Review of "Polar stratospheric nitric acid depletion surveyed from a decadal dataset of IASI total columns" by Ronsmans et al.

This manuscript analyzes 10 years (2008–2017) of IASI HNO_3 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 very valuable, and the novel approach of using the minimum in the second derivative of the HNO_3 total column with respect to time to identify the onset of HNO_3 uptake into PSCs is useful. Although I was a co-author of a Short Comment posted on the originally submitted manuscript, I was not one of the referees for it, nor have I read any of the previous revised manuscripts. However, I have now been asked to serve as an official reviewer of the current draft, so I am reading the paper for the first time in that formal capacity. Hence, despite the fact that the manuscript has already been revised twice in response to previous comments, some issues are being raised now for the first time. Moreover, some of the concerns expressed in the original reviews have not yet been adequately addressed, and new issues have been introduced through the revision process. Thus, although the manuscript has been much improved, I feel that further corrections and clarifications, as detailed below, are necessary before the paper can be accepted for publication.

Below both major substantive issues and minor points of clarification, wording suggestions, and grammar / typo corrections are listed together for each Section in sequential order.

Respectfully,
Michelle Santee

Abstract:

- L19: in the mid-stratosphere --> in the lower-to-mid stratosphere; also, delete "causal"
- L23: This wording is very awkward; I suggest: evolution of the pair HNO_3-temperature --> evolution of HNO_3 together with temperature
- L24: The meaning of "in the cycle of IASI" is not clear – do the authors mean "annual cycle"?
- L27-28: delete "differentiating"; add "to be differentiated" after "profile"
- L28: Two different values for the average drop temperature are given in the text on p. 6 (L265 and L290). These values need to be reconciled and the correct one quoted here.
- L32-33: It is a bit of an exaggeration to say that "this paper highlights the capability of IASI to monitor the long-term evolution of polar stratospheric composition and processes involved in the depletion of stratospheric O_3". It would be more accurate to state that "this paper highlights … evolution of polar stratospheric HNO_3, a key player in the processes involved …".

Introduction:

- In a number of places in the presentation of background material (e.g., L39, L42, L45, L51), a few citations are given for very well-established concepts, but many other equally suitable papers could have been cited instead of or in addition to the ones listed. Obviously not all relevant papers can be referenced for these points, but "e.g." should be added in these lines to avoid giving readers the impression that the selected references are the only appropriate ones.

- In addition, the recently published PSC review paper by Tritscher et al. (Rev. Geophys., 2021) covers all of this background material and should also be cited in several places in this section.
- L44-45: Technically, denitrification delays only the reformation of $ClONO_2$ – HNO_3 is not required for HCl production, thus it is not quite correct to say "chlorine reservoirs" here.
- L46-47: PSCs surface --> PSC surfaces; PSCs particle --> PSC particles
- L47-48: three 3different --> three different
- L56: delete comma after "(T_{ice})"
- L60: A reference should be provided for the point that NAT nucleation was thought to require temperatures below T_{ice}.
- L73: the PSCs formation --> PSC formation
- L74: add a comma after "2019)"
- L74-75: This sentence ("The influence of HNO_3 in modulating O_3 abundances in the stratosphere is furthermore underrepresented in CCMs") is out of place and potentially confusing for many readers, who will naturally assume that it has something to do with PSC processes in the lower stratosphere as it comes at the end of a long paragraph discussing nothing but PSCs. Kvissel et al. (2012), however, describe HNO_3 enhancements between 10 and 1 hPa induced by energetic particle precipitation. It's not clear why this sentence has been included here; perhaps it is meant to provide motivation for this study, but I doubt that IASI total column measurements with maximum sensitivity around 50 hPa could shed much light on modest (< 6 ppbv) HNO_3 enhancements in the middle and upper stratosphere. In fact, this issue is not explored in the manuscript. Thus this sentence should be deleted.
- L77: measure --> have measured
- L79-82: This poorly composed sentence is grammatically incorrect and hard to interpret. I'm not sure what is meant by "follow their formation mechanisms". I suggest alternative wording, but I may have misunderstood the intent: Spaceborne instruments such as the CALIOP/CALIPSO lidar and MIPAS/Envisat measuring in the infrared are capable of detecting and classifying PSC types, allowing their formation mechanisms to be investigated (Lambert et al., 2016; Pitts et al., 2018; Spang et al., 2018, and references therein); these satellite data complement in situ measurements (Voigt et al., 2005) and ground-based lidar (Snels et al., 2019).
- L84: the PSCs formation --> PSC formation; Urban 55 et al. --> Urban et al.
- L86: depends --> depend
- L92: similar to the limb --> similar to that from the limb
- L94: where the PSCs cloud form --> where PSCs form
- L95: 10-years --> 10-year
- L96: for providing --> to provide

Section 2:

- L104: embarked on --> onboard ("embarked" is not the right word)
- L117: FWHM --> full width at half maximum (FWHM)
- Figure 1 caption:
  - L446: molec.cm-^{2}/molec.cm-^{2} --> molec.cm^{-2}/molec.cm^{-2}
  - L446: after "molec.cm^{-2}/molec.cm^{-2}" add "; colored lines, with the altitude of each kernel represented by the colored dots)"
  - L447: Height --> Heights; 169,40 --> 169.40

- L122: Height --> Heights
- L125: Height --> Heights
- L126-127: Above (in L119) it was stated that the largest sensitivity of IASI is from ~70 to ~30 hPa, and here it says "the altitude of maximum sensitivity (at around 30 hPa for this case)". This is confusing because the grey line in the Arrival Heights panel of Fig. 1 depicting the "sensitivity profile" peaks at ~75 hPa, not 30 hPa. I believe that it is the total column averaging kernel, not the "sensitivity profile", that determines the altitude of maximum sensitivity? The authors should take into consideration that not all readers of this paper will be experts on IASI data or will have read Ronsmans et al. (2016). Please clarify the meaning of the "sensitivity profile" (grey curve) and the total column averaging kernel (black curve) and how they relate to the altitude where IASI provides the most information on HNO_3.
- L127: At Lauder on the contrary, --> In contrast, at Lauder
- L128: "larger range of maximum sensitivity" – again, is this conveyed by the "sensitivity profile" (grey curve) or the total column averaging kernel (black curve)?
- L129-130: The statement "from around 3% at … polar latitudes to 25% above the cold Antarctic surface during winter" is confusing, because of course the Antarctic is at polar latitudes. Please be more precise in the wording here.
- L130: DOFS --> degrees of freedom for signal (DOFS)
- L132: "lower than 12%" is ambiguous here; assuming that the values are biased low by more (not less) than 12%, this should be rewritten as "low bias (exceeding 12%)"
- L135: FTIR measurements which is not possible during --> FTIR measurements, which cannot be made during
- Figure 2 caption:
  - L454: It should be explicitly stated here (not only in the text) that the MLS total column estimates were obtained by extending the MLS partial stratospheric column values using the FORLI-HNO3 a priori information.
  - L454: 2.5 x 2.5 --> 2.5° × 2.5°
  - L454: middle --> bottom
  - L455: Figure 2 does not include a panel showing 30°–50°S EqL
- L137: 2.5 x 2.5 --> 2.5° × 2.5°
- L139: The meaning of "the averaging kernels … were considered" will not necessarily be clear to all readers. It would be better to state explicitly that the IASI averaging kernels were applied to the co-located MLS profiles.
- L140-141: "column profiles" is an oxymoron. Please specify the MLS retrieval pressures over which the partial stratospheric column was calculated. Thus, rather than "… grids, then converted into column profiles. They were also extended down to the surface by considering the …", it would be better to rephrase along the lines of "… grids, and partial stratospheric columns above xxx hPa were calculated. MLS total columns were then estimated using the …".
- L155-165: In their Short Comment, Manney & Santee pointed out: "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." In my opinion, the authors have not adequately

addressed this point. I understand that 530 K falls in the region of the atmosphere where IASI has maximum sensitivity to HNO_3. Nevertheless, it is simply not credible that changes in the size, shape, or location of the vortex over the 30–70 hPa domain primarily covered by the measurements had no effect on the results on any day in the 10-year IASI record. At this point, I am not suggesting that the authors redo their analyses, but I would like to see added to the manuscript an explicit acknowledgment of the fact that relying on the determination of the vortex boundary on a single potential temperature surface for interpretation of total column measurements inevitably introduces some uncertainty into the results because it fails to account for the possibility of changes in the size, shape, or location of the vortex with altitude.

- L158: closing ")" missing after "2016)" – it should be "2016))"
- L158: "starts a few degrees or slightly below" is awkward and confusing – "slightly" could mean "a few degrees". It would be clearer to say: "starts within a few degrees below"
- L161: It is not quite correct to say "to identify the PSCs-containing regions" – regions with temperatures below the threshold do not necessarily contain PSCs. It would be better to say "to identify regions of potential PSC existence".

Section 3:

- L170: delete the comma after "S)"
- L170: This wording – "over the whole period of measurements (2008–2017)" – seems to imply that IASI has taken no data since 2017, which I do not believe is the authors' intention.
- L172: 2018) where the contribution of the PSCs into --> 2018), where the contribution of PSCs to
- L173: delete "here"
- L175-176: The wording "along the HNO_3/temperature cycle" is not clear. I think the authors mean "within the HNO_3/temperature annual cycle". Or maybe "during" rather than "within"
- L184: "R1 in Figures 3a and b" – since the regimes are clearly labeled in the panels of Figure 3, I'm not sure that this statement is needed. Moreover, the label "R1" is also used in Figure 3c.
- L189: deployment --> development ("deployment" is not the right word); also, add a comma after "vortex"
- L190-191: lower latitudinal airmasses --> lower-latitude airmasses
- L193: A problem with the definition of R2 is that, as shown later in the paper, the onset of strong HNO_3 depletion actually begins in mid-May in most years.
- L193: add commas after "R2" and "October"
- L195: the HNO_3 total columns average below --> average HNO_3 total columns are below
- L198-199: To avoid the potential for confusion with SSWs: Despite the stratosphere warming with 50 hPa temperatures up to 240 K --> Despite 50 hPa temperatures increasing up to 240 K
- L202: by the PSCs sedimentation --> by sedimentation of PSCs
- L203: add "e.g." to the list of references
- L203-204: add commas after "2012)" and "2016)"
- L204-205: The meaning of "can hardly be inferred" is ambiguous. I expect that the lack of sensitivity of IASI total column HNO_3 to the LMS precludes detection of renitrification from those measurements. Thus: can hardly --> cannot
- L206: where --> when
- L210: occurs --> occurred

- L212: HNO_3 total columns in 2010 were higher in September as well as in July and August.
- L219: add "and" after "(R2),"
- L220: Based on Figure 3a, the plateau of low HNO_3 abundances begins in July, not August.
- Figure 3c:
  - Figure 3a clearly shows that the "strong and rapid HNO_3 depletion" (as stated also in L219) occurs mainly during June, so why is the steep drop in HNO_3 in Figure 3c labeled "Jun-Aug"?
  - I do not find the regime markers (R1, R2, R3) on this panel particularly helpful – these labels are essentially floating in semi-arbitrary positions on the plot and convey no real meaning.
  - It is stated that this panel "highlights the interannual variability in total HNO_3" (L221). But interannual variability is much easier to interpret in Figure 3a. For example, the anomalous behavior in July–September 2010 so evident in panel (a) does not stand out in panel (c). In fact, I would argue that it is not necessary to color-code the lines by year in panel (c), as the details of individual years are better seen in panel (a) anyway. Just having 10 separate lines would still illustrate the interannual spread even without distinguishing the specific years.
  - If the lines are not color-coded by year, that would allow them to be color-coded in a different manner. For example, 12 different colors could be used to indicate the portions of the curves corresponding to each month. This would allow the interannual variability in different months to be compared at a glance. In addition, different line styles (e.g., solid, dashed, dotted) could be used to differentiate the three regimes. Reformulating the plot along these lines would make this panel much more useful than it currently is.
  - Please add minor tick marks on the axes (x and y) of all of the panels in Figure 3.
- L221-222: The two parts of this sentence ("highlights the interannual variability in total HNO_3" and "shows a strong consistency in the onset of the depletion between each year") seem contradictory. If the behavior is consistent from year to year, then interannual variability is small. The sentence should be re-written using more careful language. The wording is also awkward: shows a strong consistency --> is very consistent; between each year --> every year.
- L223-224: Given the span of PSCs formation over a large range of altitudes --> Given that PSC formation spans a large range of altitudes
- L224: et al., 150 2006 --> et al., 2006
- L225: that of maximum IASI sensitivity to HNO_3 --> that IASI has maximum sensitivity to HNO_3
- L229: larger temperatures --> higher temperatures
- L230: The corresponding R2 temperature at 50 hPa to which the ~180 K value at 30 hPa and the ~185 K value at 70 hPa are being compared is not clear in this discussion.
- L234: enabling the PSCs formation --> enabling PSC formation
- L234-235: The "onset of the strong total HNO_3 depletion" in Figure 3c clearly occurs more than 5 K below 195 K (i.e., well to the left of the vertical red line). So I do not see how this statement about the consistency between 195 K and the onset of HNO_3 depletion is supportable. Perhaps the authors are accounting for the fact that HNO_3 starts to condense at temperatures 2–4 K below T_{NAT}, but (a) if so they need to state that explicitly, and (b) the difference in Figure 3c is larger than 4 K.

Section 4:

- L251: areas --> area; also, the fact that the PV value specified is for 530 K should be stated
- L252: regions --> region

- L253: total HNO_3 depletion occurs --> the largest depletion of total HNO_3 occurs
- L258: I have no idea what "as the daily second-difference HNO_3 total column" means. The rest of the sentence makes sense, so perhaps this part could simply be deleted. Otherwise, if it is supposed to convey important information, then it has to be rephrased for clarity.
- Figure 4 and its caption:
  - Why is the red line at 195 K dashed? Similar lines in Figure 3 were solid, which would be a better choice here too as the temperature time series is also shown as a red dashed curve.
  - L482: temperature --> 50 hPa temperature
- L262: around the 195 K threshold --> around the time that temperatures drop below the 195 K threshold
- L262: "some days" suggests to the reader "a few days", whereas 23 days is more than 3 weeks. Thus: within some days --> within a few days to a few weeks
- L265-266: The drop temperature in 2014 does stand out to some degree, but nevertheless I do not think that it can simply be excluded from the IASI-mission average just because it is a bit of an outlier. Strong justification is needed to exclude any individual year from the climatological mean, otherwise the authors risk being perceived as "cherry-picking" their results.
- L269-270: It is very good to remind readers of the meaning of the term "drop temperature", but this definition should come earlier in this section since the term has already been used above (L264, L266).
- L270: PSCs formation temperature --> PSC existence temperature
- L271: could reflect the preponderance by one --> could reflect variations in the preponderance of one
- L276-278: The average drop temperatures for 30 and 70 hPa appear to have been calculated over the full 10-year record, which supports my contention that 2014 should not be excluded from the 50-hPa average drop temperature calculation.
- L284: zonal distribution --> climatological zonal distribution
- L285: Does Figure 5 use geographic or equivalent latitude? If the former, the difference from other figures (which use EqL) needs to be explained, but in any case it should be made clear.
- L287: It is stated that one isocontour of 50 hPa temperature is overlaid, but actually two are shown in Figure 5.
- L289: lines indicates --> line indicates
- L290: The average drop temperature is given here as 194.2 ± 3.8 K, but above in L265 it was stated to be 194.1 ± 2.8 K. These values need to be reconciled.
- L290: rate in --> rate of
- L291: delete "of some days"
- Figure 5 and its caption:
  - Figure 5b is referred to in L284, but there is no further discussion of this panel in the text. The relationship between IASI total column HNO_3 and 50 hPa temperature has already been investigated in connection with Figures 3 and 4; moreover, Figure 5a includes two contours of 50 hPa temperature. Thus, in the absence of specific discussion about it in the text, Figure 5b seems superfluous and should be deleted.
  - L493: As noted above for the main text, whether this is geographic or equivalent latitude needs to be made clear.

- L496: The meaning of "The vertical grey dashed lines encompass the period of the second derivative minima" is unclear – the steepest rate of decrease occurs on a particular date each year, not over a prolonged period. What I believe the authors are trying to say is that the grey lines mark the earliest and latest dates for the drop temperature in the 10-year record, but this statement needs to be written more clearly.
- Panel 5b: the date labels (which are nearly illegible) for the overlaid lines indicate that the earliest drop temperature date is 12 May, whereas in L263 it is stated to be 11 May.

- L295: The PV contours are specified at 530 K, not 50 hPa.
- Figure 6 and its caption:
  - In the case of Figure 6, panel (b) is not even mentioned in the text. It should be deleted.
  - The x-axis date labels are illegible.
  - L501: Again, whether this is geographic or equivalent latitude needs to be made clear.
- L302-305: I'm a little confused about how the -8×10^{-5} $Km^2kg^{-1}s^{-1}$ contour of PV comes into the analysis in this section. It is a bit jarring to state in one sentence that the figure examines the region delimited by the −8 PV contour and then in the next sentence characterize the −10 PV contour as delimiting "the region of interest". I presume that the authors want to investigate a region larger than that of the strong depletion in total HNO_3 (encircled by the −10 PV contour) while excluding the collar region (−5 PV contour), but that rationale should be stated explicitly.
- Figure 7:
  - Similar maps of the corresponding dates are not shown, but they would be a useful addition and would help to clarify some points in the discussion, as noted below.
  - The dark green color for the -10×10^{-5} $Km^2kg^{-1}s^{-1}$ PV contour does not show up well (at least not on my monitor). A brighter green would work better.
  - Why are some of the temperature contours not closed?
  - The fonts for the years in the map titles in the top row look odd.
- L303: delete the comma after "s^{-1}"
- L303-307: Switching back and forth between temperature and PV makes these lines harder to follow. It would be better to discuss both the averaged and the minimum PV contours together and then move on to the two temperature contours.
- L306: The PV contour is specified at 530 K, not 50 hPa.
- L307: Add a comma after "period".
- L308-309: I have several comments on this part of the analysis:
  - The range of 50 hPa drop temperatures found here is considerably broader than that found in connection with Figure 4 in Section 4.1 (~180–210 K vs ~189–203 K), as is the range of corresponding dates (mid-May to mid-July vs mid-May to early June). These differences between the results based on vortex averages and those based on 1°×1° bins should be commented on in the text. Do the two approaches agree in terms of which years show generally lower/higher drop temperatures or earlier/later dates? The discrepancies are of concern because the only advantage that the IASI HNO_3 total column measurements bring over vertically resolved HNO_3 data sets (for which vortex averages can be calculated) is their dense horizontal sampling.
  - The high extremes in the drop temperature are attributed to issues with the retrievals over eastern Antarctica that are not fully screened out by quality control measures. But Figure 7 shows that unrealistically high drop temperatures are not confined to eastern Antarctica.

- As mentioned above, maps of the corresponding dates would be illuminating. What is the spatial distribution of late dates vs early dates? Do those patterns match the variations in drop temperatures, or do they bear no relation to the temperatures?
- The authors need to directly confront in the paper the fact that dates as late as mid-July for the onset of HNO_3 depletion in the Antarctic are even more implausible than drop temperatures as high as 210 K. These findings will likely cause many readers to dismiss their analysis methodology and/or the use of IASI data for studying PSCs.

- L311: surface --> surfaces
- L313: surface --> surfaces
- L318: Some years have sizeable regions with drop temperatures below 195 K that are well outside the averaged 195 K contour. Hence, encircles well --> encircles fairly well.
- L319: bins inside that area characterize air masses --> bins inside that area include air masses
- L322: show --> shows
- L325: show --> shows
- L327: is also enclosed --> is also typically enclosed
- L330-331: The weakest minima in the second derivative of total HNO_3 (not shown) observed in that area indicate a weak and slow --> The fact that the weakest minima in the second derivative of total HNO_3 (not shown) are observed in that area indicates a weak and slow
- L333-334: Although I think it is good to point out the possible impact of mixing on these results, it should also be acknowledged that previous studies have shown that in the Antarctic mixing between the edge region and the vortex core is generally weak (e.g., Roscoe et al., JGR 2012).
- L333: reflect a mixing with strong --> reflect mixing with strongly
- L334: The mixing --> Mixing; explained --> explain
- L335-336: I'm confused by this sentence. The drop temperature is defined by the onset of HNO_3 depletion, so how can it be that high drop temperatures are detected "after the HNO_3 depletion occurs"?
- L338-339: What is meant by "the range of maximum sensitivity of IASI to HNO_3"? Elsewhere in the manuscript, it is stated that IASI has the largest sensitivity to HNO_3 in the 30–70 hPa range, but how is that altitude information relevant to the spatial variations in the maps of Figure 7?
- L339-346: The second half of the sentence in L339 ("while biases …") starts a new discussion on reanalysis temperature and is followed by several related sentences, so it should be a separate sentence (not starting with "while"). In addition, it is not clear to the reader why all of the detail presented in the following sentences is really necessary. It would be better to either end the paragraph by stating explicitly that ERA-I temperature biases of the magnitude noted in these lines could not possibly account for the large range of calculated drop temperatures, or simply delete some of the details.
- L339: for explaining --> to explain
- L344: just to be clear, add "in modern reanalyses" after "reduced".
- L348: drop 50 hPa temperature --> 50 hPa drop temperature; delete the comma after "195 K"
- L349: PSCs nucleation --> PSC nucleation
- L351: on the type of formation mechanisms --> on the specific formation mechanism (i.e., the type of PSC developing)
- L353: coverage of IASI that allows capturing the rapid and critical depletion phase --> coverage of IASI, which allows the rapid and critical depletion phase to be captured in detail

Section 5:

- L357: columns dataset --> column dataset
- L358: since other IASI instruments are mentioned later: Metop --> Metop-A
- L363-364: I find this wording unclear. I suggest: level over a range where --> level, which lies in the range where; process occur --> processes occur
- L367: delete "various"
- L368: delete "and described along the cycle" (this wording is confusing and unnecessary)
- L369: delete "at play"
- L370: Only Antarctica is considered here, thus: in the poles --> over Antarctica
- L370: As mentioned earlier when this regime was defined, R2 starts in June but the uptake of HNO_3 into PSCs starts in mid-May, as shown in this paper and in previous studies.
- L371-372: PSCs nucleation --> PSC nucleation
- L372: between each year --> from year to year
- L372: R3 is actually defined (L198, Figure 3a) to begin in October, not November.
- L373: until March --> through March
- L374: PSCs sedimentation at --> PSC sedimentation to
- L376: found particularly --> found to be particularly
- L377: condensation to --> condensation into
- L379: 2.8 --> 2.8 K; also, as noted previously, the inconsistency in the average drop temperature values given in the text (L265 and L290) needs to be fixed and the correct one quoted here.
- L380: As noted above in Section 4.1, I do not think that the omission of 2014 from the climatological average is justified.
- L381: demonstrated --> demonstrate; PSCs formation --> PSC formation
- L384: PV at 50 hPa --> PV at 530 K
- L386: highest minima --> lowest minima
- L388: "from year to year" is not the right phrase; perhaps the authors mean "in some years"
- L388: As mentioned earlier, not all of the unrealistically high drop temperatures were calculated over eastern Antarctica
- L390: found in line --> found to be in line; PSCs nucleation --> PSC nucleation
- L395-396: It likely results --> These likely result; a mixing --> mixing
- L399: over the whole polar regions --> over the whole Antarctic region
- L401-403: I do not see how the authors could make the statement that the IASI dataset offers a new observational means to monitor the relation of HNO_3 to temperature and PSC formation because it can make measurements in darkness. It is certainly understandable that they want to tout IASI's excellent spatial coverage and its potential for a long record. Those are indeed very valuable contributions. But it is simply not acceptable to ignore decades of HNO_3 measurements made "throughout the year (including the polar night)" by numerous satellite instruments (e.g., LIMS, UARS MLS, Aura MLS, CLAES, MIPAS, SMR, ILAS, SMILES).
- L401-402: delete "capturing"; add "to be captured" at the end of the sentence
- L404: allow to investigate --> allow investigation of