

Response to S. Solomon

First, we would like to thank Prof. Solomon for her insightful comments and suggestions. They have all been taken into account and they have allowed improving the paper. Each of the comments is addressed on a point-by-point basis hereafter, with the original comment reproduced in blue first.

- 1) There is a lot of discussion of the apparent onset of denitrification and its association with a 'threshold' $T < 195\text{K}$. I think the paper would be stronger if this were accompanied by a discussion of the uncertainties in the temperatures used to identify this relationship (i.e., ERA-interim). Three questions arise a) Could it really be 194K within uncertainties? Why or why not? How sharp is this threshold within uncertainties? b) what about the influence of small scale waves that may not be resolved by the reanalysis? c) would there be value in simply doing a scatter plot of local HNO_3 versus local temperature in winter, as opposed to the current approach of binning by eqlat?

We agree with the referee that the temperature of 195 K for the formation of PSCs can vary, namely depending on the conditions of the atmosphere, and that small scale waves would likely influence the local formation of PSCs. We plotted, as suggested by the referee, the HNO_3 values versus the temperature for each grid cell between 70 and 90°S of equivalent latitude for the winter months (May to October, see Figure here below). The appearance of low HNO_3 values occurs as expected around the 195 K threshold (red line), albeit with some variability, namely between 190 and 200 K. These variations are of interest when studying the precise dynamics of PSC formation and the results below suggest that IASI could make a strong contribution in monitoring the processes at play. We thank the referee for having triggered us to look into this in more details; we believe a forthcoming work could be undertaken regarding this question. However, for the present study, we believe that an average temperature of 195 K is a reasonable value to work with. We have mentioned these potential temperature variations in the text: *"It should be noted that while this temperature is a widely accepted approximation for the formation threshold for NAT (type I), its actual value can be different depending on the local conditions (Lowe and MacKenzie, 2008; Drdla and Müller, 2010; Hoyle et al., 2013)."* (p. 4, line 16-18)

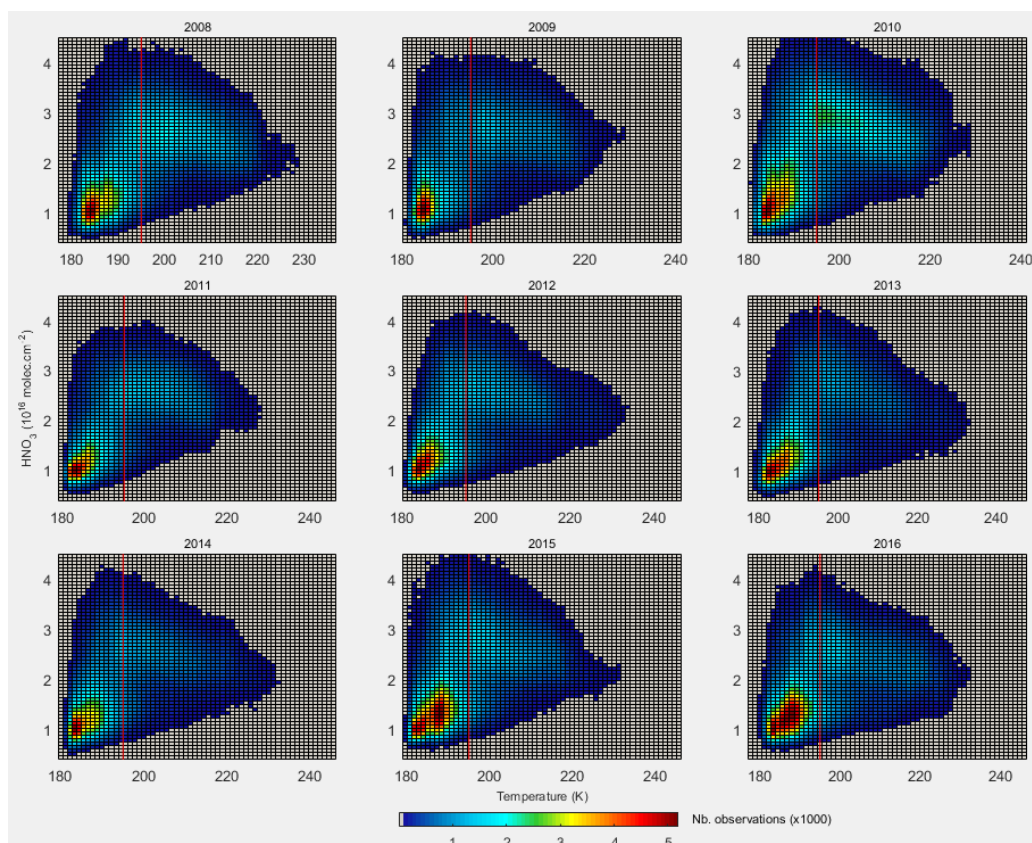


Figure 1. Total columns of HNO_3 ($10^{16} \text{ molec.cm}^{-2}$) versus temperatures (K) for the 70-90 S eqlat band, and between May and October. The color scale indicates the number of measurements per bin (with grey areas for bins with less than 80 measurements), and the red line is the 195 K threshold.

- 2) Figures, 2, 3, and 4 are key results of this study, showing very well the seasonal cycles in the two hemispheres. A minor comment on Fig 2 is that heavy ticks are needed for January, so one can see the exact mapping in time more readily.

We thank the reviewer for the positive feedback on the Figures. The ticks have been changed and they are now heavier for the months of January. This indeed makes the reading of the figure easier.

A substantive comment is that the time lag for recovery of the HNO₃ column in the southern hemisphere is striking. The paper has some good discussion on this but I wonder if more could be done. In particular, if the mechanism replacing the HNO₃ is mainly via transport, then HNO₃ and O₃ would show very similar post-final warming increases – do they? This could be shown, and would make the paper more useful.

We would like to thank Prof. Solomon for this useful comment. The behaviour of O₃ (retrieved from the same IASI observation) was compared against that of HNO₃, as shown with the Figure below. It appears that the recovery of O₃ at the end of the winter happens much sooner than that of HNO₃ (see Figure below). Note that the O₃ columns are stratospheric columns. We agree with the referee that this is a good indication that the mechanism for HNO₃ recovery is not due to transport and mixing but more with chemistry, as addressed in the next point.

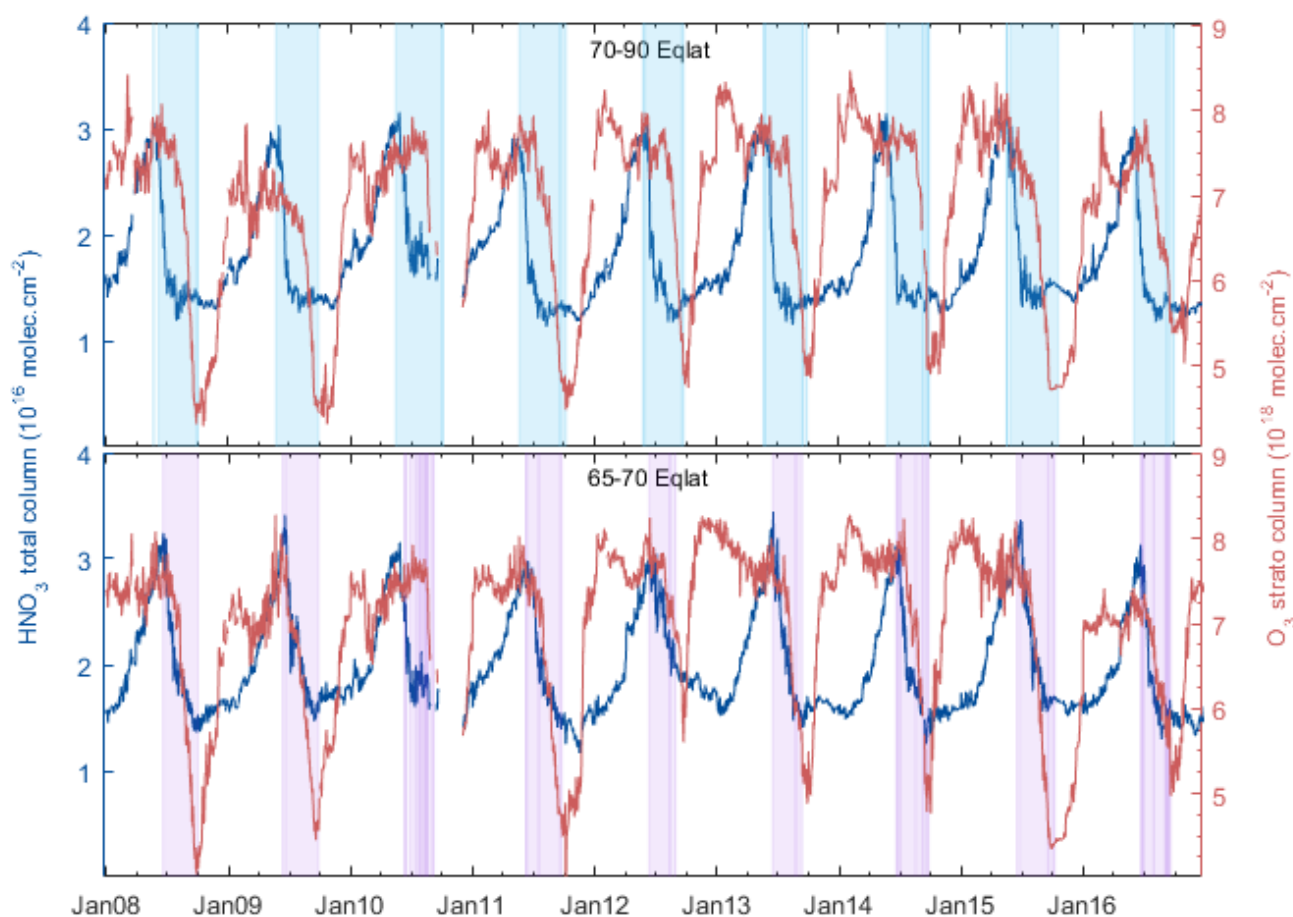


Figure 2. Total columns of HNO₃ (blue) and stratospheric columns of O₃ (red, both expressed in molec.cm⁻²) for eqlat bands 70-90 S and 65-70 S. The vertical shaded areas in each panel are the periods during which the temperature was equal to or below 195 K in that band.

Further, I suspect that the very late recovery of HNO_3 may have more to do with chemistry, in particular the fact that perpetual sunlight means NO_3 is photolyzed effectively throughout the polar summer so that little of it can end up in N_2O_5 and hence to HNO_3 . This may do a better job explaining the ramp from Mar-May (e.g., in Fig 4) than other explanations, and could be probed fairly simply using chemical kinetic equations considering length of night, temperature, and ozone near 20-25 km. Zonal means should suffice, despite the potential for vortex meandering, which is likely to be small in summer.

Further discussion was added in the text in view of the Figure above and the very useful explanation of the referee in order to clarify the late recovery of HNO_3 columns after the winter. We now explain, also relying on literature, that up until the end of September (end of the winter), the low columns are attributable to the formation of PSCs with the low temperatures and further sedimentation of PSCs, which heavily denitrifies the stratosphere. The persistence of low columns in spring and summer is then rationalized by very effective photodissociation of HNO_3 and NO_3 during the periods of prolonged sunlight, i.e. late spring and summer. Also, local minima of temperatures that stay below 195 K could cause a slower recovery too. This section in the text now reads: “... with previous studies by McDonald et al. (2000) and Santee et al. (2004) for earlier years. However, the recovery of the HNO_3 total columns is very slow compared to other species, namely O_3 , for which concentrations return rapidly to usual values almost as soon as PSCs disappear. In fact, the HNO_3 columns stay low well after the September equinox and are only subject to a slow increase 2 months later (in early December). While more persistent local temperature minima staying below 195 K could explain part of this late recovery, we make the hypothesis that it is due mainly to a combination of two factors: (1) a significant sedimentation of PSCs towards the lower atmosphere during the winter, yielding only small amounts of it to release HNO_3 under warmer temperatures (Lowe and MacKenzie, 2008; Kirner et al., 2011; Khosrawi et al., 2016), and (2) the effective photolysis of HNO_3 and NO_3 in spring and summer under prolonged sunlight conditions, i.e. mainly at the highest latitudes, which respectively increase the HNO_3 sink and reduce the chemical source (because NO_3 cannot react with NO_2 to produce N_2O_5 , Solomon, 1999; Jacob, 2000; McDonald et al., 2000). The increase observed in March, at the start of the winter, is in turn explainable by a much reduced number of hours of sunlight, implying less photodissociation. It is worth noting that the two regions previously mentioned (inner and outer vortex)...” (p.5 line 23-35)

- 3) The authors often (not always, but often) refer to low HNO_3 columns as synonymous with denitrification, but what about, for example, high ClONO_2 columns associated with the collar implying that NO_y may be in that form at times? Similarly, the low HNO_3 during mid-summer seen in Figure 4 and discussed above may have to do in part with perpetual sunlight meaning high NO and NO_2 amounts, with less in HNO_3 but not necessarily less total NO_y . Some further discussion is merited.

We thank the referee for pointing this out. We have taken care in the revised manuscript to not mix denitrification with loss of HNO_3 , and replaced the abusive “denitrification” terms with “loss of HNO_3 ”, “decreasing HNO_3 columns”, “decrease in HNO_3 ”, ...