

Interactive comment on "Spatio-temporal variations of HNO₃ total column from 9 years of IASI measurements – A driver study" *by* Gaétane Ronsmans et al.

Anonymous Referee #2

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This paper presents 9 years of daily global total column HNO3 measurements from IASI and, for the first time, performs a detailed MLR analysis on them to statistically characterize the relative contributions of several explanatory variables in controlling the stratospheric HNO3 distribution. The manuscript is generally well organized and well written, and the production quality of the figures is also high. In my opinion, the study makes a valuable contribution and warrants publication. I do, however, have a number of mostly minor substantive comments (detailed below) that I would like to see addressed before the manuscript is accepted for publication.

General comment:

C1

* One general – though minor and easily rectified – comment is a pervasive lack of adequate referencing throughout the manuscript. PSC formation and denitrification, and their roles in chlorine activation and chemical ozone loss, are extremely well-studied phenomena, and obviously it is not possible (or even desirable) to cite every paper on these topics published in the last 30 years. But in many places the authors have chosen to cite only a few papers for well-known points, without prefacing the list with "e.g.". This may seem like a petty point, but not only does their selection of which papers to reference often come across as arbitrary, but also their approach may give non-expert readers the impression that only those few highlighted papers are of relevance. So I suggest going through the manuscript and adding "e.g." in front of the list of cited papers in many places. Some specific examples of where this is needed include: p2, L4; p2, L6; p2, L8; p2, L19; p2, L23; p8, L2; p8, L4; p8, L7; p8, L17; p8, L18; p8, L24. Similarly, although the source (typically a URL) for each proxy is given in Table 1, I feel that it would be appropriate to provide a general citation in each sub-section of Section 4.3 where a given proxy is introduced. For example, references to published literature are needed on p7, L27 for F10.7, p8, L14 for MEI, and p8, L21 for AO and AAO.

Specific substantive comments and questions:

*p2, L26: I do not think it is true that "most often" MLR studies use an iterative selection procedure to identify relevant explanatory variables. In fact, I believe that only a handful of the many MLR ozone studies have done so. (And it seems strange to say "most often" and then cite only one reference.)

*p3, L30: Does the cloud screening of IASI data include PSCs?

*p4, L9: The PSC formation threshold is stated to be 195 K. It is fine for the purposes of this kind of analysis to use a constant value to indicate the likely presence of PSCs, but it should be acknowledged that the temperature at which NAT forms varies with altitude and time over the season, and thus this value is approximate.

*p5, L4-5: It is true that these IASI results confirm earlier findings, and references are

needed here.

*p5, L9-11: I find this part of the discussion confusing. First, it is stated that the "delayed denitrification" in the 65-70S band is attributable to "the later appearance of PSCs" and "the mixing of these air masses with the denitrified air masses from the center of the vortex". Are the authors asserting that some of the decrease in HNO3 observed in the 65-70S band does not arise directly from PSC sedimentation within that band, but rather from dilution of HNO3 abundances through mixing with denitrified air masses from deeper in the vortex core? In that case, the decrease in HNO3 should not be called "denitrification". More importantly, is this suggestion consistent with the findings of Roscoe et al. [JGR 117, 2012] that the broad vortex edge region is only weakly mixed with the deep core during the winter? Second, the next sentence states that these "two processes lead to the total columns in both eqlat bands being in the same range of values by the end of December". The Antarctic vortex is breaking down (or has mostly broken down) by the end of December, so of course mixing at this time homogenizes the high-latitude HNO3 distribution, but it doesn't make sense to be talking about the later appearance of PSCs in this context.

*p5, L16-18: I also find these sentences confusing. It is stated that the columns in the 55-65S band keep increasing during the low-temperature periods, but cold intervals are not marked for that eqlat band. Are the authors referring to periods that are cold at higher latitudes? If so, then this statement is not entirely correct, as HNO3 values at 55-65S start to decline from their peak values while temperatures are still low in the 70-90S and 65-70S bands. The maximum in HNO3 values in June-July is attributed to "less sunlight compared to lower latitudes", but the comparison shouldn't be to lower latitudes but rather midwinter vs summer (at the same latitude). In addition, the role of confined diabatic descent inside the vortex should be mentioned, as it is a major factor leading to strongly enhanced wintertime HNO3 abundances in the lower stratospheric layer to which the IASI column amounts are most sensitive.

*p5, L23-24: The statement that temperatures in the northern high latitudes rarely

C3

reach the PSC formation threshold is much too general. While that is true for the polar-cap (70-90N) average being considered here, temperatures in the Arctic lower stratosphere certainly do drop below PSC formation thresholds in localized regions in almost every year. Moreover, it is not the *average* temperature – which is what I believe is being shown in Figure 2, although it's not clear – that is important for PSC formation. It is the *minimum* temperature that is important. In fact, if indeed Figure 2 is showing eqlat band average temperature, then it should be reformulated to correlate HNO3 behavior with the minimum temperatures in that band. In any case, the exact nature of the temperatures being shown should be specified (at the beginning of Section 3 and in the caption).

*p6, L4-5: For ease of reference, the lack of IASI data in September-December 2010 should be first noted in Section 2, where the data set is described. It seems to me that this interval is also noticeable in Figure 2, so I suggest removing the data during this period in that Figure as well.

*p6, 7-9: It is hypothesized that the anomalous behavior in July-August 2010 seen in IASI HNO3 data was a consequence of descent induced by the midwinter minor warming. It seems to me that a more obvious explanation is that the SSW caused lower stratospheric temperatures to rise sufficiently that PSC formation was temporarily inhibited. It is worth noting in the manuscript that a similar evolution of HNO3 was recorded by Aura MLS in that winter, as shown in Figure 3-6 of the 2014 WMO Ozone Assessment. The 2014 WMO Report also showed that in 2010 VPSC (based on MERRA) remained well below the 1979-2012 Antarctic average and less denitrification than typical occurred.

*p10, L20-21: I find this discussion confusing. First, a *delay* in the drop in HNO3 concentration in the fit for the 65-70S band is noted, but then it is stated that it "happens *earlier* than in the IASI observations since the VPSC proxy is based on temperatures and composition *north* of 70". Figure 6 does show that the fitted midwinter peak in HNO3 slightly precedes that observed, so I assume that "earlier" is correct and "delay"

must be a typo. However, it is also true that the HNO3 decline is more gradual in the model than in the data, so that in late winter the fit line lags the observations. Exactly which behavior is being discussed should be clarified. Also, since it is the Antarctic that is being talked about here, "north" should be "poleward".

*p10, L27-28: The deep minima in HNO3 in the northern polar regions in October 2014 and 2016 almost certainly have nothing to do with denitrification during the preceding Arctic winters. Any signature of denitrification gets completely obliterated when the vortex breaks down at the end of winter. Even in the Antarctic, where denitrification is severe every winter, its signature is not still visible in the high-latitude HNO3 abundances the following fall. The extremely low 70-90N HNO3 values in October 2014 and 2016 (and also 2012, when the residuals are particularly large) are indeed quite interesting, but they cannot be ascribed to denitrification. It's possible that the low HNO3 observed in boreal fall 2016 may have been linked to the QBO disruption [e.g., Tweedy et al., 2017].

*p11, L14-15: It is noted that parts of Eurasia stand out with a low percentage of observed variability explained by the model. Could this be related to the low sensitivity of IASI data in this region, where the elevated terrain of the Tibetan Plateau reduces the signal-to-noise of the retrieval (e.g., Luo et al., ACPD 2017)?

*p11, L16-21: The low fraction of explained HNO3 variability in the tropics and subtropics is attributed to lightning NOx production. In addition to sources, unaccounted-for sinks of HNO3 should also be considered, such as scavenging in convective updrafts and cirrus clouds.

*p11, L27-28: If the signal over southern Africa induced by NO2 from biomass burning is being carried by the annual term in the model, then shouldn't the coefficients a1 or b1 be larger in that region in Figure 11 (which is not the case)?

*p12, L1-2: It might be good to mention the issues with the retrievals caused by elevated terrain here as well.

C5

*p12-13, Section 4.4.3: I appreciate that the authors limited the number of figures, showing only the regression coefficient for each proxy (Figure 11) and not the fraction of HNO3 variability it explains. But the accompanying discussion frequently refers to the percentage contribution from specific proxies. Although some sense of their relative importance in different regions can be obtained from Figure 11 (and also Figure 8), I suggest either adding "(not shown)" everywhere a percentage contribution is discussed in this section or adding (and referring to) another figure containing this information.

*p12-13, Section 4.4.3: I would have liked to have seen a bit more discussion of whether these results for HNO3 are consistent with previous MLR analyses of ozone data that included similar terms. In particular, the SF results are not put into the context of previous findings. In addition, the positive signal above the southern polar region is characterized as "weak", but in fact the largest positive MEI regression coefficients are found over Antarctica. Is that in line with expectation? Previous studies looking at the influence of AO/AAO on ozone are alluded to on p13, L8, but no references are given there, and it is not clear whether the citations in the next sentence are relevant for this point (e.g., the 2009 paper by Wespes et al. is about HNO3 and does not discuss the AO/AAO). The influence of the QBO in the equatorial regions is noted, but no mention is made of the fact that the coefficients are much larger at northern high latitudes.

*p14, L32-33: I do not wish to take away from the value of the IASI HNO3 measurements, whose dense spatial coverage and long-term record are obviously of great benefit, as this study has shown. But I would ask for a bit more care in the language used here. Although the novel statistical nature of these results is mentioned, I think that some readers could take away from these lines the message that this analysis has revealed the profound influence of PSC formation and denitrification on the HNO3 distribution, when in fact the crucial role of those processes has been known for decades. In truth, it is not obvious to me what additional knowledge about the variability of HNO3 in the polar regions has been gained from this study that had not been demonstrated previously using limb measurements with much coarser horizontal but much greater vertical resolution.

*p25, Figure 2: Minor tick marks on the y-axis would be helpful. As mentioned earlier, it might be good to remove the sparse measurements during the September-December 2010 interval from this plot as well. Why do some of the vertical lines, especially (but not only) in the purple 65-70 eqlat region, appear to be thicker? Is it because temperatures are hovering around the PSC threshold at those times, so the shading is being turned off and on multiple times in guick succession?

*p30, Figure 7: Why is there a break in the without-VPSC fit curve in the 70-90S panel in October-November 2014? Such a break does not appear in the similar panel for the with-VPSC fit (or in Figure 6).

*p33, Figure 10 caption: The wording of the caption ("Time evolution of IASI HNO3 (red) and NO2 (green)") implies that the NO2 data are from IASI, but the reference cited is for GOME-2 data. Please clarify.

Minor points of clarification, wording / figure suggestions, and grammar / typo corrections:

*p1, L13: "PSCs" should be defined in the abstract as well as the main body of the paper.

*p1, L23: inexsitent -> nonexistent

*p2, L3: "PSCs" was already defined on p1, L20

*p2, L8: and further -> followed by

*p2, L11, L14: These acronyms (UARS, MIPAS, ACE-FTS) should probably be spelled out. Also, "AURA" -> "Aura" and "ODIN" -> "Odin" (they are not acronyms, just names)

*p3, L10: bi-daily -> twice daily ("bi-daily" could be interpreted to mean every two days)

*p3, L15: The university name should be spelled out here

C7

*p3, L24: Can 15-20 km really be considered the "low-middle" stratosphere? This seems more like just the lower stratosphere to me.

*p3, L30: higher fractional cloud cover than 25% –> fractional cloud cover higher than 25%

*p4, L19: Further than -> Beyond

*p4, L22: delete "columns" (some of the previous studies were based on HNO3 profiles, not columns)

*p5, L14: delete "itself"

*p5, L20: more -> longer

*p5, L26: It would be good to add "Arctic" in front of "winters" and "over a broader area" after "threshold"

*p5, L35: "polar" -> "potential"

*p6, L1: it's not clear why only one contour is noted here, when 3 contours of PV are shown in both hemispheres

*p6, L4: EUMETSAT should be in all capital letters (as on p3, L29)

*p6, L12: dentrification -> denitrification

*p6, L14: What does "more stable" mean in this context? More constant over the season, or more uniform from year to year?? And what is the comparison against – wintertime values in the NH, or summertime values in the SH?

*p6, L21: I assume that "Cst" in Eqn (1) is a constant term, but it should be defined

*p6, L29: Kyrola et al. [2010] seems like an odd reference for such a general statement about the BDC. Wouldn't the Butchart [2014] review paper (already cited elsewhere) be a better choice?

*p7, L13: further -> below

*p7, L19: ENSO should also be mentioned here

*p8, L13: meridonal -> meridional

*p8, L23: I assume it is meant that AO/AAO are considered only in the high latitudes of the hemisphere they are related to, since they are both applied in equatorial regions.

*p8, L29: delete "(from 195 K or TNAT for the formation of nitric acid trihydrate particles)"

- *p8, L30: "PSCs" has already been defined
- *p8, L31: delete "either"
- *p9, L2: NAT has already been defined
- *p9, L3: gaz -> gas
- *p9, L13: dynamic -> dynamics
- *p9, L23: Section 3.2 -> Section 4.2
- *p9, L26: add "bands" after "90S"
- *p9, L29: add "major" after "Most"
- *p9, L30: RMSE is used here for the first time but not defined until p11, L31
- *p10, L6: denitrifications -> denitrification seasons
- *p10, L11: delete "here"
- *p10, L17: better -> improved
- *p10, L23: dynamic -> conditions
- *p11, L5: The reference to Section 4.4.1 is incorrect (this part of the discussion is itself

C9

in Section 4.4.1)

- *p11, L11: most -> much; also add "generally" in front of "between"
- *p11, L19: between -> in
- *p11, L21: emitted -> produced
- *p11, L25: oxydation -> oxidation
- *p11, L34: desertic -> desert

*p12, L30: For MEI, "south of Africa" the results are not significant. I think "west of South Africa" would be better here.

- *p13, L5: Groenland -> Greenland
- *p13, L10: largely -> strongly
- *p13, L21: further -> subsequent
- *p13, L25: add "proxy" after "VPSC" and "HNO3" in front of "variability"
- *p14, L7: reveal -> reflect
- *p14, L14: between -> in

*p14, L23: but still allow improving significantly the model-to-observation agreement -> but accounting for PSCs still significantly improves the model-to-observation agreement

*p29, Figure 6: The font for the year labels on the x-axis seems to be disproportionately large

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