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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.

Anonymous Referee #2

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The content of this study is an evaluation and comparison of satellite measurements from CALIOP and MLS and model results from ATLAS with the focus on Polar Stratospheric Clouds (PSC) and the heterogeneous chemistry which take place on these PSCs. The scientific focus is the PSC evolution depending on different temperature histories of air parcels and the chlorine activation on these PSCs.

Nakajima et al. calculate trajectories calculated with the Lagrangian chemistry and transport model ATLAS for the Arctic winter 2009/10 and compare the development of different PSC types from CALIOP and HCl, ClO and HNO3 from MLS with results from simulations with ATLAS on these trajectories.

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Discussion Paper

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.

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.

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.

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 doesn′t 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

ACPD

15, C8046–C8052, 2015

(page 22156, line 3-7). In this case I would suggest to skip the HNO3 and also the O3 comparisons.

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.

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.

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 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.

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.

22143, 4: If you mention that STS is H2OÂůH2SO4ÂůHNO3, perhaps you should also mention that NAT is HNO3(H2O)3

22143, 21: Voigt et al., 2005 and Hoyle et al., 2013 only assume heterogeneous nu-

ACPD

15, C8046–C8052, 2015

cleation 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.

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

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.

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

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

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

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

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

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

22151, 18: . . . of 10 %?

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

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

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

ACPD

15, C8046–C8052, 2015

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

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

22154, 25: In my opinion the temperatures are never below 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.

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.

22156, 22-23: Cl2 can′t be photolyzed to ClO_x because Cl2 is part of ClO_x.

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.

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

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

22157, 13-15: Again I don′t see really an ozone depletion.

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?

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

ACPD

15, C8046–C8052, 2015

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.

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

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

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

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?

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).

22162, 11: Please write here ClO instead ClO_x.

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.

Figures:

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

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

ACPD

15, C8046–C8052, 2015

blue in one case instead of blue.

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.

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.

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

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

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

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).

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