Atmos. Chem. Phys. Discuss., 15, C12142–C12152, 2016
www.atmos-chem-phys-discuss.net/15/C12142/2016/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Polar Stratospheric Cloud evolution and chlorine activation measured by CALIPSO and MLS, and modelled by ATLAS" *by* H. Nakajima et al.

H. Nakajima et al.

nakajima@nies.go.jp

Received and published: 1 February 2016

We would like to thank Referee #2 for his/her time, constructive and helpful comments and suggestions.

<General comments:>

The authors examine in their study different topics with the help of several trajectories calculated with ATLAS. In my opinion this method is innovative and appropriate for the performed study.

Thank you very much.

From about 30 calculated trajectories, they choose eleven for their study. Unfortunately it is not clear, what kind of criteria they use to choose these eleven trajectories. For example number 3 and 4 are very similar. That issue should be cleared by the authors.

The way of selecting the trajectories is described in Section 3.2 in detail. The selected eleven cases are chosen to cover several different PSC composition classifications and different temperature histories. We agree that case #03 and #04 are rather similar. Therefore, we deleted case #03 from the draft.

In the result section 5.1 (with the help of Figs. 5 to 7) Nakajima et al. discuss the dependence of PSC classification on their temperature history with the help of CALIOP measurements on different chosen trajectories. The authors conclude that the kind of formation of the PSC types depends on the temperature history. In cases of rapid temperature decrease first STS is formed, followed by NAT/STS clouds. When temperatures dropped below the frost point, ice clouds formed, and then transformed into NAT/STS mixture when temperature increase above the frost point. This part of the manuscript I find very interesting and the results are in my eyes relevant for publication.

Thank you very much.

In section 5.2 (with the help of the Figs. 8 to 11 and 13) the authors compare the results of the ATLAS model with MLS and CALIOP measurements. They performed for this comparison three different model runs: “STS+NAT” (I think this is the standard run), “STS” and “CALIPSO constrained”. The comparison of HCl, and ClO between ATLAS and MLS is in general very good, although ATLAS in many cases doesn0t match correctly the PSCs from CALIOP and the authors can show that chlorine activation is limited by the amount of available ClNO3. But the comparison of HNO3 fits not very well and the O3 comparison is difficult because there are only small variations in ozone on the selected trajectories. The authors write that the ALTAS model cannot reproduce the denitrification for a single trajectory (page 22156, line 3-7). In this case I would

ACPD
15, C12142–C12152, 2016

suggest to skip the HNO3 and also the O3 comparisons.

We agree that denitrification is difficult to simulate for a single trajectory. Since HNO3 is not the essential part of this paper, we deleted all the discussion and panels on HNO3 from the draft. Also, simulated and modeled O3 depletion on the trajectory is small, because no enough sunlight is available in this period of year. Therefore, we also deleted the discussion and panels on O3.

In general I would also recommend to examine one or two less trajectories studies, because there are in my opinion too much similar figures to consider. Maybe it would also be a solution to add a supplement with more trajectory cases.

We disagree to reviewer's comment on this point. Each trajectory case shown in the draft represents different temperature history and different PSC classification history on the course of trajectory. Therefore, we kept all figures in the current draft.

Moreover I don't see really the relevance of the both sensitivity runs ("STS" and "CALIPSO constrained"). In my opinion there is no real improvement shown through the results of these sensitivity runs. Especially if the setup regarding the number density of NAT and ice particles in the "STS+NAT" run and "STS" run is different as in the "CALIPSO constrained" run (Sect. 4.2) it should be considered to skip all results of the sensitivity runs from the figures and remove the corresponding discussion in the publication.

We agree to the reviewer's comment on this point. There is not enough differences between the three sensitivity runs. Moreover, the number densities used for these runs are different, because different PSC scenarios were used for these runs. Therefore, we used only "STS+NAT" run results, and deleted results and sensitivity runs part (Section 4.2) from the draft.

In Section 5.3. (with the help of Figs. 12 and 14) Nakajima et al. confirm that the formation of PSCs are very temperature dependent with the help of two another sensitivity

ACPD
15, C12142–C12152, 2016

runs (+1K and -1K temperature runs). The authors claims that the run with decreased temperatures fits better with the observations. But in my opinion this is at the most valid for HCl. With focus to the surface area density, ClO or HNO3 I don't see really an improvement. The conclusion that the formation of NAT is temperature dependent is in my opinion not really new. I would suggest also to remove this subsection or at least to choose different trajectories with better results.

The comparison with HNO3 is difficult by ATLAS model as is explained above, and we deleted HNO3 panels from the draft. The purpose of showing Figs. 12 and 14 is to represent that chlorine activation is very sensitive to temperature. Therefore, we keep these figures and discussion in the draft.

<Specific comments:>

Page 22142, 16-17: In my opinion the ATLAS model results only agree well with the observations in the case of HCl and ClO. Please add this here.

Since we deleted discussion on HNO3 from the draft, we think that the current description is OK.

22143, 4: If you mention that STS is H2O-H2SO4-HNO3, perhaps you should also mention that NAT is HNO3(H2O)3

We added the suggested description there.

22143, 21: Voigt et al., 2005 and Hoyle et al., 2013 only assume heterogeneous nucleation of NAT on meteoritic dust. Biermann et al., 1996 showed in laboratory experiments that heterogeneous nucleation rates on micrometeorites are too low to enhance freezing of polar stratospheric clouds above the frost point.

We added description on Biermann et al. (1996) there as you suggested.

22143, 24: "(and hence strongly on temperature)".

Changed as suggested.

ACPD
15, C12142–C12152, 2016

22144, 7-8: I don't know if you really can answer the second question with your study. It's really difficult to evaluate the sensitivity of chlorine activation or ozone depletion on different PSC types with your study. Moreover there is in my opinion no chemical ozone depletion shown on your trajectories neither by the simulation results nor in the observations.

You are right that it is difficult to study on the difference of chemical ozone depletion to different PSC composition by our current study. We deleted description for ozone from the text.

22146, 1-2: What is the reason that you use only three instead of at least four categories (STS, Mix1, Mix2 and ice)?

Mix1 and Mix2 are the categories both of which include so-called "NAT" PSC. Therefore, we used conventional three PSC categories, STS, NAT, and ice.

22147, 6-11: In my opinion you don't have to describe the legend to the figures in the caption and in the text.

We deleted some sentences from the text.

22150, 3: What is the meaning of supersaturation of 10? 10 percent?

We have changed "about" to "of a factor of".

22150, 7-9: What criteria do you use to choose these eleven trajectories?

The way of choosing the eleven trajectories is to cover several different PSC composition classifications and different temperature histories. We added this description in Section 3.2.

22150, 22: Please cite Dee et al. 2011 for the ERA-Interim reanalysis.

Cited as suggested.

22150, 23: . . .are allowed to form in parallel. . . (?)

ACPD

15, C12142–C12152, 2016

Changed as suggested.

22151, 16: . . . the maximum particle number density?

The number density is constant. See Wohltmann et al., 2010.

22151, 18: . . . of 10 %?

See reply to comment to page 22150, 3.

22152, 12: Please explain why do you use here a much smaller NAT number density as in the “STS” and “STS+NAT” runs?

Since the “CALIOP constrained” run used different PSC scenario compared with other (“STS” and “STS+NAT”) runs, their assumed PSC number densities were different. As is pointed out by your General comments, we found that there is not enough differences between the three sensitivity runs, so we decided to show only “STS+NAT” run results in the draft.

22152, 15: Here the same: why do you use here 1 cm-3 instead of 0.01 cm-3?

Please see the reply above.

22154, 3: In table 1 are eleven selected trajectories not nine

We discussed nine (now eight, deleted one case in new Fig. 5 following a reviewer’s comment that old case #03 and #04 are very similar) cases from table 1 for PSC evolution by temperature history. Other three cases (#02, #03, and #09) are used for temperature sensitivity study (Figs 11/12, and Figs 13/14).

22154, 9: “When temperatures warmed above T_{NAT} in trajectory case #1. . .”

Changed as suggested.

22154, 23: Please add a citation for this “old theory” of NAT PSC formation

Added Koop et al. (1995) as a citation for this “old theory”.

ACPD

15, C12142–C12152, 2016

22154, 25: In my opinion the temperatures are never below T_{ice}.

We changed “below T_ice” to “near T_ice”.

22154, 25-28: The first sentence is maybe okay, but I don’t know if this is really a proof for the accuracy of the ERA Interim temperatures. I would say maybe a hint. But you have to delete in any case the second sentence. It is presumptuous to claim that the ERA Interim temperatures have an uncertainty of 1 K, because your model fits better with 1 K lower temperatures.

Your comment is right. We have deleted the second sentence.

22156, 3-7: If it not possible to simulate the denitrification with ATLAS on your trajectories, why do you use ATLAS then for this study? Or why do you show then comparisons of HNO3 and also of O3 between the model and the measurement? That make no sense in my opinion.

We agree. We deleted panels and discussion related to O3 and HNO3.

22156, 22-23: Cl2 can’t be photolyzed to ClO_x because Cl2 is part of ClO_x.

Cl2 is photolyzed to Cl, and then form ClO, which is a part of ClOx. We rephrased in the text. We mistook the legend of ClOx in panels (f) (new panels (e)) of Figures 8 to 14. We changed the species shown in panels in Figures 8 to 14.

22156, 27: I don’t see a chemical ozone depletion neither in ATLAS nor in the measurement. ClO is the product of O3+Cl -> ClO, Cl is the reason for O3 depletion, not ClO.

You are right. There is very small O3 depletion modelled by ATLAS at around day 2.8 and 3.5, when small solar illumination was available, which was shown by orange dots on the top of panel (i) in Figure 9. However, O3 depletion is not the main issue of the focus of this paper, we deleted description related to O3 from the draft.

22157, 11: Cl2 is photolyzed to Cl, not to ClO.

ACPD

15, C12142–C12152, 2016

See reply to comment 22156, 22-23. The text was rephrased.

22157, 12: I would say “slightly” is the wrong word for this depletion.

All the discussion on HNO3 is removed from the manuscript.

22157, 13-15: Again I don’t see really an ozone depletion.

We deleted description related to O3 from the draft.

22157, 28-22158, 2: Why do you have such a decrease in HCl and ClNO3 in the STS run (and also in the STS-NAT run), although you simulate only a very small PSC surface area?

Surface area density does not directly translate into reaction rates. The reaction rate is also dependent on the amount of ClONO2 available and temperature. E.g., in Figures 8-10, the model just runs out of ClONO2. In this case, it does not matter that the surface area densities are much larger.

22158, 12: Is the result of the -1 K model run really better than the standard model run? In the first simulation you have in average smaller values as measured, in the second simulation you have in average higher values as measured. In my opinion both runs fit well with MLS regarding HCl. But in both runs the simulated PSCs don’t match the observations and also the denitrification is not really better in the -1 K run.

Since there is not enough difference between three PSC model runs, we only show “STS+NAT” run results in the draft. We believe that temperature difference of 1 K is important for the simulation result.

22158, 13-15: That is surely a correct suggestion, but not really a new result, or?

We modified the text to refer earlier result by Carslaw et al. (1994).

22158, 24-26: You have also to delete this sentence. There is really no proof in your study for any uncertainties in ECMWF temperatures.

ACPD

15, C12142–C12152, 2016

The sentence was deleted as suggested.

22159, 1: Surely the HCl results are better in this case with the -1 K model run, but quite well?

We want to keep as it is.

22160, 13-15: This is also my opinion. What is the reason that you show the results of all these scenario runs in this publication?

We modified to show only “STS+NAT” result in the draft.

22161, 20-23: If you write as on page 22159, 18-19, that your study “suggest the possibility of heterogeneous nucleation of NAT on solid particles” I can live with it, but here you mention again the meteoritic dust, although you write on page 22159, line 16-17 that this case is unlikely (Biermann et al., 2006).

We added the description on Biermann et al. (1996) here as well as on page 22159.

22162, 11: Please write here ClO instead ClO_x.

Changed as suggested.

22162, 24-27: It is not true that the temperature explain most discrepancies between model and observation. Please correct this sentence. Also in the -1 K run you don’t match PSC periods from CALIOP or the HNO3 MLS measurements.

We rephrased here to more focus on modeled and measured HCl discrepancies. The PSC field is created by taking the closest CALIOP measurement, and sometimes not represent the fine structure of actual PSC field. (Please see our reply to other referee (#1) major comment 3.)

< FIGUES >:

Fig.1: last sentence: “.. and the Type II (ice) frost point temperature ..”

Changed as suggested.

ACPD

15, C12142–C12152, 2016

Fig.3: Can you please also plot the colour bar which indicates the different PSC types in the panels b and c. This is in my opinion important because the colour code is different to panel a (otherwise it is confusing). If it possible it would also be great to write the longitudes and latitudes to the horizontal axis of panel b and c (in the same way as in panel a). In panel c you have two blue lines, maybe you can choose light blue in one case instead of blue.

Color bar which indicates the different PSC types were added in panels (b). The longitudes and latitudes was not added to panels (b) and (c), because it is not a very useful information for the reader. The color of PSC saturation temperature was modified as suggested.

Fig.4. to 14: Same as in Fig. 3: Please add a colour bar with the PSC types and the latitudes and longitudes of the trajectories.

Changed as suggested. The latitudes and longitudes of the trajectories were not added.

Fig.5: a) Can you please add in the caption at least the information about time and altitude and/or pressure for the trajectories cases? b) Why is the sequence #1, #4,#3 and not #1,#3,#4 c) Why do you show #3 and #4 both, this trajectories have more or less the same conditions due to the very similar starting point? d) Again information about the latitudes and longitudes of the trajectories and a colour bar would be great.

The time and altitude information was added to the caption of Fig. 5. The sequence was re-ordered. The case #03 was removed. The color bar was added. Latitudes and longitudes of the trajectories were not added.

Figs.6. and 7: Same as in Fig.5: More information would be great and the sequence is strange.

Cases were re-ordered.

Fig.8: a) Temperature of trajectory case #3 is only displayed in Fig.5 not in Figs.5-7. b)

ACPD

15, C12142–C12152, 2016

It's difficult to read the titles and the units in the panels. c) In general the panels are very small (maybe this is better in the final paper?). For example the line of the grey curve of the STS run is extremely difficult to see. d) In Fig. 8h I see also a dashed blue line. What is the meaning of this line or is this a mistake?

Since we deleted old panels (c) Rates, (h) HNO3, and (i) O3, the new figures 8-14 consists of six panels. We hope this will improve the readability of each line. The HNO3 panel (h) was deleted.

Fig.9: There is again this dashed blue line in panel h.

The old HNO3 panel (h) was deleted.

Fig.12: a) There are again dashed blue lines in panel h. b) The comparison of measurements and model is in the -1 K run not very good. The surface area density and the HNO3 don't match the observations. Why do you select this trajectory? Is this really the best comparison you have (you write you analysed more than 30 trajectories).

The old HNO3 panel (h) was deleted.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C12142/2016/acpd-15-C12142-2016-supplement.pdf

---

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 22141, 2015.