Dear Dr. Khosrawi,

Thank you for your letter associated to our manuscript submitted to ACP. We attached our point-to-point responses to the latest referee report to the submission. Here you can find our responses to your concerns described in your letter. References added here can be found below this response.

Re your questions 1 and 4:

1. For me also the answer why you cannot compare WACCM to MLS is not satisfiying. I compared ECHAM5/MESSy simulations to both instruments without encountering any problems (Khosrawi et al. 2017, 208). Both instruments are suited for comparisons with model data. Generally, I would say that the decision is rather made by which data set is available and maybe also to some extent which fits better to the model data. The comparisons in Khosrawi et al. (2018) showed that e.g the HNO3 distribution looks generally the same, but the absolute values are quite different, which I think will in case of your study change the results to some extent.

4. Since there are large differences between the instruments I doubt that you can find a general solution for the "best" NAT density value by just comparing with one instrument. There we are again back at comment 1, I am not convinced by your arguments that it is technically not possible to perform the same study with MLS data. You need better arguments why you only use one instrument and why only MIPAS.

It is important to note here that the comparison with MIPAS in the manuscript is based on a sampling of the model at exactly the same locations and times as the MIPAS measurements. Therefore, this is

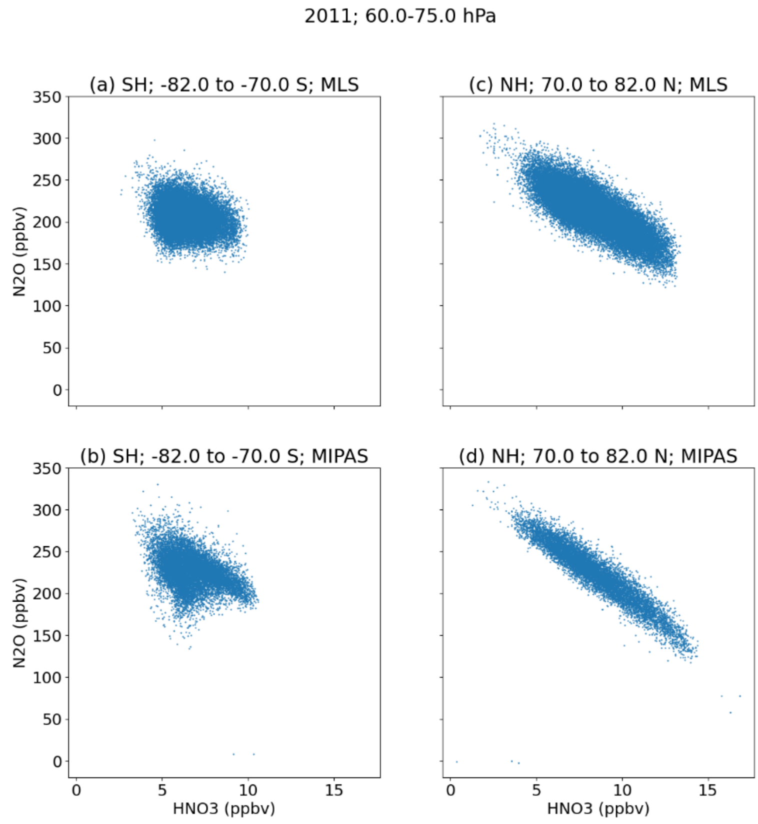

Figure 1: MLS and MIPAS HNO3 VMRs vs. N2O VMRs at high latitudes for both hemispheres during the local fall months in 2011. Data screening as recommended by the MLS documentation has been applied to the MLS data. The pressure range 60 to 75 hPa is shown.

based on single point comparisons, which means that precision of the instrument is crucial when applying our method. As stated e.g. by Piccolo and Dudhia (2007) and Sheese et al. (2016), the precision of MIPAS (about 0.2 ppbv or even lower) during the local spring months is substantially better than MLS (about 0.6 ppbv, see MLS data documentation , Livesey et al., 2022). In addition, the vertical resolution of MIPAS is about 3 km in the stratosphere which is higher than for MLS. MIPAS was able to measure profiles to latitudes up to 87 °N/S which is not possible with MLS. Figure 1 shows MLS and MIPAS HNO_3 vs. N_2O for both hemispheres (70 to 82 °N/S) at around 68 hPa during local fall months and it can be seen that the scatter is considerably larger for MLS than for MIPAS. For all these reasons, we decided to compare our results with MIPAS. Note that MIPAS is not the only dataset used but we also use ozonesondes for comparison with the same message as the MIPAS comparisons, which supports the conclusions of our study.

Re your question 2:

2. I cannot understand why one NAT density values needs to be used for both hemispheres. There are so many differences between the hemispheres in PSC occurrence and formation, would it not then be wiser to use different values and thus optimise these for the two hemispheres separately? At the moment it looks a bit like with every new comparison the NAT density value is adjusted (where we are back at my first comment), meaning for each (extreme) winter, satellite data set a new "best value" is defined.

The "real world" NAT density and subsequent denitrification can vary significantly based on air-parcel history (i.e., the Lagrangian trajectory) of temperature magnitude and water-vapor & HNO_3 abundance. The "model" choice to use a "one" number representation is not to suggest that the current parameterization is going to be directly comparable to the observed representation of NAT particle density. The point of changing this parameter in a Eulerian CCM (i.e., like WACCM6) is to get a reasonable representation of the observed gas-phase HNO_3 distributions in both hemispheres. Getting the right abundance of HNO_3 (g) is necessary for adequately representing ozone loss. The previous choice of the "model" NAT density (0.01 particles cm^{-3}) greatly underestimated the denitrification impact in the Northern Hemisphere, especially in cold winters (e.g., 2011 and 2020). Using the new particle density value (5×10^{-4} particles cm^{-3}) we were able to improve the Northern Hemisphere HNO_3 abundance and subsequent ozone loss. It also slightly improved the Southern Hemisphere HNO_3 distribution. The Southern Hemisphere is less sensitive to the “model” NAT density parameter choice due to the long period of cold temperatures where near complete denitrification can occur.

Re your question 3:

3. You provided some measured values of the NAT density which is important. However, how does your value fit into the range of measured values? This should also be shortly discussed. If it does not fit into the range of measured values you have a serious problem, because then it looks like you just find the perfect NAT density value to tune your model. By looking at the values, however, I had the feeling you where at the lower end of the measured values.

We would like to emphasize here (as also stated in our responses to the referees) that the NAT density does not necessarily need to be comparable to measured values. As we discussed in the responses to the first review, the NAT density in the model parameterizes more than only the physical quantity of a NAT density due to the gross simplifications in the NAT parameterization. It is a tuning parameter which can be used to lead to the goals: realistic global distributions of gaseous HNO_3. Nevertheless, we added a sentence to the conclusions about its correspondence to the measured values.

We hope we have addressed your concerns with these responses.

Yours sincerely and on behalf of all authors,

Michael Weimer

References

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Dear Ingo Wohltmann,

Thank you for the review of our revised manuscript. Please find below our responses to your comments.
Thanks again for your review and on behalf of all authors,
Michael Weimer

1 Responses to major comments

1.1 1.4 I acknowledge your decision not to use MLS data, but I think your argumentation in your reply is not valid. You are either showing vortex means (new Fig. 1), pdfs or a scatter plot based on the profiles inside the vortex (new Figs. 2, 3, 4, 5). That means that the statistical error (precision) of MLS or MIPAS data is negligible in your "final product", either because of the average of many profiles (Fig. 1) or because these errors cancel out in a pdf (or scatter plot) and don't change the pdf significantly (except maybe at the tails). That leaves the systematic error (accuracy). For HCl, this is for instance 0.2 ppb in the considered altitude range in the version 5 data, which is reasonably low (similar for other species). Consequently, MLS has been used in many studies for comparison to model data (I hope I don't have to give citations, you probably can easily come up with a list).

The precision of individual measurements are key to the distributions that are of interest here. MLS data distributions are compared to MIPAS in a separate letter to the editor and shown to be less precise.

1.2 1.15: Do I interpret you correctly that you would like to say that some of the chlorine originally in HCl at the start of the winter shows up in ClONO2 at the end of the winter? It might help to phrase it like this in the manuscript. In the moment, the formulation is not really to the point.

Thank you, we reformulated this sentence.

2 Specific comments

(Please note that the line numbers refer to the "Author's Track Changes")

2.1 96-97, comment of other reviewer: I think the main reason why this reaction should be kept is that it also happens on the background aerosol and not only on PSCs, and is important for the NOx partitioning. Without this reaction, the chemistry in the model would be completely unrealistic. This is partly stated in your sentence, but could be made clearer.

Thank you for this comment. We added this explanation to the sentence.

2.2 166-168: If this sentence is meant to serve as a justification for using the pdfs, you can only argue with the years here and not with the pressure range. The pressure range stays the same in Fig. 2 and Fig. 3.

From our point of view, Figure 2 demonstrates that there is a correlation between WACCM and MIPAS HNO3 when using the recommended NAT density. As stated in the comment, the same pressure ranges are used which is why we think that this also demonstrates that we can use the whole pressure range in the following figures.

3 Technical corrections

3.1 46: "solid HNO3 of NAT" : "HNO3 contained in NAT particles"

Corrected.

3.2 48: "stadard" : "standard"

Corrected.

3.3 71: I would write "NAT threshold temperature", because this is not necessarily the formation temperature in reality (supersaturation).

Corrected.

3.4 90: Is "decrease" the right word? "shrink", "evaporate" etc.

We corrected this to "change their size over time" instead of "grow or decrease over time".

3.5 125: "sigificant" : "significant"

Corrected.

3.6 134: "illustration" : "discussion"

Corrected.

3.7 Fig. 1 font size could be a little larger. Very hard to read. For the labels on the y axis (pressure), I would prefer 40, 60 and 100 hPa instead of the exponential notation.

We adapted the figure accordingly.

3.8 148: "deactivated through this mentioned reaction" sounds a little odd. "deactivated by the reaction into ClONO2" or something like this.

Corrected.

3.9 152: "decreasing" : Maybe "HNO3 VMR decreasing with time"?

Corrected.

3.10 155: Logic: "...do not get large enough for the simulated HNO3 to compare well..."

Corrected.

3.11 Fig. 2 caption "corresonds" : "corresponds"

Corrected.

3.12 163: "concise correlation" : "compact correlation"

Corrected.

3.13 164: "show a larger spread" : "show a larger scatter"?

Corrected.

3.14 164-165: Formulation: Suggestion: "The simulation HetAll.1e-2 shows HNO3 VMRs that are too large compared to the observations because of NAT particles that are too small."

Corrected.

3.15 165: Split sentence: Start new sentence: "As already suggested by Fig.1..."

Corrected.

3.16 165: Rephrase the sentence starting in 165 in a way similar to my suggestion 164-165.

We rephrased the sentence.

3.17 Fig. 7 : Font sizes are a bit small.

We increased the font sizes in the figure.