acpd-2020-347

Referee comments on the revised version or on the author responses are in green type.

Referee comments on the original version are in black type.

Author responses are in blue type.

The revised manuscript includes a qualitative comparison with contemporaneous HNO3 measurements by MLS. This analysis should have been taken further to provide a more useful quantitative comparison with the IASI HNO3 column data. Additionally, incorporating CALIOP data would have enabled much better insight into the interannual variations of the drop temperatures and their relation to the distribution of PSC types. Overall, the revised manuscript does not put forward a compelling case for the scientific utility of the "drop temperatures". I do not find that the authors have addressed previously raised concerns in the on-line referee comments sufficently well to recommend publication. Detailed comments are given below.

Line numbers from here correspond to the revised version ...

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19 good vertical sensitivity in the mid-stratosphere (around 50 hPa),

The IASI HNO3 nadir measurements can not be considered as having "good vertical sensitivity" . Prospective data users need to know the vertical resolution of the measurements and that is conveyed by the standard practice of quoting the full width at half maximum (FWHM) of the averaging kernel. There is no reason not to do this for a nadir sounder e.g. Maddy and Barnett, Vertical Resolution Estimates in Version 5 of AIRS Operational Retrievals, IEEE Trans. Geosci. Remote Sens., 46, 8, 2375-2384, 2008.

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29 nitric acid trihydrate (NAT) formation temperature

For reasons explained elsewhere the "formation temperature" can be better expressed as an "existence thresold".

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26 Although the measured
27 HNO3 total column does not allow differentiating the uptake of HNO3 by different types of PSC particles
28 along the vertical profile, an average drop temperature of ˜194.2 +/- 3.8 K, consistent with the nitric acid
29 trihydrate (NAT) formation temperature (close to 195 K at 50 hPa)

The averaged "drop temperature" disregards the considerable interannual variability in the early stage formation of different types of Antarctic PSCs and the role played by the exposure of liquid PSCs to low temperatures in the formation of NAT i.e. many studies have shown that NAT is not uniquely constrained to nucleate at TNAT and some supersaturation is generally needed leading to a lower temperature for NAT formation (as in fact you discuss in the text L55-L75). Therefore, stating that the drop temperature is "consistent with TNAT", which implies that PSCs are mainly NAT forming at TNAT, is invalid. On line 28 the " " sign should be deleted since a specific value and its uncertainty is quoted.

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30 potential vorticity lower than -10x10-5 K.m2.kg-1.s-1

Some corresponding indication of the equivalent latitude range would be useful here.

==

30 vorticity lower than -10x10-5 K.m2.kg-1.s-1. The spatial distribution and inter-annual variability of the
31 drop temperature are investigated and discussed in the context of previous PSCs studies.

However, the study presented here does not include any observed data on PSCs and is therefore not a "PSC study".

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92 profile of HNO3 similar to the limb sounders, IASI provides reliable total column measurements of
93 HNO3 characterized by a maximum sensitivity in the low-middle stratosphere around 50 hPa (20 km)
94 during the dark Antarctic winter (Ronsmans et al., 2016, 2018) where the PSCs cloud form (Voigt et al.,

Please give the fullwidth half-max (FWHM) of the vertical response in km and not just the height of maximum sensitivity.

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126 In order to expand on the comparisons against FTIR measurements which
127 is impossible during the polar night, Figure 1 (top panel) presents the time series of daily IASI total
128 HNO3 columns co-located with MLS VMR measurements within 2.5x.5 grid boxes at three pressure
129 levels (at 30, 50 and 70 hPa), averaged in the 70 - 90 S equivalent latitude band. Similar variations in
130 HNO3 are captured by the two instruments with an excellent agreement for the timing of the strong
131 HNO3 depletion within the inner vortex core. IASI HNO3 variations generally match well those of MLS
132 HNO3 in each latitude band (see Figure 1 bottom panel for the 50 - 70 S equivalent latitude band; the
133 other bands are not shown here).

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251 The 50 hPa drop temperatures are detected between 189.2 K and 202.8 K, with an average of
252 194.2 +/- 3.8 K (1 standard deviation) over the ten years. Knowing that TNAT can be higher or lower
253 depending on the atmospheric conditions and that NAT starts to nucleate from 2–4 K below TNAT (Pitts
254 et al., 2011; Hoyle et al., 2013; Lambert et al., 2016), the results here demonstrate the consistency
255 between the 50 hPa drop temperature,

The software bug that was fixed in the revised version has changed the drop temperatures such that the year with 202.8K (previously 190.6K) is a significant outlier since it lies 8K higher than the 10-year mean drop temperature and is almost as much above the assumed 50hPa TNAT (195K). Therefore it does not support the statement on L365-366 that the 10-year range "demonstrated the good consistency between the 50 hPa drop temperature and the PSCs formation temperatures in that altitude region".

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295 Note that the high extremes in the drop temperature, which are found in some case above
295 eastern Antarctica, should be considered with caution:. they correspond to specific regions above ice
296 surface with emissivity features that are known to yield errors in the IASI retrievals (Hurtmans et al.,
297 2012; Ronsmans et al., 2016). Indeed, bright land surface such as ice might in some cases lead to poor
298 HNO3 retrievals. Although wavenumber-dependent surface emissivity atlases are used in FORLI
299 (Hurtmans et al., 2012), this parameter remains critical and causes poorer retrievals that, in some
300 instances, pass through the series of quality filters and could affect the drop temperature calculation.
...
371 Except for extreme drop
372 temperatures (˜210 K) that were found from year to year above eastern Antarctica and suspected to
373 result from unfiltered poor quality retrievals in case of emissivity issues above ice

It is not clear why this data quality problem has not been addressed in the revised submission. Measurements that are known to be bad must be screened out.

==
332 Except above some parts of Antarctica which are prone to larger retrieval errors, the overall range in the
333 drop 50 hPa temperature for total HNO3 inside the isocontour for the averaged temperature of 195 K,
334 typically extends from ˜187 K to ˜195 K, which falls within the range of PSCs nucleation temperature
335 at 50 hPa: from slightly below TNAT to around 3-4 K below the ice frost point - Tice - depending on
336 atmospheric conditions, on TTE and on the type of formation mechanisms (Pitts et al., 2011; Peter and
337 Grooß, 2012; Hoyle et al., 2013).
...
373 the range of drop
374 temperatures is interestingly found in line with the PSCs nucleation temperature

Why is the discussion in L302-338 and L367-376 limited to nucleation of NAT and ice PSCs with no mention of STS? There is no nucleation barrier to STS formation and it generally forms in advance of ice nucleation except possibly under very fast cooling e.g. in mountain waves. STS is not even mentioned in the paper after the introduction in L55-72.

Line numbers from here correspond to the original version ...

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429 Major comments
430
431 [The description of the polar HNO3 variation presented in the paper is already well known from
432 numerous other studies.]

433 The purpose of this paper is to demonstrate the interest of IASI for HNO3 stratospheric studies
434 (Ronsmans et al., 2018) after having undergone a rigorous validation exercise (Ronsmans et al., 2016).
435 If limb measurements allows resolving the HNO3 profile in the stratosphere, the potential of IASI lies in
436 its exceptional spatial and temporal sampling. We demonstrate here that despite its limited vertical
437 resolution forcing us to consider one total column, the information content that actually lies in the low
438 and middle stratosphere offers potential to expand on previous polar stratospheric denitrification studies,
439 usually performed using limb sounder measurements, and to continue the long-term records of HNO3
440 started with the latter. We have tried in this paper not to repeat too much of our earlier work but some
441 duplication was unavoidable; in particular, with respect to vertical sensitivity and errors (these are two
442 aspects that referee1 finds in fact insufficiently described here).

==
444 [The lack of vertical resolution in the IASI HNO3 measurements severely limits the interpretation of the
445 results and precludes differentiation between denitrification and renitrification e.g. consider the effect of
446 the vertical integration through depleted higher layers overlaying lower enhanced layers.]

447 We understand that the referee sees this as a limitation. However, despite the lack of vertical resolution,
448 which is recognized in the paper and which forces us to consider total HNO3 columns, IASI is
449 characterized by a good sensitivity to HNO3 at specific levels, in particular, in the range between ˜70
450 hPa to ˜30 hPa in the southernmost latitude in winter and as such it provides an adequate means to

investigate the stratospheric processes in the polar nights.

In order to justify this further, we would like to refer to the figure 3 (top and bottom panels) of Ronsmans et al. (2016) that presents global distributions of the degrees of freedom for signal (DOFS, top panels) and of the altitude of maximum sensitivity of IASI to the HNO3 profile (bottom panel), separately for January (left) and July (right) 2011, when the strong HNO3 depletion occurs within the cold Antarctic winter. It shows clearly that the altitude of maximum sensitivity of the total columns is invariant at equatorial and tropical latitudes, whereas it varies with seasons at middle and polar latitudes. Above the Antarctic, the altitude of maximum sensitivity varies between ˜9 km in summer and ˜22 km in winter. The variations of the altitude of maximum sensitivity follow the altitude variations of maximum HNO3 concentrations.

We agree that the IASI sensitivity was insufficiently put forward in the text. We made it more explicit at several places in the revised manuscript; e.g. in Section 1: “IASI provides reliable total column measurements of HNO3 characterized by a maximum sensitivity in the low-middle stratosphere around 50 hPa (20 km) during the dark Antarctic winter (Ronsmans et al., 2016; 2018) . . . ” and in Section 2: “. . . the largest sensitivity of IASI in the region of interest, i.e. in the low and mid-stratosphere (from 70 to 30 hPa), where the HNO3 abundance is the highest (Ronsmans et al., 2016).”

The response does not address the specific case example of where IASI views HNO3 depleted higher layers that overlay lower enhanced layers. How does the IASI column HNO3 measurement change if the HNO3 is redistributed in the vertical coordinate by denitrification and renitrification? A further question would be how does downwelling of higher values of HNO3 affect the HNO3 column?

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[Although the IASI HNO3 data has much better 2D horizontal resolution than any other measurement this has not been developed as a tool to provide information beyond that of satellite instruments that measure only along the orbit track.]

We do not fully agree. The determination of the drop temperature using the second derivative exploits the large dataset of daily IASI measurements. Furthermore, the spatial distributions of the drop temperature calculated at 50hPa, which are presented in the figure 5 of the manuscript, do actually take advantage of the excellent spatial/temporal resolution of IASI to provide information throughout the entire vortex and outside. This would probably not be feasible with other types of measurements.

As a further example of the 2D potential, could IASI be used to image the HNO3 field to show depletion in the cold phases of mountains waves e.g. near the Palmer peninsula (similar to the wave structures seen in AIRS brightness temperatures) or is that defeated by the vertical integration caused by the poor vertical resolution?

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[CALIOP PSC information is available for the same time frame, why was this not used? Certainly, PSC volumes vs time would be helpful in providing the underlying interannual variability of PSC types (NAT, STS, ice) to compare with the resulting drop temperatures derived from IASI. Similarly, at least some comparisons of the IASI HNO3 column with integrated column calculated from Aura MLS are necesssary to establish the validity of the measurements in the most severly depleted inner vortex core.]

Thank you for this comment. It is certainly a good idea to use the CALIOP measurements in support but this goes beyond the goal of this paper, which is to demonstrate the capability of IASI to measure HNO3 columns that are relevant for stratospheric studies. Using CALIOP PSC information and, in particular, comparing the spatial distributions of IASI derived drop temperatures (Figure 5 of the revised paper) with maps of CALIOP PSC would be very interesting in order to go a step further in the analyses of the underlying HNO3 condensation processes, but it will be challenging and add significant complexity given the high variability in the distribution of PSC types.

Regarding the second point on a comparison with MLS, we fully agree that this is highly relevant; it was also a request of referee“#1. We provide here below a comparison with observations by MLS in three equivalent latitude bands (see Figure 1). We would like to point out that we here compare total columns measured by IASI with VMR measured by MLS at several pressure levels that cover the highest sensitivity of IASI (at ˜50 hPa, ˜70 hPa and ˜30 hPa for the sake of the comparison). Hence, the comparison of IASI columns with MLS measurements is mostly qualitative at this stage and differences are expected for this reason. Note also that we have preferred comparing IASI HNO3 columns with VMR measured by MLS at specific levels instead of integrated columns calculated from MLS, given the difference in the sensitivity profile between IASI and MLS, the non-negligible IASI sensitivity to HNO3 in the troposphere where MLS does not measure HNO3 etc, which makes the integrated columns from IASI vs MLS not directly comparable. It should be pointed out finally that part of the differences between IASI and MLS are likely due to the different number of co-located data within the 2.5“deg x 2.5“deg grid cells considered here for the comparison, with a much larger number of observations for IASI (through the quality filtering) than for MLS.

Despite this, the comparison shows similar spatial and seasonal variations between IASI total HNO3 columns and MLS VMR between ˜70 and 30 hPa in the different latitude bands, in particular, in the southernmost equivalent latitudes (see top panel). The strong HNO3 depletion is well captured by both IASI and MLS measurements with a perfect match for the onset of the depletion. It further supports the good sensitivity of IASI to HNO3 in the range of these pressure levels, justifying the methodology used in this study.

The cross-comparison with MLS is indeed insightful and gives further credit on the IASI observations during the polar night. That comparison figure between IASI and MLS has therefore been included in Section 2 of the revised manuscript and the text was changed to:

”In order to expand on the comparisons against FTIR measurements which is impossible during the polar night, Figure 1 (top panel) presents the time series of daily IASI total HNO3 columns co-located with MLS VMR measurements within 2.5x2.5 grid boxes at three pressure levels (at 30, 50 and 70 hPa), averaged in the 70 “deg S–90 “deg S equivalent latitude band. Similar variations in HNO3 are captured by the instruments with an excellent agreement for the timing of the strong HNO3 depletion within the inner vortex core. IASI HNO3 variations generally match well those of MLS HNO3 in each latitude band (see Figure 1 bottom panel for the 50 “deg S–70 “deg S equivalent latitude band; the other bands are not shown h

“CALIOP measurements ... this goes beyond the goal of this paper, which is to demonstrate the capability of IASI to measure HNO3 columns that are relevant for stratospheric studies”. That goal was largely achieved already by Ronsmans et al (2016) and published in Atmos. Meas. Tech. This paper is under review for Atmos. Chem. Phys. and should relate more to a science investigation rather than a technical description. The comparisons with MLS are a welcome improvement, but unfortunately fall short of the analysis I was expecting. Surely the tropospheric contribution of HNO3 to the IASI column is not all that much (you could estimate the effect to confirm). I expected the MLS profile to be integrated with the IASI response function for a more direct comparison. That would facilitate a quantitative interpretation of the differences in the variation of the column data from the two instruments.

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[Regarding the sensitivity of the IASI column HNO3 measurements, I suggest presenting a few examples of vertical HNO3 profiles (from a model or data), ranging from non-depleted to extreme depletion with calculations of the corresponding calculated integrated IASI column. This would help to indicate the sensitivity of the column measurement to changes in the vertical distribution of HNO3 ... i.e. generate profiles of the change in the IASI column HNO3 wrt the actual change in HNO3 at a level, j, d(column)/d(HNO3)j.]

This is an example of information reported in earlier work and that we have tried not to repeat extensively here. To summarize, the validation study of Ronsmans et al. (2016) provides a complete characterization of the IASI HNO3 retrievals: it shows example of vertical HNO3 profiles along with the total retrieval error, the a apriori profiles and associated averaging kernels profiles (d(HNO3ret)i/d(HNO3true)j), along with the total column averaging kernel (d(columnret)/d(HNO3true)j) and the sensitivity profile (d(HNO3ret)i/d(columntrue)), were already given in Figures 1 and 2 of that study. Note that the averaging kernel profile describes how the true state changes the estimate at a specific altitude, i.e. how the retrieval smooths the true profile. The sum of the elements of an averaging kernel characterizing the retrieval at a specific altitude returns the sensitivity of the retrieval at that altitude, i.e. to which extent the retrieval at that specific altitude comes from the spectral measurement rather than the apriori, while the sum of the averaging kernels indicates how the true state at a specific altitude changes the retrieved total column, i.e. the altitude to which the retrieved total column is mainly sensitive/representative.

Figure 3 (top and bottom panels) of Ronsmans et al. (2016) further presents the global distributions of the degrees of freedom for signal (DOFS, top panels) and of the altitude of maximum sensitivity of the retrieval to the HNO3 profile (bottom panel), separately for January (left) and July (right) 2011, when the strong HNO3 depletion occurs within the cold Antarctic winter. It clearly shows that above the Antarctic, the altitude of maximum sensitivity varies between ˜9 km in summer and ˜22 km in winter (˜ 50 hPa) on average.

To address the comment of the referee without repeating too much of the earlier results, we have carefully verified the manuscript with regard to unclear or incomplete statements about vertical sensitivity. The following has been added in Section 1: “IASI provides reliable total column measurements of HNO3 with a maximum sensitivity in the low-middle stratosphere around 50 hPa (20 km) during the dark Antarctic winter (Ronsmans et al., 2016; 2018) . . . ” and in Section 2: “. . . the largest sensitivity of IASI in the region of interest, i.e. in the low and mid-stratosphere (from 70 to 30 hPa), where the HNO3 abundance is the highest (Ronsmans et al., 2016).

In order to convince the referee that IASI measurements capture the expected variations of HNO3 within the polar night, we provide in Figure 1 below examples of vertical HNO3 profiles retrieved within the dark Antarctic vortex (above Arrival Height) and outside the vortex (above Lauder). The retrieved profiles are shown along with their associated total retrieval error and averaging kernels (the total column AvK and the so-called “sensitivity profile” are also represented). Above Arrival Height during the dark Antarctic winter, we clearly see depleted HNO3 levels in the low and mid-stratosphere and the altitude of maximum sensitivity at around 30 hPa. At Lauder on the contrary, HNO3 levels larger than the a priori are observed in the stratosphere with a larger range of maximum sensitivity.

I also wanted to see specific depleted vs non-depleted cases (one with a re-nitrification layer would be good also) generated along with the simulated IASI columns and the calculated columns. I suggest that the figure provided on the averaging kernels etc could be added to a supplemental material section with a description tailored to the cases studied here in addition to just referring readers to a prior publication.

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[L10: 191K is also consistent with STS temperatures (192 K) and is actually closer than TNAT (195 K)]

Indeed but as stated in the manuscript: “. . . recent observational and modelling studies have shown that HNO3 starts to condense in early PSC season in liquid NAT mixtures well above Tice (˜4 K below TNAT, close to TSTS). . . ”. The NAT nucleation temperature at 50 hPa range from slightly below TNAT to around 3-4 K below Tice, depending on atmospheric conditions, on TTE and on the type of formation mechanisms (Pitts et al., 2011; Peter and GrooS, 2012; Hoyle et al., 2013).

Note that in replying to referee#1 we have identified a bug for the automatically detection of the drop temperature, as well as for the detection of the corresponding dates in the figure 2 of the manuscript. It has been corrected. The position of the drop temperatures does now perfectly match the yearly minima of the total HNO3 second derivative. An average drop temperature over the ten years of IASI of 194.2 +/- 3.8 K is now calculated, which is even closer to TNAT.

Finally, as requested by referee #1, we also now clearly mention in Section 4.1 of the manuscript the range of drop temperatures when calculated at two other pressure levels to better judge on the uncertainty of the drop temperature at 50 hPa (see Figure 3 here below):
. . . Nevertheless, given the range of maximum IASI sensitivity to HNO3 around 50 hPa, typically between 70 and 30 hPa (Ronsmans et al., 2016), the drop temperatures are also calculated at these two other pressure levels (not shown here) to estimate the uncertainty of the calculated drop temperature defined in this study at 50 hPa. The 30 hPa and 70 hPa drop temperatures range respectively over 185.7 K – 194.9 K and over 194.8 K – 203.7 K, with an average of 192.0 +/- 2.9 K and 198.0 +/- 3.2 K (1 standard deviation) over the ten years of IASI. The average values at 30 hPa and 70 hPa fall within the 1 standard deviation associated with the average drop temperature at 50 hPa. It is also worth noting the agreement between the drop temperatures and the NAT formation threshold (TNAT ˜193 K at 30 hPa and ˜197 K at 70 hPa) (Lambert et al., 2016).”

CALIOP PSC data (Pitts et al 2013, doi:10.5194/acp-13-2975-2013) have been used to show that different PSC types exist in different temperature regimes, with ice PSCs detected close to the frost point, STS follows the expected equilibrium curve and NAT exhibits two preferred mode below the NAT existence temperature. The analysis presented here is not constrained by the simultaneous presence of known PSC types and in fact there may not even be any PSCs in the atmospheric path sampled. Therefore, it is too simplistic to compare the drop temperatures to TNAT. The proximity of the 10-year mean drop temperatures to TNAT does not constitute a validation as is claimed here. Individual years could be expected to show a variation in drop temperature because of interannual atmospheric differences. For instance, the years domimated by STS should necessarily show lower drop temperature than years dominated by NAT. The highest drop temperatures are far above PSC temperatures (e.g. 202.8K at 50hPa in one particular year) and deserve more scrutiny and should be investigated thoroughly. Interannual comparisons of the drop temperature may benefit from using (T-Tice) as the temperature coordinate (rather than absolute temperature) as this removes variations due to changes in H2O partial pressure (see Fig 2 of Pitts et (2013)). There is a fundamental problem with making an assessment of the potential future scientific utility of the drop temperatures when they have only been evaluated in the absence of knowledge of the different types of PSCs present.

[L18: add more recent references e.g. Peter and Gross (2012). L28: Much more has been done in the past decade with MIPAS and CALIOP that should be referenced]

Thank you for this suggestion. Peter and GrooS (2012) was cited elsewhere in the manuscript but has been added here as well. Note that the goal of the introduction is not to provide an exhaustive list of all studies related to the PSC thermodynamics. Several general reference papers are cited and we have decided to put more focus here on HNO3.

[L59: This section should explain what is meant by ”maximum sensitivity” etc.]

See our responses to the second major comment and specific comments above.

[L79: Information on the data quality for IASI HNO3 is poor. Is the value of bias and uncertainty the same for depleted and non-depleted conditions?]

The reader is here invited to refer to the figure 4 of Ronsmans et al. (2016) which illustrates the global distribution of the total retrieval error for HNO3 (integrated over 5 to 35 km) separately for January (left) and July (right) over the period of the IASI measurements. The mid- and polar latitudes are characterized by low total retrieval errors of around ˜3-5% - which corresponds to a reduction by a factor of 18-30 compared to the prior uncertainty (90%) and indicates a real gain of information – except above Antarctica during wintertime where the errors reach 25%. They are explained by (1) a weaker sensitivity (i.e. a larger smoothing error which represents in all cases the largest source of the retrieval error) above such cold surface (DOFS of ˜0.95 within the dark Antarctic vortex – see figure 3 of Ronsmans et al., 2016) and by (2) a poor knowledge of the wavenumber-dependent surface emissivity above ice surface, which also varies in time (Hurtmans et al., 2012).). This is made more explicit in Section 2 of the revised manuscript:

“The total columns are associated with a total retrieval error ranging from around 3% at mid- and polar latitudes to 25% above cold Antarctic surface during winter (due to a weaker sensitivity above very cold surface with a DOFS of ˜0.95 and to an poor knowledge of the seasonally and wavenumber-dependent emissivity above ice surfaces which induces larger forward model errors), and a low bias (lower than 12%) in polar regions over the altitude range where the IASI sensitivity is the largest, when compared to ground-based FTIR measurements (see Hurtmans et al., 2012; Ronsmans et al., 2016).”

The response does not address the specific case of whether there are differences in bias and uncertainty for depleted and non-depleted conditions.

[L82: Yet, problems with the retrievals because of cloud contamination seem to remain even after the 25% cloud fraction filter is applied.]

We do not understand the referee comment here. In this section of the manuscript, we only describe the quality flags used in our analysis.

Even after all the quality controls are applied there are apparently still cases with poor retrievals that could be removed.

==

[L83: Cloud contamination? Tropospheric cloud only or also thick ice PSCs?]

The clouds that have most impact are clearly tropospheric water clouds. Cirrus clouds or PSCs are mostly transparent in the IR; thick cirrus however show up in the longwave part of the IASI spectrum, below 900 cm-1. We have added “tropospheric cloud contamination” in the text.

Note that the threshold of 25 % cloud cover was carefully chosen after a series of tests, which have shown that these scenes could be treated as cloud-free without significant impact on the retrievals (Hurtmans et al., 2012).

Thick ice PSCs have been detected by AIRS, TOVS HIRS2 and AVHRR (see Stajner et al. and refs therein, https://doi.org/10.1029/2007GL029415). Do these have an effect on the HNO3 retrieved by IASI?

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[L102: Why was 2011 chosen?]

As expected from figure 1c, any other year could have been chosen instead of the year 2011 to illustrate the HNO3 total columns versus temperatures (at 50 hPa) histogram in figure 1b. It is now clearly mentioned in the revised manuscript:

“Similar histograms are observed for the ten years of IASI measurements (not shown).”

==

[L106: Heterogeneous hydrolysis of N2O5 requires aerosol particles. So this process starts with cold binary aerosols (i.e. sulfates) before the formation of STS?]

Indeed, previous studies have shown enhanced HNO3 columns during autumn in Antarctica and have attributed them to decreasing sunlight and conversion of N205 to HNO3 by the reaction of N205 with background aerosols, before the formation of polar stratospheric clouds (e.g. Keys et al., Nature, 1993). At these temperatures, the conversion may occur on binary sulfuric aerosols.

The sentence has been rewritten as follows:

“These high HNO3 levels result from low sunlight, preventing photodissociation, along with the heterogeneous hydrolysis of N2O5 to HNO3 during autumn before the formation of polar stratospheric clouds (Keys et al., 1993; Santee et al., 1999; Urban et al., 2009; DeZafra et al., 2001). This period also corresponds to the onset of the deployment of the southern polar vortex which is characterized by strong diabatic descent with weak latitudinal mixing across its boundary, isolating polar HNO3-rich air from lower latitudinal airmasses."

[L129: The onset of depletion seems to start when the temperatures fall substantially below 190K from inspection of Fig 1(c) and quite far below the red line marked at 195K.]
The onset of HNO3 depletion starts in June at around 190K, which is in agreement with figure 1a.

[L136-137: Why are two temperatures (180 and 185 K) quoted for 30hPa? Why is the actual value from Fig1(c) (I estimate this as about 188K) for the 50hPa temperature not given in L129?]

The sentence has been rewritten for clarity:
"The results (not shown here) exhibit a similar HNO3-temperature behaviour at the different levels with, as expected, lower and larger temperatures in R2, respectively, at 30 hPa (down to 180 K) and at 70 hPa 145 (down to 185K), but still below the NAT formation threshold at these pressure levels (TNAT =193 K at 30 hPa and 197 K at 70 hPa) (Lambert et al., 2016)."

[L138: "characterized by" seems the wrong description for the chance occurrence that the maximum sensitivity of IASI HNO3 falls in the same altitude range as the PSCs.]

Changed to: ". . . the altitude range of maximum IASI sensitivity to HNO3 (see Section 2) is characterized by temperatures that are below the NAT formation threshold at these pressure levels, enabling the PSCs formation and the denitrification process."

[L139-146: This section rather seems to belong in the conclusions.]

L150-154 of the revised manuscript has been moved to the conclusions.

[L148: Clearly this does not "go beyond the vertically integrated view" since the column HNO3 is all that is available. It could be reworded as "To identify the spatial and temporal variability of the column HNO3 ..."]

Corrected as suggested.

[L165-169: Denitrification is the term used to describe the permanent removal of some HNO3 from the gas phase by sedimentation of PSCs. Sequestration is the term used to describe the uptake of HNO3 from the gas phase into PSCs. Denitrification by STS is a lengthy process compared to NAT since the smaller STS particles sediment slowly. STS can (and frequently does) form without the prior nucleation of NAT. IASI alone cannot discriminate between these processes and it should not be assumed that what is observed is the "onset of HNO3 denitrification".]
We thank the referee for this remark. We are of course aware of the definition of the so-called "denitrification". We agree that, from IASI, we can only measure a "removal from the gas phase", caused

by sequestration into particles with or without sedimentation. Careful attention has now been made in the manuscript to avoid abusive use of the term “denitrification”. Hence, “onset of HNO3 denitrification” has been changed to “the onset of HNO3 depletion” in L.169 and where appropriated in the revised manuscript and he title has also been changed accordingly to:
“Polar stratospheric HNO3 depletion surveyed from a decadal dataset of IASI total columns”.

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[L185-187: 210K is much too high for PSC formation, but could possibly be NAT that is in process of melting? If these are observed over ocean then they warrant further investigation. However, why are specific regions with emissivity features not flagged as such? They should be discarded rather than ”used with caution”.]

Bright land surface such as desert or ice might in some cases lead to poor HNO3 retrievals due to a poor knowledge of the wavenumber-dependent emissivity above such surfaces, which can alter the retrieval by compensation effects (Wespes et al., 2009). FORLI relies on the monthly climatology of surface emissivity built by Zhou et al. (2011) from several years of IASI measurements on a 0.5x0.5 grid and for each 8461 IASI spectral channels when available, or on the MODIS climatology that is unfortunately restricted to only 12 channels in the IASI spectral range; see Hurtmans et al. (2012) for more details. Although wavenumber-dependent surface emissivity atlases are used in FORLI, it is clear that this parameter remains critical and causes poorer retrievals that, in some instances, pass the posterior filtering. The total HNO3 columns over eastern Antarctica which show drop temperatures much above 195K might precisely be related to this. We have made this clear in Section 4.2 of the revised version:

“. . . emissivity features that are known to yield errors in the IASI retrievals. Indeed, bright land surface such as ice might in some cases lead to poor HNO3 retrievals. Although wavenumber-dependent surface emissivity atlases are used in FORLI (Hurtmans et al., 2012), this parameter remains critical and causes poorer retrievals that, in some instances, pass through the series of quality filters and could affect the drop temperature calculation.”

We refer on the good agreement with MLS (suggested by the referee) to underline the potentiality of IASI to detect the HNO3 variations as well within the Antarctic winter (see general comment and Figure 1 here below).

What is the fraction of data that is affected by surface emissivity?

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[L189: Modern reanalysis temperatures (e.g. ERA-I) do not ”feature large uncertainties” large enough to account for a 195K to 210K shift. L195-L201: The limitations of the reanalysis temperatures seems to be an accuracy of better than 1K and clearly this in no way limits the derivation 760 of the ”50hPa drop temperature” which simply necessitates finding the 50hPa reanalysis temperature that corresponds to the second derivative wrt time minimum in column HNO3.]

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We agree with the referee’s comment; the discussion about the potential role of the uncertainty of the ECMWF reanalysis temperature on the drop temperature has been removed from the section, hence, this paragraph has been strongly revised accordingly:

“. . . while biases in ECMWF reanalysis are too small for explaining the spatial variation in drop temperatures. Thanks to the assimilation of an advanced Tiros Operational Vertical Sounder (ATOVS) around 1998–2000 in reanalyses, to the better coverage of satellite instruments and to the use of global navigation satellite system (GNSS) radio occultation (RO) (Schreiner et al., 2007; Wang et al., 2007; Lambert and Santee, 2018; Lawrence et al., 2018), the uncertainties have been vastly reduced. Comparisons of the ECMWF ERA Interim dataset used in this work with the COSMIC data (Lambert and Santee, 2018) found a small warm bias, with median differences around 0.5 K, reaching 0–0.25 K in the southernmost regions of the globe at ˜68–21 hPa where PSCs form.”

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[What is meant by ”spatial variability”? The plots in Fig 5 show the spatial distribution of the drop temperature over a number of years but what variability is being considered? Interannual? Why have these spatial maps of drop temperatures not been compared with published maps of PSC types made by CALIOP or MIPAS. Wouldn’t some correlation be expected according to the arguments made here? i.e. NAT PSCs at the higher temperature e.g. the highest temperatures (orange) appear downstream of the Palmar Peninsula in the ”NAT ring” structure described by Hopfner et al (2006).]

Corrected: “Figure 5 shows the spatial variability” .. “Figure 5 shows the spatial distribution”.

We do not understand the referee’s comment here. Figure 5 of the manuscript shows the spatial distribution of the drop temperature calculated inside a region enclosed by an isocontour PV of -8x10-5 K.m2.kg-1.s-1, which, hence, encircles a region larger than the inner vortex core (see Figures 3 and 4 of the manuscript). The drop temperatures much above the NAT formation temperature, which are mostly found outside the averaged isocontour PV of -10x10-5 K.m2.kg-1.s-1, do not correspond to high minima (¿-0.5 x1014 molec.cm-2.d-2) in the second derivative of HNO3 total column with respect to time. We cannot argue that it corresponds to the NAT belt of Höpfner et al. (2006) downstream of the Antarctic Peninsula, which was enclosed inside the region of the NAT threshold temperature; the highest drop temperatures from IASI are found on the contrary outside the isocontour of the NAT threshold temperature (see figure 5 of the revised manuscript). In addition, comparing the distributions of drop temperatures from IASI with PSC information from CALIPSO/MIPAS remain difficult given the difference in spatial coverage and, most importantly, the highly variable distribution of PSC types and of the NAT belt, temporally (daily) and spatially (Höpfner et al., 2006; Lambert et al., 2012).

Finally, in response to G. Manney and M. Santee, the contour of -10 x 10-5K.m2.kg-1.s-1 based on the minimum PV encountered at 50 hPa over the 10 May to 15 July period as well as the isocontours of 195 K at 50 hPa for the averaged temperatures and the minima over the same period are also now represented in the revised Fig.5 and the distribution of the drop temperatures is much better described and explained in the revised version:

“The averaged isocontour of 195 K encircles well the area of HNO3 drop temperatures lower than 195 K, which means that the bins inside that area characterize airmasses that experience the NAT threshold temperature during a long time over the 10 May – 15 July period. That area encompasses the inner vortex core (delimited by the isocontour of -10 x 10-5K.m2.kg-1.s-1 for the averaged PV) and show pronounced minima (lower than -0.5 x1014 molec.cm-2.d-2) in the second derivative of the HNO3 total column with respect to time (not shown here), which indicate a strong and rapid HNO3 depletion.

The area enclosed between the two isocontours of 195 K for the temperatures, the averaged one and the one for the minimum temperatures, show higher drop temperatures and weakest minima (larger than -0.5 x1014 molec.cm-2.d-2) in the second derivative of the HNO3 total column (not shown). That area is also enclosed by the isocontour of -10 x 10-5K.m2.kg-1.s-1 for the minimum PV, meaning that the bins inside correspond, at least for one day over the 10 May - 15 July period, to airmasses located at the inner edge of the vortex and characterized by temperature lower than the NAT threshold temperature. The weakest minima in the second derivative of total HNO3 (not shown) observed in that area indicate a weak and slow HNO3 depletion and might be explained by a short period of the NAT threshold temperature experienced at the inner edge of the vortex. It could also reflect a mixing with strong HNO3-depleted and colder airmasses from the inner vortex core. The mixing with these “already” depleted airmasses could also explained the higher drop temperatures detected in those bins. Finally, note also that these high drop temperatures are generally detected later (after the HNO3 depletion occurs, i.e. after the 10 May – 15 July period considered here – not shown), which supports the transport, in those bins, of earlier HNO3-depleted airmasses and the likely mixing at the edge of the vortex.”

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[L205: Nothing has been presented that demonstrates PSC occurrence. For that you would need to compare to actual data on PSCs from CALIOP and/or MIPAS.]

Corrected: “PSCs occurrence” .. “NAT formation temperature”

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[L224: Again, the suspect data should be discarded because of the detrimental impact on the scientific analysis. Also, if you cannot manage to work out and apply adequate quality control to your own data then you have no reason to expect anyone else to do so.]

See our response to comment [L185-187] above.

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[L230: ”To the best of our knowledge, it is the first time that such a large satellite observational data set of stratospheric HNO3 concentrations is exploited to monitor the evolution HNO3 versus temperatures” In fact you cite several papers that have done exactly this, but let’s take the one published over two decades ago by Santee et al (1999) titled ”Six years of UARS Microwave Limb Sounder HNO3 observations : Seasonal, interhemispheric, and interannual variations in the lower stratosphere”. https://doi.org/10.1029/1998JD100089. Not only does this paper compare HNO3 with UKMO temperatures we are referred to a more complete paper on this topic on p8241 ... ”The correlation of the HNO3 behavior with temperature during this time period, and its implications for PSC phase and composition, is explored in detail by Santee et al (1998). I noticed that the outside edge of the ”HNO3 collar region” at 465K was defined by these authors as inside the 0.25 x E-4 K m2 kg-1 s-1 PV contour. This seems at odds with the 1E-4 value that is used for the second derivative minimum calculation in this paper and seemingly places the boundary quite far equatorward. Santee et al (1998) also includes a description of the heterogeneous hydration of N2O5 that would be helpful in response to the question above on L106.]

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We here simply refer to the unprecedented potential of IASI in terms of its exceptional spatial and temporal sampling. Ronsmans et al. (2018) also referred to the IASI dataset and correlations with temperature were done but in a lesser extent. In order to avoid overselling, the sentence has been rewritten: “We show in this study that the IASI dataset allows capturing the variability of stratospheric HNO3 throughout the year (including the polar night) in the Antarctic. In that respect, it offers a new observational means to monitor the relation of HNO3 to temperature and the related formation of PSCs.”

In this study, we use the PV fields taken from the ECMWF ERA Interim Reanalysis dataset at the potential temperature of 530 K (corresponding to ˜20 km where the IASI sensitivity to HNO3 is the highest), while Santee et al. (1998) considered 465K. We clearly see from Figures 3a and 4 of the manuscript that PV contours at -0.5e-4 K m2 kg-1 s-1 and at -0.8e-4 K m2 kg-1 s-858 1 encompass the so called HNO3 collar region. The PV value of -1e-4 K m2 kg-1 s-1 is used in this study to calculate the drop temperature based on the second derivative minimum as it clearly encompass the regions inside the inner polar vortex (see Figure 3a and 4 of the manuscript).

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[L231: ”It could constitute a new accurate climatological parameter that could be inserted in the PSCs classification schemes.” The analysis presented does not support this statement. Specifically, how could the HNO3 column amount be used in a classification scheme?]

This sentence has been removed.

The conclusion of the paper is that ability to monitor the polar atmosphere over several decades with current and planed IASI instruments “will provide an unprecedented long-term dataset of HNO3 total columns”. The drop temperature is defined as the 50hPa temperature corresponding to the greatest rate of decline of the column HNO3 with respect to time. However, even with a record now extending over a decade the scientific utility of this dataset has not been demonstrated.