

## ***Interactive comment on “Nitrification of the lowermost stratosphere during the exceptionally cold Arctic winter 2015/16” by Marleen Braun et al.***

### **Anonymous Referee #2**

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This paper presents interesting observations of hno<sub>3</sub> from aircraft observations in the Arctic winter of 2016 indicating nitrification of the Arctic polar vortex in the 10-014 km range, the maximum altitude of the observations. While the observations are of interest and the overall analysis convincing, that nitrification of the lower most stratosphere occurred, the paper is poorly written, much too long, and should not be accepted in this form.

Throughout the descriptions of the individual flights, which are used to introduce the observations, the authors claim that the observations show redistribution, enhanced hno<sub>3</sub> layers and nitrification. This is before the methods are explained, or the reference profiles discussed. The reader has to guess how these conclusions are made. The description of the figures often resorts to a recitation of numbers in the figures. The

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authors specify vortex air in the figures by stating what is not vortex air. Figure caption 2 confuses by not describing the panels in order. Waypoints marked in figures are not used. Reference is made to  $\text{NO}_y^*$ , but it is not used further, or defined. The claims of “distinct differences,” with some of the CLAMS sensitivity tests, are not well supported by the figures.

The paper could be published, but only after major revision. The analysis should begin by describing that ozone will be used as a tracer for the air sampled and to describe why that works for 2016. Thus the majority of section 5 should appear before any aircraft data are shown. Only two detailed aircraft profiles need to be shown, first the reference profile in December which currently is not shown, and an example of the measurements in January. These two examples are enough to set the stage for the relative normalized frequency distribution (RNFD) discussion, and Figure 6, which is the key figure of the paper. A figure showing an example of RNFDs would also be of interest.

While the authors seem to be keen on showing all of the aircraft data in detail, this really distracts from the main point of the paper. The authors should find another venue to do that and to stick here to the science which can be obtained from the data.

Further detailed comments follow by page and line number.

3.6-8 What is meant by stating that “the vertical  $\text{HNO}_3$  redistribution may be saturated”? How is a vertical redistribution saturated?

5.28-29 and Fig 1 caption. In the text “the identified vortex region (indicated by non-shaded areas in Fig. 1a)” and the figure caption, “light grey shading: areas that are not associated with the polar vortex”, are at best confusing and at worst contradictory. The figure caption’s version is more consistent with the figure, but then there is a meandering split of the vortex into a western and eastern half. The uniform width of this split seems to indicate more than a filament of non-vortex air.

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6.1-2. “Only above the British Isles, southern Scandinavia and north-west of Norway patches of air masses do not fulfil this filter criterion.” How is the reader to interpret this statement? Is it an apology that these regions aren’t also included as vortex air, thus doubting the Nash criterion? Is “southern Scandinavia” meant to indicate the southern Baltic Sea? The aforementioned region of air nearly splitting the vortex cannot be characterized as “patches of air masses”. And with the criterion indicated as vortex a majority of the a/c observations are in this “patch”.

6.10-7.2 “In summary, the observed HNO<sub>3</sub> structures exhibit a much larger spatial variability than those observed in the ozone distribution, indicating their formation due to redistribution processes.” This statement is not acceptable. First the figure does not show a “much larger spatial variability in HNO<sub>3</sub> than in ozone. Between 11:30 and 13:00 in the flight data, both gases vary: HNO<sub>3</sub> from 3-7 ppbv, a factor of 2, ozone from 0.4 -1.2 ppmv, a factor of 3. Second if there were a difference how does that immediately lead to the conclusion of a redistribution process? There must be some additional explanation to make this leap this early in the paper.

7.11-12 “during in . . .” “Nash criterion, a relatively”

7.13 redundant with 7.11, please don’t repeat.

7.15-16 “a number of GLORIA observations where sorted out by cloud-filtering” What does this mean? Sorted out and put where? Do the authors mean remove? I do not understand what sorting out means.

7.20 NO<sub>y</sub>\* has not been explained and there is no reference and it is not used again. Is it important?

7.20-23 What is particulate HNO<sub>3</sub>? Perhaps these are particles vaporized in an inlet and the hno<sub>3</sub> gas measured, or ??? Why are the data not corrected for enhancement efficiency, insufficient information, small correction, . . .? Is it important that they are not corrected? Why do the presence of HNO<sub>3</sub> containing PSC particles need to be

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confirmed? Confirming compared to what? What other kind of PSC particles could be present besides hno<sub>3</sub>-containing particles? The first particle maximum doesn't coincided with a GLORIA maximum, but should it, if the hno<sub>3</sub> has condensed?

7.30 What is meant by “band-like structures”?

7.33 “distribution, indicating their formation by redistribution of HNO<sub>3</sub>” The authors again jump to their major conclusion without presenting any reasons.

7.34-35 Here for the first time the authors make an argument for their conclusion, but it is very brief and none of their data has included temperature relative to equilibrium temperatures with respect to NAT. Such information would help the reader understand why in some regions there are particles and in other regions gas phase hno<sub>3</sub>? In the regions where GLORIA data are shown, should the reader assume these are cloud free?

Figure 2 caption. The panels need to be described in the order in which they appear. A figure caption is so readers can understand what is in the figure. Why confuse them by listing the contents of each panel out of the order in which they appear? Since the interest is in polar vortex air, why do the authors state, here and elsewhere, what is not polar vortex air, rather than what is polar vortex air?

8.1-6 While the hno<sub>3</sub> mixing ratios for CLAMS agree with GLORIA, CLAMS does not show anything like the altitude tilted features appearing in the hno<sub>3</sub> GLORIA data. What particle information does CLAMS contain? Does that reproduce the in situ particle measurements? Figure 3 Why is waypoint A marked on the map and then not on the panels and not mentioned in the text.

9.8 Why call the hno<sub>3</sub> mixing ratios “enhanced” and “strongly enhanced”? This language assumes the authors' pre-determined conclusions prior to the arguments being made. The hno<sub>3</sub> mixing ratios are what they are, without this qualifying language.

10.1-2 “Since those structures between waypoints C and E vary significantly from those

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observed in the ozone concentrations they most likely originated from nitrification” Is this now the argument to be pursued, using ozone as a tracer? But in fact Figure panels 3b) and 3c) do not support the statement. The structure and the relative magnitudes of ozone and hno3 are quite similar. How do they “vary significantly”?

10.3-4 Now the CLAMS hno3 is “enhanced” even though the maximums and structure of the high hno3 regions do not match the observations. What is the significance of pointing out the very narrow high regions of hno3 in the CLAMS hno3? The last sentence describes well how the model and observations compare.

11.6-9 The enhanced language is used again and the claim of structures indicative of nitrification, as if this point is obvious for almost all of the hno3 values measured by GLORIA greater than some number. Perhaps if the reader were shown “non-enhanced” measurements of hno3 they may agree with the authors about the language, but all we see are hno3 values in the range of 5-10 ppbv pretty much in every flight segment shown.

11.22- It would be helpful to show some of the relative normalized frequency distributions. These could be more interesting for the reader than so many flight profiles, and the pointing out of small features in the flight profiles, which now may be removed as outliers, when the data analysis is finally explained.

12.5- Here finally information on the reference profile which motivated the previous language about enhancements, etc. is offered to the reader. Considering its importance the paper would be better served by showing this nominal reference data for comparison with the more dynamic data later. I am not sure the point of sentences listing the numbers of the maximums. Tables are good for numbers. Text is good for describing general features of the figures such as the progression of the descent of the hno3 over January and how CLAMS doesn’t capture this, nor the magnitude of the hno3.

12.14 An ozone loss of 15% significantly reduces the nitrification, so it needs to be clarified whether this was possible. Was such ozone loss occurring in the LMS? CLAMS

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should be able to at least estimate this.

12.16 I thought the measurements were filtered from non-vortex air in several ways. Are we now to assume that these correlation plots may be affected by non-vortex air?

13.4-9 Another paragraph pointing out the numbers which the reader can more easily obtain from the figure, or if they are important could be put in a table. Text should be reserved for something more interesting. This whole paragraph and others like it could be mostly removed and the paper would be better for it.

13.10-12 Yes but CLAMS completely misses the continuing descent of the hno3 observed by GLORIA.

13.27-29 “The comparison is based on the RNFDs depicted for the individual flights in Fig. 6.” And that is all there is to say about the sensitivity analysis of CLAMS compared to the flight data? Amazing, after the paragraph above describing all the different scenarios to test the sensitivity of CLAMS, no discussion of the results which indicate that the CLAMS results are almost insensitive to these perturbations. Figure 6 the flight dates should be added to the figure panels for reference later.

13.30-16.6 Without much of a difference observed in the summary RNFD plots for the CLAMS sensitivity tests the authors proceed to discuss the flight 6 cross section and its sensitivity simulation in detail, pointing out fine features/differences in Figure 7. But what is the conclusion reached from this detailed discussion? Line 14.8, “However, the overall structure in the RNFD is similar to the reference simulation.” Exactly, which is clear from Figure 6a. With this diversion back to detailed discussions of cross sections I gave up on the paper, assuming the same was going to be done for each subsequent flight. Although this is not done, neither is a general discussion of figure 6 offered. Is it really important to go through each model sensitivity difference for each flight when there are so few differences with the reference? A more helpful discussion of the figure would organize it by model sensitivity, and only discuss those sensitivities which make a significant difference with the reference in the direction of the observations. Based

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on Figure 6 this criteria would shorten the discussion considerably. Figure 7 and the current sections 6.1-6.4 should be removed.

17.11-19 Claiming distinct differences is an overstatement. The improvement of the temperature fluctuation for 31 Jan. is only evidenced by increased hno3 near 800 ppbv, otherwise it matches the CLAMS reference and all simulations lay significantly higher than the observations. The improvements in the T-1K simulation are generally hardly outside the reference except for 20 Jan. The aspherical particle case provides only a slight difference on 12 Jan. If some estimates of precision were placed on the CLAMS reference, most sensitivity simulations would be hardly outside. The sensitivity simulations simply do not support the claim of distinct differences. Which sensitivity should be chosen to improve the overall agreement with observations over the campaign? There is none.

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