of PSC formation. Similarly, the range in 50 hPa drop T is quoted as 188.2 K to 196.6 K in L164, whereas here drop Ts vary over a wider range, from 180 to 210 K. The values at both extremes of this range are unrealistic. Indeed, the date and T ranges found in connection with Fig. 5 call into question the analysis method.
- L189-196: The questionable results derived from this analysis cannot be pinned on biases in the ERA-Interim data. The statement is made that “Reanalysis data sets are, indeed, known to feature large uncertainties”, but the uncertainty in modern reanalysis temperatures (typically less than ~ 1 K) is by no means large enough to account for drop Ts as extreme as 180 and 210 K. The reliability of reanalysis temperatures in the polar lower stratosphere (including those from ERA-Interim) has been conclusively demonstrated in several recent papers, notably by Lawrence et al. [2018] and Lambert and Santee [2018]. Although both papers are cited here, their implications have apparently been overlooked.
- L197-199: This sentence is confusing and its intended meaning is unclear. It appears to be comparing apples (the spatial variability in drop T seen in the maps in Fig. 5) to oranges (“natural” variations in PSC nucleation T, TTE, and PSC formation mechanism). Perhaps the authors meant the spatial variability in those parameters (and not the values themselves), but that is not how the sentence is constructed. In any case, further discussion of comparisons of Fig. 5 with previously published results is warranted.
- L199-200: A number of other satellite data sets have captured gas-phase HNO₃ depletion (from both sequestration and denitrification) on similarly large scales.

Conclusions

- L225-226: It is stated that “the range of drop temperatures is interestingly found in line with the PSCs nucleation temperature that is known, from previous studies, to strongly depend on a series a factors”. In fact, the derived range (180–210 K) is so large that it is arguably not in line with previous work, and it is therefore difficult to see how the IASI total column HNO₃ measurements provide added value (as stated in L203) to studies of

Antarctic PSC formation and the interannual variability therein beyond that obtained from other satellite HNO₃ datasets.

- L230-231: The statement that this paper represents “the first time that such a large satellite observational data set of stratospheric HNO₃ concentrations is exploited to monitor the evolution HNO₃ versus temperatures” is wholly unsupported. In fact, there is a substantial body of literature on the relationship between HNO₃ and temperature, including studies of long-term vertically resolved datasets. In particular, Lambert et al. [2016] (which is cited in a number of places in this manuscript, but only in passing) examined 10 years of Aura MLS HNO₃ in the Antarctic winter vortex and its relationship to T – including temperature history (a factor that has been largely ignored here) and T with respect to T_{ICE} – as well as PSC composition as determined by CALIOP. In general, discussion of how the current results fit into the context of the findings from Lambert et al. [2016] and other relevant prior studies is inadequate.
- L233-234: More explanation of how HNO₃ total column amounts could be used to inform PSC classification schemes is needed to justify this statement, especially given how spatially heterogeneous and layered PSCs have been shown to be.

Finally, in addition to the serious substantive issues enumerated above and in the formal reviews of the official referees, the manuscript suffers from the poor quality of the writing. If this paper were to be eventually accepted for publication, it would require extensive copy-editing to improve the English.