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 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 500 K 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).

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 NOx 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, is 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 uncertainty in simulated Arctic ozone loss to uncertainties in modelled 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.

"In this study we investigate the sensitivity of simulated Arctic ozone loss to the amount of water vapour that enters the stratosphere through the tropical tropopause."

"The water vapour changes in the tropical tropopause led to about 1.5 ppm less and 2 ppm more water vapour in the Arctic polar vortex compared to the ERA-Interim, respectively. The change induced in the water vapour concentration in the tropical tropopause region was seen as a nearly one-to-one change in the Arctic polar vortex."

- abstract, l 13-15: I think this is an important finding of this paper.

We further streamlined the text to highlight this finding.

"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.

- 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 the 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 NOx 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.

"Several studies have discussed Antarctic ozone depletion and its recovery (see e.g. Eyring (2010), Dameris (2014), Solomon et al. (2016), Chipperfield et al. (2017), Kuttippurath and Nair (2017) and Strahan and Douglass (2018). "

"Kuttippurath and Nair (2017) recently showed that Antarctic ozone has begun to recover based on ozone balloon soundings and total ozone data from satellite instruments. Based on profile data from the Aura Microwave Limb Sounder (MLS) of HCl and ozone Strahan and Douglass (2018) showed a decline in lower stratosphere chlorine and a corresponding decline in ozone depletion for the period 2013-2016 compared to the period 2004–2007. However, a recovery of ozone to 1980 ozone levels is projected not to occur until around 2025–2043 in the Arctic and 2055–2066 in the Antarctic (Dhomse et al., 2018)."

- 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 Dameris 2014, Solomon et al. (2016), Chipperfield et al. (2017), Kuttippurath and Nair (2017) and Strahan and Douglass (2018) were added. See also previous comment.

- 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). I 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.

"However, cooling of the stratosphere could at least partially offset the effect of the increased PSCs by slowing down the second-order reactions in ozone loss cycles (Rosenfield et al., 2002 and Revell et al., 2012). This effect is mainly seen in NOx and HOx induced loss outside the polar vortex, while the effect on PSCs from temperature is seen within the vortex"

- 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.
We modified the text in the Introduction slightly. In addition we reorganised the discussion about NOx induced loss in the Results section to more clearly separate the discussion of the NOx and chlorine caused ozone loss. We were not able to attribute any change in NOx chemistry to changes in water vapour.

"In the winter 2012/13 the polar vortex was more unstable and a vortex split occurred early January due to a sudden stratospheric warming (SSW), and NOx rich air from the mesosphere descended to the upper stratosphere and led to ozone loss there. Thus the effect on Arctic ozone depletion from changes in SWV will depend on the meteorological conditions, and the dynamical stability in a given winter."

- 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 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?

CH3O2 is not included in our chemistry scheme, and therefore we are unfortunately not able to discuss the importance of the CH3O2 + 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 γ) 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 ClONO2, BrONO2 and N2O5, as well as the reaction of HCl between ClONO2 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 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.

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 (e.g., Damski et al 2007, Thölix et al., 2010, Salmi et al., 2011 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.

"The FinROSE-CTM has previously been used to study the impact of meteorological conditions on water vapour trends (Thölix et al., 2016), ozone and NO x chemistry in the mesosphere (Salmi et al., 2011), Arctic polar ozone loss (Karpechko et al., 2013) and the impact of the driver data on the model transport (Thölix et al., 2010). Long term trends of Arctic and Antarctic ozone losses, past and future, have been investigated by using driving data from a chemistry–climate model (Damski et al., 2007a). The model results showed good agreement with satellite and ground based observations. The FinROSE water vapour was compared to observations of water vapour profiles from the Microwave Limb Sounder (MLS) and frost point hygrometer soundings from Sodankylä Thölix et al. (2016). The extent of ICE PSCs simulated by FinROSE was compared to Cloud-Aerosol Lidar and Infrared Path finder Satellite Observation (CALIPSO) data in Thölix et al. (2016).The total ozone distribution was compared to data from Total Ozone Mapping Spectrometer (TOMS) and the Ozone Monitoring Instrument (OMI) satellite instruments in Damski et al. (2007a), Thölix et al. (2010) and Karpechko et al. (2013). Salmi et al. (2011) compared the NO x and ozone profiles in FinROSE to data from the tmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) instrument."

- 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 at 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 N2O with observations would be helpful here.

Please note that validation of FinROSE-ctm have been done earlier (e.g., Damski et al 2007, Thölix et al., 2010, Salmi et al., 2011 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. See also previous cmment.

- 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 area. 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_3 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 Cl2."

- p 9, l 25: which reservoir species?

In early March mostly ClONO2 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 an 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.

"The chlorine activation in 2015/16 winter seems to be less dependent on water vapour content, probably due to the ICE PSCs that appear in all simulations. Therefore, the conditions were favourable for high chlorine activation in all simulations. The difference in the fraction of activated chlorine between simulations is only few percents, only when the deactivation starts (in the end of February) the difference is more than 5 %. "

- 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.

"The timing of the changes in the partitioning of the chlorine species correlates well with the occurrence of PSCs, e.g. the chlorine reservoir species start to transform into intermediate species when the STS PSCs appear (Fig. 5)."

- 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 means of activated chlorine to the table, and now the timing of the chlorine activation in different winters can be seen from the table. The text was modified according the new table values.

"Table 2 shows the vortex averaged ClO x as a cumulative sum over the whole winter, and as monthly mean concentration. The sums are integrated from November to April. The cumulative sum has information about both the duration of the chlorine activation period and the concentration of ClO x , while the monthly average concentration shows the timing of chlorine activation. The cumulative chlorine activation was largest in winter 2010/11 and the smallest in 2012/13. The activation started in November every year, but remained small until December. The winter 2010/11 differs from the others, with high chlorine activation from January to March, giving the largest cumulative sum of the studied winters. Even in April the ClO x concentration remains elevated. The warm winter 2012/13 had the smallest cumulative chlorine activation, significant chlorine activation was seen only in December and January. The changes in water vapour between the Min/Interim/Max simulations had the largest effect on the cumulative ClOx in moderately cold winters (2010/11 and 2013/14), where the increase from Interim to Max was 3 to 6 % and change from Interim to Min was -1 to -11 %. In the cold winter 2015/16 the respective changes were only 3 and -2 %, and in the warm winter 2012/13 the changes were 4 and -1 %."

- 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) and Pommereau et al. (2013), related to the effect of water vapour on Arctic ozone depletion.

- 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?

The text was rearranged, and the NOx-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 NOx -rich air to the polar vortex than usual. Hence the ozone loss in the 2012/13 winter was produced mostly by NO x 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.

"An additional sensitivity experiment showed that the difference compared to other studies can be, at least partly, due to the coarse horizontal resolution in FinROSE (3x6), which is not sufficient to fully capture the deepest ozone loss. Specifically, repeating Interim simulation for winter 2010/11 with higher resolution (1.5x3) than in the original simulation showed larger ozone loss by 15 DU."

- p 12., l 7: “without het. chemistry”: it should be clearly stated what has been assumed regarding the heterogeneous reaction N2O5 + H2O, which is not temperature dependent but would be important here.

No separate assumptions have been made for the N2O5 + H2O 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 HNO3. The N2O5 + H2O reaction is practically not temperature dependent, but the rate will be affected by the smaller surface area available. The possible effect on e.g. NOx induced loss was not considered here. We altered the description of the setup for the no-hetero simulations.

"In the second simulation the formation of PSCs was limited by setting the air temperature passed to the heterogeneous chemistry module to 200 K, similarly to what was done in Karpechko (2013). This setting allows some heterogeneous processing on binary aerosols and some STS that are very dilute in HNO3, and due to the temperature limit the surface area densities will remain quite small."

- p 12., l 13: this is not true for the het. reaction N2O5 + H2O

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. See also previous comment.

- 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.

"In the Interim simulation with full chemistry in 2010/11 about 90 DU ozone was depleted, of which the heterogeneous chemistry caused 56 DU depletion, i.e. about 62 % of the total ozone loss. Heterogeneous chemistry due to PSCs destroyed 30 DU ozone, which was about 33 % of the total loss. The result indicates that chlorine activation on the binary aerosols has a significant role in ozone depletion. Some studies 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). The increase of water vapour (Max simulation) did not increase the ozone loss, but in the Min simulation there was 6 DU less ozone depletion."

- 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.

"If the winter is cold enough, the increase is less important, because the PSCs may form even at low water vapour concentrations, and the chlorine activation is already nearly complete in Arctic vortex, therefore the water vapour increase is less important. As expected, heterogeneous chemistry is more important in cold winters."

- 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 in early winter is probably due to the effect of water vapour on the composition of binary aerosols (and STS). A higher water vapour concentration gives a more dilute aerosol and larger surface area, which increase the uptake coefficient and reaction rate. We added discussion of the early chlorine partitioning.

"The water vapour concentration seems 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 during November and December. The water vapour concentration affects the composition of binary aerosols an STS. When more water condenses to the particles the uptake coefficients for the heterogeneous reactions and the surface area increase, i.e. the reaction rates increase."

- 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.

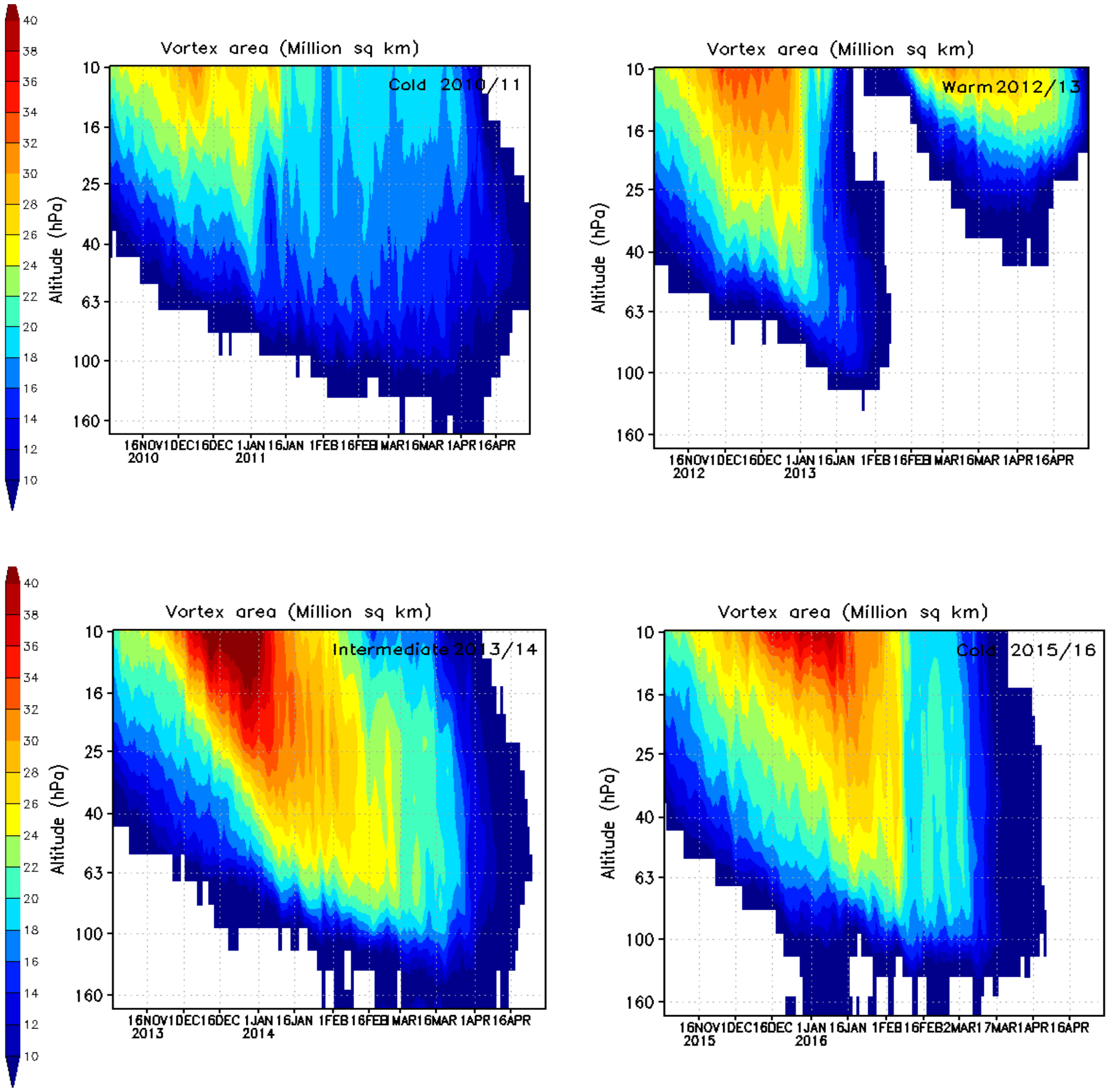

Figure 1. Vortex area (Million sq meters).

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Anonymous Referee #2

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), negelecting 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 $ClONO_2$, $BrONO_2$ and N_2O_5, as well as the reaction of HCl between $ClONO_2$ 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 an 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 NOx chemistry in warm winters. I would appreciate plots showing this, e.g., altitude-time plots of vortex-averaged NOx or / and altitude-time plots of the corresponding reaction rates ($NO_2 + O \rightarrow$ ... for NOx chemistry, and, for comparison, $Cl_2O_2 + h\nu \rightarrow$ ... and perhaps $ClO + O \rightarrow$ ... for ClOx 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 NOx-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 NOx -rich air to the polar vortex than usual. Hence the ozone loss in the 2012/13 winter was produced mostly by NO x 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 NOx chemistry on ozone. Can the model results be used to answer the question whether the heterogeneous reaction $N_2O_5 + H_2O \rightarrow 2HNO_3$ reduces NOx and thus NOx -driven ozone loss in cold years (compared to other years)?

We were not able to attribute any change in NOx chemistry to changes in water vapour, but we modified the text to more clearly separate the discussion of the NOx 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 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 NOx 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 $ClO + ClO + M \rightarrow Cl_2O_2$, 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 NOx and HOx 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.

"However, cooling of the stratosphere could at least partially offset the effect of the increased PSCs by slowing down the second-order reactions in ozone loss cycles (Rosenfield et al., 2002 and Revell et al., 2012). This effect is mainly seen in NOx and HOx induced loss outside the polar vortex, while the effect on PSCs from temperature is seen within the vortex."

- 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 x (ClO, Cl 2 O 2 and Cl)."

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

The reservoir species are continuously formed through the ClO+NO2 and Cl+CH4 reactions, but as long as there are PSCs the ClOx is regenerated. We added some text to clarify this part.

“Active chlorine goes back to reservoir species through reactions with NO2 and CH4, however if PSCs are present the regeneration of ClOx 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 ClOx shorter than in winter 2010/11.”

- 10/17: “differs significantly ... during the period with high $\mathrm{ClO_x}$ ": 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 intermediatly 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. It is difficult to find a robust definition and we therefore removed the phrase. See also previous comment.

"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.”

- 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 $\mathrm{ClO + ClO + M \rightarrow Cl_2O_2 + M}$ and $\mathrm{BrO + ClO \rightarrow}$ ... .

In extreme cases a negative bias could limit the ozone loss, 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.

"FinROSE seems to underestimate the ozone loss; for example Sinnhuber et al. (2011) and Manney et al. (2011) simulated 120 DU ozone loss and Pommereau et al. (2013) even 170 DU in winter 2010/11."

- 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 missleading, 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 temperatures 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 a formation threshold temperature of 202 K for STS. We changed the text accordingly.

"The area was calculated by summing the areas of model grid boxes containing PSCs. STS is assumed to exist in the gridbox if there is more than 0.6 ppt liquid HNO3."

- 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

Linking ~~the~~ uncertainty in simulated Arctic ozone ~~losses~~ loss to ~~modelling of~~ uncertainties in modelled tropical stratospheric water vapour

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Abstract. Stratospheric water vapour influences the chemical ozone loss in the polar stratosphere via controlling the polar stratospheric cloud formation. The amount of water entering the stratosphere through the tropical tropopause differs substantially between chemistry–climate models (CCM). This is because the present-day models, e.g. CCMs, have difficulties in capturing the whole complexity of processes that control the water transport across the tropopause. As a result there are large differences in the stratospheric water vapour between the models.

In this study we investigate the sensitivity of simulated Arctic ozone loss to the amount of water ~~, which~~ vapour that enters the stratosphere through the tropical tropopause. We used a chemical transport model, FinROSE-CTM, forced by ERA-Interim meteorology. The water vapour concentration in the tropical tropopause was varied between 0.5 and 1.6 times the concentration in ERA-Interim, which is similar to the range seen in chemistry climate models. The water vapour changes in the tropical tropopause led to about 1.5 ppm less and 2 ppm more water vapour in the Arctic polar vortex compared to the ERA-Interim, respectively. The change induced in the water vapour concentration in the tropical tropopause region was seen as a nearly one-to-one change in the Arctic polar vortex.

We found that the impact of water vapour changes on ozone loss in the Arctic polar vortex ~~depend~~ depends on the meteorological conditions. ~~Polar stratospheric clouds form in the cold conditionswithin the Arctic vortex, and~~ 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~~lead~~ , leading to ozone loss. ~~If the cold conditions persist long enough (e.g. in~~ The effect was less pronounced in cold winters such as 2010/11 ~~), the chlorine activation is nearly complete~~ 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 ~~increased the formation of ICE clouds, but did~~ 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_x induced. ~~In intermediately cold conditions, e.g. 2013/14, the effect of added water vapour was more prominent, and resulted in 2-7 % more ozone loss than in the colder winters.~~ The results show that the simulated water vapour concentration in the tropical tropopause has a significant impact on the Arctic ozone loss and ~~deserves attention~~ and therefore needs to be well simulated in order to improve future projections of ozone layer recovery.

1 Introduction

Water vapour in the stratosphere is a minor constituent with typical mixing ratios of 3–6 ppmv (e.g., Randel et al., 2004). It plays, however, an important role in radiative and chemical processes~~. Especially~~ , especially in the upper troposphere/lower stratosphere (UTLS) where changes in the water vapour concentration result in significant changes in radiative forcing of the ~~atmosphere~~ troposphere (Riese et al., 2012). A warmer climate in the troposphere ~~increases~~ is suggested to increase stratospheric water vapour (SWV) through increases in the water vapour entering through the tropopause, which ~~warms the climate further (Dessler et al., 2013).~~ 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. Photodissociation of water vapour is an important source of odd hydrogen HO_x ($H+OH+HO_2$). Catalytic cycles involving HO_x contribute to chemical ozone loss in the stratosphere (Dvortsov and Solomon, 2001). ~~Changes in affects the chlorine partitioning, which may lead to even more efficient ozone destruction (e.g., Dvortsov and Solomon, 2001).~~ Water vapour contributes to the formation of stratospheric aerosols including polar stratospheric clouds (PSCs), i.e. liquid and solid particles in combination with H_2SO_4 and HNO_3, or ice particles ~~. In addition, the water vapour concentration affects the composition of liquid PSCs, i.e. an increase in water vapour may increase the heterogeneous reaction rates (e.g. ?).~~ (e.g., Solomon, 1999; Khosrawi et al., 2016).

Heterogeneous reactions in or on PSC particles can lead to massive ozone depletion inside the polar vortices when atmospheric concentration of halogens is sufficiently high (Solomon et al., 1986; Wohltmann et al., 2013). Since the formation of PSCs requires very low temperatures (below about 195 K), significant polar ozone depletion takes place only occasionally in the Arctic (Rex et al., 2006; Manney et al., 2011; Müller et al., 2008; Chipperfield et al., 2015), while it ~~is~~ has been a yearly phenomenon in the Antarctic since about the mid 1980s ~~(Dameris et al., 2014)~~(e.g., Dameris et al., 2014). The stratospheric abundance of chlorine will remain elevated for decades, and polar ozone losses will therefore be seen also in the future. ~~A recovery to~~ Equivalent effective stratospheric chlorine (EESC) is a proxy that is frequently used to describe the combined effect of chlorine and bromine on stratospheric ozone. The level of EESC in 1980 ~~ozone levels will not occur until ca. 2025–2030~~ is commonly taken as a level that needs to be reached to achieve a recovery of stratospheric ozone. According to a recent study by Engel et al. (2018) the EESC would return to the 1980 level in 2077 for a mean age of 5.5 years, which is representative for polar winter conditions. Several studies have discussed Antarctic ozone depletion and its recovery (see e.g. Eyring et al. (2010); Dameris et al. (2014); Solomon et al. (2016); Chipperfield et al. (2017); Kuttippurath and Nair (2017); Strahan ar Kuttippurath and Nair (2017) recently showed that Antarctic ozone has begun to recover based on ozone balloon soundings and total ozone data from satellite instruments. Based on profile data from the Aura Microwave Limb Sounder (MLS) of HCl and ozone Strahan and Douglass (2018) showed a decline in lower stratosphere chlorine and a corresponding decline in ozone depletion for the period 2013–2016 compared to the period 2004–2007. However, a recovery of ozone to 1980 ozone levels is projected not to occur until around 2025–2043 in the Arctic and ~~2050–2070~~ 2055–2066 in the Antarctic ~~(Eyring et al., 2010)~~(Dhomse et al., 2018). Both colder air and increased SWV can increase the formation of PSCs, which could release more active chlorine and cause severe ozone depletion although future chlorine loadings will be smaller. All these suggest that SWV is a critical factor affecting ozone chemistry.

The majority of the previous studies addressing impacts of SWV on Arctic ozone depletion considered the effects of observed ~~(?) and projected (Eyring et al., 2007)~~ and projected increases in SWV concentrations ~~(Kirk-Davidoff et al., 1999; Dvortsov and Solomon,~~ For example Vogel et al. (2011) used a chemistry–transport model (CTM) and studied the effect of increased SWV on Arctic ozone loss for meteorological conditions from the cold Arctic winter 2004/05. They found that increasing SWV by 0.58 ppm, which is a typical amount simulated by chemistry climate models (CCMs) by the mid-21 century (Eyring et al., 2007), would lead to an additional 6 DU of ozone loss under cold winter conditions. Sinnhuber et al. (2011) used a CTM driven by meteorological conditions for the cold Arctic winter 2010/11 and assumed a uniform increase of SWV of 1 ppm. For such conditions they reported a 25 DU increase in ozone loss, i.e. about 20 % of their simulated total ozone loss for that winter.

Smalley et al. (2017) studied future trends in the tropical lower stratospheric water vapour and provided a regression model for analysing the factors driving the trends and variability in the 21st-century. They found that warming of the troposphere causes a long term increasing trend in the water vapour entering the stratosphere, which can be partially offset by an increase of the Brewer–Dobson circulation with accompanied cooling of the tropical tropopause. MacKenzie and Harwood (2004) studied the effect of increasing SWV due to future increase in tropospheric methane on ozone. They simulated the year 2060 under the Intergovernmental Panel on Climate Change Special Report on Emission Scenarios (SRES) B2 scenario, where CH_4 lies approximately midway between the extremes of the SRES scenarios. They found an increase in the occurrence of PSCs, with about 20 to 25 % of the increase due to increases in the water vapour. The rest is from radiative cooling of the middle atmosphere due to changes in the concentration of several trace gases. In the simulations by MacKenzie and Harwood (2004) the increased SWV due to projected methane increases caused a 15 % (about 0.5 ppm at 465 K level) deeper Arctic ozone loss in 2060. However, cooling of the stratosphere could at least partially offset the effect of the increased PSCs by slowing down ~~some gas-phase reactions involved in the catalytic~~ the second-order reactions in ozone loss cycles ~~(e.g., Revell et al., 2012).~~ (Rosenfield et al., 2002; Revell et al., 2012). This effect is mainly seen in NO_x and HO_x induced loss outside the polar vortex, while the effect on PSCs from temperature is seen within the vortex.

Also Revell et al. (2016) studied the effect of future methane changes on SWV under different RCP-scenarios. The contribution of methane to the SWV was found to be highly dependent on the projected methane concentration, altitude and latitude. Under RCP 6.0 between 1960 and 2100 the SWV was projected to increase by approximately 1 ppm throughout most of the stratosphere, excluding the Antarctic lower stratosphere. The largest increase was seen following the RCP 8.5, with 60 % additional water vapour in the extratropical upper stratosphere, and ca. 35 % in the Arctic lower stratosphere. The largest contribution from methane to the SWV change was about 50 % under RCP 8.5, which assumes a rather extreme methane increase scenario, and the smallest about 4 % under RCP 2.6.

Recently Sagi et al. (2017) studied Arctic ozone losses between years 2002 and 2013 using data assimilation of Odin/Sub-Millimetre Radiometer (SMR) atmospheric observations. They found that the largest ozone losses were caused either by halogens or by the NO_x-family, and the dominating process for ozone destruction is determined mostly by the temperatures inside the polar vortex. The very stable and cold polar vortex in the Arctic winter 2010/11 led to remarkable halogen driven ozone loss with 2.1 ppm ozone destroyed at the 450 K level. In the winter 2012/13 the polar vortex was more unstable and a vortex split occurred early January due to a sudden stratospheric warming (SSW)~~. Thus~~, and NO_x rich air from the mesosphere descended to the upper stratosphere and led to ozone loss there. ~~It is therefore likely that~~ Thus the effect on Arctic ozone depletion from changes in SWV ~~will~~ depend on the meteorological conditions, and the dynamical stability in a given winter.

The main source of SWV is the upward transport from the troposphere through the tropical tropopause in the upwelling branch of the Brewer–Dobson circulation. The concentration of SWV is controlled by the coldest temperature met by the ascending air parcels (i.e. cold point temperature). Gettleman et al. (2010) analysed 16 state-of-the-art CCMs and demonstrated large discrepancies between simulated SWV in these models, which were closely related to the simulated cold point temperatures. The "entry" value of SWV in these models ranged between 2 and 6 ppm, compared to the observed value of 3–4 ppm. These intermodel differences by far exceed the magnitude of the projected water vapour increases in the 21 century used so far in the studies of ozone loss sensitivities to SWV. One may wonder what are the implications of these discrepancies for stratospheric ozone losses simulated by the CCMs? This question is difficult to address by analyzing CCM outputs because there are other differences between the models which affect simulated ozone losses, such as differences in transport. Therefore a more controlled experiment is needed in order to assess ~~the impacts~~ impact of these SWV ~~differences for ozone losses~~changes on ozone loss.

In this study we address the question of what the implications of the differences in simulated tropical stratospheric water vapour between chemistry–climate models are for the simulated Arctic ozone loss. Similar to Vogel et al. (2011) and Sinnhuber et al. (2011) we address this question by performing CTM simulations using different SWV concentrations. The principal differences in our methodology from the previous studies are (1) the boundary conditions of perturbed water vapour experiments resulting in a different spatial pattern of SWV anomalies and (2) the magnitude of SWV perturbation, which is larger than in the Vogel et al. (2011) and Sinnhuber et al. (2011) studies, but in the range of Revell et al. (2016) and MacKenzie and Harwood (2004). We also analysed seven different winters, whose dynamical circumstances such as the as the evolution of the temperature and polar vortex were different (see Section 3 for more details).

2 Modelling and data

A global off-line chemistry–transport model for the middle-atmosphere, FinROSE-CTM, was used for simulating the effect of the SWV on Arctic ozone depletion. The FinROSE-CTM is described in detail in Damski et al. (2007b). In this study, the model has a horizontal resolution of 3° × 6° (latitude × longitude). It has 40 hybrid-sigma levels up to 0.1 hPa (about 65 km). The temperature, winds and surface pressure are from the European Centre for Medium-range Weather Forecasts (ECMWF) ERA-Interim reanalyses (Dee et al., 2011).

The model transport is computed using a flux-form semi-Lagrangian transport code (Lin and Rood, 1996). The chemistry scheme of the model comprises 36 species and includes about 150 reactions. In addition to gas-phase chemistry, the model includes a PSC scheme with liquid binary aerosols (LBA), supercooled ternary solution of sulfuric acid, nitric acid and water (STS, type Ib), solid nitric acid trihydrate (NAT, type Ia) and ice (ICE, type II) PSCs. The heterogeneous chemistry includes altogether 30 reactions and is based on the calculation of the composition and volume of sulphate aerosols and PSCs, as well as the partitioning of species between gas phase and condensed phase. The number density profile is prescribed for each PSC type (Damski et al., 2007b) and the sulphuric acid distribution is based on 2-D model data (Bekki and Pyle, 1992). Absorption cross-sections and rate coefficients follow the recommendations by Sander et al. (2011) and for some heterogeneous chemistry reactions the recommendations by Atkinson et al. (2007), see details in Damski et al. (2007b). ~~Photodissociation coefficients~~ 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 $\mathrm{ClONO_2}$, $\mathrm{BrONO_2}$ and $\mathrm{N_2O_5}$, as well as the reaction of HCl between $\mathrm{ClONO_2}$ and HOCl (Sander et al., 2011). 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.

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 ~~photolysis scheme of PHODIS radiative transfer model and are used through a look-up table (Kylling et al., 1995).~~ approximation (Dahlback et al., 1991).

The tropospheric concentrations of the chemical species are prescribed via model boundary conditions. The boundary conditions of water vapour and ozone are taken from the ECMWF ERA-Interim reanalysis (Dee et al., 2011) except for the water vapour boundary conditions in the sensitivity experiments which are described below. The concentration of tropospheric methane ($\mathrm{CH_4}$) is from Global view-data (http://www.esrl.noaa.gov/gmd/ccgg/globalview/ch4), nitrous oxide ($\mathrm{N_2O}$) concentration is from Agage data (Prinn et al., 2000), and halogens concentrations in the troposphere (Cly and Bry) are from Montzka et al. (1999) updated data. The carbon dioxide ($\mathrm{CO_2}$) concentration is based on global annual mean trend data (ftp://aftp.cmdl.noaa.gov/products/trends/co2). At the upper boundary ($0.1\,\mathrm{hPa}$) climatological values of water vapour and ozone averaged over 2005–2013 from MLS data were used.

The FinROSE-CTM has previously been used to study the impact of meteorological conditions on water vapour trends (Thölix et al., 2016), ozone ~~/~~and $\mathrm{NO_x}$ chemistry ~~(Salmi et al., 2011) and ozone chemical loss (Karpechko et al., 2013) , and the model results showed good agreement with observations. Also future ozone losses~~in the mesosphere (Salmi et al., 2011), Arctic polar ozone loss (Karpechko et al., 2013) and the impact of the driver data on the model transport (Thölix et al., 2010). Long term trends of Arctic and Antarctic ozone losses, past and future, have been investigated by using driving data from a chemistry–climate model (Damski et al., 2007a). The model results showed good agreement with satellite and ground based observations. The FinROSE water vapour was compared to observations of water vapour profiles from the Microwave Limb Sounder (MLS) and frost point hygrometer soundings from Sodankylä Thölix et al. (2016). The extent of ICE PSCs simulated by FinROSE was compared to Cloud-Aerosol Lidar and Infrared Path finder Satellite Observation (CALIPSO) data in Thölix et al. (2016).The total ozone distribution was compared to data from Total Ozone Mapping Spectrometer

(TOMS) and the Ozone Monitoring Instrument (OMI) satellite instruments in Damski et al. (2007a), Thölix et al. (2010) and Karpechko et al. (2013). Salmi et al. (2011) compared the $\mathrm{NO_x}$ and ozone profiles in FinROSE to data from the tmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) instrument.

For this study, three simulations covering the Arctic winters between 2009/2010 and 2015/2016 were performed. The simulations differed from each other by the prescribed water vapour concentration in the tropical tropopause region (stratosphere between 21° S–21° N, below 80 hPa), where it was prescribed as follows: (1) water vapour taken from ERA-Interim (Interim simulation), (2) ~~1.6× Interim (Max),~~ increased water vapour (Max simulation), and (3) ~~0.5× Interim (Min). The~~ decreased water vapour (Min simulation). Specifically, the SWV lower boundary conditions for Min and Max simulations were obtained by ~~scaling the reanalysis data in the tropical tropopause layer (TTL), around~~ multiplying values from ERA-Interim between tropopause and 80 hPa, and between 21° ~~S-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 ~~CCMVal-2 models~~CCM, as determined by SWV values at the tropical tropopause, across models analyzed by (Gettleman et al., 2010). This construction allows us to isolate the influence of the tropical water vapour on stratospheric chemistry while keeping all other factors fixed, and thus to estimate the contribution of processes controlling tropical water vapour entry values to Arctic ozone loss. Eight simulated years before 2009 are considered spinup and were not analysed. Ozone was initialised with ERA-Interim ozone in every year, in the beginning of December. The water vapour was not adjusted and allowed to evolve freely through the whole period of integrations. Ozone and water vapour observations from ~~the Microwave Limb Sounder (MLS) abroad Aura satellite~~ MLS (Lambert et al., 2007) were used to validate the reference simulation. MLS data is shown as 5 day averages because of the small amount of data covering the polar vortex in some cases.

3 Results

Model simulations were made for seven winters (2009–2016), but only four of them are discussed here. The four selected Arctic winters, 2010/11, 2012/13, 2013/14 and 2015/16 differ from each other with respect to the stratospheric temperatures and polar vortex strength. They provide examples of different role of SWV in ozone loss in mild (2012/13) cold (2010/11, 2015/16) and intermediate (2013/14) stratospheric winter conditions.

3.1 Temperature and water vapour

The boundary condition at the tropical tropopause for the reference simulation was evaluated by comparing simulated water vapour concentrations with observed ones from MLS. The top panels in Fig. 1 show daily mean water vapour at 80 hPa averaged between 21° S and 21° N for two representative years 2013 and 2014. The temperature for the same region is shown in the lower panels. The cold point, where SWV boundary conditions were prescribed, is just below the 80 hPa level. The temperature shows the typical annual cycle in the TTL with minimum in northern hemisphere (NH) winter and maximum in NH summer. The temperature in the TTL controls how much water vapour enters the stratosphere by freeze drying the upwelling air (e.g., Fueglistaler et al., 2005). As a result the maximum water vapour concentration occur in the NH autumn and minimum in early NH spring. The effect of interannual variability and shorter term variations in the temperature on stratospheric water vapour can also be seen, e.g. the low temperature in early 2013 results in 0.5–1 ppm less water vapour than during the same time in 2014.

The Interim simulation produces water vapour concentrations comparable to the amount seen by MLS (Fig. 1), which shows that the boundary condition is reasonable. However, Interim variability leads that of MLS by 3–4 weeks~~suggesting that the~~. 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 ~~could be too fast (?Schoeberl et al., 2012; ?)~~(Schoeberl et al., 2012). 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. ~~A closer look at the SWV differences between the simulations in the tropics suggests that while the~~ These differences correspond to the ratio between Max/Interim ~~ratio is between~~ of approximately 1.55~~and 1.6~~–1.6 and about 0.55–0.6 between Min/Interim, i.e. ~~very close to 1.6 and in line~~ they are consistent with the prescribed boundary ~~condition, the Min/Interim is about 0.55–0.6 suggesting that the Min run gains a small amount of water while transporting air upward~~conditions.

We next describe the meteorological condition in the Arctic stratosphere during the analysed winters. Figure 2 shows the daily average temperature in the Arctic polar vortex in winters 2010/11, 2012/13, 2013/14 and 2015/16. 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). The winter 2010/11 represents a cold winter with vortex average temperatures below 200 K and minimum temperatures below 195 K, sufficient for formation of NAT/STS PSCs, through most of the winter, from December to the beginning of April with only a brief interruption by a warming in early January. Minimum temperatures in the vortex were record cold and below 190 K even in the end of March (Manney et al., 2011). The winter 2012/13 is an example of a warm Arctic stratospheric winter. Vortex average temperatures below 195 K were seen for only a few days in December in the ERA-Interim data, and the minimum temperature was below 195 K until mid January. A SSW occurred in early January followed by a weakening and a break up of the polar vortex in the lower stratosphere already in February. The winter 2013/14 was intermediate with average temperatures inside polar vortex being close to long-term climatological mean through most of the winter, until late March when a final SSW occurred. There were only a few days in late December when the average temperature was below 195 K. The 2015/16 winter was as cold, or even colder, as the 2010/11 winter during December–February with minimum vortex average temperatures below 195 K. However, a minor SSW occurred in early February and the final warming came in early March, ending the cold period and reducing ozone depletion potential much earlier than in the 2010/11 winter.

Figure 3 shows the five day running mean concentration of water vapour at 54 hPa averaged over the Arctic polar vortex for winters 2010/11, 2012/13, 2013/14 and 2015/16. The gaps in the 2012/13 MLS curve are due to undefined vortex (or a too small vortex with only few observations) after the SSW. The water vapour concentration in the Interim simulation is comparable to the MLS data. However, the variability in water vapour is smaller in FinROSE than in the MLS data. Typically there is a stronger increase in the water vapour towards the spring in MLS than in the simulation. This is most evident in winter 2013/14 when MLS concentrations increased by more than 1 ppm between November and April while the simulated increase was only 0.3 ppm. Although an increase by spring is expected due to downward transport of air with higher SWV concentration by the ~~BD circulation~~ Brewer–Dobson-circulation the increase seen in MLS observations in 2014 is unusual. For example the observed increase in January 2013 after the SSW associated with downward transport of water-rich air from above was about 0.3 ppm and that increase was reasonably well reproduced by FinROSE. Note that the MLS observations within the polar vortex are sparse, which adds some noise to the MLS vortex average. Also note that FinROSE vortex-mean values are calculated using all data points inside the vortex even if MLS data are not available for each point. This approach increases the robustness of model estimates but at the same time complicates direct comparison with MLS. Interestingly, when looking at the 60–90° N average, which includes also air from outside the polar vortex, there is no similar spring increase in MLS data as seen in ~~(~~Fig. 3~~)~~, and the agreement between FinROSE and MLS improves (not shown). In all winters, the Max simulation has about 2 ppm more water vapour in the Arctic polar vortex than the Interim simulation, and the Min simulation is about 1.5 ppm drier than the Interim simulation. This indicates that the simulated differences in the polar vortex water vapour are about the same as the differences in the boundary conditions for the tropical tropopause (Fig. 1), despite the average increase in SSW between the TTL and the polar vortex of about 1.5 ppm in each run.

There are also several SWV decreases seen in Fig. 3 which are due to the formation of ICE PSCs and possibly also to dehydration due to sedimentation of ICE particles. The most pronounced one is in the winter 2015/16 when, during a very cold period (Fig. 2), the observed concentrations decreased from 5.2 ppm to 4.7 ppm and remained low until late February. A relatively small decrease of only about 0.2 ppm was simulated in the Interim run. This decrease corresponded to formation of ICE PSCs in the model (see Section 3.2 for discussion of PSC results) and therefore at least a part of the decrease could be explained by sedimentation. A much larger decrease of about 1 ppm was seen in the Max simulation starting from late December, which is consistent with larger amounts of ICE PSCs simulated in this run. Another, much smaller, decrease of about 0.2 ppm can be seen in the MLS observations during mid-January 2011 corresponding to a cold period. The decrease is almost undistinguishable in the Interim simulation, but is pronounced in the Max simulation, which is a result of a larger amount of ICE PSCs.

3.2 Polar stratospheric clouds

Figure 4 and 5 show PSC type 2 (ICE) and PSC type 1 a (STS) and b (NAT~~/STS) volumes between 15 and 37~~) areas at 55 ~~(about 375 and 950~~hPa (about 20 km) in the Arctic polar vortex for winters 2010/11, 2012/13, 2013/14 and 2015/16. The ~~volume~~ area was calculated by summing the ~~volumes~~ areas of model grid boxes containing PSCs. ~~NAT/STS PSC~~ STS is assumed to exist in the gridbox if there is ~~liquid more than 0.3~~more than 0.6 ppt ~~.~~ liquid HNO_3.

In the winter 2010/11 the polar vortex was stable and cold, but not extremely cold. The ICE PSC ~~volume~~ area (Fig. 4) in the Interim simulation was mostly moderate except for a period in late January with cold temperatures and large ICE PSC ~~volumes~~areas. The ICE PSCs lasted longer in the spring than in other winters. It is unusual that ICE PSC:s occur after January, but in 2011 ICE PSCs were seen through February even in the Interim simulation. In the Max simulation the

Table 1. Sum of the ICE~~and~~, NAT ~~/~~and STS PSC ~~volumes~~ areas (~~$10^6 km^3$~~ $10^6 km^2$*day) at 55 hPa.

	Year	2010/11	2012/13	2013/14	2015/16
ICE	Interim	~~94~~ 24	~~17~~ 3	~~27~~ 9	~~499~~100
	Min	~~5~~ 1	0	0	~~105~~23
	Max	~~425~~ 78	~~160~~ 33	~~183~~ 47	~~1150~~183
NAT ~~/~~	Interim	680	280	590	760
	Min	490	210	400	630
	Max	730	310	670	810
STS	Interim	~~19487~~ 1830	~~9642~~ 946	~~20417~~ 2110	~~22128~~2030
	Min	~~17751~~ 1770	~~8600~~ 850	~~18220~~ 1900	~~20939~~1890
	Max	~~19919~~ 2010	~~10007~~ 990	~~21474~~ 2230	~~22034~~2110

ICE PSCs lasted until mid March. Also ~~the Cloud-Aerosol Lidar and Infrared Path finder Satellite Observation (CALIPSO) (Pitts et al., 2007)~~ CALIPSO (Pitts et al., 2007, 2018; Spang et al., 2018) observed PSCs in the 2010/11 winter. The observed ICE PSC areas are comparable to the FinROSE modelled ICE areas (Thölix et al., 2016). Also the duration of ICE clouds is comparable. However, Khosrawi et al. (2018) reported that the comparison between the PSC volume densities simulated with model are several orders of magnitude smaller than the observed ones.

In the winter 2012/13 the polar vortex was very cold in December, and some ICE PSCs were simulated. However, after the SSW in early January no ICE PSCs were simulated, not even in the Max simulation. The 2013/14 winter was moderately cold with some ICE PSC occurrence in late January. ~~For example too dry models may not be able to simulate a large Arctic ozone loss such as of 2010/11.~~ The winter 2015/16 started as very cold in December and January, and the ICE PSC ~~volume~~ area was large through January. The maximum ICE PSC ~~volumes~~ areas in the Interim simulation were ~~more than 50~~about 70 % larger compared to the other cold winter 2010/11. Dörnbrack et al. (2017) and Khosrawi et al. (2017) also reported unprecedented and widespread ICE PSC formation seen in CALIPSO observations in 2015/16. This was also the only winter with a significant ICE PSC ~~volume~~ area in the Min simulation, with a water vapour concentration of ~~only about 3.7~~less than 4 ppm.

The water vapour concentration had a strong effect on the ICE PSC formation: in the Max simulations the ICE PSC ~~volume~~ area increases significantly in all winters. For instance in 2010/11 the largest PSC ~~volume~~ area is more than twice as large in Max as in the Interim simulation. In the warm winters (2012/13 and 2013/14) the relative increase in the ICE PSC ~~volume~~ area due to additional water vapour was even larger than in the cold winters (2010/11 and 2015/16). The amount of water vapour was an important factor for the extent of ICE PSC occurrence also in winter 2015/16, however, the relative increase between the Interim and Max simulation was smaller than in other studied winters, that were warmer. PSC starts to form about two weeks earlier in the Max simulation compared to the Interim simulation. In the Min simulation the stratosphere is too dry for ICE PSC formation in nearly all years.

Figure 5 shows the ~~volume of NAT /~~area of NAT and STS PSCs in the Arctic vortex. ~~NAT/STS volumes~~ Both NAT and STS areas are always significantly larger than the ICE ~~volumes~~ areas because type 1 PSCs form at warmer temperatures than ICE PSCs. Type 1 PSCs typically start to form in early November and ICE PSCs in mid to late December. The simulated peak values in the ~~NAT/STS volume range from 260 to 340~~ STS area range from 18 to 24 million ~~,~~ km^2, NAT area range from 11 to 16 million km^2 while the ICE ~~volume~~ area peaks range from ~~4 to 30~~ 1.5 to 5.5 million km^2 in the Interim simulation. As expected the type 1 PSCs occur later in the spring than ICE PSCs, e.g. in the winter 2010/11 ~~NAT/~~both NAT and STS PSCs were simulated until late April, more than a month later than the ICE PSCs.

In the cold winter 2010/11 ~~NAT/~~STS PSCs persisted for more than five months, and NAT almost five months, from December to mid April. An increase in moisture (Max simulation) had only a minor effect on the NAT ~~/STS volume~~and STS areas. In the Min simulation the maximum ~~NAT/STS volume was usually about 20~~ STS (and NAT) area was about 2 million km^2 smaller than in Interim simulation, while the difference between Max and Interim was much smaller. In the 2012/13 winter ~~NAT/~~both NAT and STS PSCs were simulated only in the beginning of the winter and by early January all the NAT PSCs disappeared due to warm conditions. The maximum ~~values of NAT/STS volumes~~ value of STS area in 2012/13 winter were ~~large, 330~~ the largest among the simulated years, 24 million ~~, and the difference between Interim and Min simulation is more than 30~~ km^2. The maximum NAT area was also the largest, about 13 million km^2. The increase of water vapour in the Max simulation did not change the PSC ~~volume much~~ area as much as the decrease. In the early ~~2013~~2012/~~14~~ 13 winter the NAT ~~/STS volume was~~ and STS areas were even larger than in 2010/11, but warmer temperatures in the vortex in February caused the ~~PSC volume~~ PSCs to diminish more rapidly. The effect of water vapour in 2012/13 was the largest among the simulated years, the increase in NAT ~~/STS volume~~ and STS areas between Min and Interim simulations ~~was 30–40~~ were about 3 million ~~. The NAT~~km^2. In winter 2013/~~STS volumes in~~ 14 both NAT and STS maximum areas were nearly as high as in 2012/13 (about 23 million km^2 and 11 million km^2, respectively), but large areas of PSCs persist much longer than in winter 2012/13. The maximum difference between Interim and Min simulation was about 2 million km^2. Also in the 2015/16 ~~winter~~ the NAT and STS area were larger than in 2010/11, but the PSCs did not persist as late as in 2010/11. The increase in type 1 PSC ~~volume~~ area due to increased water vapour was smaller than in 2013/14, about ~~25~~ 1.5 million km^2.

Table 1 shows cumulative ICE~~and NAT /STS PSC volumes between the altitudes 15 and 37~~, NAT and STS PSC areas at altitude 55 hPa. The largest cumulative ICE ~~volumes~~ areas are always seen in the Max simulations. In the Min simulations there are only very small or no ICE PSC ~~volumes~~areas, with the exception of the winter 2015/16, when considerable ICE PSC ~~volume~~ area was present in all runs. However, in 2015/16 the ICE PSCs occurred mainly in December and January, while in 2010/11 the ICE PSCs occurred from January until the end of February. The timing of the PSCs is important for chlorine activation and ozone loss as discussed later. The effect of water vapour on the cumulative ICE PSC ~~volume~~ area is larger in warm years than in cold years. The NAT ~~/STS volume also strongly depends~~ and STS areas also strongly depend on winter temperatures – the maximum ~~volume is~~ NAT and STS areas are simulated in the coldest winter 2015/16 in every simulation, while the smallest ~~volume is~~ areas are simulated during the warmest winter 2012/13. However, unlike ICE PSC, the NAT ~~/~~and STS clouds are formed in every winter. The formation of type 1 PSCs is less sensitive to changes in water vapour concentration than the ICE PSCs. The relatively large changes in water vapour between different simulations results in relatively small ~~small~~ changes in the cumulative NAT ~~/STS volume.~~ and STS area.

3.3 Chlorine activation

Table 2. Vortex-mean mixing ratio of ClO_x integrated over the whole winter ~~of activated chlorine in Arctic vortex in different runs~~ (ppb*day) and as monthly mean concentration (ppb) in the Min, Max and Interim simulations. Percentage in parentheses indicate the effect of SWV concentration change compared to the Interim simulation.

	Year	2010/11	2012/13	2013/14	2015/16
		(Cold)	(Warm)	(Intermediate)	(Cold)
Winter	Interim	~~148~~ 152	~~69~~ 73	~~110~~ 113	~~115~~119
	Min	~~141~~ 145 (-5 %)	~~68~~ 72 (-1 %)	~~97 (-12~~101 (-11 %)	~~113~~ 117 (-2 %)
	Max	~~157~~ 161 (+6 %)	~~72~~ 76 (+4 %)	~~116~~ 120 (+~~5~~6 %)	~~119~~ 123 (+3 %)
November	Interim	0.12	0.12	0.11	0.12
	Min	0.14	0.13	0.12	0.14
	Max	0.13	0.13	0.12	0.13
December	Interim	0.35	0.74	0.28	0.43
	Min	0.41	0.79	0.38	0.48
	Max	0.36	0.72	0.34	0.43
January	Interim	1.4	0.94	1.28	1.59
	Min	1.49	1	1.43	1.61
	Max	1.36	0.98	1.34	1.55
February	Interim	1.5	0.22	1.03	1.23
	Min	1.59	0.23	1.22	1.28
	Max	1.56	0.23	1.17	1.25
March	Interim	1.21	0.17	0.52	0.41
	Min	1.35	0.18	0.71	0.48
	Max	1.31	0.18	0.66	0.46
April	Interim	0.26	0.14	0.14	0.052
	Min	0.38	0.15	0.14	0.054
	Max	0.34	0.14	0.14	0.053

In early winter chlorine is present as reservoir compounds (HCl and $ClONO_2$), which do not destroy ozone. ~~When PSCs start to form in~~ In the cold conditions within the polar vortex the chlorine species are transformed, through heterogeneous reactions, into intermediate species such as Cl_2. ~~These species are easily dissociated when the~~ When sunlight reaches the polar vortex ~~in the spring~~ these species are easily dissociated to form active chlorine species that participate in the catalytic ozone depletion cycles, i.e. ClO_x (ClO, Cl_2O_2 and Cl). ~~PSCs sustain~~ Active chlorine goes back to reservoir species through reactions with NO_2 and CH_4, however if PSCs are present the regeneration of ClO_x is sustained.

Figure 6 shows the fraction of reservoir, intermediate and active chlorine species at ~~54~~55 hPa in the Min and Max simulations. The results from the Max simulation are represented by the upper limit for the intermediate (magenta) and active species (green), and by the lower limit for the reservoir species (black). The chlorine fractions from the Interim simulation always fit within the range from the Min and Max simulations. The timing of the changes in the partitioning of the chlorine species correlates well with the occurrence of PSCs, ~~both NAT/STS and ICE.NAT/STS PSC volume starts to grow at the same time when chlorine starts to transform from reservoirs to intermediate species~~ e.g. the chlorine reservoir species start to transform into intermediate species when the STS PSCs appear (Fig. 5).

In the 2010/11 winter chlorine activation starts in the latter half of December and the fraction of ClO_x is large through the January–Mach period, reaching ~~the~~ a maximum of about 85 %. The active chlorine starts to transform back to reservoirs (mainly $ClONO_2$) in the beginning of March. In early April when the PSCs disappear the active chlorine rapidly decreases to ~~the~~ background values.

In the 2012/13 winter chlorine activation starts slightly earlier than in the other years, but already in the beginning of February most of the chlorine has converted back to reservoir species due to a SSW. The maximum fraction of ClO_x is about 75 %, and it is reached already in the end of December. The ~~period with active chlorine lasted only for a short period, the active chlorine~~ active chlorine decreased during January, and in the beginning of February the concentration reached nearly background values.

The beginning of the winter 2013/14 winter was very cold, chlorine activation started in mid December, and the maximum chlorine activation ~~is~~ was reached already in the end of January. After that the vortex warmed and chlorine transformed back to reservoir species. The maximum fraction of ClO_x ~~is~~ was slightly lower than in cold winters, about 70 %.

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_x ~~rather short.~~ shorter than in winter 2010/11. In the end of February the vortex warmed up and chlorine transformed back to reservoir species. The maximum fraction of the activated chlorine of about 80 % was reached by the beginning of January.

The water vapour concentration ~~seem~~ seems 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~~ during November and December~~, when NAT/STSPSCs start to form.~~. The water vapour concentration affects the composition of binary aerosols an STS. When more water condenses to the particles the uptake coefficients for the heterogeneous reactions and the surface area increase, i.e. the reaction rates increase. The difference between Min and Max simulations can be up to 30 % when about half of the reservoir chlorine have transformed to intermediate species, just before the concentration of active chlorine species starts to increase. The ~~concentrations of the active and reservoir chlorine species differs significantly~~ difference in concentration of active chlorine and reservoir species between the Min and

Max simulations ~~during the period with high , except for the 2016 spring. The~~ 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 ~~intermediate chlorine species during the chlorine activation period~~chlorine partitioning in cold winters. In the cold spring 2011 the difference in chlorine activation between Min and Max simulations was about 5 % on average, it reached nearly 20 % in the beginning of April, when the chlorine deactivation was fast. In the warm winter 2012/13 the ~~difference~~ change was less significant, about 5 % during the ~~whole short activation period. In~~ winter. In the 2013/14 winter the difference in chlorine activation between Min and Max simulations reached 10 % in the latter half of January~~. Between mid February and~~, from mid February to mid March the difference ~~is~~ was 15–18 %. The chlorine activation in 2015/16 winter ~~seems~~ seemed to be less dependent on water vapour content~~. The difference~~, probably due to the ICE PSCs that appear in all simulations. Therefore, the conditions were favourable for high chlorine activation in all simulations. The difference in the fraction of activated chlorine between simulations is only few percents, only when the deactivation starts (in the end of February) the difference is more than 5 %.

The effect of increased water vapour seems to be large in moderately cold years, i.e. when the chlorine activation is not so complete. The ~~amount~~ start and end of chlorine activation correlate with the ~~volume of NAT/STS PSCs. Also the end of chlorine activation depends on the existence of NAT/STS PSCs. ICE PSC volume instead does not correlate with~~ appearance of STS and NAT. The ICE PSCs did not significantly increase the chlorine activation. For example in 2012/13 and 2013/14 winters there were no ICE PSC in the Min simulation, but the chlorine activation ~~is~~ was nearly as high as in the Max simulation, which ~~have~~ had ICE PSCs.

Table 2 shows the ~~cumulative sum of activated chlorine within the polar vortex. The~~ vortex averaged ClO_x as a cumulative sum over the whole winter, and as monthly mean concentration. The sums are integrated from November to April. The cumulative sum has information about both the duration of the chlorine activation period and the concentration of ClO_x, while the monthly average concentration shows the timing of chlorine activation. The cumulative chlorine activation was largest in winter 2010/11 and the smallest in 2012/13. The activation started in November every year, but remained small until December. The winter 2010/11 differs from the others, with high chlorine activation from January to March, giving the largest cumulative sum of the studied winters. Even in April the ClO_x concentration remains elevated. The warm winter 2012/13 had the smallest cumulative chlorine activation, significant chlorine activation was seen only in December and January. The changes in water vapour between the Min/Interim/Max simulations ~~have~~ had the largest effect on the cumulative ClO_x in moderately cold winters (2010/11 and 2013/14)~~. The~~, where the increase from Interim to Max was ~~+5~~ 3 to 6 % and change from Interim to Min was ~~-5 to -12~~-1 to -11 %. In the cold winter 2015/16 the respective changes were ~~+~~only 3 and -2 %, and in the warm winter 2012/13 the changes were ~~+~~4 and -1 %.

3.4 Ozone loss

Figure 7 shows the mean chemical total ozone loss within the polar vortex for all the studied winters 2010/11, 2012/13, 2013/14 and 2015/16. The total column chemical ozone loss was calculated by subtracting the passive transported total ozone from the modelled total ozone. ~~However,~~ In Fig. 7 the polar vortex is defined ~~here~~ using the potential vorticity limit 36 PVU only at the

475 K level. The figure shows the chemical ozone depletion in the Interim, Min and Max simulations as well as the difference in the loss between the Min and Max simulations. The passive ozone tracer was initialized every year on December 1^{st}, when it was set equal to the ozone in the model. Chemical processes start to reduce ozone already in December, but they have minor effect on the total wintertime ozone loss. In January the chemical processes become more intensive, when the chlorine activation increases (see Fig. 6).

In general the ozone loss is larger in cold years. The largest ozone loss was simulated in the beginning of April 2011 when about 90 DU ozone had been destroyed according to our model. FinROSE seems to underestimate the ozone loss~~, possibly due to a general 10 % negative bias in total ozone,~~ ; for example Sinnhuber et al. (2011) and Manney et al. (2011) simulated 120 DU ozone loss and Pommereau et al. (2013) even 170 DU in winter 2010/11. If we look at maximum ozone losses instead of the polar vortex mean losses, then the numbers are larger. The maximum ozone loss in 2010/11 within the polar vortex was 128 DU, which is comparable to the Sinnhuber et al. (2011) value.

The ozone loss in the warm winter 2012/13 differs from the loss in colder winters (2010/11 and 2015/16). ~~In winters when~~ The maximum average ozone loss in the polar vortex ~~is unstable and small or disturbed the Brewer–Dobson circulation brings more -rich air to the polar vortex than usual. Hence the ozone loss~~ in the 2012/13 winter was ~~produced mostly by chemistry as shown previously by e.g., Sagi et al. (2017). The total ozone column loss in this winter remained smaller than in cold years, when the ozone depletion is driven by halogens~~only 23 DU, because the polar vortex was unstable and small. By the ~~end of~~ mid April 2014 the simulated vortex mean ozone loss was ~~about 75~~79 DU in the Interim simulation. Before mid February, i.e. during the coldest period, there was very little effect from the changes in SWV. A relatively small ozone loss of ~~60~~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 ~~early.~~in the beginning of March, i.e. earlier than in 2010/11 and 2013/14.

Figure 7 also shows the difference of polar vortex averaged chemical ozone loss between Min and Max simulation. It tells how much the water vapour concentration change affects the ozone loss. The difference is largest (~~nearly 16~~about 15 DU) in 2013/14, a moderately cold winter, with significant ozone depletion. Another winter, 2010/11, with significant ozone loss and cold, but not extremely cold conditions showed the second largest effect from addition of water vapour, about 10 DU. In 2012/13, when the ozone loss ~~was mostly caused by chemistry~~stopped very early, the difference between Min and Max simulations was about ~~7.5~~5 DU. The 2015/16 winter started as very cold, but warmed early. The difference in ozone loss between the simulations remained very small up to mid February, by mid March the difference was about ~~8~~7.5 DU. A reduction in the water vapour decreased ozone loss in every winter. In the Interim simulation the deepest ozone losses were about ~~3–8~~2–9 DU (~~5–10~~6–11 %) deeper than in the Min-simulation. The effect from an increase in water vapour from Interim to Max was about same. In 2010/11 winter the loss increased by about 7 %, while Sinnhuber et al. (2011) and Vogel et al. (2011) reported an increase of ozone loss by 20 % and 10 % (respectively) with water vapour increase being of about the same magnitude as here. Thus, our estimates are slightly smaller than those by Vogel et al. (2011). ~~The reason for the difference is not clear, but FinROSE's horizontal resolution~~ An additional sensitivity experiment showed that the difference compared to other studies can be, at least partly, due to the coarse horizontal resolution in FinROSE (3° × 6°)~~may be too coarse to~~, which is not sufficient to fully capture the deepest ozone loss. Specifically, repeating Interim simulation for winter 2010/11 with higher resolution (1.5° × 3°) than in the original simulation showed larger ozone loss by 15 DU.

The changes in the amount of water vapour is not very important for ozone loss in cold years, within the range that was tested here. In the 2010/11 winter the chlorine activation was nearly complete in the Arctic polar vortex, and additional water vapour did not increase chlorine activation and thus not the ozone depletion. Increasing water vapour concentration (compared the Interim simulation) strengthen ozone loss at least 4 DU at other winters except for 2011 when the increase is not significant.

Table 3. Maximum polar vortex-mean ozone loss produced by full chemistry, heterogeneous chemistry and separately the ~~NAT/~~STS, NAT and ICE part in Min, Max and Interim simulations (DU). Percentages show the fraction due to each part relative to the full chemistry.

	Year	2010/11 (Cold)	2012/13 (Warm)	2013/14 (Intermediate)	2015/16 (Cold)
Min	Full chemistry	84	~~54~~ 21	~~75~~ 70	~~58~~53
	Heterogeneous part	50 (60 %)	~~11 (21~~13 (62 %)	~~28 (38~~32 (45 %)	~~37 (64~~33 (62 %)
	~~NAT/STS~~STS, NAT and ICE	20 (24 %)	~~5 (10~~3 (15 %)	~~11 (15~~13 (19 %)	~~14 (24~~11 (21 %)
Interim	Full chemistry	90	~~56~~ 23	~~83~~ 79	~~62~~56
	Heterogeneous part	56 (62 %)	14 (~~25~~63 %)	~~36 (44~~40 (51 %)	~~41 (66~~35 (63 %)
	~~NAT/STS~~STS, NAT and ICE	30 (33 %)	~~8 (15~~5 (24 %)	~~21 (25~~23 (30 %)	~~20 (32~~17 (30 %)
Max	Full chemistry	91	~~61~~ 25	~~90~~ 85	~~66~~59
	Heterogeneous part	56 (62 %)	~~18 (30~~15 (62 %)	~~42 (47~~45 (53 %)	~~44 (66~~37 (63 %)
	~~NAT/STS~~STS, NAT and ICE	34 (37 %)	~~11 (18~~7 (28 %)	~~27 (~~30 (35 %)	~~24 (36~~19 (33 %)

To better understand the mechanism of SWV influence on ozone loss, simulations without heterogeneous chemistry were performed. From those simulations ozone loss caused by heterogeneous chemistry can be separated by subtracting the total ozone simulated without heterogeneous chemistry from that simulated in the full chemistry run. Two different set-ups were used for testing the effect of the heterogeneous chemistry. In the first gas-phase chemistry simulation the heterogeneous chemistry was not included at all. In the second simulation the formation of PSCs was limited by setting the air temperature passed to the heterogeneous chemistry module to 200 K, similarly to what was done in Karpechko et al. (2013). ~~This setting has little influence on the reactions on the background aerosols , but prohibits formation of STS , NAT and ICE PSCs~~The increase of water vapour (Max simulation) did not increase the ozone loss, but in the Min simulation there was 6 DU less ozone depletion. This setting allows some heterogeneous processing on binary aerosols and some STS that are very dilute in HNO_3, and due to the temperature limit the surface area densities will remain quite small. Table 3 summarises ozone loss characteristics during the studied years and shows the loss produced by full chemistry, heterogeneous chemistry and separately the NAT~~/~~, STS and ICE PSCs in Interim, Min and Max simulations.

In the Interim simulation with full chemistry in 2010/11 about 90 DU ozone was depleted, of which the heterogeneous chemistry caused 56 DU depletion, i.e. about 62 % of the total ozone loss. Heterogeneous chemistry due to ~~ICE and NAT/STS~~

PSCs destroyed 30 DU ozone, which was about 33 % of the total loss. The result indicates that chlorine activation on the binary aerosols has a significant role in ozone depletion. Some studies 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). The increase of water vapour (Max simulation) did not increase the ozone loss, but in the Min simulation there was 6 DU less ozone depletion. This is consistent with the results by Kirner et al. (2015), who argue that the contribution of ICE PSCs to the ozone loss is always less than 5 % in the Antarctic spring, where the chlorine activation is nearly complete.

In the warm 2012/13 winter the ozone loss is only 23 DU, and the heterogeneous part is ~~only 25~~63 % ~~. NAT/~~of it. NAT, STS and ICE PSCs caused only a ~~very~~ small part of the total heterogeneous chemistry driven ozone loss (24 %). The loss caused by heterogeneous chemistry increased with increasing water vapour, but remained small even in the Max simulation.

In 2013/14 the heterogeneous chemistry ~~destroyed about 36~~caused about 40 DU ~~(44~~ozone destruction, which is about 51 % ~~)~~ of the ozone ~~and ICE and NAT/STS about 21~~loss and NAT, STS and ICE about 23 DU (~~25~~30 %), when the total ozone loss was ~~83~~79 DU in the Interim run. The increase in SWV from Interim to Max increased the ozone loss by about ~~7~~6 DU and the decrease in SWV from Min to Interim decreased the ozone loss by ~~8~~9 DU. So, water vapour changes have larger role than in colder year. The ozone depletion due to heterogeneous chemistry increased with water vapour, even though the fraction due to heterogeneous chemistry was smaller than in 2010/11 and 2015/16.

In the 2015/16 winter the heterogeneous part was largest when compared to other simulated years, reaching even ~~66~~63 % of the ozone loss, and also the ~~ICE and NAT/STS~~STS, NAT and ICE part was large, ~~32~~30 %. The total ozone loss is however only ~~62~~56 DU~~about the same as in 2012/13 winter~~. When the water vapour content was increased from Interim to Max simulation, the fraction due to the heterogeneous chemistry remained the same, but the fraction due to ~~NAT/STS~~STS, NAT and ICE PSCs increased.

Based on the results in Table 3 it can be concluded that nearly all SWV impact on ozone loss is through heterogeneous chemistry. For example in 2010/11 the ozone loss without heterogeneous chemistry was 34 DU in Interim, 34 DU in Min and 35 DU in Max simulation and only the heterogeneous part changed from model run to model run. In ~~2012/13 and~~ 2013/14 the non-heterogeneous contribution is ~~42 and 47~~about 39 DU~~respectively~~, and in 2015/16 about 21 DU, i.e. in warm years it is larger than in cold years. In 2012/13 the non-heterogeneous contribution is only 9 DU, but also the total ozone loss is very small. However, the heterogeneous part is about 62 % of the total ozone loss in all other winters than 2013/14 and also with both increased and decreased water vapour. The fraction of STS, NAT and ICE driven chemistry changes with eater vapour concentration.

Finally we analyse the vertical distribution of the ozone loss and the effect of SWV on ozone loss, which is shown in Fig. 8. The largest ozone loss was simulated in 2010/11, when the ozone destruction in the Interim run with normal SWV was about 1.4 ppm between 60–30 hPa. The ozone depletion increased by 0.2 ppm between the Min and Max simulations. In 2012/13 the maximum ozone reduction is almost the same as in 2010/11, but it occurs at higher altitude ~~(induced)~~ and lasts for shorter period than in 2011. The effect of the increase in water vapour from Min to Max simulation had only a minor effect on the ozone depletion in 2012/13. The heterogeneous chemistry and chlorine activation did not have an important role in the warm conditions ~~. In the~~ and the ozone loss between 60–30 hPa remains very weak. In winters when the polar vortex is unstable and small or disturbed the Brewer–Dobson circulation brings more $\mathrm{NO_x}$-rich air to the polar vortex than usual. Hence the ozone loss in the 2012/13 winter was produced mostly by $\mathrm{NO_x}$ 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 total ozone column loss in this winter remained smaller than in cold years, when the ozone depletion is driven by halogens. In the 2014 spring the conditions in the polar vortex remained favourable, but the temperature was not as low as in 2011. The ozone loss developed steadily, but remained moderate. The two winters 2010/11 and 2013/14 with the most favourable conditions for halogen driven ozone depletion showed the largest increase in ozone loss with water vapour. The effect was more clear in 2013/14, which was the warmer of the two winters. The winters 2010/11 and 2015/16 look similar during January–February, but the ozone loss became much more severe in 2010/11 due to favourable conditions in March–April. In 2015/16 there was a very cold period, but it occurred too early to have a large impact on the ozone depletion, and therefore the water vapour increase had only a moderate effect later in the spring. In 2013/14 the largest ozone loss is about ~~1.2~~1.1 ppm between 60 and 30 hPa while in 2015/16 it is ~~only~~ about 1 ppm at the same altitude. Livesey et al. (2015) and Sagi et al. (2017) showed results from 450 K level, and their ozone losses were about 2 ppm in winter 2011. In winter 2013 Sagi et al. (2017) had about 1.5 ppm ozone loss, which is about the same as we found.

4 Discussion and conclusions

Warmer climate in the troposphere in the future leads to increasing water vapour concentrations in the stratosphere (Dessler et al., 2013), which further warms the ~~climate~~ troposphere due to water vapour feedback. Khosrawi et al. (2016) showed that an increase in SWV and a cooling of the stratospheric temperature ~~enhances~~ enhance each other, the ~~volume~~ area of PSCs increases and they last longer in the vortex. The ozone loss can thus increase although the halogen loading has been decreased. In this study, rather than artificially changing the temperature we used meteorological fields from seven winters during the period 2010–2016 with different temperatures and dynamical conditions in the stratosphere. We changed the water vapour content in the tropical tropopause region according to the CCMVal-2 simulations. The water vapour entry concentration is controlled by the cold point in the TTL, and the distribution of SWV is largely determined by this entry concentration together with the transport and the contribution from methane oxidation. Results show that, as expected, wetter/drier tropical tropopause leads to wetter/drier Arctic polar vortex and also the size of polar ozone depletion changes along the water vapour changes~~. For example too dry models may not be able to simulate a large Arctic ozone loss such as of 2010/11, which can be seen from the Table 3~~, which affects also the Arctic ozone loss.

A reduction in SWV decreases the ozone loss due to heterogeneous processes by decreasing the PSC formation. An increase in SWV instead makes the heterogeneous chemistry more important~~by increasing PSCs.~~. As expected, heterogeneous chemistry is less important in warm winters. I.e. in 2012/13 only 14 DU ozone depletion was initiated by heterogeneous chemistry in the Interim simulation. The corresponding loss in the winter 2010/11, with persistent cold conditions, was 56 DU. The increase in loss with water vapour was small, i.e. the loss increased from 50 to 56 DU from the Min to the Max simulation. In the winter 2012/13 the corresponding losses were 32 and 45 DU, i.e. the water vapour concentration had the largest impact in moderately cold winters. If the winter is cold enough, the increase is less important, because the ~~PSC volume is large anyway~~PSCs may form even at low water vapour concentrations, and the chlorine activation is already nearly complete in Arctic vortex~~. As expected, heterogeneous chemistry is more important if the winter is cold and PSC volumes are large. In winters 2010/11 and 2015/16 over 62 % of the ozone loss is initiated by heterogeneous chemistry, and in the warm winter 2012/13 about 21 %. In winter 2010/11 Pommereau et al. (2013) got 120 ozone loss due to heterogeneous reactions, which is about 70 % of the total loss.~~ , therefore the water vapour increase is less important.

Winters in the stratosphere are often divided into cold, or dynamically inactive, and warm, or dynamically active. In the cold winters the polar vortex is stable and more PSCs are formed and halogens can destroy ozone. Warm conditions in the winter stratosphere are often due to SSW, which allows NO$_x$-rich air masses from the mesosphere to enter the vortex and take part in the ozone depletion (Sagi et al., 2017). Cold winters differ from the warm winters ~~when looking~~ regarding the ozone loss and the fraction of ozone loss initiated by heterogeneous chemistry~~. Also the PSC volumes~~ , during cold winters the PSC areas are larger and thus chlorine activation ~~are in higher level during cold winters~~within the polar vortex is more complete. A lack of water leads to less ICE PSCs, and therefore to less ClO$_x$. However, the ICE PSC ~~volume~~ area is not the only explaining factor for ozone loss. The type 1 PSCs that form at higher temperatures are responsible for a large fraction of the chlorine activation. The formation of STS and NAT is limited by the partial pressure of nitric acid, sulfuric acid and water and hence the concentration of water vapour is not the only thing affecting the NAT ~~/STS volume~~and STS areas. However, the dry conditions in the Min simulations have some limiting effect on the peak NAT ~~/STS volume~~and STS areas.

The cold winter 2010/11 differs from the others by an especially long chlorine activation period, which lead to large ozone depletion. In the warm winter 2012/13 the polar vortex was weak; however it was shifted to south where it was exposed to sun-light earlier than usually, and thus ozone loss could start earlier. The ozone loss was however weak because chlorine activation remained very low. The ozone depletion in 2012/13 occurred at higher altitudes than in the other years, because of the NO$_x$ induced ozone loss. The 2013/14 winter was moderately cold, and the ozone depletion was second largest among the considered winters. In this winter the effect of water vapour changes on ozone loss was the largest across the studied winters. Winter 2015/16 started as extreme cold (Matthias et al., 2016; Manney et al., 2016), but the stratosphere warmed early terminating chlorine activation and leaving ozone loss relatively low, despite the fact that the cumulative ICE ~~(and NAT/STS) volumes~~ areas were extremely large.

Chemical ozone destruction inside the Arctic vortex varied between ~~56~~ 23 and 90 DU in the Interim-simulations, ~~61~~ 25 and 91 DU in the Max-simulations and ~~54~~ 21 and 84 DU in the Min simulations. We find that the meteorological conditions are more important for the ozone depletion than the concentration of water vapour. Also the fraction of heterogeneous chemistry in the ozone loss is more dependent on the temperature than on the water content. Livesey et al. (2015) arrived to similar conclusion, when investigating ozone loss based on the MLS observations.

MacKenzie and Harwood (2004) showed from their chemistry–climate model simulations, that the increase of water vapour increases the ~~volume~~ area of PSCs both by microphysical effects and due to lowering the stratospheric temperatures. The microphysical processes cover about 20 % of the increase and the rest is due to cooling of the stratosphere. In our study ~~the volume of ICE PSCs increased by 20 %, but only due to the microphysics. The increase could have been larger with~~

~~temperature changes in the simulations.~~ we only changed the water vapour concentration. However, the temperature effect can be seen by investigating different years. The ~~difference in cumulative ICE volumes~~ cumulative ICE areas between studied years ~~was as large as factor 20~~varied by a factor of 30, and in cumulative NAT ~~/STS volumes it varied by factor 2.~~ and STS by 2.7 and 2.2, respectively. MacKenzie and Harwood (2004) got about 15 % more ozone loss at 465 K level with less than 1 ppm additional water vapour without changing temperature. In our study the ozone loss increased by 1 DU (1 %) in 2010/2011, ~~5~~2 DU (9 %) in 2012/2013, ~~7~~6 DU (8 %) in 2013/2014 and ~~4~~3 DU (~~6.5~~5 %) in 2015/2016 when the water vapour concentration was increased by about 2 ppm. When the water vapour was instead decreased by about 1.5 ppm, the ozone loss decreased by 6 DU (~~6.7~~7 %), 2 DU (~~3.6~~9 %), ~~8~~9 DU (~~10~~11 %) and ~~4~~3 DU (~~6.5~~5 %), respectively. The small contribution due to water vapour increase in winter 2010/11 can be compared to the results of MacKenzie and Harwood (2004) in the Antarctic vortex. There the chlorine activation is nearly complete in every winter. In winter 2010/11 also in Arctic vortex the chlorine activation was nearly complete, and additional water vapour did not change the activation and, thus not either the ozone depletion.

Note that effects of changing water vapour concentration on air temperature, not accounted for here, would probably have increased the water vapour impact on ozone loss. The indirect impact comes through water vapour radiative impact on stratospheric temperatures. Tian et al. (2016) estimates that a 2 ppm increase of water vapour would cool the stratosphere by approximately 2 K, while Rex et al. (2004) estimates that a 1 K cooling could increase ozone loss in the Arctic by 15 DU. Thus based on these estimates a water vapour increase of 2 ppm, similar to the difference between Interim and Max runs, could result in up to 30 DU additional ozone loss. This estimate ~~suggest~~ suggests that the direct water vapour impact on ozone loss quantified in our experiments may account for only about ~~20 % of~~ one fifth of the total ozone loss, but in order to confirm this estimation a designed experiment with a chemistry–climate model would be needed.

In summary, we find that variability of stratospheric water vapour of 3.5 ppm, comparable in magnitude to uncertainty in simulated water vapour concentration near the tropical tropopause, results in differences in simulated Arctic ozone loss up to 15 DU, i.e. more than ~~10~~15 % of the total chemical ozone loss in the Arctic vortex. Better understanding of tropical processes contributing to the stratospheric water vapour concentration, and thus constraining stratospheric water vapour, would therefore reduce the uncertainty in Arctic ozone loss and improve future projections of ozone layer recovery.

Competing interests. The authors declare that they have no conflict of interest.

Data availability. Data from the FinROSE simulations is available from the authors upon request. MLS data is available at https://mls.jpl.nasa.gov.

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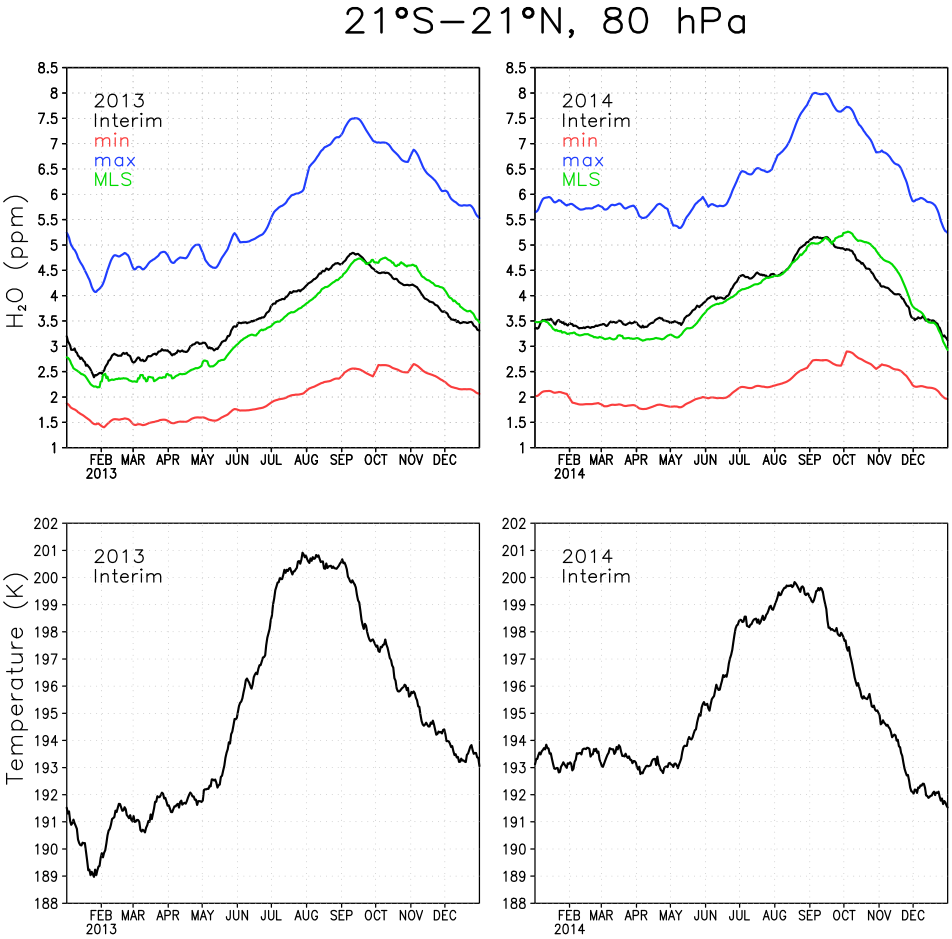

Figure 1. Water vapour and temperature around the tropical tropopause between 21° S and 21° N at level 80 hPa in 2013 and 2014. Green line is Interim, blue Max, red Min simulation and black is MLS.

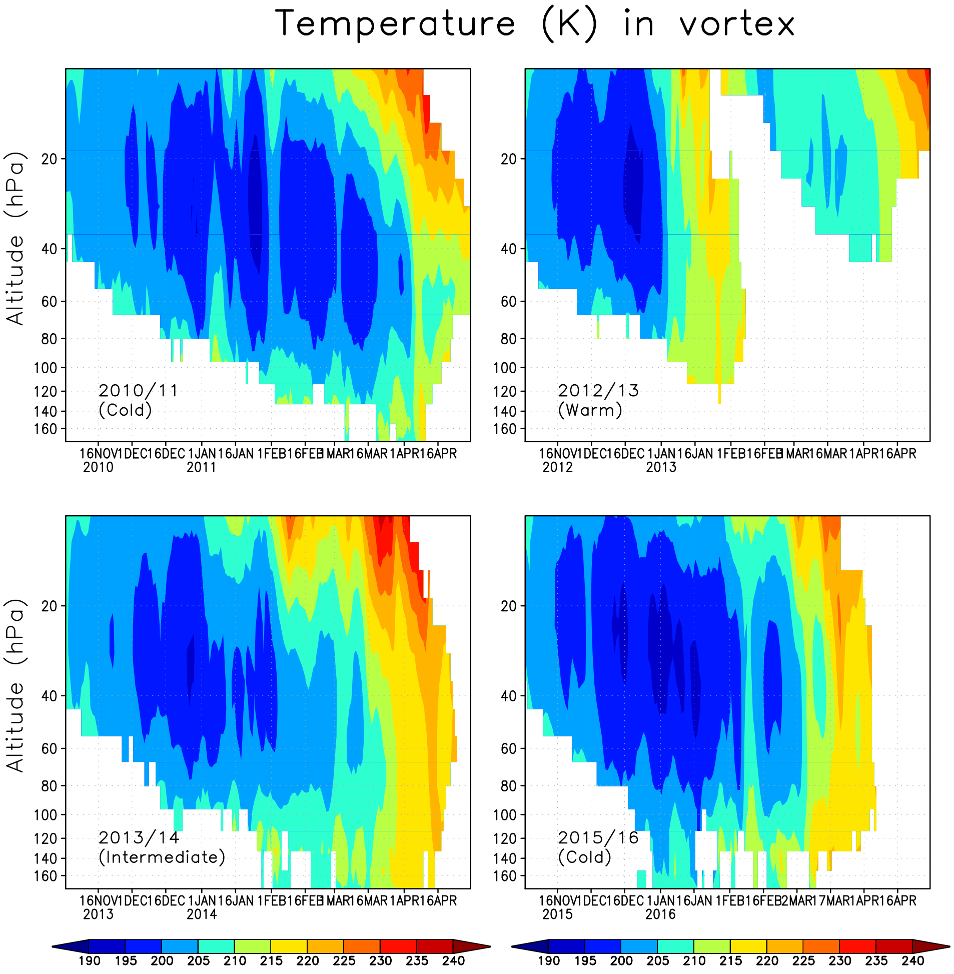

Figure 2. Vortex average temperature within the Arctic polar vortex between altitudes 170 and 10 hPa.

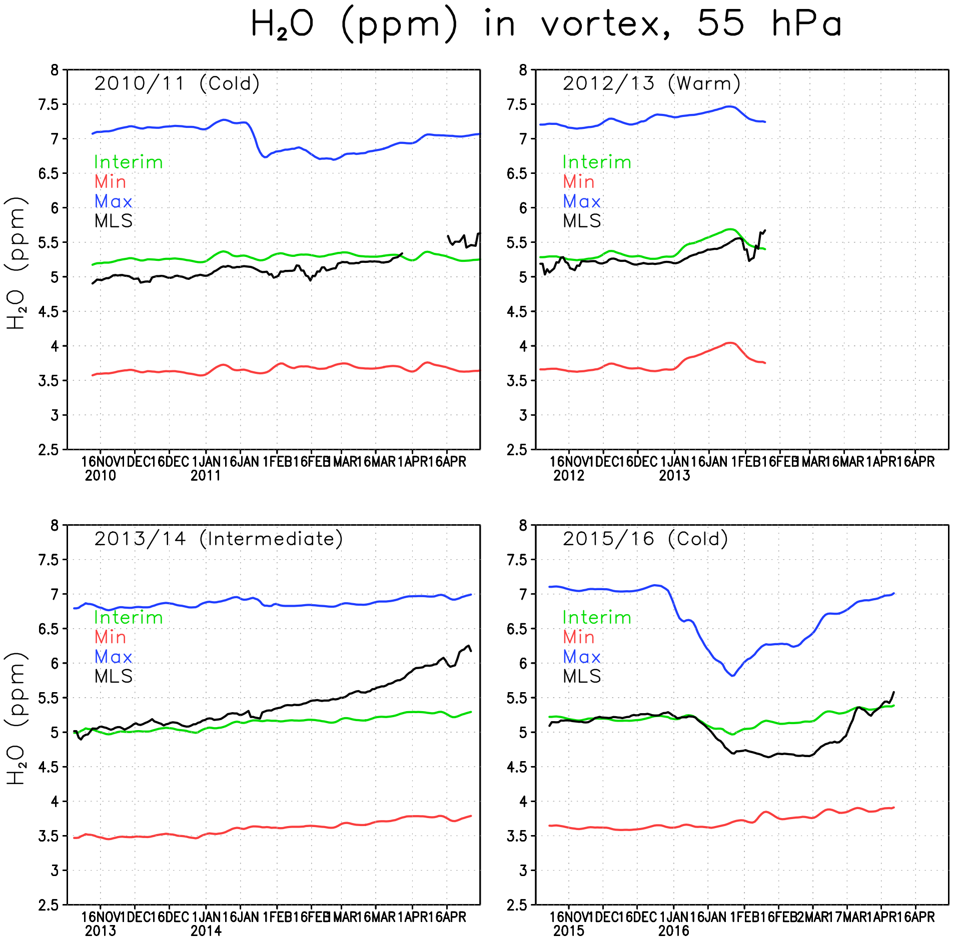

Figure 3. Water vapour concentration (ppm) at 54 hPa within the Arctic polar vortex. Green line is Interim, blue Max, red Min simulation and black is MLS.

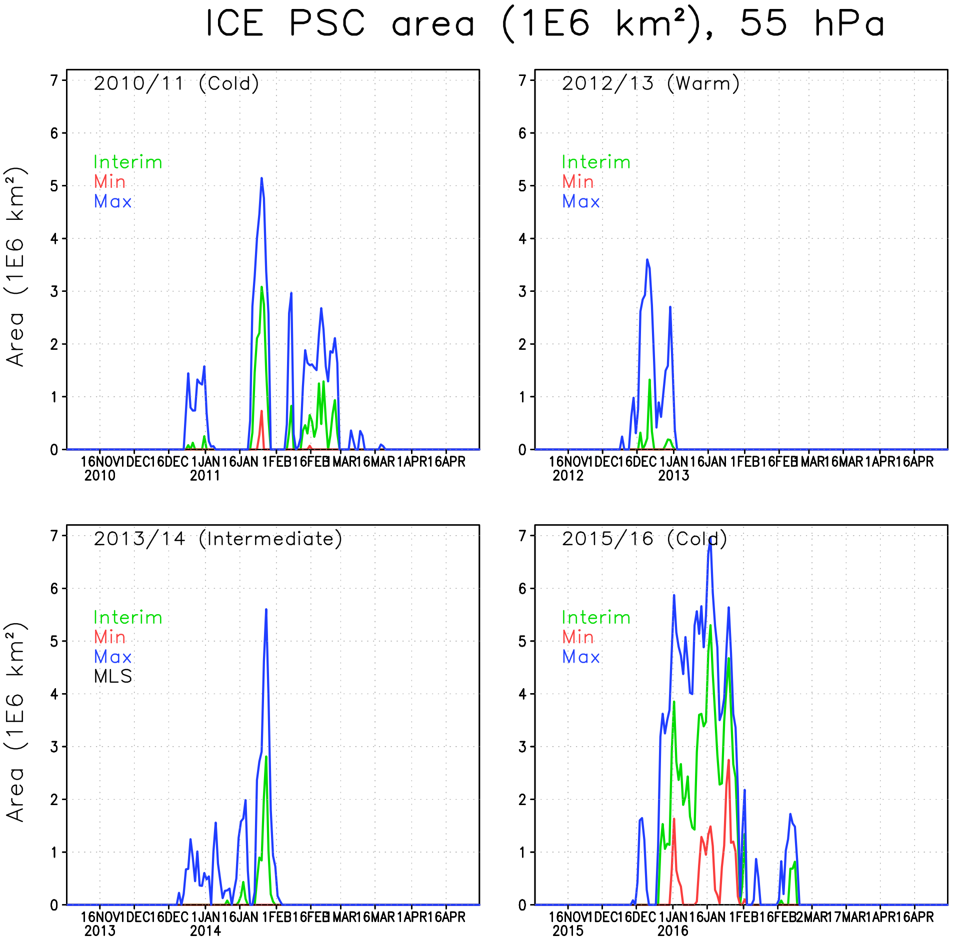

Figure 4. The ares of ICE PSCs ($10^6\mathrm{km}^2$) within the Arctic polar vortex in the FinROSE simulations at 55 hPa. Green line is Interim, blue Max and red Min simulation.

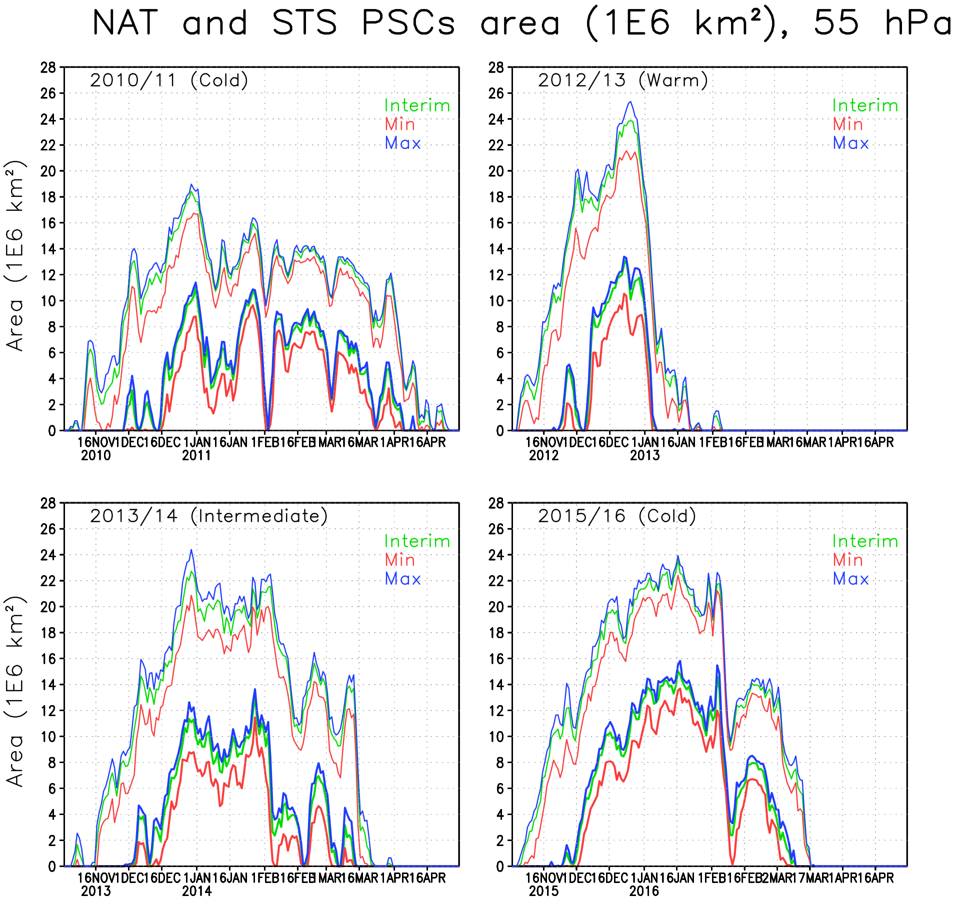

Figure 5. The area of NAT (thick lines) and STS PSCs (thin lines) ($10^6 \mathrm{km}^2$) within the Arctic polar vortex in the simulations at 55 hPa. Green line is Interim, blue Max and red Min simulation.

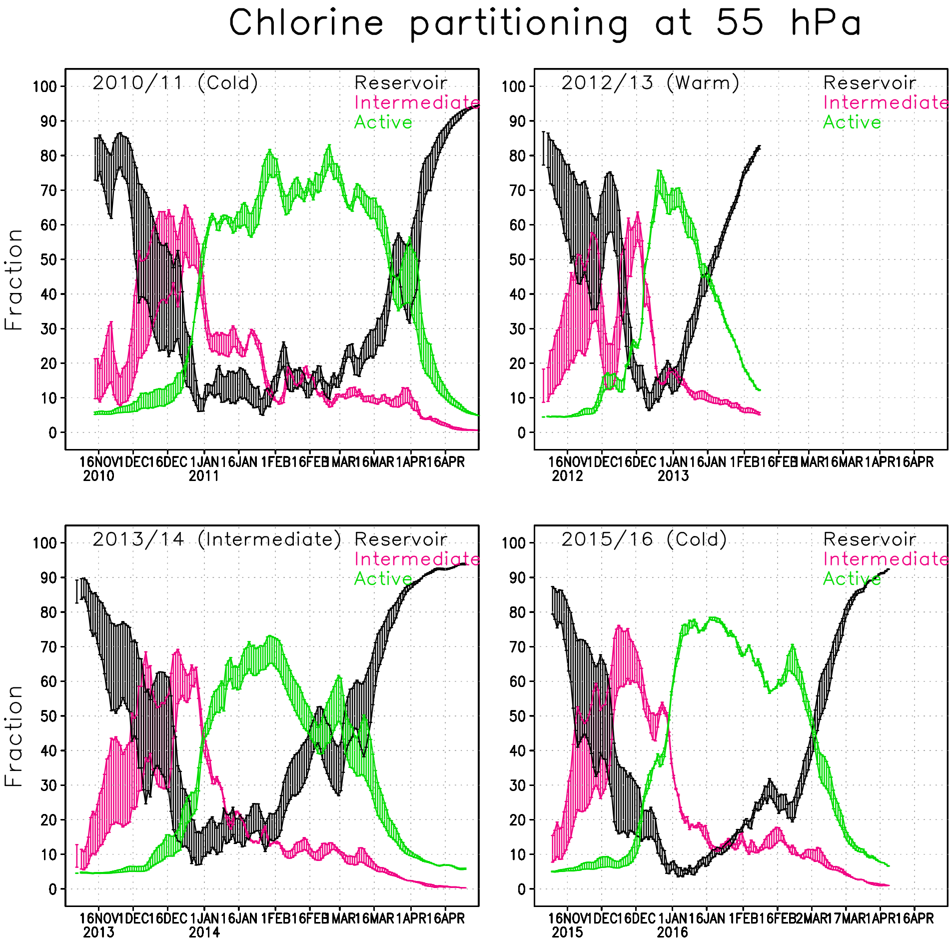

Figure 6. Chlorine partitioning (%) within the Arctic polar vortex at 54 hPa in the Min and Max simulations. Active form (green) is $Cl+ClO+2*Cl_2O_2$. Intermediate (magenta) contains $2*Cl_2+HOCl+OClO+BrCl+ClNO_2$ and reservoir chlorine (black) $HCl+ClONO_2$.

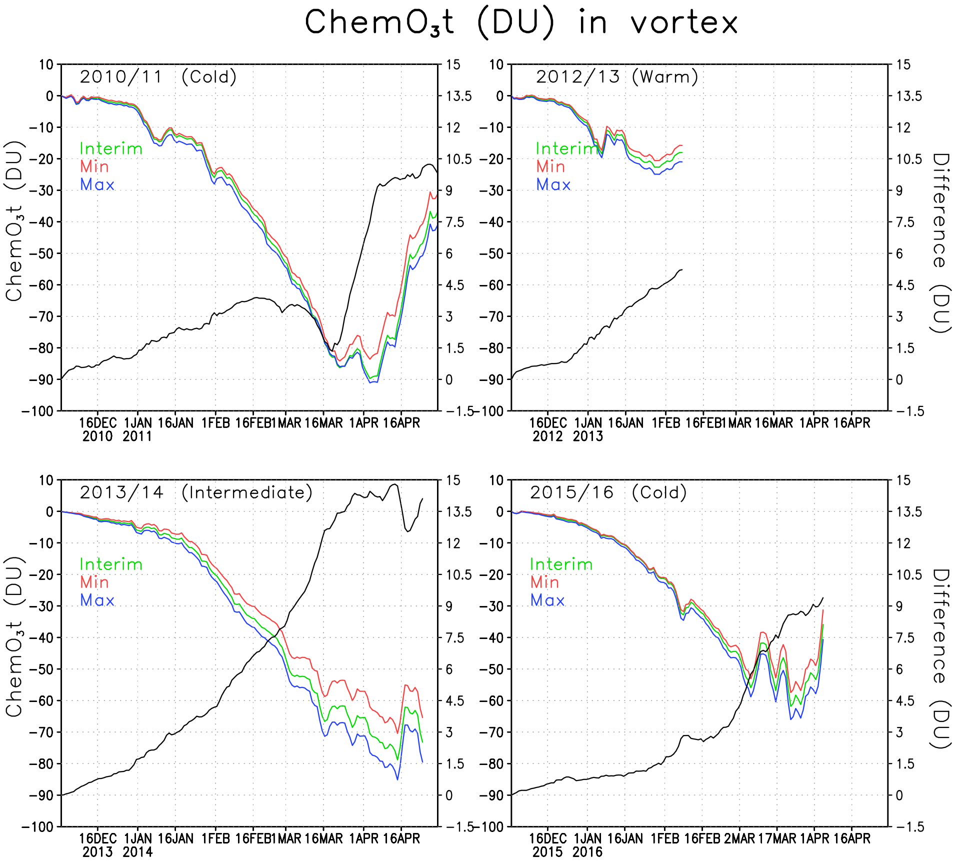

Figure 7. Chemical total ozone loss (DU) and difference between ozone loss in the Min and Max simulations within the Arctic polar vortex. Green line is Interim, blue Max and red Min simulation. The difference is in black.

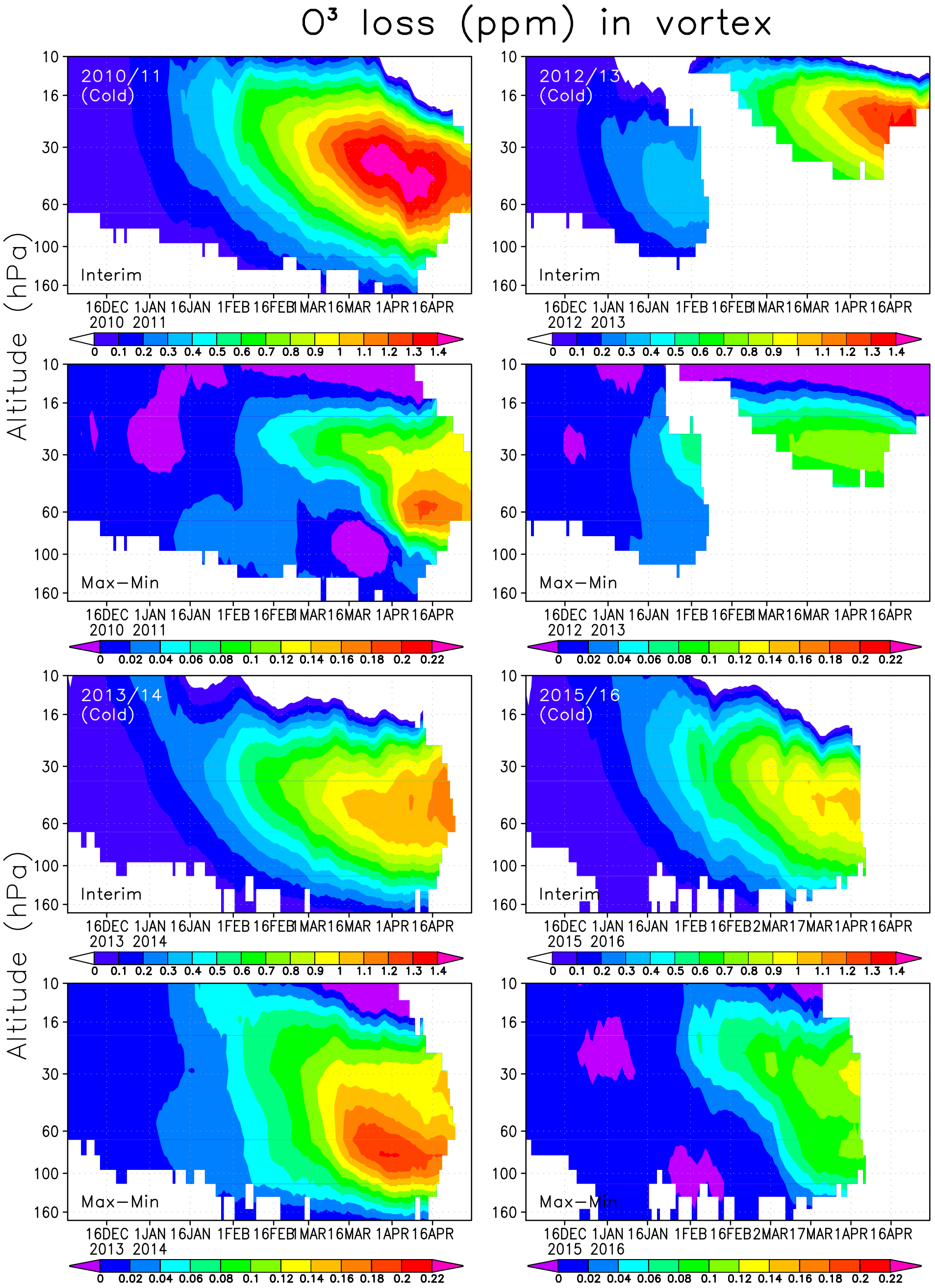

Figure 8. Averaged chemical ozone loss (ppm) in the Interim simulation (upper panels) and the difference between Max and Min simulations (lower panels) within the Arctic polar vortex.