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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A nitric acid dataset from IASI for polar stratospheric denitrification studies" *by* Gaetane Ronsmans et al.

Anonymous Referee #3

Received and published: 31 August 2020

The paper presents an analysis of a 10-year IASI column HNO3 record (2008-2017) for the Southern Hemisphere in conjunction with ERA-I reanalysis temperatures. The sequestration of HNO3 into PSCs is extremely temperature sensitive and it's unlikely that a single temperature at 50hPa is sufficent to capture anything but the most basic features. The description of the polar HNO3 variation presented in the paper is already well known from numerous other studies. The lack of vertical resolution in the IASI HNO3 measurements severely limits the interpretation of the results and precludes differentiation between denitrification and renitrification e.g. consider the effect of the vertical integration through depleted higher layers overlaying lower enhanced layers. 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

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of satellite instruments that measure only along the orbit track. 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 comparisions 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.

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.

I do not recommend publishing this paper in ACP without attention to the points raised in this review.

Specific comments:

L2: "good vertical sensitivity" ... only column HNO3 measurements are discussed here - there is no vertical resolution in the measurements.

L8: in [the] Antarctic

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

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

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L53: Studies of HNO3 depletion and PSC formation predate the sensors named in the paragraph e.g. the Santee et al (1999) reference used UARS/MLS launched in 1991, measurement using balloons should have been be referenced here.

L59: This section should explain what is meant by "maximum sensitivity" etc.

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?

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

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

L102: Why was 2011 chosen?

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

L108: extends

Figure 1 caption: Each figure title in 1(b) needs to state the year e.g. "January - December 2011 or put a label "2011" above the whole figure.

Figure 1 caption: 50 hpa => 50 hPa

Figure 1 caption: it is not clear to what 0.1E16 molec. cm-2. This low value is not even on the y-axis of the figures.

Figures 1(a) and 1(c): Are the HNO3 and temperature structures (localized peaks and valleys) visible in the time series in 1(a) quite well correlated when plotted as a scatter diagram as in 1(c), but without the 7-day averaging?

L123: 7-day

L124 and Figure 1 caption: "in the range of" : only one value is given and not a range of values

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.

L130: Supplementary material - this does not appear to be available from the ACP website.

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?

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.

L139-146: This section rather seems to belong in 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 ..."

L164: drop temperatures

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".

Figure 3 caption: sumperimposed => superimposed

L170: Figures 3a and b

L171: three isocontour levels

L174: lines indicate

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".

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 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.

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).

L200: It underlines ... What does "it" refer to? The subject of the previous sentence is "the spatial variability" but that has not been defined.

L201:critical denitrification phase

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.

L205: to PSCs occurrence to PSCs ??

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.

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.

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?

L240: "All authors contributed to the writting of the text and reviewed the manuscript."

writting => writing

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