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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Lagrangian simulation of ice particles and resulting dehydration in the polar winter stratosphere" *by* Ines Tritscher et al.

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We would like to thank the anonymous reviewer for reading this manuscript and offering suggestions for improvements. In the following, we respond to his/her comments.

Major concern (1) *In addition to the general agreement of the model and the observations, which is impressive, the deviations of the model and the observations could be addressed in a more comprehensive way in order to highlight and fill gaps in scientific knowledge on PSC microphysics. This discussion can help to answer the following questions: Are implemented nucleation schemes for NAT and ice sufficient to explain all observations? How could the nucleation*

schemes be corrected or extended to better cover dehydration and denitrification measured by MLS? Shortcomings in the implemented NAT nucleation pathways lead to deviations in NAT particle size distributions and therefore to biases in the representation of denitrification in the model. Which processes or modifications could reduce those deviations? Similarly deficits in the ice nucleation schemes lead to smaller coverage of ice PSCs in the model and related reduced dehydration. Which processes could reduce shortcomings of the implemented ice nucleation schemes with respect to coverage with ice PSCs? This discussion could help to increase the scientific relevance of the paper and extend science beyond the state of the art. The results of the discussion should clearly be summarized in the abstract.

We considered your first comment as very important and tried to improve the different parts of the paper (Results, Discussion, and Abstract) accordingly. We have also taken into account your comment on "processes", which are better discussed in the revised version now. However, a detailed study about the importance of individual PSC formation mechanisms is beyond the scope of this paper. This publication is primarily meant as an introduction to the new CLaMS PSC ice module. We already have in mind to come up with a further study analyzing and (hopefully) understanding PSC formation in more detail. However, here is one example with changes to the discussion of the existing manuscript:

"However, the comparisons with CALIOP also shows differences regarding NAT occurrence. Cloud free areas, next to or surrounded by PSCs in the CALIOP data, are often populated with NAT particles in the CLaMS simulations. Looking at the temporal and vortex averaged evolution of HNO_3, CLaMS shows an uptake of HNO_3 from the gas into the particle phase which is somewhat too large and happens too early in the NH season. The permanent redistribution of HNO_3 in the NH is smaller compared to the observations. Also the Antarctic model run shows too little denitrification at lower altitudes towards the end of the winter compared to the observations. These findings point to shortcomings in the simulation of NAT particle sizes in combination with number densities namely that NAT particle sedimentation should be more efficient in CLaMS. Further studies should try to find better combinations of NAT number densities and sizes with the potential to denitrify the stratosphere more precisely. Heterogeneous NAT nucleation on foreign nuclei and preexisting ice particles is already implemented and covers most currently discussed routes to form NAT. However, NAT clouds downstream of mountain waves may act as “mother clouds” and individual NAT particles falling out of these clouds in low number densities can grow to large sizes of up to 10 μm (Fueglistaler et al., 2002). So far, CLaMS comprises the development of high number density NAT clouds on ice surfaces. No attention has been paid to the “mother cloud” theory, which could be a step forward to resolve deviations seen in the NAT simulations.“

Major concern (2) *A quantification of the deviation of the model results with respect to observations will strengthen the discussion of model agreement / disagreement with observations and will help to quantify uncertainties in the observations. In particular, quantitative deviations in stratospheric water vapor and nitric acid distributions between MLS und CLaMS could be added in a new the panel in Figures 5 and 10. Further a quantitative discussion of the agreement of model results and PSC observations by CALIOP and MIPAS could help to increase the value of the so far more qualitative discussion. Adding contour lines of TNAT and Tice to Figures 3, 7 and 8 could give further insights in data quality from observations and model. Could delta TNAT (or delta Tice) instead of temperatures be shown in Figures 3, 7 and 8 lowermost panel to get independent information on PSC phase or ambient conditions?*

As suggested by the reviewer, we added new panels to Figs. 5 and 10 showing the deviations between MLS and CLaMS. For clarity reasons, we removed the panel showing CLaMS results without the MLS averaging kernel. Moreover, we added contour lines of T_{NAT} and T_{frost} to the upper row of Figs. 3, 7, and 8 and set the color coded temperatures in the scatter plots in relation to the frost point.

Major concern (3) *The abstract could be rephrased to specifically highlight the results of the study with respect to ice PSCs and dehydration. The first 3 sentences of the abstract are too general and do not cover the content of the manuscript and therefore could be shifted to the introduction. If needed in the abstract, a more specific motivation could be given why Lagrangian simulations of ice PSCs and sedimentation are important. The scope of the abstract is to present the scientific results of this study. Comments with respect to previous work or campaigns (without explanation) could be omitted or shifted in the introduction unless it is urgently needed for a specific result. Quantitative descriptions of model agreement with observations should be given and disagreement could be discussed in sight of missing processes. Comments (1) and (2) will help to enhance the quality of the abstract, which is rather descriptive at the moment.*

We agree that the abstract was quite descriptive so far and reformulated it completely:

”Polar stratospheric clouds (PSCs) and cold stratospheric aerosols drive heterogeneous chemistry and play a major role in polar ozone depletion. The Chemical Lagrangian Model of the Stratosphere (CLaMS) simulates the nucleation, growth, sedimentation, and evaporation of PSC particles along individual trajectories. Particles consisting of nitric acid trihydrate (NAT), which contain a substantial fraction of the stratospheric nitric acid (HNO_3), were the focus of previous modeling work and are known for their potential to denitrify the polar stratosphere.

Here, we carried this idea forward and introduced the formation of ice PSCs and related dehydration into the sedimentation module of CLaMS. Both processes change the simulated chemical composition of the lower stratosphere. Due to the Lagrangian transport scheme, NAT and ice particles move freely in three-dimensional space. Heterogeneous NAT and ice nucleation on foreign nuclei as well as homogeneous ice nucleation and NAT nucleation on preexisting ice particles are now implemented into CLaMS and cover major PSC formation pathways.

We show results from the Arctic winter 2009/2010 and from the Antarctic winter 2011 to demonstrate the performance of the model over two entire PSC seasons. For both hemispheres, we present CLaMS results in comparison to measurements from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS), and the Microwave Limb Sounder (MLS). Observations and simulations are presented on season-long and vortex-wide scales as well as for single PSC events. The simulations reproduce well both the timing and the extent of PSC occurrence inside the entire vortex. Divided into specific PSC classes, CLaMS results show predominantly good agreement with CALIOP and MIPAS observations, even for specific days and single satellite orbits. CLaMS and CALIOP agree that NAT mixtures are the first type of PSC to be present in both winters. NAT PSC areal coverages over the entire season agree satisfactorily. However, cloud free areas, next to or surrounded by PSCs in the CALIOP data, are often populated with NAT particles in the CLaMS simulations. Looking at the temporal and vortex averaged evolution of HNO_3, CLaMS shows an uptake of HNO_3 from the gas into the particle phase which is too large and happens too early in the simulation of the Arctic winter. In turn, the permanent redistribution of HNO_3 is smaller in the simulations than in the observations. The Antarctic model run shows too little denitrification at lower altitudes towards the end of the winter compared to the observations. The occurrence of synoptic-scale ice PSCs agree satisfactorily

between observations and simulations for both hemispheres and the simulated vertical redistribution of water vapor (H_2O) is in very good agreement with MLS observations. In summary, a conclusive agreement between CLaMS simulations and a variety of independent measurements is presented."

Minor comments

P2 l19 *Which knowledge gaps exist? Be more specific.*

We rephrased this general statement to be more specific:

"Due to unknown processes in the formation of solid PSC particles, large differences in the parameterization of PSCs in global models exist."

P2 l27 *Which gaps, weaknesses and uncertainties exist? Be more specific.*

We improved this text passage as well:

"Non satisfying agreement between models and observations as well as fundamental differences e.g. in the NAT nucleation exist even in advanced PSC schemes, which further motivated the research presented in this paper."

P5 l26 *Water equilibrium depends in water partial pressure and ice crystals concentrations/surface areas.*

We agree with this statement and added one more sentence to the manuscript:

"Water equilibrium depends on gas-phase water partial pressure and water vapor pressure of the aerosol particles."

P6 l25 *Are the temperature fluctuations used for the NAT nucleation pathway, too?*

Yes, they are used for both nucleation pathways, NAT and ice. We clarified this at the end of Section 2.2.

P10 l8 *More information on MLS data and uncertainties could be given.*

We added information about the A-Train satellite constellation and MLS uncertainties.

P11 l14 *What causes the MIPAS NAT observations/interference $<$ 15 km altitude? Polar cirrus are not measured by MIPAS.*

We further improved Figs. 2 and 6 of the manuscript by including cirrus data from CALIOP and introducing an additional temperature threshold to the CLaMS data of 200 K. Moreover, we added the following paragraph to the manuscript to explain the differences at low altitudes in more detail. This paragraph should also answer your next comment.

“The disagreement between CALIOP, MIPAS, and CLaMS at altitudes below 15 km is noticeable. CALIOP observes cirrus clouds throughout the entire 2009/2010 season at altitudes below 15 km (Pitts et al., 2018). Further, CALIOP NAT at low altitudes is likely cirrus that has been misclassified. Volcanic aerosol from the Sarychev (48.1° N, 153.2° E) eruption in June 2009 were transported into the polar region producing an enhancement in the background aerosol at altitudes below about 18 km. MIPAS is highly sensitive to the presence of this volcanic aerosol which biases the MIPAS PSC detection (Spang et al., 2018) causing a striking maximum of PSC areal coverage at low altitudes in the early winter period. The widespread volcanic aerosol does produce an enhancement in the CALIOP estimate of the background levels, but will not significantly affect the PSC product since it is based on outlier detection. Both, cirrus and volcanic aerosols introduce a bias in the MIPAS PSC detection and are misclas-

sified as NAT (Spang et al., 2018). In CLaMS, we do not simulate clouds other than PSCs. The origin of the larger PSC area at low altitudes seen in the CLaMS PSC area panel can be explained by the altitude independent fixed detection threshold of 3.3 $\mu\,m^2\,cm^{-3}$ for STS droplets. At altitudes around 12 km, the stratospheric aerosol layer becomes visible as well. To reduce the large “PSC area” in CLaMS at low altitudes, we introduced a temperature threshold to this plot. Only data points with temperatures less than 200 K are considered. This temperature threshold reduces the maximum values of PSC areal coverage slightly.“

P11 l18 *Could you comment on the CALIOP and CLAMS results of total PSC, ice and NAT areas below 13 km altitude?*

Please see my comment to **P11 l14** above.

P11 l26 *Could you comment/quantify the agreement/disagreement between CLaMS and CALIOP?*

We understand that our discussion about the shown figure is on the short side. Therefore, we added more details to our explanation of Fig. 3 in the manuscript.

P11 l26 *Could you comment on the deviations in EnhNAT between CLaMS and CALIOP (Figure 3).*

We added the following paragraph:

”Enhanced NAT mixtures represent PSCs heterogeneously nucleated in wave ice PSCs. The CALIOP criteria defining enhanced NAT mixtures are conservative and therefore, the enhanced NAT mixtures subclass is not all-inclusive (Pitts et al., 2018). On this particular day, we expect no NAT PSCs downstream of wave ice clouds. Whereas this area is not populated in the CLaMS data, single scattered measurement points from CALIOP fall into this class, likely due to measurement noise."

P11 l29 *What causes the spread in CALIOP data (Figure 3, lowermost row) with respect to CLaMS results?*

We added the following information:

"The spread in the CALIOP data is caused by measurement noise. Although measurement noise is mimicked and added to the modeled data, the spread in the modeled data is slightly less than for the observed values. Those data points are still more confined and do not fill the whole space of the diagram."

P12 l33 *NAT PSCs do not follow due to data gaps, maybe rephrase.*

We rephrased this sentence.

P12 l35 *Could you comment on the disagreement in PSC occurrence below 15 km altitude between CLAMS and CALIOP and MIPAS?*

Please see my comment to **P11 l14** above.

P13 l1 *Explanation of results from Figure 7 are missing. Again there are similarities but also differences in the NAT and ice PSC occurrence in the upper panel and in the scatter in the lowermost panel in Figure 7.*

We extended the description of Fig. 7 in the manuscript.

P14 *General agreement is reasonable or good. Please now explain in detail deviations between model results and observations in sight of current PSC formation*

schemes. Which processes are not understood or not covered in the model that help to resolve the deviations?

Please see my comment to your **Major concern (2)**.

Figure 5 and 10 *Could a new panel be added in Figures 5 and 10 that quantifies the agreement/deviations between MLS and CLaMS?*

Done. Please see Fig. 1 below.

Figure 3, 7 and 8 *Could the TNAT contour lines be given in Figure 3, 7 and 8? This could help to decide on a bias in NAT occurrence by CLaMS or the observations. Could the Tice contour lines be given in Figure 3, 7 and 8? This could help to decide on a bias in NAT occurrence by CLaMS or the observations. Could delta TNAT (or delta Tice) instead of temperatures be shown in Figures 3, 7 and 8 lowermost panel to get independent information on PSC phase and to be independent on altitude/H_2O and HNO_3 partial pressures?*

Please see Fig. 2 below.
We added contour lines for T_{NAT} and T_{frost} and adopted also the temperatures shown in the lowermost panels of the corresponding figures. However, we would like to note that those temperatures depend on temperatures, pressure levels, and vapor concentrations from CLaMS and could therefore easily be wrong by some few Kelvin as well.

References

Fueglistaler, S., Luo, B. P., Voigt, C., Carslaw, K. S., and Peter, T.: NAT-rock formation by mother clouds: a microphysical model study, Atmos. Chem. Phys., 2, 93–98, https://doi.org/

10.5194/acp-2-93-2002, 2002.
Pitts, M. C., Poole, L. R., and Gonzalez, R.: Polar stratospheric cloud climatology based on CALIPSO spaceborne lidar measurements from 2006 to 2017, Atmos. Chem. Phys., 18, 10 881–10 913, https://doi.org/10.5194/acp-18-10881-2018, 2018.
Spang, R., Hoffmann, L., Müller, R., Grooß, J.-U., Tritscher, I., Höpfner, M., Pitts, M., Orr, A., and Riese, M.: A climatology of polar stratospheric cloud composition between 2002 and 2012 based on MIPAS/Envisat observations, Atmos. Chem. Phys., 18, 5089–5113, https://doi.org/10.5194/acp-18-5089-2018, 2018.

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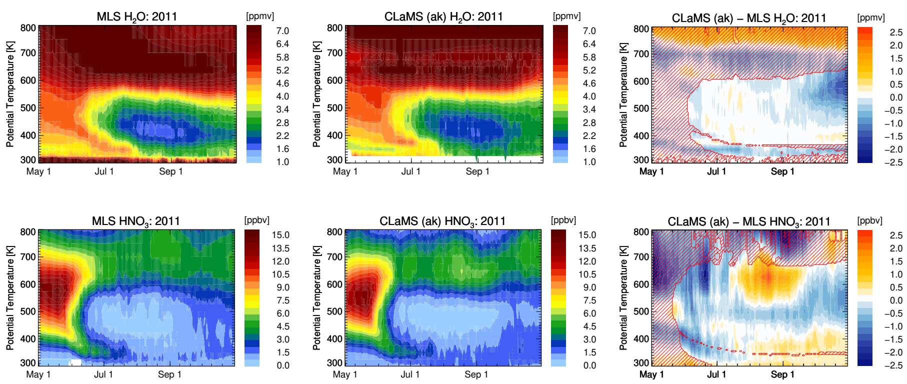

Fig. 1. Temporal evolution of water vapor (top row) and nitric acid (bottom row) are shown as an average inside the core of the polar vortex...

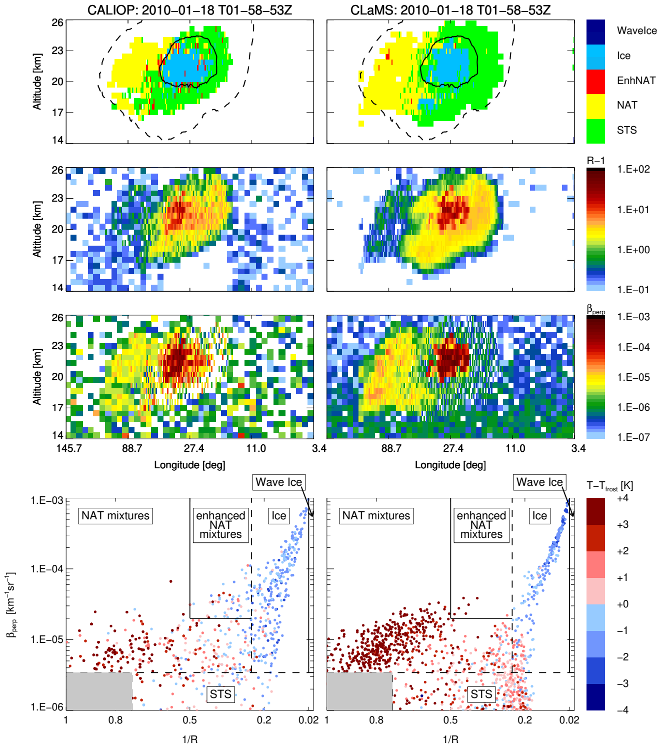

Fig. 2. 18 January 2010: CALIPSO orbit track 2010-01-18T01-58-53Z. CALIOP measurements are shown in the left column, corresponding model results in the right column...