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Interactive comment on "Linking the uncertainty in simulated arctic ozone losses to modelling of tropical stratospheric water vapour" *by* Laura Thölix et al.

Anonymous Referee #1

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General

The topic of this paper is Arctic ozone loss in general and the sensitivity of Arctic ozone loss on the polar water vapour concentrations in particular. If the authors agree, they need to remove all the discussion of mid-latitude ozone loss and the related cycles. Further, discussion/citation of upper stratospheric ozone loss is not helpful.

The topic of the paper (water vapour sensitivity) is of importance and the the approach using a CTM to focus on the impact of water vapour on PSCs on heterogeneous chemistry is good. On the other hand, the results on the sensitivity will gain on impact if the representation of PSCs in the model, the simulation of chlorine activation and ozone

loss are clearly demonstrated in the paper. I do not think that this is the case in the present version of the paper (the cited papers are not sufficient, see below).

For example, the impact of humidity on heterogeneous chemistry should be different for different types of PSCs. If in this model the PSC types are dominated by a different type that in reality (or even in different models) carrying over the information on sensitivity deduced here can be problematic. There is some comparison with ERA-interim, but this is not the real world. Also there i.e. very little information on chlorine chemistry (except in Fig.6, where there is no distinction between HCl and ClONO2 for example, see also below).

The paper addresses a second case, namely a warm and dynamically unstable winter in the Arctic stratosphere. In such a case, in contrast to the halogen induced ozone loss in cold Arctic winters, there is little ozone loss below 500âĂŕK and ozone loss in the middle stratosphere (NOx-induced destruction) becomes important (e.g., Konopka et al., 2007; Sagi et al., 2017). However, this is a completely different chemical mechanism, which will have a very different dependence on water vapour (clearly no impact of heterogeneous chemistry). Therefore this second case needs to be very clearly discriminated from the "halogen" case throughout the paper.

In summary, I recommend focusing the paper and a providing a better basis and justification for the work presented. I also suggest a better balance of the cited literature (as stated in the quick review, I am not suggesting to cite all the references mentioned here). I am sorry for the many critical remarks but I believe that a revised paper taking into account these comments would be much stronger than the present version.

General

The discussion of the HOx chemistry in mid-latitudes and in the upper stratosphere (and the associated references) in the introduction is confusing. In the Arctic, in cold winters, an increase in water vapour is expected to enhance ozone loss (assuming that a substantial amount of stratospheric chlorine is present) by enhancing heterogeneous reactions (Shi et al., 2001; Drdla and Müller, 2012). The situation can actually be more complicated as there is no simple argument that “more PSCs” means “more ozone loss”. This is also a finding of this paper, if one considers the case of the cold Arctic winter 2010/11. The latter point should be more emphasized in the paper I suggest.

Recently, climatologies of PSC occurrence have become available from observations (Spang et al., 2017; Pitts et al., 2018). The question how important the discrimination between different PSC types in a model is for a successful simulation of polar ozone loss is not trivial. This paper could contribute substantially to this issue; perhaps more than visible in the present version of this paper. This aspect could be very relevant for the discussion of the sensitivities on water vapour (which will be different for different types of PSCS). Finally, models might misrepresent PSC volume density compared to observations; Khosrawi et al. (2018) report that the comparison between the PSC volume density as simulated with EMAC and the one derived from Envisat/MIPAS observations shows that the simulated PSC volume densities are several orders of magnitude smaller than the observed ones.

I also think that the demonstration that FinROSE is successful in modelling polar ozone loss is not sufficient. Very little information is given in this paper. Fig. 6 only give a summary (e.g. I cannot judge whether or not the recovery of active chlorine into ClONO2 is convincing), Other models have done comparisons with observations in details (see e.g., Wohltmann et al., 2017, and references therein). Here the reference to Karpetchko et al. (2013) is given, but in this study FinROSE is used with PSCs “switched off”, so this paper is not valid to support the performance of the model for Arctic ozone loss in cold winters (see also below). In section two of the paper a description of the initialization of the model should be given. How is total chlorine (Cly) initialized? What ware the initial values for HCl, ClONO2, N2O5 etc – this information would be helpful to interpret the results of the paper. How good is ERA-I ozone in comparison to MLS ozone? How well does FinROSE simulate downward transport in the Arctic (compare to MLS N2O?).

One important driver of chemical ozone loss in cold Arctic winters is the stratospheric chlorine loading. I suggest making this point and briefly discuss the temporal development of stratospheric chlorine (e.g., Engel et al., 2018).

Details

- title: the title is too general, is sounds like *the uncertainty* in simulated Arctic ozone is due to water vapour. But there are more reasons for uncertainties
- abstract, l. 8: the point here is that there is a one-to-one correspondence between entry value and polar water vapour conditions – correct? This point could be made more clearly.
- abstract, l 13-15: I think this is an important finding of this paper.
- abstract, l 17: 2–7% more: this is a very important result of this paper. But can be anything worked out what the mechanism is? It would be important to state this finding in the abstract.
- p.2, l. 1: Temperature may show a warming in the troposphere but a significant cooling near the tropopause in connection with deep convection (Kim et al., 2018), so the connection between climate change and tropopause temperature may not as straightforward as suggested here. I suggest more discussion of this point which is important for this paper.
- p.2, l. 3: “affect chlorine partitioning” but how? Is this relevant for polar chemistry?
- p.2, l. 7: this citation is for the upper stratosphere; I do not think it is appropriate here.

- p.2, l. 12: One could also mention the observations discussed by Kuttippurath and Nair (2017) here.
- p. 2, l. 14: do you have references to more recent studies to back up this statement on recovery?
- p. 2, l. 17: remember that the focus here is on *Arctic* ozone.
- p. 3, l. 3: I do not think this statement is correct. Please consider the temperature dependence of the main polar ozone loss cycles (von Clarmann, 2013; Canty et al., 2016; Wohltmann et al., 2017). I you do not agree, please specify “some”.
- p.3, l 17/18: this is more than “likely” if temperatures are high, there are no PSCs and thus there is no impact of water vapour on PSCs and heterogeneous reactions. Under these conditions NOx chemistry indeed is relevant (but we are missing a discussion here of the impact – if any – of water vapour on the NOx chemistry in question). In any even (see also other points in this review) these two cases must be clearly separated. You are looking at different processes here.
- p. 3, l 28: the idea of a “controlled” experiment is good! also change to “impact of . . . on ozone loss”.
- p. 4., l 9: this is not an important point, but is has recently been argued that the reaction CH3O2 + ClO is important for polar ozone loss (Müller et al., 2018); is this reaction taken into account in the chemical scheme used here?
- p. 4, l. 16/17: As it is very important for this paper: describe here how the dependence on water vapour of the heterogeneous reactions (i.e. the γ) is determined.
- p. 4, l. 19: As it is very important for chemistry simulations at the poles: describe here if(how) spherical geometry is properly taken into account in the photolysis code.

- p. 4, l. 29: the reference to Karpetchko et al. (2013) is important here as this is the only citation give to support the performance of FinROSE in simulation polar ozone loss. However, in Karpetchko et al. (2013) FinROSE is used with PSCs “switched off”, so this paper is not valid to support the performance of the model for the issues discussed in this paper.
- p. 5, top: It is not clear to me what was done exactly here: what is “Interim(MAX)”? Why did you not simply shift the water vapour values up and down by some value preserving the variability?
- p 5, l 9: The comparison to MLS ozone would be important. It should be shown and discussed in the paper in detail. Further, MLS HCl (and possibly other measurements of Cl-species could be helpful to validate the model).
- p. 6, l 7: which reference theta level was used for mPV? How is 36 PVU chosen?
- p. 6, l. 23: Here (and perhaps elsewhere) the question arises if downward transport in the FinROSE model in the Arctic polar vortex is simulated appropriately. This issue has an impact of water vapour in the polar vortex. A comparison of simulated N2O with observations would be helpful here.
- p. 7., l 5: when dehydration occurs the initial water vapour and thus the Max/Min scenarios will not be relevant any more, as the polar water vapour is set to the equilibrium value over ice – correct? This issue should be discussed in the paper.
- p. 7, l 15: what is meant by NAT/STS volume, Just adding both PSC types? But the impact of increased water vapour on NAT and STS will be different. So why has this been done; I suggest separating NAT and STS.
- p. 7, l. 22: is there some impact of sedimentation on the duration of ice PSCs?

- p. 7, l. 23/24: Note that the Calipso PSC product has more recent information now (Spang et al., 2017; Pitts et al., 2018); I suggest using the most recent information. For example the estimated ice area might change.
- p. 8, l 4: “too dry models” this point sounds very speculative
- p 8, l. 25: One question that arises here how well the model simulates the size of the vortex – as this point might be relevant for assessing the PSC area.
- p 9, l 10: small twice
- p. 9, l 14: chlorine activation does not require PSCs, it starts on cold binary aerosols, but is also humidity dependent (e.g. Solomon, 1999; Wegner et al., 2012).
- p 9, l 25: which reservoir species?
- p 10, top: the start of activation is one thing, but not really what determines how much ozone loss happens in a particular winter.
- p 10, l 12: which process is responsible here?
- p 10., l 23: but why?
- p 10., l 26: I am not convinced that this statement is correct, is this really a cause and effect relation?
- p 10., l 30: I do not think that table 2 is a good summary of the chlorine activation simulation in the model run, too many important details are missing. (See also other comments in this review).
- p 11, sec 3.4: it would helpful to have more comparisons to ozone loss from simulations of other models.

- p 11, l 18: is this statement on NOx also true for FinROSE? If yes, what is the evidence from the model simulation for this statement?
- p 12, l 2: this might be true, but this is an example where speculation could be replaced by statements based on the actual simulations. Has a simulation been attempted with a better resolution?
- p 12., l 7: “without het. chemistry”: it should be clearly stated what has been assumed regarding the heterogeneous reaction $N_2O_5 + H_2O$, which is not temperature dependent but would be important here.
- p 12., l 13: this is not true for the het. reaction $N_2O_5 + H_2O$
- p 12., l 16-18: this statement is really confusing: if I understand correctly, only 30 DU ozone loss is caused by NAT/STS/ICE PSCs in Arctic winter 2010/11: This is in contrast to statements elsewhere in the paper and also to literature and our general understanding of Arctic ozone loss. rent types of PSCs are for the results of this section.
- p. 14., section 4: My suggestion would be to not combine discussions and conclusions. Have a separate discussion section to focus on the relations of the results of this study with what is available in the relevant literature and a separate conclusion section to focus on the main conclusions of *this study*. But this is up to the authors.
- p 14., l 15: this is an example where the analysis of the paper could be more focused and more detailed. What means “larger”? It will be important to which altitude regions and how much towards the vortex edge the PSC area extends. If these details are analyzed more can be learned about the processes responsible for the model results.

- Fig. 6: In general I think that there is not enough information on chlorine species in this paper. Specifically: 2012/13: why is there a range in reservoir species in early winter in this case for Min/Max? Can this be explained/understood? In 2010/11 there seems to be full depletion of HCl. It is not obvious that this can be easily achieved in models. Is it understood how this happens in FinROSE (reformation of ClONO2 and HOCl is necessary for this). What is the role of HOCl in chlorine activation through heterogeneous reactions?
- Fig. 7: Is there really an established Arctic (!) vortex (not only small remainders of the vortex) for the entire period shown here? Until mid-April! Show a time series of the size of the vortex at least in the reply or in a supplement.

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