Answer to Referee Comment 1

June 11, 2019

We thank referee 1 for valuable comments and suggestions. Our answers are given below. The original referee comment is repeated in **bold**, changes in the manuscript text are printed in *italic*.

General Comments: The authors have reported the observed HNO3 (and O3) from~8km up to 14 km from GLORIA during the PGS aircraft campaign took place from December 2015 to March 2016. The unique aircraft data will be useful for the atmospheric chemistry community. They have mainly focused on four flights data and also used a chemical transport model CLaMS to investigate the nitrification of the lowermost stratosphere for Arctic winter 2015/16. It is clearly shown that there are still large variabilities of measured HNO3 (and O3) in the LMS along the flight track and CLaMS seems to simulate HNO3 quite well though the model is not perfect to capture some fine structures and also underestimates the observed HNO3. Therefore, the authors have also done four sensitivity experiments to try to understand the discrepancies. Overall, the manuscript is well structured. The data analysis and model results are reasonable.

We thank referee 1 for this positive statement.

However, there are some important messages still missing or misleading in the current version. These need to be clarified.

Specific Comments:

1) Selection of aircraft data. It has been mentioned that 18 research flights were carried out between December 2015 and March 2016, but only five flights data are used. Some of other aircraft data may be not suitable for this work, but the authors have not mentioned why they chose these specific 4-5 flights data?

We added information on the flight selection in section 2:

The selection of the flight data was guided by the availability of long continuous "chemistry mode" measurements (see Sect. 2.2) in order to show how patterns in the lowermost stratospheric HNO$_3$ distribution change during the winter. We furthermore focus on flights in January, where PSCs extended down to the LMS and where the most notable changes are found in the observed HNO$_3$ distributions. Since we use ozone as a stratospheric tracer to quantify nitrification, flights in January are preferable since only little chemical ozone loss was diagnosed at this time of the winter when compared to February and March (see Johansson et al., 2019). Further GLORIA "chemistry mode" observations can be found in the supplementary information of Johansson et al. (2018) and at the HALO Database (https://halo-db.pa.op.dlr.de/).

2) ClONO2. I think the comparison of ClONO2 between GLORIA and CLaMS would help since GLORIA has also measured ClONO2 (Johansson et al., 2019) and CLaMS simulates ClONO2.

We agree that a comparison of ClONO$_2$ would be of interesting since this species also contributes significantly to NO$_y$. This aspect is part of the study by Johansson et al. (2019). We added the reference to this study in the manuscript (Page 3, Lines 26-28).

3) Abstract is not well written and some key points are not supported anywhere (for example the sentence in Lines 10-11,

Here we refer to P7/L29-34 and P16/L23-24 of the original manuscript in ACPD where the statements in lines 10-11 are discussed.

I am also not sure if the conclusion in the Lines 11-12 is a fair statement because other satellite has measured HNO3 in this region).

While we agree that other observations (in situ and satellite, see introduction) observed significant nitrification in the LMS region before, we are not aware of any studies reporting such high levels of HNO3 within the LMS due to nitrification as reported here. However, we would like to mention the separate study by Ziereis et al., which addresses this aspect using in situ observations during the same winter. However, we agree that this statement is somewhat strong and modified the abstract as follows:
Overall, *extensive* *nitrification of the LMS between 5.0 ppbv and 7.0 ppbv at potential temperature levels between 350 and 380 K is estimated.* ~~*This extent of nitrification has never been observed before in the Arctic lowermost stratosphere.*~~

What are missing in CLaMS when the authors conclude the model underestimates....(Lines 15-16).

The reasons for this deficiencies in the model are yet unclear. It should be noted that the distribution of tracers depends critically on vertical transport and that the vertical velocities are difficult to model. Another sources of uncertainty are the physical parametrisations and their implementation in the model. Therefore, a point-to-point agreement between model and observations is not expected. However, recent studies (Grooß,2018; Tritscher, 2019) showed that the in principle, the vertical HNO_3 (and H_2O) redistribution is reproduced well.

What is the implication for this work to improve HNO3 simulation in the lowermost stratosphere though some has mentioned in the Introduction?

There is no obvious fix. However, we point out a possible strategy assess our model understanding. We stated in the introduction that we test how well different parameterizations within the model reproduce the GLORIA observations. Based on our results we conclude that the sensitivity simulations show differences and lead to a sometimes improved agreement. However, no sensitivity simulation can be identified that generally improves the model. Therefore, we conclude that more extensive modifications of the model parameters addressed by our study are required and/or that important processes are still missing in the model. For clarification, we modified the abstract at P1/L19ff as follows:

… waves) slightly improve the agreement with the GLORIA observations of individual flights. However, no parameter could be isolated which results in a general improvement for all flights. Therefore, we conclude that a more comprehensive change in the model representations is required. Still, the sensitivity simulations suggest that details of particle microphysics play a significant role for…

4) Section 3.2 (Page 5). I am confused with the description. If CLaMS can save daily output at 12:00 UTC, why it can not save the model output along flight track (time, locations etc)? I am not sure why CLaMS needs to re-run forward/backward trajectory for the flight track though I understand CLaMS is based on trajectory calculations..

Using a standard forward integration, interpolation would always be required. Regardless how the flight paths are constructed, there would be no guarantee for a trajectory ending on the flight path at the right time. Certainly it could be done online with a slightly improved temporal resolution; however, the standard product for a longer term forward integration is stored in snapshots and processed afterwards.

5) Results explanation. Sometimes it is very hard to follow. For example, Page 7 Lines 9-10. Maybe the coarse vertical resolution is one factor. That will be easier to confirm by increasing the model vertical resolution in the LMS.

We investigated this effect and could not confirm the coarse vertical resolution as a dominating factor. However, since this flight was removed in context of suggestions by referee 2, this aspect is not mentioned any more in the revised manuscript.

Pages 9 and 10: What are the key points here? Sorry it is hard to understand the Lines 1-2 in Page 10.

We revised the discussion of the vertical cross sections to better highlight the key points (see revised manuscript). Further, we removed the statement given in lines 1-2 (see comments to referee 2).

Lines 15-18 in Page 12. Not so sure the points of the estimation of lower limit nitrification (though there is an almost linear relationship from the reference in Figure 5).

For clarification, in the revised figure (now Fig. 4) we constructed the same correlation neglecting a potential ozone loss of 15 % (i.e. the ozone mixing rations are scaled accordingly), which now clearly shows potential impacts of ozone loss on the presented tracer-tracer analysis.

6) Sensitivity experiments in the Page 13. The descriptions of the model sensitivity experiments are too general. Some of these can only be understandable by the people who are familiar with CLaMS. For example, 'ice settling' simulation, the authors just have one extra criteria to consider in the model (Line 17-18), but we don't know how settling velocity is calculated in the standard CLaMS model. 1.5 times settling velocity for the whole altitude range or something like that needs to add.

We added a reference for the settling velocity and changed the manuscript for clarification:

Therefore in the 'ice settling' simulation the computed ice settling velocity (computed as described by Tritscher et al., 2019) was increased by a factor of 1.5 at all locations where the saturation ratio of ice, S_{Ice}, is larger than 1.2.

For the temperature offset, why decrease global temperature by 1K rather than 1.5 or 2 K? Just simple say "NAT formation is T dependent" seems not enough.

We chose this sensitivity test based on the study by Hoffman et al., 2017 [ACP 17, 10.5194/acp-17-8045-2017, 2017]. This study found that temperatures from ERA Interim have a bias of 0.8K in comparison with those from Concordiasi long-duration balloon measurements in the Antarctic. For MERRA-2 they find a bias of 1K. We assumed that errors in the Arctic are unlikely to be much larger. Therefore, this number seems useful to us to investigate the impact of a potential temperature bias.

7) Discussion and Conclusion. Can you add more why the nitrification for Arctic winter2015/16 has much more than previous work as you mentioned in the Lines 20-26 in the Page 17?

We added following sentences to the manuscript:

During the Arctic winter 2015/16 exceptionally low stratospheric temperatures occurred and the vortex was sufficiently stable to allow formation of PSCs down to lowest stratospheric altitudes. Those conditions were the prerequisites for the strong nitrification observed and presented here.

Technical corrections:

1) Abstract, Page 1 Line 1, change "cold" to "low".

We changed the manuscript according to the referee's suggestion.

2) Page 1Line 5, why it is only spatial resolution? Does high temporal resolution matter for this case?

We emphasize the aspect of high spatial resolution, since our measurement technique allows particularly for a high spatial resolution. With regard to temporal resolution, the GLORIA data show continuous observations (i.e. 1 profile is measured in ~13 s), but of different air masses along entire flights or flight sections over periods of several hours. Thus we are focusing on overall conditions that are representative for several hours, while fast developments (i.e. at timescales of seconds or minutes) are not in focus of our study and would require a different experimental/flight design (e.g. "self-match flights" or a slowly moving carrier such as a balloon).

3) Page 1 Line 9. Are you sure about 11 pbbv of HNO3 is observed at 11 km from GLORIA? The only one I can see from Figures 4 and 5 but it occurs above 12 or13 km (?)

Thank you for pointing this out. We agree and changed it to 12 km.

4) Page 3 Line 7-8. What do you mean "mesoscale temperature is not well known"?

We changed the manuscript to: *mesoscale temperature modulations (e.g. by gravity waves) are…*

5) Page 3 Lines 18-20. This is too general.

We changed the manuscript to: *We compare the GLORIA data with simulations by the Chemical Lagrangian Model of the Stratosphere (CLaMS; Grooß et al., 2014, references therein). To test how well different parametrizations within the same model reproduce the GLORIA observations, four sensitivity studies were performed. Those sensitivity simulations investigated the impact of (i) enhanced sedimentation rates in case of ice supersaturation, (ii) a global temperature offset, (iii) modified growth rates and (iv) temperature fluctuations.*

6) Page 4 Line 2. "spectra and spectra"?

For better understanding we rephrased to: *… are transformed into spectra. The spectra from horizontal detector rows …*

7) Page 4 Line 25-26. A reference is needed. Is the same reference as Tritscher et al. (2018)?

Yes it is, we added the reference to the manuscript.

8) Page 5 Line 20. Add a reference for MERRA2. Why not to use ECMWF ERA interim because you have also done the model simulations based on the meteorological conditions.

We added a reference for MERRA -2. Data products from MERRA-2 were available, too. However, since we assume the MERRA2 and ECMWF datasets to be of comparable quality, we used both datasets.

9) Page 5 Line 22. Better to use "x" rather than . after"1.2"

We changed the manuscript according to the referee's suggestion.

10) Page 6 Line 3. Better to add an altitude range after 1.2 ppmv.

We removed this flight from the manuscript, based on the suggestions of referee 2.

11) Page 7Lines 8-9. Can you make "the enhancement at low altitude" clear? Is it enhancement of HNO3 inside the vortex region compared with outside vortex. Or you mean 2-3 ppbv HNO3 inside the vortex.

We removed this flight from the manuscript, based on the suggestions of referee 2.

12) Page 12. The unit in the text should be consistent with the figure

We changed the units of the figure to ppmv.