Dear Ingo Wohltmann,

thank you very much for your efforts in performing your comprehensive review of our manuscript and for your helpful remarks and corrections.

Here are our answers to all your comments:

*This study deals with the question what are the relative contributions of the different types of stratospheric clouds to the activation of chlorine. This is still an open question and part of a recent, at times controversial debate (e.g. Drdla and Mller, 2012, Ann. Geophys. and the preceding discussion in Atmos. Chem. Phys. Discuss., 10, 28687, 2010, or Solomon et al., PNAS, 111, 17, 6220, 2014). The study adds evidence to this ongoing discussion and as such, is worth publishing.*

*The scientific methods are sound, the used model is state-of-the-art and the manuscript is short and concise. In general, it is easy to understand and well structured.*

Thank you.

*My main comment is that you really need to compare your model results to observations (see Major comments for a detailed explanation). Another issue is, that while I am a supporter of short and concise manuscripts, a little bit more of detail wouldnt hurt in some places. Sometimes, information that is necessary to understand what you have done is missing. Some more discussion would also help to assess your results in comparison to other results. I will try to give specific advice for these issues in the specific comments below.*

*The manuscript has some deficiencies in terms of the correct use of the English language. Some parts (especially towards the end of the manuscript and in the abstract and introduction) could be improved in grammar, wording and style. I will try to give specific advice in the technical corrections, but I would recommend that a native speaker proof-reads the manuscript before publication (since I am no native speaker, my corrections may be wrong).*

We will try to optimize our manuscript. Our detail comments further below in the sections Major and Specific comments.

## *Major comments*

- *Your results are of less value than they could be if you dont compare the results of your EMAC model runs to observations. If the model runs compare well to observations, it would strengthen your conclusions, and if there are discrepancies, the discrepancies (and their potential impact on your conclu-*

*sions) need to be discussed. Since this is a nudged version of EMAC, it is similar to a CTM and should be treated as such, which includes a validation of the simulated mixing ratios of the key species by observational data. You are free to choose the observational data that is most appropriate for you (or to cite a reference if these model runs are validated somewhere else). Just as a suggestion, I would recommend MLS satellite data for comparison: They are available for the complete time period that is covered by your model runs (and in polar night), many of the key species (ozone, HCl, HNO3, ClO) are included in the data set and data are freely available.*

We agree and will compare our results with MLS observations. Unfortunately this observations are first available from August 2004. If we compare our results with MLS we think it is better also to change the Fig. 1, because there are no $HNO_3$ data from MLS for the Antarctic winter 2004 available. We will choose another Antarctic winter for Sect. 5, create new plots for Fig. 1 and compare the $HNO_3$ results with MLS data.

- *You do not provide all necessary information on the model setup that is needed to reproduce your model runs and to interpret the results, even supposed the reader has consulted Kirner et al. (2011). Several pieces of information are missing, e.g. which of the both NAT schemes of the submodel PSC you use, parameters for the calculation of surface area densities are missing etc. See the specific comments for details. It may also be helpful to repeat some additional basic information from Kirner et al. in the manuscript, so that the average reader does not have to look it up there. E.g. which types of clouds are allowed to exist at the same time, do NAT clouds form from ice clouds or not etc.*

  Okay you a right here. We will add more informations regarding our PSC schemes, submodel and used parameters. It is really a mistake that we don't mention the used NAT parameterisation. Thank you for this remark.

### Specific comments

- *Abstract: I think it would make sense to mention that you are using a nudged version of the model. You basically use a CTM here and you can expect the temperature and wind fields to follow the actual meteorological evolution of the winters, which is not the case with a CCM. This is important information and would help the reader to know what to expect in the following. That is, I think it will attract more readers if people know that this is not taking place in a model world with temperature biases etc., but is based on real conditions, or more precisely based on an ECMWF world.*

  Okay we will add this in the abstract.

- *Page 14834, line 7: Can you shortly explain what the difference between the sensitivity runs is? It would help to have this information in the abstract.*

  OK.

- *Page 14835, lines 10-11: In addition to the reference to Kirner et al. (2011), I suggest to put a reference to your Section 3 here.*

  OK.

- *Page 14835, line 16: While it is true that assuming only liquid binary particles leads only to a moderate reduction, this is a hypothetical case (we know that ternary liquids exist in reality most of the time). I am not sure, but maybe it is better to skip that.*

  Yes, assuming only liquid binary particles is a hypothetical case. Actually, in the Antarctic, for extended time periods, 'STS only' is also a hypothetical case, because of the dominance of heterogeneous reactivity on ice particles. However, we agree that for the present paper, the discussion of the liquid binary particles can be dropped. Instead, we mention here now the recent contribution to this discussion by Solomon et al. (2014). The section in question reads now:

  "Drdla and Müller (2012) have raised the question of the necessity of STS, NAT and ice particles for reaching full polar stratospheric chlorine activation. In contrast, Solomon et al. (2014) have argued that in sunlit air in the polar stratosphere, deactivation, i.e. the reformation of HCl and $ClONO_2$ increases the rate at which heterogeneous reactions must proceed to keep pace with deactivation."

- *Page 14836, line 10: What is the approximate vertical resolution in the stratosphere? Please add to the text.*

  The verical resolution in the stratosphere is between 1.5 and 2.5 km decreasing upwards. We will integrate this information into the manuscript.

- *Page 14836, line 18 to page 14837, line 6: Please only cite the submodels which are relevant for the results in the stratosphere. Citing tropospheric submodels is not only confusing, it also makes the impression that you are collecting citations for your colleagues here ;-) . This is a nudged version of the model, where the tropospheric submodels will play a very minor role for the results.*

  It is a standard recommendation from the MESSy consortium to describe the used model configuration and setup as comprehensive as possible (see http://www.messy-interface.org under MESSy Licence). But it is not absolutely necessary. Therefore we will cite only the submodels which are relevant for the stratosphere.

- *Page 14837, lines 7-8: What are the rate coefficients that are not taken from Sander et al. (2011)? Are they important for this study? If the answer is yes (and if it requires not too much additional discussion), it could make sense to cite the references here.*

Beside the rate coefficients from Sander et al. (2011) there are also some relevant coefficients from:

Atkinson, R., Baulch, D. L., Cox, R. A., Crowley, J. N., Hampson, R. F., Hynes, R. G., Jenkin, M. E., Rossi, M. J., and Troe, J.: Evaluated kinetic and photochemical data for atmospheric chemistry: Volume III - gas phase reactions of inorganic halogens, Atmos. Chem. Phys., 7, 981-1191, 2007

We will additional cite this reference.

- *Page 14837, lines 18-24: I suggest to shortly mention the alternative parameterizations (Abbatt and Molina for NAT, Shi et al. for liquids) and to give reasons for your choices.*

  We agree and have changed the paragraph in question accordingly. The text in the revised version of the paper now reads:

  "Heterogeneous reaction rates and their temperature dependencies on NAT were calculated according to the parametrisation of Carslaw and Peter (1997) based on the measurements of Hanson and Ravishankara (1993). Carslaw and Peter (1997) also developed a parametrisation for heterogeneous reactions on NAT based on the measurements of Abbatt and Molina (1992). Using the latter parametrisation results in substantially lower (by a factor of more than 50) heterogeneous reactivity on NAT and in a reduced chemical ozone loss Carslaw et al. (1997); therefore this latter parametrisation was not considered here.

  The heterogeneous reaction rate coefficients on liquid particles are taken from Hanson and Ravishankara (1994) and Hanson et al. (1994). An alternative parametrisation for reactions on liquid particles was reported by Shi et al. (2001), which results however in a heterogeneous reactivity very close to the one employed here (e.g., Wegner et al., 2012).

  The heterogeneous reactivity of ice particles is much larger than that of other types of PSCs because of the much larger surface area of ice PSCs; the uptake coefficients and reaction probabilities for ice particles are taken from Sander et al. (2011)."

- *Page 14838, line 1: What is the value of 50% based on? Measurements? Give a reference or explanation.*

  The value is based on Tabazadeh et al. (1997) and Peter and Grooß (2012). We will add these references into our manuscript.

- *Page 14838, line 3-4: It is nice to know that there are two parameterizations for NAT particles, but unfortunately you dont tell us which one is used here :-)*

Thank you very much for this remark. We really forgot to mention that we used the kinetic growth NAT parameterisation. We will integrate this information into the manuscript.

- *Section 3 (comment 1): Given that you use the thermodynamic NAT parameterization: NAT seems only to be allowed to form from ice clouds (if I understand Kirner et al., 2011, correctly). That could limit the occurence of NAT clouds. Since NAT clouds have been observed under conditions where the formation from ice clouds can be excluded (e.g. Voigt et al., 2005, Pitts et al., 2011), this assumption should be discussed.*

  As mentioned above we didn't used the thermodynamic NAT parameterization, but we can nevertheless mention that homogeneous NAT nucleation is possible.

- *Section 3 (comment 2): Some of the necessary information about number densities and particle radius that is required to calculate surface area densities seems to miss here (even if referring to Kirner et al., 2011). If you use the thermodynamic NAT scheme, provide the free parameters rmin and Nmax. By the way, maybe I am wrong, but is there a typo in Eq. 17 in Kirner et al.? The way it is written, Nmax is a minimum and not a maximum number density. Please clarify. If you use the kinetic NAT scheme, it would not hurt to shortly describe how it works and what are the parameters (initial radius, initial number density). Please also provide the necessary parameters for ice. It would also not hurt to shortly repeat how the surface area density for liquids is calculated.*

  Unfortunately you are right with the typo in Eq. 17 in Kirner et al. Thank you for the precise reading of this paper. It have to be "min" instead of "max".

  We will integrate a short description of the kinetic NAT parameterisation (surface growth factor and the growth of NAT particles over size bins).

- *Section 3 (comment 3): Maybe I missed that when reading Kirner et al. (2011), but what happens if STS and NAT clouds exist at the same time? How do they compete for the available HNO3? Please discuss.*

  First the calculation of NAT take place and then the calculation of STS.

- *Section 3 (comment 4): It seems that there are several sedimentation schemes implemented according to Kirner et al. (2011). Please indicate which scheme is used.*

  We used the trapezoid scheme. We will add this information into the manuscript.

- *Section 3 (comment 5): Same for the other parameters from Table 4 of Kirner et al., which cannot be deduced from the existing information in the manuscript.*

  OK.

- *Page 14838, line 17: Of course, it makes discussion easier to only switch off the heterogeneous chemistry (e.g. only one Fig. 2 needed), but not to change the formation of the clouds. But this also has a disadvantage: If you would switch off the NAT clouds completely, the HNO3 condensed in the NAT clouds would be available for STS clouds, i.e. you would expect that the surface area density of STS clouds increases. In practice, the effect may be small, since NAT clouds play a small role relative to STS clouds in your model setup. But the more realistic assumption would be to switch of the clouds completely (i.e. we know that once we have NAT clouds, there will be reactions on them (provided we dont believe in the Abbatt and Molina rates). What we dont know exactly from measurements are the relative surface area densities of STS clouds and NAT clouds). I dont want you to change the model runs here (I know they are expensive), but I suggest to discuss this.*

  Thank you for this remark. We agree and will discuss this topic in our manuscript.

- *Page 14839, line 1: May make sense to give this information earlier (i.e. in Section 2). How well does the nudging work (temperature bias to ECMWF)?*

  We will move this information to Sect. 2. The evaluation of the nudging technique was for example done in van Aalst (2005). The temperature bias within the nudging area from ground until 1 hPa is very small if you use the relaxations coefficients mentioned in our reply to Review 2.

- *Page 14839, line 6: Do you only use tropospheric boundary conditions and wait some years until a realistic distribution in the stratosphere is reached? Or do you initialize the stratospheric fields of O3, N2O, CH4, HCl etc. with some data to reduce the spin-up period of the model? What is the maximum Cly and Bry that is observed in the model runs (i.e. do you consider short-lived bromine species)?*

  We initialize all chemical spezies with the help of data from older EMAC model runs with earlier starting points (for example 1960). The maximum values in the year 2005 for $\mathrm{Cl}_y$ are approx. 3.4 ppbv and for $\mathrm{Br}_y$ approx. 19.5 pptv. We don't consider the VSLS, but added 6 pptv bromine to $\mathrm{CH_3Br}$ (according to WMO (2011)). We will add a corresponding sentence into our manuscript.

- *Page 14839, line 15 and Fig. 1 (bottom row): This is a good place to compare your model results to observations. I suggest to show maps of HNO3 from satellite data similar to the bottom row in Fig. 1. Put another way: You say that PSCs are simulated very reasonably in line 14. But how do you know that if you dont compare to observations?*

  We will integrate this $\mathrm{HNO_3}$ comparison into our paper. But as mentioned above we have to choose another year in Fig. 1, because the MLS data are in the winter 2004 not available.

- *Page 14839, line 19: Please briefly compare to observed number densities from other studies and discuss (e.g. Northway et al., 2002, Voigt et al., 2005, Pitts et al. 2011). Please change the unit to cm-3, since this is common usage. It seems to me that the simulated number densities are at the very low end of observations, which helps to explain the low influence of your modelled NAT clouds on chlorine activation. I suggest to write something along these lines in the manuscript.*

  We will discuss our number densities with the mentioned references. Our NAT number densities are based on Fahey et al., 2001. We will integrate this topic into the discussion.

  We will also change the unit to $cm^{-3}$ (also in the plots of $N_{NAT}$ and $N_{ICE}$ in Fig. 2).

- *Page 14839, line 25: It is probably no good idea to use a dot between the chemical species. To my knowledge, that implies that exactly one molecule H2SO4 is bound to one molecule H2O etc. Use a slash or hyphen.*

  OK.

- *Page 14839, line 26 to page 14840, line 1: Probably a good idea to take an average over the 5 years here, since interannual variability will not be too large and the manuscript gets clearer. Can you show to me that this approach is justified (e.g. by attaching some figures for the single years and the different quantities to your reply)?*

  We added the Figs. 3 - 7 to this reply.

- *Figure 2 (caption and text page 14840, line 1): Add a remark to the text and figure caption that the plots are scaled very differently.*

  OK

- *Fig. 2: There is a strange artifact here: In the plots for the number density and surface area densities, the values seem to be cut at exactly 200 hPa. Please explain.*

  The reason is a prescribed PSC area in our model. The lower boundary of this PSC region is 180 hPa.

- *Page 14839, line 27 to page 14840, line 1: Why do you not use equivalent latitude or PV here for latitude? As can be seen in Fig. 1, you are in danger of averaging over areas which are not in the polar vortex.*

  We agree that in principle, the use of equivalent latitude would be preferable here. However, for technical reasons, this is rather difficult to do at this point in time. More importantly however, the vortex core, poleward of 80°S for the rather stable Antarctic vortex, will not likely be significantly influenced by out of vortex air. This is particularly true here, as we are not considering individual cases, but rather a multi-year climatology.

- *Page 14840, line 1-3 and Fig. 2 (caption): Can you put the information that the figure is valid for all runs in the figure caption, too?*

  OK.

- *Page 14840, line 4-5: lower temperatures. Lower than what? Probably you mean low here. But since this is very unspecific, you can skip that.*

  OK, we will skip that.

- *Page 14840, line 6 and line 9, Fig. 2: Please change the unit to cm-3.*

  OK.

- *Page 14840, line 10: At first, it may look like a contradiction that the ice surface area density is high and still not relevant for activation. I suggest to add a note that by the time that the ice clouds form, all chlorine is already activated and additional activation by ice clouds does not matter anymore (maybe in Section 6).*

  We will add in about the following sentence:

  “It might be surprising that the enormous increase of heterogeneous reactivity provided by the frequent occurrence of ice PSCs has rather little impact on chlorine activation in the simulations presented here. However, a stronger heterogeneous reactivity does not necessarily translate into stronger chlorine activation. Frequently, chlorine activation through heterogeneous chemistry is rate limited by the gas phase formation of reaction partners for heterogeneous reactions (citations). This is in particularly true for the initial activation step when HCl and $ClONO_2$ are titrated against each other on the first occurrence of heterogeneous reactivity when temperatures sink below the threshold for chlorine activation. If under such circumstances, likely in polar night, ice particles form days or weeks after the initial activation step, they will have very little impact on chlorine activation. ”

- *Fig. 3 (add additional figure following Fig. 3): This is a good place to compare your results to observations again. Since you dont have ClOx from observations, I suggest to compare ClO from satellite and model in a map similar to Figure 1 in an additional figure (to get the diurnal cycle right).*

  We will integrate ClO from our Standard simulation and valdiate the results with ClO from MLS.

- *Fig. 3: Only a minor remark: In the upper right panel, the difference between the NoHet and Liquid runs is shown. Why is there a difference between these runs outside of the area where polar stratospheric clouds form? It is difficult* *to see how large the difference is from the contour plot and there may be some numerical reasons (diffusion?), but it catches your attention that e.g. in the troposphere, ClOx seems not be zero in at least one of the runs.*

We created two plots (Fig. 1) with the $ClO_x$ difference between the simulations **Liquid** and **NoHet** in 2 hPa and 700 h. It is visible that the differences are extremely small. In 2 hPa the differences are between -0.006 and 0.006 ppbv and in 700 hPa between $-4{\cdot}10^{-6}$ and $2.4{\cdot}10^{-5}$ ppbv. As you mentioned the reason is most likely the diffusion.

- *Fig. 3: The different scales are really confusing. Even the upper two plots dont have the same scale. Add a remark to the text and figure caption that the plots are scaled very differently. And I would suggest to use the same scaling for the upper two plots (yes, I know, some values are cut this way, but it causes more confusion the way it is in the moment, because it it easy to miss the fact that they are scaled differently).*

  The upper two plots in Fig. 3 are different. The left one is the $ClO_x$ development in the **Standard** simulation. The right one is the difference between the simulations **Liquid** and **NoHet**. Hence in the right panel we have very small negative values (see above), but in the left one we have no negative values. But nevertheless we can adapt the colour scale in both plots. We would suggest to adapt the scale in the left and right panel to -0.2 to 3.0 (see Figure 2).

- *Fig. 4 (add additional figure following Fig. 4): And again, good place to compare to observations. Following Fig. 4, I suggest to show exactly the same plot as in the upper left corner, just with satellite data for ozone. You would make me perfectly happy if you could add a new figure showing HCl both from satellite and your model as an example for a reservoir gas and discuss the figure.*

  OK.

- *Fig. 1, Fig. 5: Just for consistency, you could omit the black lines between the areas of different color in the contour plots. There are no lines in Figs. 2-4.*

  OK.

- *Page 14843, line 1-3: I would express it a little bit more carefully. You dont really definitely confirm a major contribution of liquid particles, since these are still model runs, and the occurence of liquid and solid clouds is not sufficiently constrained by measurements. I would write something like suggest or if you want to put more emphasis on it strongly suggest.*

  OK.

***Technical corrections***

*I am happy that you implemented most of the technical corrections I suggested in the pre-review, but you did not get the grammar and wording right in some cases.*

- *Page 14840, line 19: It has to be subtracting (verb) and not subtraction (noun), see pre-review. Same in line 21 and 23.*

  Okay, sorry.

- *Page 14841, line 14-15: Sounds a little bit better now, but I suggested to start the sentence with The contribution is. . . . Normally, it is a good idea to start sentences with the subject, followed by verb and object. You often tend to start with the object or dependent clauses, which is difficult to understand and read.*

  OK.

- *Page 14841, line 17: Not quite my suggestion from the pre-review. A suggestion for the complete sentence, in (hopefully) correct English: The NAT particles show only a relevant contribution to chlorine activation in mid-May in the region between 20 hPa and 50 hPa, with contributions of up to 10 sure where to place only.*

  OK.

*Additional technical corrections. Some parts at the end of the manuscript and in the abstract and introduction can be improved in grammar, wording and style. I will give suggestions below.*

- *Page 14834, 7-10: Suggestion: The results of these simulations show that the significance of heterogeneous reactions on NAT and ice particles for chlorine activation and ozone depletion in Antarctic winter and spring is small in comparison to the significance of reactions on liquid particles. (i.e. replace subordinate by small, change order, regarding was not correct)*

- *Page 14834, 12-15: Suggestion: Liquid particles alone are sufficient to activate almost all of the available chlorine, with the exception of the upper PSC regions between 10 and 30 hPa, where temporarily ice particles show a relevant contribution. Shortly after the first PSC occurence, NAT particles contribute a small part to the chlorine activation. (i.e. change the order of the clauses, omit the before ice particles, shorten have a relevant contribution to the chlorine activation, change have to show, split into two sentences, during following with the execption of makes no sense logically)*

- *Page 14834, line 16: I would suggest to move In the model simulations to the very end of the sentence.*

- *Page 14834, line 17: I would suggest: Heterogenous chemistry on ice particles causes only up to 5 DU of additional ozone depletion in the column. . . (and so on)*

- *Page 14834, line 25: I would suggest to write . . . is essential for the correct simulation of chlorine activation. . .*

- *Page 14834, line 26 to Page 14835, line 5: This is a very long sentence. In addition, it is not a correct English sentence (which cause . . . for the denitrification). I would suggest to split the sentence into two parts, e.g. The liquid (cold binary aerosols and STS particles) and solid particles (NAT and ice particles) allow heterogeneous reactions to proceed, which cause the activation of chlorine reservoirs and the production of chlorine radicals leading to ozone destruction. In addition, it causes stratospheric denitrification, resulting in a delay in the deactivation of active chlorine in polar spring. . . (activation to chlorine radicals sounds odd, omit this preceding active chlorine)*

- *Page 14835, line 6: Delete To consider. . . and start with A new algorithm. . .*

- *Page 14835, line 25: Insert comma following However*

- *Page 14835, line 27: Insert comma following Therefore*

- *Page 14836, line 5: I would write . . . and to determine the corresponding ozone depletion. . . .*

- *Page 14836, line 9: atmospheric in correspondence to tropospheric?*

- *Page 14836, line 11: Sounds odd. Suggestion: to link source codes from different institutions*

- *Page 14836, line 15-16: Spell out approx.*

- *Page 14837, line 14: Do you mean surface areas here?*

- *Page 14837, line 15: Since you introduced the abbreviations NAT and STS in the introduction, you may want to shorten the sentence here.*

- *Page 14837, line 23: Add . . . for ice particles are taken from. . .*

- *Page 14838, line 3: You can omit Besides. . . and start with There are. . . without loosing information.*

- *Page 14838, line 9: Insert comma following In both cases*

- *Page 14838, line 17: Insert comma following In the three sensitivity simulations*

- *Page 14839, line 1: Skip in all four EMAC simulations or move to the end of the sentence. You could also move the clause to simulate. . . to the end of the sentence.*
- *Page 14839, line 7: Split into two sentences: Boundary conditions for ozone depleting substances are from. . .*
- *Page 14839, line 7: Delete hyphen following NAT*
- *Page 14839, line 15: Insert comma following For example*
- *Page 14839, line 17: Insert comma following NAT formation*
- *Page 14839, line 19: Delete up*
- *Page 14839, line 19: Split sentence: Simultaneously,. . .*
- *Page 14839, line 20: Replace hyphen by space in NAT-PSC*
- *Page 14839, line 20: Starting with In HNO3 sounds odd. Delete and end the sentence with . . . in the HNO3 fields of the model. or similar.*
- *Page 14839, line 24: Suggestion: In addition to NAT particles, ice particles and. . .*
- *Page 14840, line 1: The illustrated results. Better: The results shown in Fig. 2*
- *Page 14840, line 3: In average. . . sounds very odd. Suggestion: NAT is visible from mid-May until October in the averaged fields of the EMAC simulations.*
- *Page 14840, line 5: Simplify a little bit: . . . increase of the number density to values of more than 200 m-3. . .*
- *Page 14840, line 6: Suggestion: . . . differences in the timing of the first NAT appearances. . .*
- *Page 14840, line 10-11: I would suggest surface area densities*
- *Page 14840, line 16: Suggestion: The contribution of heterogeneous reactions on liquid, NAT and ice particles to chlorine activation and the corresponding ozone depletion in polar spring is determined by comparing the standard and sensitivity simulations. (start sentence with The contribution. . . and shift the clause by. . . to the end. Replace reactivity of by reactions on, avoid possible (you are really doing it, and you repeat possible in the next sentence))*
- *Page 14840, line 18: Suggestion: The impact of the liquid particles is assessed by. . . (and see far above for subtraction)*

- *Page 14840, line 24: I would first introduce the figures (i.e. sentence Page 14841, line 1) and then talk about the averaging.*
- *Page 14840, line 26: It doesnt get quite clear here that you show several zonal means as a function of latitude.*
- *Page 14841, line 1: Suggestion: In Fig. 3, the evolution of ClOx in the standard simulation and the sensitivity simulations is illustrated. The standard run is shown in the top left, the liquid run is shown in. . . . A little bit more organized this way.*
- *Page 14841, line 4, line 6-7: Replace due to by by*
- *Page 14841, line 5: Insert of following contributions*
- *Page 14841, line 8: Sounds odd. Suggestion: Ice particles only show a relevant contribution in the upper PSC region. . .*
- *Page 14841, line 8: Replace extra by additional and write A maximum of about. . . .*
- *Page 14841, line 13: Suggestion: Liquid particles contribute more than 90chlorine activation for most of the time. For the rest of the paragraph, see also above (pre-review comments).*
- *Page 14841, line 20: Simply The evolution of ozone is shown in Fig. 4?*
- *Page 14841, line 21: Insert comma following October*
- *Page 14841, line 22: Sounds a little bit odd. Suggestion: The largest contribution to the ozone depletion comes from chlorine activated on liquid particles with values of more than 2500 ppv.*
- *Page 14841, line 24: Suggestion: In contrast, the contribution that can be attributed to additional chemistry on ice particles reaches values of only 170 ppbv in the upper PSC region.*
- *Page 14842, line 1: This sentence sounds very odd. Suggestion: Figure 5 shows the total effect that heterogeneous chemistry on different PSC particles has on the time development of the ozone column.*
- *Page 14842, line 3-5: Split into two sentences: The ozone column shows a decrease of more than 130 DU in early October in high southern latitudes. Liquid particles are mainly responsible for this decrease.*
- *Page 14842, line 5-7: Rearrange: The contribution. . . is at least 95% during September. . .*

- *Page 14842, line 8: Replace comma by period and start new sentence.*
- *Page 14842, line 10: Insert comma following hand*
- *Page 14842, line 17-21: Sounds odd. See my comment on the similar sentence in the abstract on page 14834, line 12-15 for a suggestion how to improve this sentence.*
- *Page 14842, line 22-23: See my comment above for page 14834, line 16.*
- *Page 14841, line 20: Simply The evolution of ozone is shown in Fig. 4?*
- *Page 14841, line 21: Insert comma following October*
- *Page 14841, line 22: Sounds a little bit odd. Suggestion: The largest contribution to the ozone depletion comes from chlorine activated on liquid particles with values of more than 2500 ppv.*
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- *Page 14842, line 3-5: Split into two sentences: The ozone column shows a decrease of more than 130 DU in early October in high southern latitudes. Liquid particles are mainly responsible for this decrease.*
- *Page 14842, line 5-7: Rearrange: The contribution. . . is at least 95September. . .*

- *Page 14842, line 8: Replace comma by period and start new sentence.*
- *Page 14842, line 10: Insert comma following hand*
- *Page 14842, line 17-21: Sounds odd. See my comment on the similar sentence in the abstract on page 14834, line 12-15 for a suggestion how to improve this sentence.*
- *Page 14842, line 22-23: See my comment above for page 14834, line 16.*

OK, we will take over your suggestions. Thank you very much for these.

### ***References***

- *Abbatt and Molina, J. Phys. Chem., 96, 7674-7679, 1992*

- *Northway et al., J. Geophys. Res., 107, doi:10.1029/2001JD001079, 2002*
- *Pitts et al., Atmos. Chem. Phys., 11, 2161-2177, 2011*
- *Shi et al., J. Geophys. Res., 106, 24259-24274, 2001*
- *Voigt et al., Atmos. Chem. Phys., 5, 1371-1380, 2005*

We will integrate these references into our manuscript and additional the references Atkinson et al. (2007), Carslaw and Peter (1997), Carslaw et al. (1997), Peter and Grooß (2012), Tabazadeh et al. (1997), and WMO (2011).

# References

Abbatt, J. P. D. and Molina, M. J.: Heterogeneous interactions of $ClONO_2$ and HCl on nitric acid trihydrate at 202 K, J. Phys. Chem., 96, 7674-7679, 1992.

Atkinson, R., Baulch, D. L., Cox, R. A., Crowley, J. N., Hampson, R. F., Hynes, R. G., Jenkin, M. E., Rossi, M. J., and Troe, J.: Evaluated kinetic and photochemical data for atmospheric chemistry: Volume III gas phase reactions of inorganic halogens, Atmos. Chem. Phys., 7, 981-1191, doi:10.5194/acp-7-981-2007, 2007

Carslaw, K. S. and Peter, T.: Uncertainties in reactive uptake coefficients for solid stratospheric particles - 1. Surface chemistry, Geophys. Res. Lett., 24, 1743-1746, 1997.

Carslaw, K. S., Peter, T., and Müller, R.: Uncertainties in Reactive Uptake Coefficients for Solid Stratospheric Particles - 2. Effect on Ozone Depletion, Geophys. Res. Lett., 24, 1747-1750, 1997.

Drdla, K. and Müller, R.: Temperature thresholds for chlorine activation and ozone loss in the polar stratosphere, Ann. Geophys., 30, 1055-1073, doi:10.5194/angeo-30-1055-2012, 2012

Hanson, D.R., and Ravishankara, A.R.: Reaction of $ClONO_2$ with HCl on NAT, NAD, and frozen sulfuric acid and hydrolysis of $N_2O_5$ and $ClONO_2$ on frozen sulfuric acid, J. Geophys. Res., 96, 22931-22936, 1993

Hanson, D.R., and Ravishankara, A.R.: Reactiv uptake of $ClONO_2$ onto sulfuric acid due to reaction with HCl and $H_2O$, J. Phys. Chem., 98,5728-5735, 1994

Hanson, D.R., Ravishankara, A.R., and Solomon, S.: Heterogeneous reactions in sulfuric acid aerosols: A framework for model calculations, J. Geophys. Res., 99, 3615-3629, 1994

Northway, M.J., Gao, R.S., Popp, P.J., Holecek, J.C., Fahey, D.W., Carslaw, K.S., Tolbert, M.A., Lait, L.R., Dhaniyala, S., Flagan, R.C., Wennberg, P.O., Mahoney, M.J., Herman, R.L., Toon, G.C., and Bui, T.P.: An analysis of large $HNO_3$-containing particles sampled in the Arctic stratosphere during the winter of 1999/2000, J. Geophys. Res., 107(D20), 8298, doi:10.1029/2001JD001079, 2002.

Peter, T. and Grooß, J.-U.: Polar Stratospheric Clouds and Sulfate Aerosol Particles: Microphysics, Denitrification and Heterogeneous Chemistry, in: Stratospheric Ozone Depletion and Climate Change, edited by Müller, R., pp. 108–144, Royal Society of Chemistry, doi:10.1039/9781849733182-00108, ISBN: 978-1-84973-002-0, 2012.

Pitts, M. C., Poole, L. R., Drnbrack, A., and Thomason, L. W.: The 20092010 Arctic polar stratospheric cloud season: a CALIPSO perspective, Atmos. Chem. Phys., 11, 2161-2177, doi:10.5194/acp-11-2161-2011, 2011.

Sander, S.P., Abbatt, J., Barker, J.R., Burkholder, J.B., Friedl, R.R., Golden, D.M., Huie, R.E., Kolb, C.E. Kurylo, M.J., Moortgat, G.K., Orkin, V.L. and Wine, P.H.: Chemical Kinetics and Photochemical Data for Use in Atmospheric Studies, Evaluation No. 17, JPL Publication 10-6, Jet Propulsion Laboratory, Pasadena, 2011.

Shi, Q., Jayne, J. T., Kolb, C. E., Worsnop, D. R., and Davidovits, P.: Kinetic model for reaction of $ClONO_2$ with $H_2O$ and HCl and HOCl with HCl in sulfuric acid solutions, J. Geophys. Res., 106, 24,259-24,274, doi:10.1029/2000JD000181, 2001.

Solomon, S., Haskins, J., Ivy, D. J., and Min, F.: Fundamental differences between Arctic and Antarctic ozone depletion, Proc. Natl. Acad. Sci., 111, 6220–6225, doi: 10.1073/pnas.1319307111, 2014.

Tabazadeh, A., Toon, O.B., and Jensen, E.J.: Formation and implications of ice particle nucleation in the stratosphere, Geophys. Res. Lett., 24, 2007-2010, 1997

Van Aalst, M.K.: Dynamics and Transport in the Stratosphere - simulations with a general circluation model, Ph.D. thesis, Institute for Marine and Atmospheric Research Utrecht, Netherlands, 2005.

Voigt, C., Schlager, H., Luo, B. P., Drnbrack, A., Roiger, A., Stock, P., Curtius, J., Vssing, H., Borrmann, S., Davies, S., Konopka, P., Schiller, C., Shur, G., and Peter, T.: Nitric Acid Trihydrate (NAT) formation at low NAT supersaturation in Polar Stratospheric Clouds (PSCs), Atmos. Chem. Phys., 5, 1371-1380, doi:10.5194/acp-5-1371-2005, 2005.

Wegner, T., Grooß, J.-U., von Hobe, M., Stroh, F., Sumińska-Ebersoldt, O., Volk, C.M., Hösen, E., Mitev, V., Shur, G., and Müller, R.: Heterogeneous chlorine activation on stratospheric aerosols and clouds in the Arctic polar vortex, Atmos. Chem. Phys., 12, 11095-11106, doi:10.5194/acp-12-11095-2012, 2012

WMO (World Meteorological Organization): Scientific Assessment of Ozone Depletion: 2010, Global Ozone Research and Monitoring Project Report No. 52, Geneva, Switzerland, 2011.

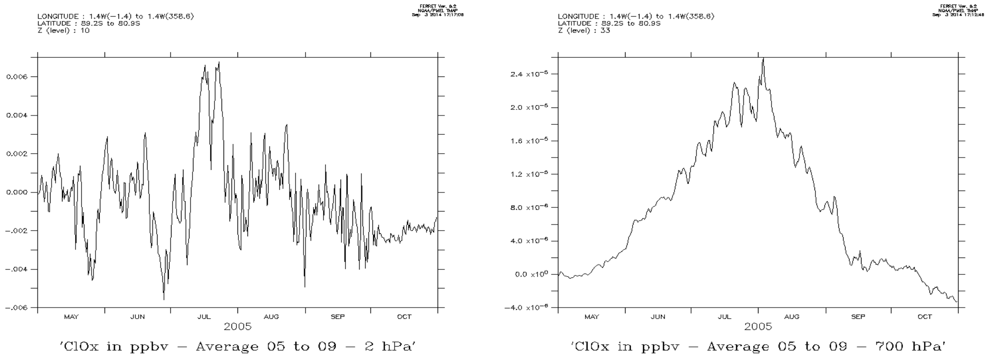

Figure 1: Difference in $ClO_x$ between the simulations **Liquid** and **NoHet**

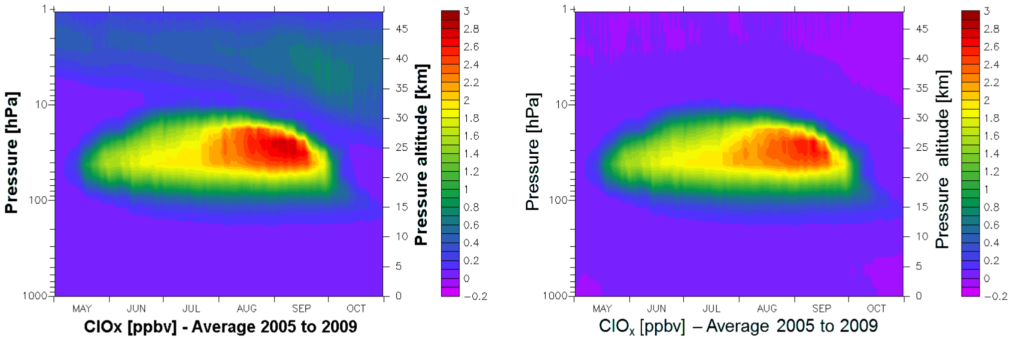

Figure 2: Chlorine activation in the **Standard** simulationn (left) and contribution of liquid particles to $ClO_x$ (right). Plot as in manuscript Fig. 3 (top).

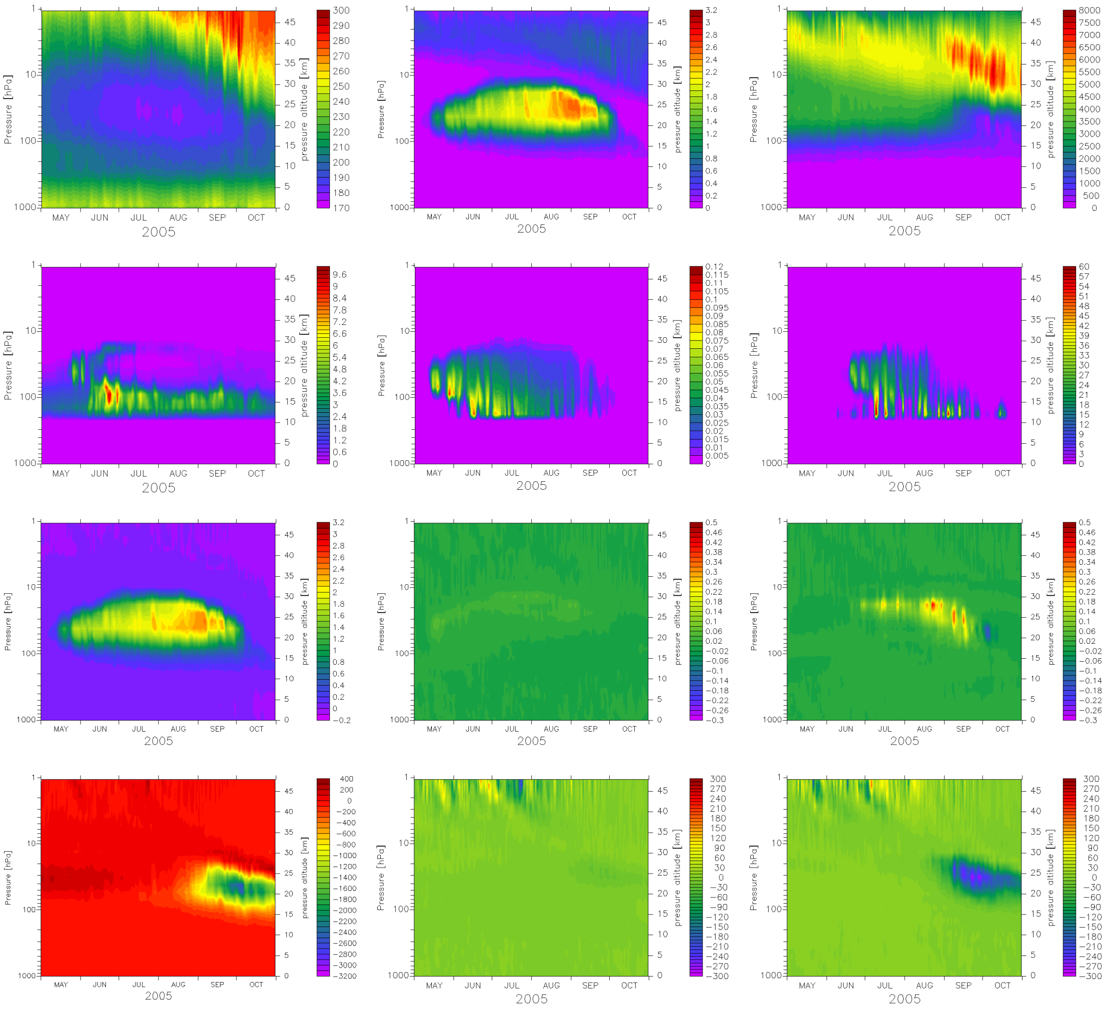

Figure 3: Time series averaged from 80° to 90°S for **2005**: First row: Temperature, $ClO_x$, and $O_3$ (all from Standard simulation); second row: Surface densities of liquid particles ($A_{LIQ}$), and NAT ($A_{NAT}$) (all from Standard simulation); third row: Contribution to chlorine activation from liquid particles, NAT, and ICE; bottom row: Contribution to ozone depletion from liquid particles, NAT, and ICE.

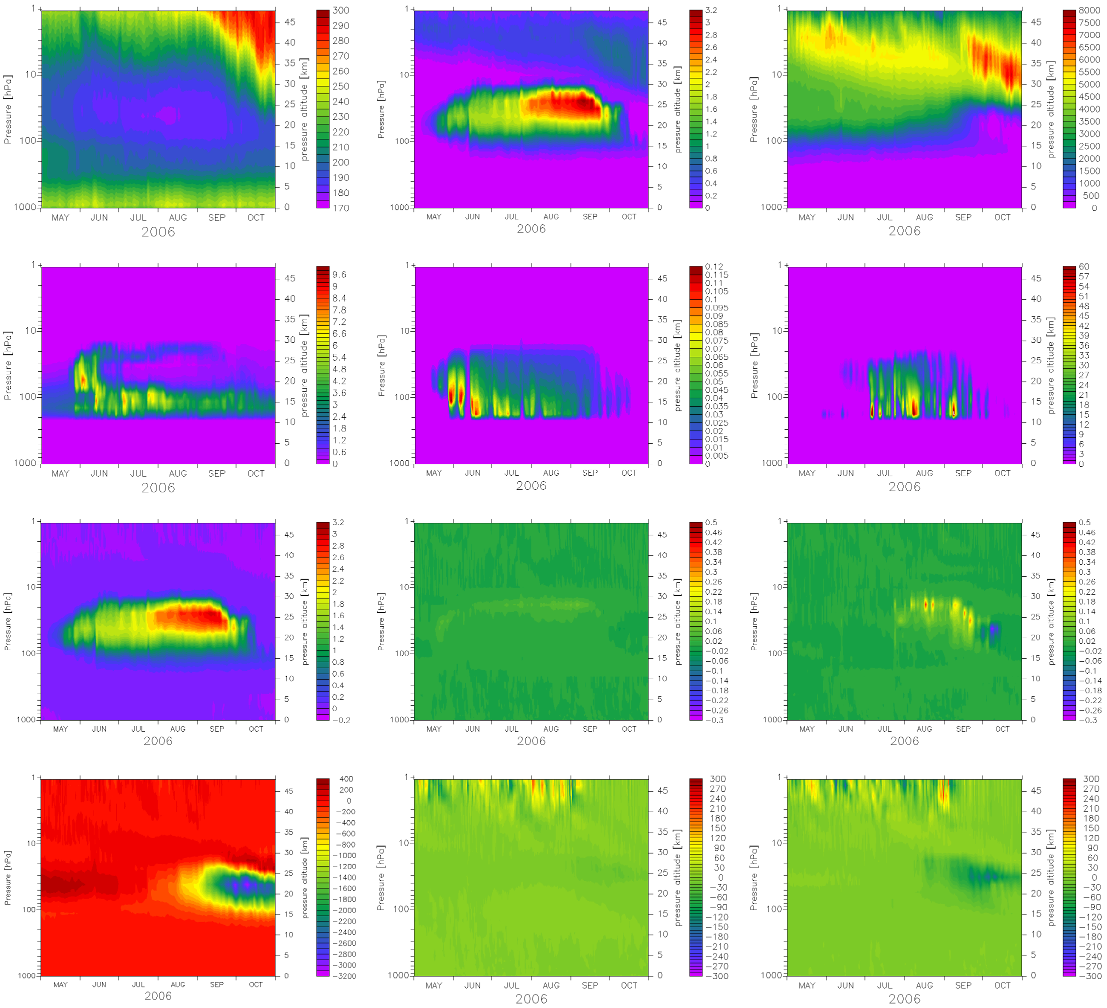

Figure 4: Time series averaged from 80° to 90°S for **2006**: First row: Temperature, $ClO_x$, and $O_3$ (all from Standard simulation); second row: Surface densities of liquid particles ($A_{LIQ}$), and NAT ($A_{NAT}$) (all from Standard simulation); third row: Contribution to chlorine activation from liquid particles, NAT, and ICE; bottom row: Contribution to ozone depletion from liquid particles, NAT, and ICE.

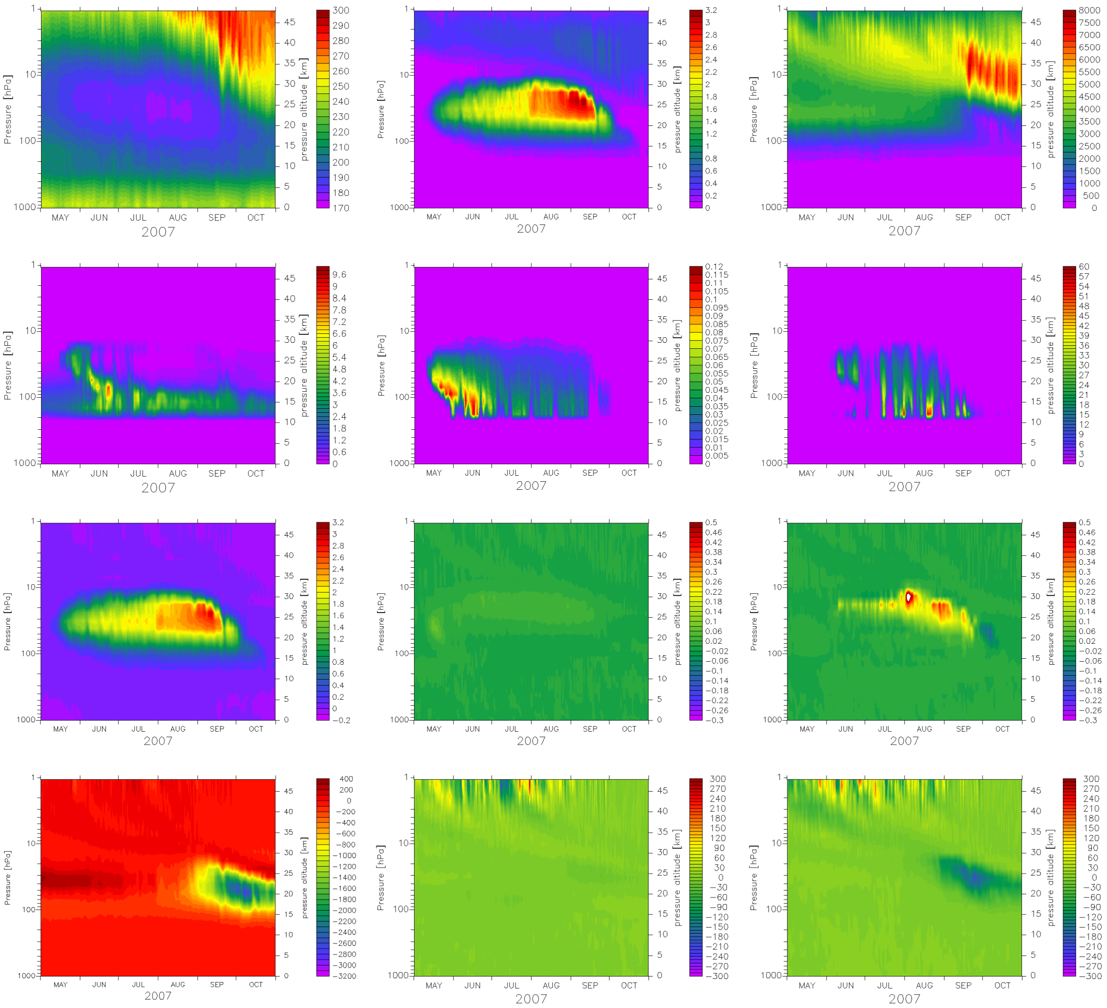

Figure 5: Time series averaged from 80° to 90°S for **2007**: First row: Temperature, $ClO_x$, and $O_3$ (all from Standard simulation); second row: Surface densities of liquid particles ($A_{LIQ}$), and NAT ($A_{NAT}$) (all from Standard simulation); third row: Contribution to chlorine activation from liquid particles, NAT, and ICE; bottom row: Contribution to ozone depletion from liquid particles, NAT, and ICE.

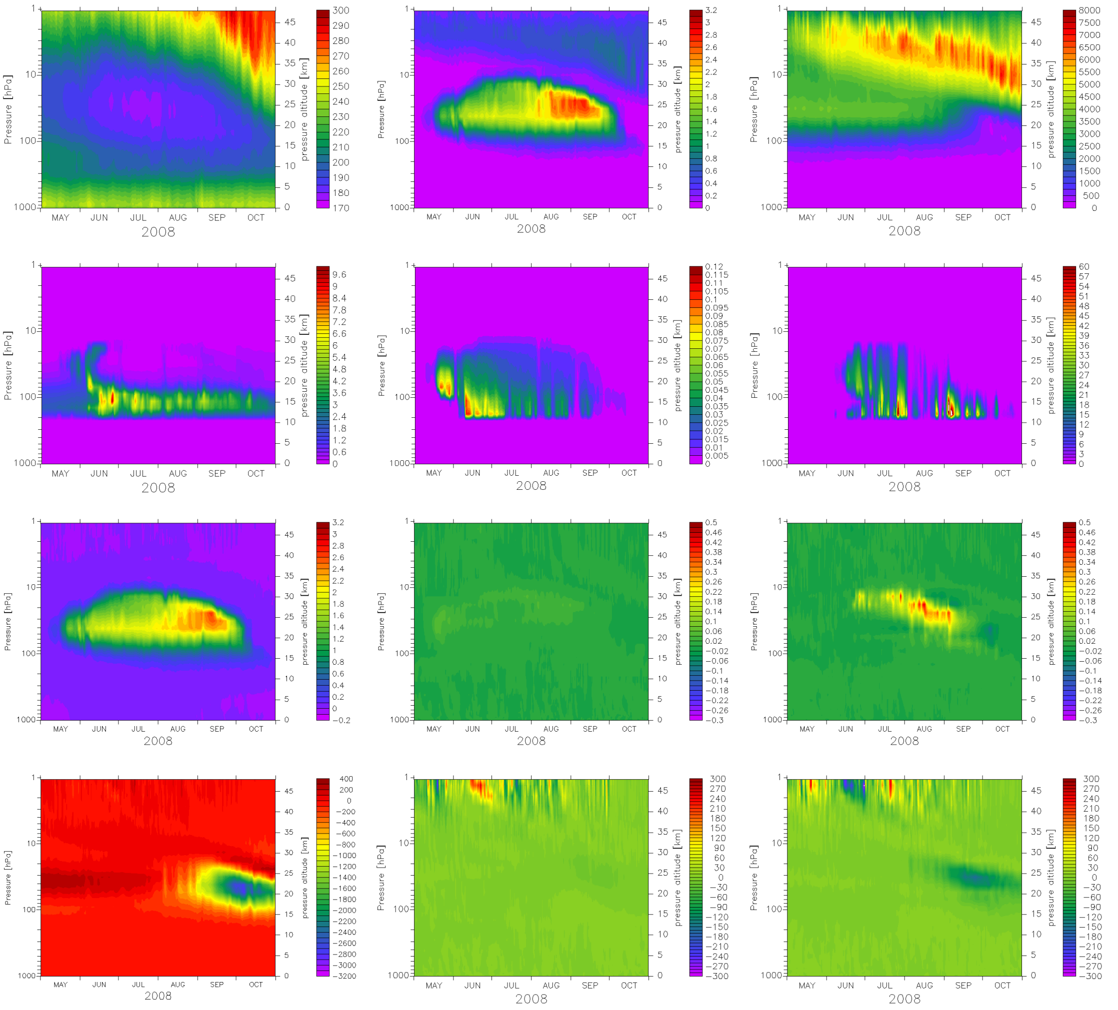

Figure 6: Time series averaged from 80° to 90°S for **2008**: First row: Temperature, $ClO_x$, and $O_3$ (all from Standard simulation); second row: Surface densities of liquid particles ($A_{LIQ}$), and NAT ($A_{NAT}$) (all from Standard simulation); third row: Contribution to chlorine activation from liquid particles, NAT, and ICE; bottom row: Contribution to ozone depletion from liquid particles, NAT, and ICE.

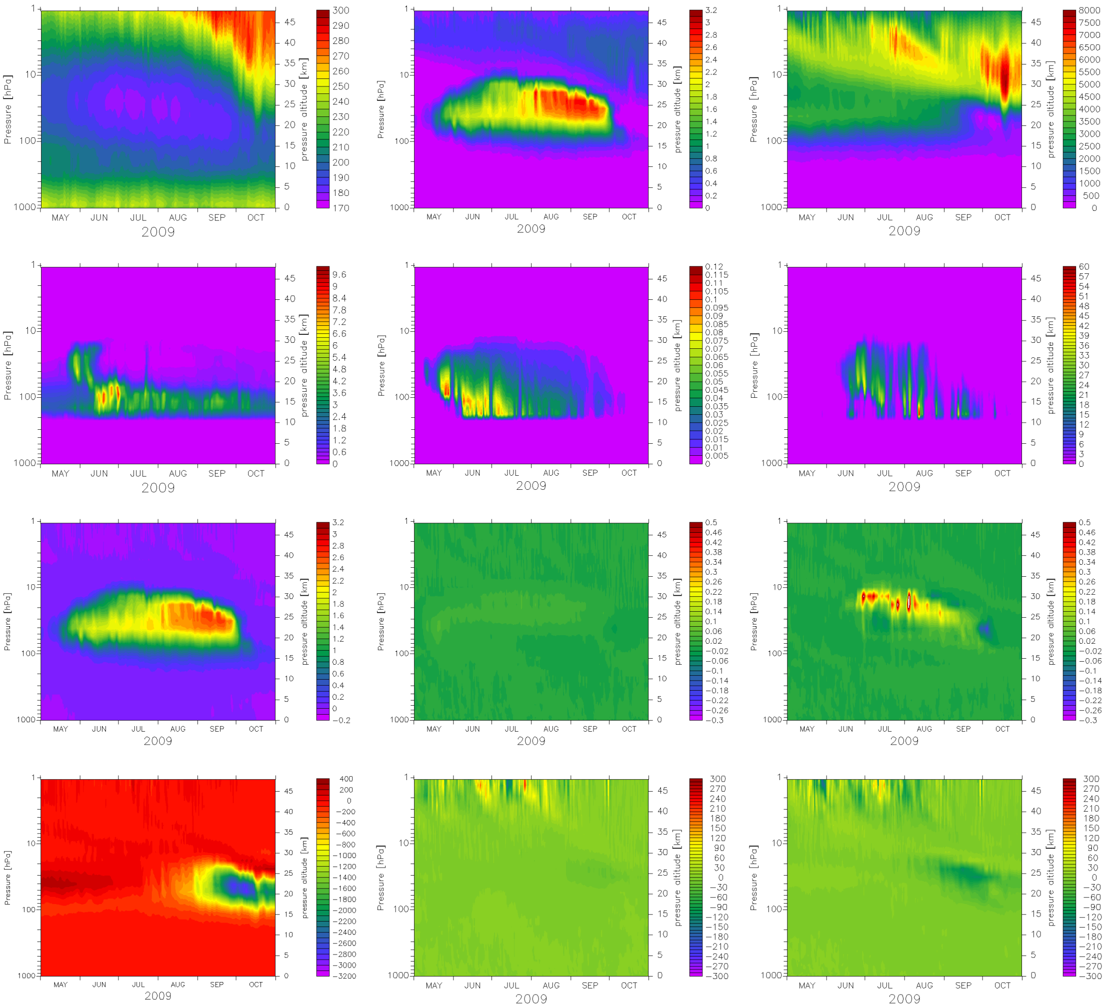

Figure 7: Time series averaged from 80° to 90°S for **2009**: First row: Temperature, $ClO_x$, and $O_3$ (all from Standard simulation); second row: Surface densities of liquid particles ($A_{LIQ}$), and NAT ($A_{NAT}$) (all from Standard simulation); third row: Contribution to chlorine activation from liquid particles, NAT, and ICE; bottom row: Contribution to ozone depletion from liquid particles, NAT, and ICE.