

## Anonymous Referee #1

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 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 is very little information on chlorine chemistry (except in Fig.6, where there is no distinction between HCl and ClONO<sub>2</sub> 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 500 K and ozone loss in the middle stratosphere (NO<sub>x</sub>-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 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 HO<sub>x</sub> 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 ClONO<sub>2</sub> 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 were the initial values for HCl, ClONO<sub>2</sub>, N<sub>2</sub>O<sub>5</sub> 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 N<sub>2</sub>O?).

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).

We thank the Referee #1 for the thorough and valuable comments. We think that the issues raised here in the introduction has been addressed in the detailed comments below, except for the discussion related to the temporal evolution of stratospheric chlorine. A discussion was added to the Introduction in the manuscript and a reference to Engel et al. (2018) was added. In addition, the references were checked and corrected and new references were added.

The discussion of the ozone loss results was changed in order to more clearly discriminate the loss caused by NO<sub>x</sub> from the loss related to halogens.

The PSC volume was changed to PSC area, which didn't change the results and conclusions, but the text was altered and some of the figures were replotted.

The definition of the start and end of the polar vortex season was defined in more detail, which changed the appearance of some of the figures, but did not change the results or conclusions.

The validation of the FinROSE model has been done earlier, therefore we didn't include validation in this study, e.g. in Karpechko et al. (2013) we have compared the full chemistry simulation to satellite observations. The ERA-Interim ozone have been validated by e.g. Dragani (2011).

## Details

- title: the title is too general, it sounds like the uncertainty in simulated Arctic ozone is due to water vapour. But there are more reasons for uncertainties

We changed the title, it now reads:

"Linking the Uncertainty of Simulated Arctic Ozone Losses to Modelling Uncertainties in the Tropical Stratospheric Water Vapour "

- 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.

The change in water vapour in the tropical tropopause region was seen nearly as a one-to-one correspondence in the Arctic polar vortex. We highlighted this finding in the text.

- abstract, l 13-15: I think this is an important finding of this paper. We further streamlined the text to highlight this finding.

"If the cold conditions persist long enough (e.g. as in 2010/11), the chlorine activation is nearly complete. In this case addition of water vapour to the stratosphere does not increase the chlorine activation and ozone destruction significantly."

- 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. We made some changes to the changed the Abstract, related also to the previous comment:

"We found that the impact of water vapour changes on ozone loss in the Arctic polar vortex depends on the meteorological conditions. The strongest effect was in intermediately cold conditions, such as 2013/14, when added water vapour resulted in 2-7% more ozone loss due to the additional polar stratospheric clouds (PSC) and associated chlorine activation on their surface, leading to ozone loss. The effect was less pronounced in cold winters such as 2010/11 because cold conditions persisted long enough for a nearly complete chlorine activation even in simulations with observed water vapour. In this case addition of water vapour to the stratosphere led to increased area of ice PSC but it could not increase the chlorine activation and ozone destruction significantly. In the warm winter 2012/13 the impact of water vapour concentration on ozone loss was small, because the ozone loss was mainly NO<sub>x</sub> induced."

- 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. We specified the text and added the reference:

"A warmer climate in the troposphere is suggested to increase stratospheric water vapour (SWV) through increases in the water vapour entering through the tropopause, which would further warm the troposphere below (Dessler et al., 2013). However tropospheric warming may also lead to a significant cooling near the tropopause in connection with deep convection (Kim et al., 2018), so that the link between warming climate and tropopause temperature is not straightforward."

- p.2, l. 3: "affect chlorine partitioning" but how? Is this relevant for polar chemistry? We removed some of the discussion that was not relevant to the polar chemistry.

- p.2, l. 7: this citation is for the upper stratosphere; I do not think it is appropriate here. The citations were checked and changed (Solomon 1999 and Khosrawi 2016)

p.2, l. 12: One could also mention the observations discussed by Kuttippurath and Nair (2017) here. The reference and discussion was added.

- p. 2, l. 14: do you have references to more recent studies to back up this statement on recovery? We added reference to a recent study of Dhomse 2018, and changed the recovery dates according to their results. Also references to WMO 2014, and Morgernstren 2017 were added.

- p. 2, l. 17: remember that the focus here is on Arctic ozone. References to Smalley 2017 and Rosenlof 2001 were removed from here.

• 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). If you do not agree, please specify “some”.

We think the text is correct in general, but it is true that the offset would probably come from slower ozone loss reactions in areas outside the vortex, which could contribute to a so called super recovery of the ozone layer. We changed the text accordingly.

• 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 NO<sub>x</sub> chemistry indeed is relevant (but we are missing a discussion here of the impact – if any – of water vapour on the NO<sub>x</sub> chemistry in question). In any event (see also other points in this review) these two cases must be clearly separated. You are looking at different processes here.

We modified the text to more clearly separate the discussion of the NO<sub>x</sub> and chlorine caused ozone loss. We were not able to attribute any change in NO<sub>x</sub> chemistry to changes in water vapour.

• p. 3, l 28: the idea of a “controlled” experiment is good! also change to “impact of . . . on ozone loss”.

Sentence was changed to:

"Therefore a more controlled experiment is needed in order to assess impact of these SWV changes on ozone loss."

• p. 4., l 9: this is not an important point, but it has recently been argued that the reaction CH<sub>3</sub>O<sub>2</sub> + ClO is important for polar ozone loss (Müller et al., 2018); is this reaction taken into account in the chemical scheme used here?

CH<sub>3</sub>O<sub>2</sub> is not included in our chemistry scheme, and therefore we are unfortunately not able to discuss the importance of the CH<sub>3</sub>O<sub>2</sub> + ClO reaction in the Arctic polar vortex.

• 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  $\gamma$ ) is determined.

The following text was added to the description of the chemistry scheme.

"The reaction rates on NAT and ICE PSCs are not directly affected by the water vapour concentration except through the available surface area, i.e. the uptake coefficients are constant. In the case of binary aerosols and STS PSCs the uptake coefficients of some reactions depend on the composition of the droplets, i.e. the hydrolysis reactions of ClONO<sub>2</sub>, BrONO<sub>2</sub> and N<sub>2</sub>O<sub>5</sub>, as well as the reaction of HCl between ClONO<sub>2</sub> and HOCl (Sander et al., 2011)."

• 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.

We changed the text and added references to Stamnes (1988) and Dahlback and Stamnes, (1991) where the pseudo-spherical approximation is discussed.

"Look-up-tables of photodissociation coefficients were pre-calculated using the PHODIS model (Kylling et al., 1995). Within PHODIS the radiative transfer equation is solved by the discrete ordinate algorithm (Stamnes et al., 1988). This algorithm has been modified to account for the spherical shape of the atmosphere using the pseudo-spherical approximation (Dahlback and Stamnes, 1991)."

• p. 4, l. 29: the reference to Karpetchko et al. (2013) is important here as this is the only citation given 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.

Please note that Karpechko et al have both simulations, with and without heterogeneous chemistry.

- 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?

Please note that the variability is not preserved across CCMs either and our approach was chosen to replicate simulations by CCM. In our approach we replicate changes in amplitude as well as shift in mean water vapour. The description was improved.

"The simulations differed from each other by the prescribed water vapour concentration in the tropical tropopause region (stratosphere between 21S–21N, below 80 hPa), where it was prescribed as follows: (1) water vapour taken from ERA-Interim (Interim simulation), (2) increased water vapour (Max simulation), and (3) decreased water vapour (Min simulation). Specifically, the SWV lower boundary conditions for Min and Max simulations were obtained by multiplying values from ERA-Interim between tropopause and 80 hPa, and between 21 S–21 N by monthly coefficients ranging between 1.46-1.7 (Max) and between 0.5-0.63 (min), so that they approximately correspond to the driest and wettest CCM, as determined by SWV values at the tropical tropopause, across models analyzed by Gettleman et al. (2010)."

- 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).

Please note that validation of FinROSE-ctm have been done earlier (Damski et al 2007, and Thölix et al. 2016) and therefore it was not included in this paper. In the revised version we provide references to our earlier studies where such validation was done.

- p. 6, l 7: which reference theta level was used for mPV? How is 36 PVU chosen?

The reference theta level is 475K. The limit 36 PVU was chosen based on publications by Rex et al. (1999) and Streibel et al. (2006), who have shown that 36 PVU is a good approximation for the vortex edge. The text was modified as follows:

"The polar vortex was identified using the modified potential vorticity (mPV) (Lait, 1994), with the 475 K potential temperature as reference level. Here the polar vortex is defined as the area enclosed by the 36 PVU isoline separately for every model level. The 36 PVU contour approximately correspond to the region of maximum PV gradient, i.e the polar vortex edge (Rex et al, 1999, Streibel et al, 2006)."

- 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 N<sub>2</sub>O with observations would be helpful here.

Please note that validation of FinROSE-ctm have been done earlier (Damski et al 2007, and Thölix et al. 2016) and therefore it was not included in this paper. In the revised version we provide references to our earlier studies where such validation was done.

- 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.

The dehydration is calculated based on the settling velocity of ICE PSC particles. In the Arctic the dehydration is typically never complete, at least not on large areas. The not enough time for all

particles to sediment out of a layer at a given grid point during the time the ICE PSCs persist, and the area cold enough for formation of ICE PSCs is typically small compared to the Arctic polar vortex. Therefore the differences in water vapour concentration persists also after dehydration has occurred. The following text was added to the Modelling and data section:

"The sedimentation of PSC particles, which can lead to denitrification and dehydration, is calculated based on the settling velocity, which takes into account the PSC particle size."

- 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.

Yes, the volume of the grid points containing either NAT or STS (or both) were summed. To better separate the impact of water vapour on the heterogeneous chemistry we now separated the NAT and STS volume. We now also use area instead of volume. The text was altered accordingly.

- p. 7, l. 22: is there some impact of sedimentation on the duration of ice PSCs?

Yes, the dehydration deepens with time. The longer the ICE PSCs persist the more of the particles have time to settle out of a given layer. See comment above related to dehydration.

- 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.

References to Spang et al., (2018) and Pitts et al., 2018 were added. The PSC volumes have changed to PSC areas at 20 km level, because of easier comparison to observations. In general the timing of PSC in our model is good, but our ICE area is somewhat overestimated compared to Calipso data. The text was modified according the new figures.

- p. 8, l 4: "too dry models" this point sounds very speculative

The sentence "For example too dry models may not be able to simulate a large Arctic ozone loss such as of 2010/11." was removed from the results section and the discussion was moved to the Discussion and conclusions section.

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.

The polar vortex is calculated using ERA-Interim data, and not altered by the model. The vortex limit has been discussed in the reply to question of p. 6, l 7.

- p 9, l 10: small twice

Corrected.

- 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).

It is true that the activation starts on binary aerosols. However, there is no hard limit between binary aerosols and PSCs. In FinROSE the switchover from binary aerosols to STS is done at 215 K. At that temperature the STS is practically a binary aerosol, the large influence from dissolved water and HNO<sub>3</sub> happens at even lower temperatures. The humidity is taken into account through the composition and uptake coefficient, as described in a previous comment (p. 4, l. 16/17). The text was modified as follows:

"In the cold conditions within the polar vortex the chlorine species are transformed, through heterogeneous reactions, into intermediate species such as Cl<sub>2</sub>."

- p 9, l 25: which reservoir species?

In early March mostly ClONO<sub>2</sub> is formed and a few weeks later HCl. This was added to the text.

- p 10, top: the start of activation is one thing, but not really what determines how much ozone loss happens in a particular winter.

The purpose here is only to describe the evolution of active chlorine for different winters.

- p 10, l 12: which process is responsible here?

The reason is probably heterogeneous reactions that occur in binary aerosols and STS, the added water vapour increases the uptake coefficients and to some extent also the surface area. The text was modified as follows:

"The water vapour concentration seem to strongly affect the transformation of chlorine from the reservoir species to the intermediate ones in the beginning of Arctic winter. The fractions of intermediate and reservoir chlorine species change significantly with water vapour concentration in November and December, when STS PSCs start to form. The water vapour concentration affects the composition of binary aerosols and STS. When more water condenses to the particles the uptake coefficients for the heterogeneous reactions increase, i.e. the reaction rate increases."

- p 10., l 23: but why?

During the coldest period the difference in chlorine activation is small probably due to the ICE PSCs that appear in all simulations. The conditions were favourable for high chlorine activation in all simulations and therefore the differences were small. A clarification was added to the text.

- p 10., l 26: I am not convinced that this statement is correct, is this really a cause and effect relation?

We agree with the referee in the sense that the statement "The amount of chlorine activation correlate with the volume of NAT/STS PSCs." is probably too broad. The correlation is more complex, we altered the text to reflect this. We now deal with the STS and NAT separately, as suggested earlier.

- 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).

We added monthly sums to the table, and now the timing of the chlorine activation in different winters can be seen from the table. The text modified according the new table values.

- p 11, sec 3.4: it would helpful to have more comparisons to ozone loss from simulations of other models.

We were not able to find additional references than Vogel (2011), Sinnhuber (2011) ja Pommereau et al. (2013), related to the effect of water vapour on Arctic ozone depletion.

- p 11, l 18: is this statement on NO<sub>x</sub> also true for FinROSE? If yes, what is the evidence from the model simulation for this statement?

The text was rearranged, and the NO<sub>x</sub>-part was moved to the discussion of the ozone profile figure (Fig 8). It can be seen from the no-hetero run that the ozone loss in winter 2012/13 is gas phase chemistry driven. The text now reads:

"In winters when the polar vortex is unstable and small or disturbed the Brewer–Dobson circulation brings more NO<sub>x</sub>-rich air to the polar vortex than usual. Hence the ozone loss in the 2012/13 winter was produced mostly by NO<sub>x</sub> chemistry as shown previously by e.g., Sagi et al. (2017), and can be seen from FinROSE results by comparing the simulations with and without the heterogeneous chemistry."

- 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?

We did an additional experiment and it confirmed our assumption. The ozone loss in 2010/11 with higher resolution (1.5 x 3) were about 10 DU larger than with FinROSE's resolution. Unfortunately, redoing all experiments with increased resolution are currently not feasible for technical reasons, therefore we report in the manuscript the results based on the original resolution.

- p 12., l 7: “without het. chemistry”: it should be clearly stated what has been assumed regarding the heterogeneous reaction  $\text{N}_2\text{O}_5 + \text{H}_2\text{O}$ , which is not temperature dependent but would be important here.

No separate assumptions have been made for the  $\text{N}_2\text{O}_5 + \text{H}_2\text{O}$  reaction. There were two different setups used, one where no heterogeneous reactions were included, and another one where the temperature for the aerosol/PSC scheme was limited to 200K. The one with the 200K limit allows some heterogeneous processing on binary aerosols and some STS that are very dilute in  $\text{HNO}_3$ . The  $\text{N}_2\text{O}_5 + \text{H}_2\text{O}$  reaction is practically not temperature dependent, but the rate will be affected by the smaller surface area available. The possible effect on e.g.  $\text{NO}_x$  induced loss was not considered here. We altered the description of the setup for the no-hetero simulations.

- p 12., l 13: this is not true for the het. reaction  $\text{N}_2\text{O}_5 + \text{H}_2\text{O}$

The idea to limit the temperature to 200K was to allow reactions on binary aerosols, but not on PSCs. It is true that a small amount of STS can form, but due to the temperature limit the surface area densities will remain quite small.

- 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.

We believe that the result shows that the binary aerosols and STS have an effect. There are studies that suggest that binary aerosols are more important for chlorine activation than PSCs (e.g. Drdla and Müller, 2012; Wohltmann et al., 2013; Kirner et al., 2015). However, the contribution of PSCs are also important (e.g. Wegner et al., 2016). We added discussion on these results.

- 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.

We did consider this suggestion, but decided not to change the structure.

- 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.

The main finding here was that if the winter is cold, the PSCs may form even at low water vapour concentrations, therefore the water vapour increase is less important.

- 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?



The range can at least partly be explained by differences in the PSCs, i.e. temperature. We added discussion of the early chlorine partitioning.

• 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.

We believe that in cold winters a well developed vortex can last until mid-April and, in extreme cases such as 1990 and 1997, potentially even until early May. See for example study by Waugh et al. (1999). In the revised version we consider vortex to be established when its area (defined by 36 PVU contour) exceeds that corresponding to 80° equivalent latitude, following Waugh et al. (1999). This new definition results in only minor changes with respect to our original version. The size of the vortex can be seen in Fig 1.

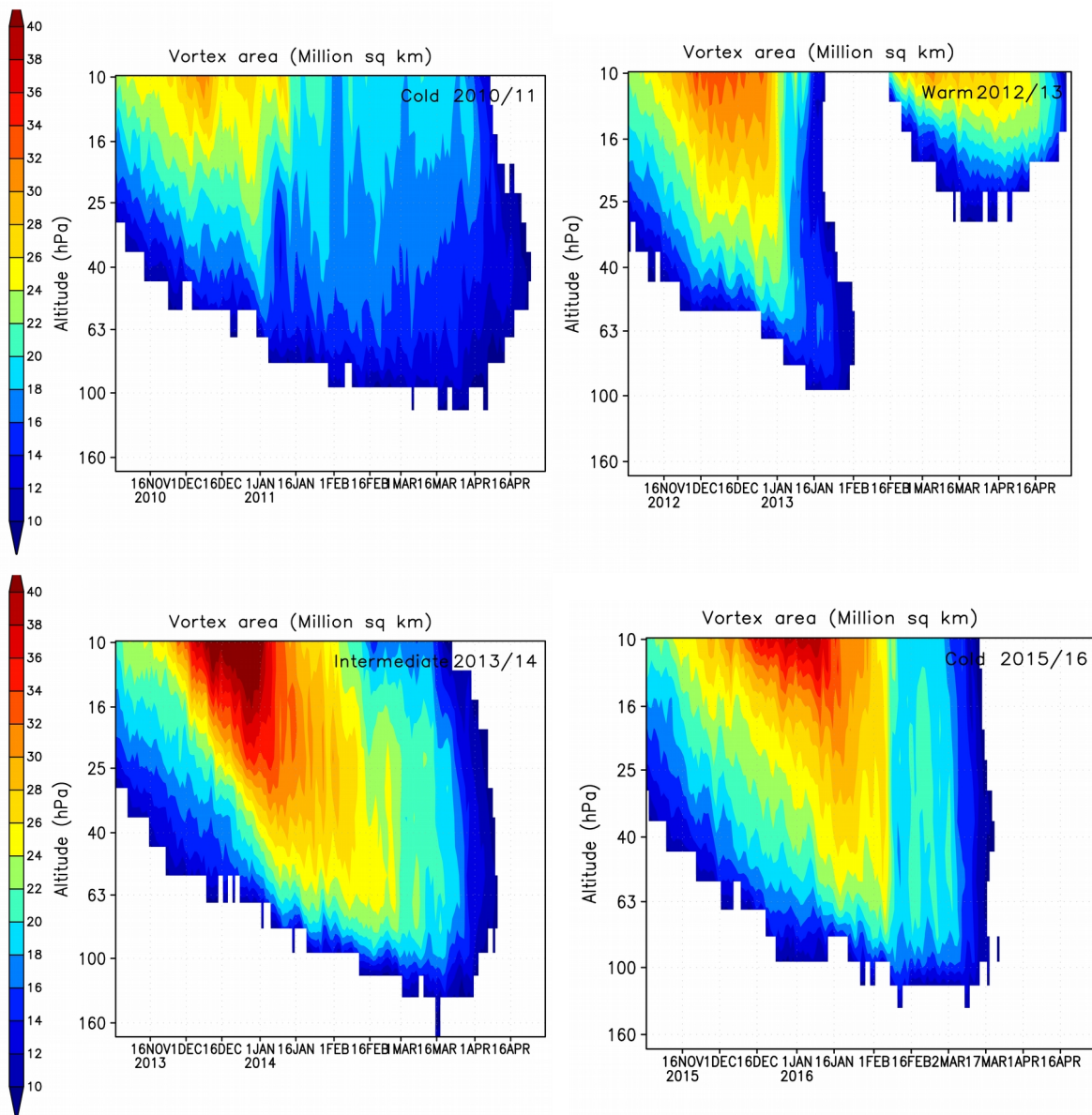


Figure 1. Vortex area (Milion sq meters).

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## General

The authors investigate the sensitivity of modelled Arctic ozone loss to the water vapour mixing ratio entering the tropical stratosphere in a chemical transport model. They guide the reader well step by step through the causal chain water vapour concentration → PSC volume → chlorine activation → ozone loss. The authors clearly state that the investigated question is different from investigating the effect of water vapour changes due to climate change (which would occur on such a timescale that also the concentration of chlorine- and bromine-containing species changes considerably). They also clearly state that they investigate only one aspect of the above-mentioned question, namely the effect of water vapour on the surface area density of Polar Stratospheric Clouds (PSCs), neglecting the (probably stronger) effect of water vapour changes on ozone loss via changes of stratospheric temperature.

## Comments

- An increase of water vapour may enhance heterogeneous chemistry by enlarging the air volume in which PSCs are formed (shown in Figs. 4 and 5) or via enlarging the surface area of existing particles (not shown). The authors seem to assume that the first effect is the dominant one. A discussion of this topic would help to complete the logic of the paper.

It is true that there also the available surface area and the composition of binary aerosols and PSCs can affect the heterogeneous chemistry, in addition to just the existence of PSCs. We therefore

1) improved the description of the chemistry scheme

“The reaction rates on NAT and ICE PSCs are not directly affected by the water vapour concentration except through the available surface area, i.e. the uptake coefficients are constant. In the case of binary aerosols and STS PSCs the uptake coefficients of some reactions depend on the composition of the droplets, i.e. the hydrolysis reactions of ClONO<sub>2</sub>, BrONO<sub>2</sub> and N<sub>2</sub>O<sub>5</sub>, as well as the reaction of HCl between ClONO<sub>2</sub> and HOCl (Sander et al., 2011).”

2) We added some discussion on the effect of water vapour on heterogeneous binary in/on aerosols and STS.

“The water vapour concentration seem to strongly affect the transformation of chlorine from the reservoir species to the intermediate ones in the beginning of Arctic winter. The fractions of intermediate and reservoir chlorine species change significantly with water vapour concentration in November and December, when STS PSCs start to form. The water vapour concentration affects the composition of binary aerosols and STS. When more water condenses to the particles the uptake coefficients for the heterogeneous reactions increase, i.e. the reaction rate increases.”

- The authors claim an important role of NO<sub>x</sub> chemistry in warm winters. I would appreciate plots showing this, e.g., altitude-time plots of vortex-averaged NO<sub>x</sub> or / and altitude-time plots of the corresponding reaction rates (NO<sub>2</sub> + O → ... for NO<sub>x</sub> chemistry, and, for comparison, Cl<sub>2</sub>O<sub>2</sub> + hv → ... and perhaps ClO + O → ... for ClO<sub>x</sub> chemistry).

From the difference between the no-hetero and the full chemistry simulations it can be seen that the ozone loss in winter 2012/13 is gas phase chemistry driven. The text was rearranged, and the NO<sub>x</sub>-part was moved to the discussion of the ozone profile figure (Fig 8). The text now reads:

"In winters when the polar vortex is unstable and small or disturbed the Brewer–Dobson circulation brings more NO<sub>x</sub>-rich air to the polar vortex than usual. Hence the ozone loss in the 2012/13 winter was produced mostly by NO<sub>x</sub> chemistry as shown previously by e.g., Sagi et al. (2017), and

can be seen from FinROSE results by comparing the simulations with and without the heterogeneous chemistry."

- The authors do not discuss the influence of heterogeneous NO<sub>x</sub> chemistry on ozone. Can the model results be used to answer the question whether the heterogeneous reaction  $\text{N}_2\text{O}_5 + \text{H}_2\text{O} \rightarrow 2\text{HNO}_3$  reduces NO<sub>x</sub> and thus NO<sub>x</sub>-driven ozone loss in cold years (compared to other years)? We were not able to attribute any change in NO<sub>x</sub> chemistry to changes in water vapour, but we modified the text to more clearly separate the discussion of the NO<sub>x</sub> and chlorine caused ozone loss.

#### Minor comments

- 1/17: "2-7% more ozone loss than in colder winters" ⇒ Does this mean "2-7% stronger increase of ozone loss than in colder winters"?

Yes. We changed the text in the abstract:

"We found that the impact of water vapour changes on ozone loss in the Arctic polar vortex depends on the meteorological conditions. The strongest effect was in intermediately cold conditions, such as 2013/14, when added water vapour resulted in 2-7% more ozone loss due to the additional polar stratospheric clouds (PSC) and associated chlorine activation on their surface, leading to ozone loss. The effect was less pronounced in cold winters such as 2010/11 because cold conditions persisted long enough for a nearly complete chlorine activation even in simulations with observed water vapour. In this case addition of water vapour to the stratosphere led to increased volume of ice PSC but it could not increase the chlorine activation and ozone destruction significantly. In the warm winter 2012/13 the impact of water vapour concentration on ozone loss was small, because the ozone loss was mainly NO<sub>x</sub> induced."

- 2/1 and 14/3: "warms the climate" means "warms the troposphere"?

We changed the text as suggested.

- 3/2-3: "cooling stratosphere ... slowing down some gas-phase reactions": Which gas-phase reactions are meant? In the polar stratosphere (during winter / spring) an important reaction is the three-body reaction  $\text{ClO} + \text{ClO} + \text{M} \rightarrow \text{Cl}_2\text{O}_2$ , the rate-constant of which increases for decreasing temperature.

It is true that the effect of temperature on future ozone is complex and our statement was too general. We meant the second-order reactions mainly in NO<sub>x</sub> and HO<sub>x</sub> ozone loss cycles. This effect is mainly seen outside the polar vortex, while the effect on PSCs from temperature is seen within the vortex. We improved the text to clarify this.

- 5/3: "around 80 hPa": In view of the discussion in 5/29-31 this should be formulated more precisely, e.g. "at the cold point, which lies approximately x hPa below 80 hPa"

We added some details to the description of the lower boundary condition for water vapour.

"The simulations differed from each other by the prescribed water vapour concentration in the tropical tropopause region (stratosphere between 21S–21N, below 80 hPa), where it was prescribed as follows: (1) water vapour taken from ERA-Interim (Interim simulation), (2) increased water vapour (Max simulation), and (3) decreased water vapour (Min simulation). Specifically, the SWV lower boundary conditions for Min and Max simulations were obtained by multiplying values from ERA-Interim between tropopause and 80 hPa, and between 21 S–21 N by monthly coefficients ranging between 1.46-1.7 (Max) and between 0.5-0.63 (min), so that they approximately correspond to the driest and wettest CCM, as determined by SWV values at the tropical tropopause, across models analyzed by Gettleman et al. (2010)."

- 5/29-31: “leads ... by 3-4 weeks ... Brewer-Dobson circulation ... too fast”: Does this mean that between the cold point and 80 hPa the ERA-Interim circulation takes 3-4 weeks less than the real circulation. Is the distance between the cold point and 80 hPa large enough to gain such a difference?

Thank you for this comment. We believe that a too fast BD transport in ERA-I likely contributes to the difference but we are not sure if it can indeed explain such a large delay. Therefore we rewrite the text as follows:

"However, Interim variability leads that of MLS by 3–4 weeks. The reason for the time lag between Interim and MLS is not clear although it could at least partly be associated with a too fast Brewer–Dobson circulation in ERA-Interim which is responsible for upward transport of the water vapour anomalies in the tropics (Schoeberl et al., 2012)."

- 6/3: “gains a small amount of water”: How, by horizontal mixing?

In the revised version this claim is removed and the sentence is rewritten. In the original version we unfortunately overlooked the fact that in Min simulation the scaling factor was varying with a seasonal cycle between 0.5-0.63, rather than fixed at value 0.5 as was stated in the original text. This point is clarified in the revised version. Therefore the difference between Min and Interim of factor 0.6 in Fig. 1 is consistent with prescribed values and there is no evidence of gained water vapour in Min simulation. The new text reads:

“The Max simulation has 2–3 ppm more water vapour in the tropics than the Interim simulation, while the Min simulation is about 1.5 ppm drier than the Interim simulation. These differences correspond to the ratio between Max/Interim of approximately 1.55-1.6 and about 0.55–0.6 between Min/Interim, i.e. they are consistent with the prescribed boundary conditions.”

- 8/4: “For example too dry models may not be able to simulate a large Arctic ozone loss such as of 2010/11”: How does this sentence relate to the preceding sentence?

The sentence “For example too dry models may not be able to simulate a large Arctic ozone loss such as of 2010/11.” was removed from the results section and the discussion was moved to the Discussion and conclusions section.

- 9/15: “in spring”: and also during southward excursions of air masses during winter

Yes, it is more related to availability of sunlight than the time of the year, the text was changed accordingly:

"When sunlight reaches the polar vortex these species are easily dissociated to form active chlorine species that participate in the catalytic ozone depletion cycles, i.e. ClO<sub>x</sub> (ClO, Cl<sub>2</sub>O<sub>2</sub> and Cl)."

- 9/16: “PSCs sustain the regeneration of ClO<sub>x</sub>”: This is only possible if both reaction partners for a heterogeneous reaction are still present.

The reservoir species are continuously formed through the ClO+NO<sub>2</sub> and Cl+CH<sub>4</sub> reactions, but as long as there are PSCs the ClO<sub>x</sub> is regenerated. We added some text to clarify this part.

“Active chlorine goes back to reservoir species through reactions with NO<sub>2</sub> and CH<sub>4</sub>, however if PSCs are present the regeneration of ClO<sub>x</sub> is sustained.”

- 10/9: “rather short”: The green curve in Fig. 6d lies above 60% for about 2 months. Is this meant by “rather short”?

We meant in comparison to the winter 2010/11, the text was corrected:

“The 2015/16 winter started similar to the cold winter 2010/11 and nearly all of the chlorine was activated at the beginning of January, but the deactivation started already in the end of January making the period with high ClO<sub>x</sub> shorter than in winter 2010/11.”

- 10/17: “differs significantly ... during the period with high ClO<sub>x</sub>”: The difference is mostly less than 10%. Is this meant by “differs significantly”?

The formulation was quite vague, we altered this and the next sentence as follows:

“The difference in concentration of active chlorine and reservoir species between the Min and Max simulations are smallest during the cold periods, due to the effective processing on the PSCs (Fig 6.). The cold winter 2015/16 shows a very small range, and the intermediately cold winter 2013/14 a wider range in concentrations. The water vapour content has less effect on the chlorine partitioning in cold winters.”

- 10/18: What exactly is meant by “chlorine activation period”: the time when most chlorine exists in active form?

The chlorine activation period was not defined exactly, it was based on a subjective interpretation of the figures. We made an effort to define it more robustly and altered the text accordingly.

- 11/4: “... only at the 475 K level”: Does this mean that the 475 K level is used for the definition of the whole vortex? If so, this should be mentioned (and perhaps be discussed) already in 6/7.

We used the PV on the 475 K level to define the polar vortex only in Fig 7. The text was modified as follows:

“In Fig. 7 the polar vortex is defined using the potential vorticity limit 36 PVU only at the 475 K level.”

- 11/11: “FinROSE seems to underestimate the ozone loss, possibly due to a general 10% negative bias in total ozone”: Why does an underestimation of the ozone concentration lead to a significant underestimation of the ozone loss? The rate of ozone loss in polar winter/spring is largely determined by the rates of reactions like ClO + ClO + M → Cl<sub>2</sub>O<sub>2</sub> + M and BrO + ClO → ... . In extreme cases a negative bias could limit the ozone loss in DU, however for Arctic conditions this is generally not the case. We don't have a clear understanding of the reason for the underestimation, and this statement is not valid here. We changed the text accordingly.

- 11/23: “stopping the catalytic ozone cycles and ozone loss early”: The ozone loss stops around the beginning of March (Fig. 7d). Is this meant by “early”?

We meant earlier than 2011. The text has changed:

"A relatively small ozone loss of 56 DU was simulated in 2015/16, which was due to the unstable polar vortex, which split and warmed, stopping the catalytic ozone cycles and ozone loss in the beginning of March, i.e. earlier than in 2010/11 and 2013/14."

- 13/4: “the heterogeneous chemistry destroyed about 36 DU of ozone”: In fact, ozone is destroyed by gas-phase reactions. Heterogeneous chemistry produces (some of) the corresponding reactants. Yes, the formulation was misleading, the text now reads:

“In 2013/14 the heterogeneous chemistry caused about 40 DU ozone destruction, which is about 51 % of the ozone loss and ICE and NAT/STS about 23 DU (30 %), when the total ozone loss was 79 DU in the Interim run.

- 14/24: “in higher level”: Does this mean at higher altitudes?



No, we meant more complete. The text has changed:

"Cold winters differ from the warm winters regarding the ozone loss and the fraction of ozone loss initiated by heterogeneous chemistry, during cold winters the PSC volumes are larger and thus chlorine activation within the polar vortex is more complete."

- Tables 1, 2, 3: As a "service" for the reader the character of the years (warm, intermediate, cold) might be added (as was done in the figures). Perhaps the winter 2015/16 might be called "initially cold", in order to distinguish it from 2010/11.

The character of the years was added to the tables as well.

### Technical details

- Please check the "s" of the plural of substantives or singular of verbs  
1/13, 2/3, 10/12, 10/16-17, 14/4, 15/29

We made the suggested corrections to the text.

- 1/22: "processes. Especially" ⇒ "processes, especially"

OK

- 2/7: "(2002)" ⇒ ", 2002"

OK

- 4/9: "lagrangian" ⇒ "Lagrangian"

OK

- 6/27: The abbreviation "BD" has not been defined before (and is used only once).

OK

- 6/33: "(Fig. 3)" ⇒ Fig. 3

OK

- 7/18: Really "0.3 ppt" (or 0.3 ppb)?

The limit we used was 0.3 ppt, it seems low but at temperature above 200K only a small fraction of the nitric acid is dissolved in the aerosols. We had a closer look at the limit and we have now increased it to 0,6 ppt, which approximately corresponds to 202 K. We changed the text accordingly.

- Table 1: Please show only 2-3 non-zero digits.

OK

- Table 2: "mixing ratio integrated over the winter of activated chlorine" ⇒ "mixing ratio of activated chlorine integrated over the winter"

OK