

AUTHORS' RESPONSE TO THE REFEREE 2 COMMENTS

We thank Referee 2 for helpful comments regarding improving our manuscript. Below are point by point replies to the particular issues raised.

General

The paper addresses an important scientific question, namely projections of future ozone levels in the Arctic. Overall the paper is well written and the discussions and arguments are clear (see below for some exceptions). The study uses a well established and well described model (the UM-UKCA model). The results on the timing of future Arctic ozone recovery are very relevant for the readership of ACP and also for future scientific ozone assessments. A particular strength of the paper is the use of ensembles and the analysis of relevant chemical and dynamical processes with a focus on the case of the simulation for winter 2063.

One major question to the models projecting the future is how well they simulate present day chemical Arctic ozone loss. Of course, this is a prerequisite for the assessment of future recovery. And it is not obvious that state-of-the-art models do a good job in all respects at present day chemical Arctic ozone loss. For example, Brakebusch et al. (2013) find a systematic high bias in ozone in the model of 18% in the lowermost stratosphere in March. They attribute most of this ozone bias to too little heterogeneous processing of halogens late in the winter and suggest that the model underpredicts ClONO₂ early in the winter and has too little activated chlorine. How is the UM-UKCA model doing in this respect? How well does the model simulate denitrification (which is important for Arctic ozone loss)?

The overall issue of model performance is addressed in the general authors' response. We have added further material to the manuscript that gives more detail on the overall performance of the model compared to observation/reanalysis data. We also note that all models exhibit biases. We also remind the reviewer that the objective here is to compare the UM-UKCA model for the present day with a period later this century, and the comparison is therefore internally consistent. In this case, model biases become somewhat less relevant.

Further, as far as I understand the UM-UKCA model uses an equilibrium NAT scheme, where NAT is formed at the NAT equilibrium temperature, which likely overestimates the onset of heterogeneous reactivity in the model. Are there earlier studies, where these points have been addressed?

Indeed, the UMUKCA simulates the NAT PSCs formation according to the equilibrium equation from Hanson and Mauersberger (1988; we now state it clearly in the manuscript). Yet, the use of equilibrium NAT scheme, although likely imperfect, is a common technique employed in CCMs (see e.g. Morgenstern et al., 2010).

In the model heterogeneous reactivity is driven by NAT and ice particles, i.e., what regards the Arctic largely by NAT. This is likely not realistic, as there are extensive observations of liquid particles in the polar regions (e.g., Pitts et al., 2013). Nonetheless, Keeble et al. (2014) obtain a reasonable simulation of the Antarctic ozone hole assuming heterogeneous reactions on NAT and ice

using the model employed here. Similarly, Grooß et al. (2011) used a set-up with a NAT dominated heterogeneous chemistry (likely not realistic) but were able to reproduce the observed extremely low ozone values in the Antarctic. Possibly, it is not necessary to get every detail of PSC formation right to obtain a reasonable representation of chlorine activation and ozone loss in the model (see also Kirner et al., 2015; Solomon et al., 2015, and references therein). But I suggest that the issue of heterogeneous reactivity is discussed in more detail in the paper (see also detailed comments below).

See below. We now acknowledge the relative simplicity of our scheme in Sect. 2.1, as well as rephrase the wording regarding the study of Keeble et al., 2014 (as suggested).

Moreover, an important theme of the paper is halogen induced ozone loss due to heterogeneous reactions and chlorine activation (and the relative role of dynamics). As sufficiently cold conditions develop almost exclusively in the polar vortex, halogen induced ozone loss is only expected to occur in the vortex. However the analysis in the paper is mostly based on geographical latitude thereby neglecting the distinction between inside and outside of the vortex (in Fig. 7a however, a vortex average is presented, see also comments below). For example, how different would Figure 1 look, if equivalent latitude would be used rather than geographic latitude?

Data at sufficiently high temporal resolution to convert diagnostics from geographical to equivalent latitudes are only available for one ensemble member. The use of only one ensemble member would limit our ability to explore the role of interannual variability, as is otherwise achieved by the use of ensemble simulations. Since the reviewer recognises the value of using a model ensemble for this study, we have retained the use of geographic latitudes. Note that we now give additional vortex averaged values for halogen induced ozone losses in the model case study years 2060/2063 in Sect 3.3.

Further, in the discussions on the ozone anomaly simulated for the year 2063, the distinction between vortex processes and out of vortex processes is not always brought across clearly (see detailed comments below). Form my reading of the discussion in the paper (top of page 14), in the model, a significant fraction of the 2063 anomaly is driven by chemistry outside of the vortex – is this correct? I suggest improving the discussion and carefully quantify the contributions of chemistry and dynamics inside and outside of the polar vortex to the simulated ozone anomaly in 2063.

These issues are addressed in general authors' response. As noted above, the vortex-average halogen induced ozone losses for the two case study years have been added to the manuscripts (Sect. 3.3). To avoid confusion, the 65-90°N passive ozone tracer diagnostic has been removed from the manuscript.

In summary, I think with respect to several issues raised in this review, the paper needs to be revised and improved. Nonetheless, I believe that this is a potentially very good paper, which could make an important contribution to improved projections of future Arctic ozone levels and in particular regarding the various processes impacting polar ozone. The paper will also be very relevant

to the upcoming new WMO ozone assessment.

Detailed comments

• p 1, l 15: This statement is confusing: to me it implies that present day spring Arctic ozone is 50-100 DU below the values expected after recovery in 2060. Is this what you want to say here? Is this true for your model simulations presented here?

We have clarified the text. The sentence implies that the extreme low ozone episodes past 2060 have similar ozone levels as the average values, where no strong polar ozone depletion occurs, routinely simulated at the present day conditions (Fig. 1a in the manuscript).

• p 1, l 20: why does an increase in downwelling lead to less consistency?

We have changed the text to "...there is less confidence in the projected temperature trends in the lower stratosphere (100-50 hPa). This is partly due to an increase in downwelling ...".

• p 2, l 2: The use of CFCs did not lead to the 'suggestion...'

We have corrected the text.

• p. 2, l 4: One should distinguish the issue raised by Molina and Rowland (1974) (upper stratospheric ozone, globally) from the ozone hole issue pointed out by Farman et al. (1985).

We do not think the text causes ambiguity.

• p 2., l9: a citation from 1997 does not really allow to say 'soon' with reference to Farman et al. (1985).

We have changed it to 'later'.

• p 3, l 1: the impact is also on ecosystems not only on human populations.

We have added that to the text.

• p 3, l 14: change 'sulphate' to 'cold sulphate'

We have corrected the text.

• p. 3, l 25: 'controlling' is perhaps to strong

We have changed it to 'substantial'.

• p 4, l 30: it might be worth pointing out that the mean age of the UMUKCA model is in relatively good agreement with the observations in the high latitudes of the Northern hemisphere (according to Chipperfield et al., 2014), which is the most important region for this study.

We have added the information about the UMUKCA age of air in the Northern Hemisphere high latitudes to the manuscript. We note it is more accurate to say here that the UMUKCA

mean age of air in the Arctic is at the lower end of the observationally derived values, as there is a spread of the mean age of air between different observational datasets (see SPARC, 2013).

• I am assuming that not only this reaction is not taken into account on liquid aerosol but all five reactions listed in on page 13707 of Keeble et al. (2014). While this is likely not realistic (there are extensive observations of liquid particles in the polar regions, e.g., Pitts et al., 2013) it should not affect the quality of the ozone loss simulations too much. Assuming that the details of heterogeneous reactivity are not essential for a good representation of polar ozone loss; see also discussion below.

As stated in the manuscript, the $\text{ClONO}_2 + \text{HCl}$ reaction is not included on liquid aerosols. The $\text{HOCl} + \text{HCl}$, $\text{ClONO}_2 + \text{H}_2\text{O}$ and $\text{N}_2\text{O}_5 + \text{H}_2\text{O}$ reactions are included in the scheme. The only other reaction listed in Keeble et al. (2014) that is not included on liquid aerosols is the $\text{N}_2\text{O}_5 + \text{HCl}$ reaction, and we have now added this information to the manuscript.

• p. 5, l 10-12: I do not think it is correct to say that Keeble et al. (2014) showed that ozone depletion can be attributed to heterogeneous reactions on NAT and ice. They obtain a reasonable simulation of the Antarctic ozone hole making this assumption. In the real world, for a long time, the heterogeneous reactivity will be dominated by ice particles. On the other hand, neglecting ice particles (and indeed NAT) does not result in a substantial change of the simulated ozone loss (Kirner et al., 2015; Solomon et al., 2015). So I think the wording should be more careful here.

We agree with the reviewer and have clarified the text.

• p 5, l 29: chemical formulas should not be in italics

We have changed the formatting.

• Section 2.3: I would suggest some more discussion of the relevance of the cycles discussed here. Could you roughly quantify what is meant with “lesser importance”. It could be close to negligible for some of the cycles I think. On the other hand close to the tropopause in the non activated region natural (e.g. HOx driven cycles might be important for ozone loss.

Some quantification of the contribution of individual cycles has been added to Sect.3.2.1. (see also our reply to the comment below).

• Figure 1: how different would this figure look, if equivalent latitude would be used rather than geographic latitude. Would this not be the better choice? What is the reason for preferring geographic latitude vs. equivalent latitude?

As explained above, the daily mean data needed to convert the diagnostics from geographical to equivalent latitudes are only available for one ensemble member. Using a single ensemble member would limit our ability to explore the role of interannual variability in our study.

• p 7, l 15: change ‘atmospheric’ to ‘stratospheric’ – Cly is not defined in the troposphere

We have corrected the text.

• p 7, l 20: *I do not agree. The paper by Haigh and Pyle (1982) does not discuss the relevant ozone loss cycles in the polar regions, in particularly the ClO-dimer cycle, which does not slow down with decreasing temperature. I think you are discussing polar ozone loss in the lower stratosphere here.*

Whilst we agree that Haigh and Pyle (1982) do not explicitly discuss the NH polar regions, the increased ozone levels in the tropical mid/upper stratosphere driven by the GHG-induced cooling would then be transported by the BDC to the high latitudes, thereby contributing to increased ozone levels there.

Nonetheless, to avoid ambiguity, we have removed the sentence in question from the manuscript.

• p 7, l 31: *If I understand correctly, this value is computed by determining the minimum ozone value poleward of 65° N each day and then computing the mean value over a month. Have you ensured that all these values are within the polar vortex? Or could some of these values stem from (dynamically caused) so-called mini-holes?*

The minimum ozone values are determined from the monthly-mean data, and we believe the wording in the text is clear. As dynamically induced ozone mini-holes are likely to be short-lived, we believe it is unlikely that these would have a substantial impact on monthly mean values.

• p 8, l 5: *What is the implication of this statement? This sentence could be interpreted as stating that under present day conditions routinely strongly depleted ozone values are found. Please clarify.*

We have attempted to clarify the text by adding ‘average’ in front of ‘values’. We believe it is clear that the word ‘routinely’ refers to average values, and does not refer to extremely low ozone events associated with the extremely cold Arctic winters.

• P. 8, l 25-28: *Difference to the results of Langematz et al. (2014); if the reason is ‘differences in the representation’, do you mean chemical or dynamical effects? If you agree with me that the difference is very likely not due to chemistry, you could state this point more clearly.*

We have changed ‘due to differences in the representation’ to ‘due to differences, likely dynamical, in the representation’ in that sentence.

• p 9, l 6: *This is a bit misleading – are there more, even less important halogen cycles? I suggest stating which of the six cycles are dominant, which play a minor role and which are negligible.*

The wording has been corrected to ‘six halogen cycles of most importance in the polar lower stratosphere’.

A general definition of the dominant and minor halogen cycles is in Sect. 2.3 where the halogen induced ozone loss diagnostic is defined. We have now added some extra information about the contribution of individual halogen cycles to the cumulative loss into Sect. 3.2.1.

• *p. 9, l 21: do you really mean ‘halogen losses’ here?*

Corrected to ‘halogen induced ozone losses’. Also, we have added an explanatory sentence at the beginning of Sect. 3.2.1 stating that the terms ‘halogen induced ozone loss’ and ‘halogen loss’ are henceforth used interchangeably in reference to this diagnostic.

• *p 9, l 26: this formulation is a bit awkward; I think you never applied the 11-year running mean rather than removing it.*

We believe the formulation is correct. For clarity, we have changed ‘11-year running mean’ to ‘11-year running ensemble mean’ (both in the text and the caption to Fig. 3b).

• *p 10, l 3: not only the amount of PSCs also the length of the cold period. This is also important (Manney et al., 2011). Further below you also make this point.*

We have added this point to the text.

• *p 10, l 18: change ‘insignificant’ to ‘not significant’*

We have corrected the text.

• *p 10, l. 19: “Similar is true” – reformulate*

We have reformulated the sentence.

• *p 10, l 27-29: Actually, the ozone levels in Antarctica are also strongly influenced by the BD-circulation; it is just that the dynamical variability is lower – correct?*

We agree and have clarified the text.

• *p 11, line 1: provide a citation and/or explanation for w*

We have now added a citation and explanation to the text.

• *p 11, l 15: ‘relatively’ to what?*

Relative to the long-term trend, as well as to the range of variability found during the preceding and following periods.

• *p. 11, l 21-22: A central issue here is also the continued presence of PSCs and thus the continued activation.*

We agree and have added this point to the text.

• *p 11, l 32: will continue to occur in the future . . .*

We have corrected the text.

• p 12, l 10: “long-term minimum of ensemble mean” – this is not quite clear? Which period is exactly considered? And what is meant is the lowest value for March mean ozone in the ensemble?

We believe the sentence is clear. As stated in the text, the period considered is the late 1990s (We have now clarified this by replacing ‘in the 1990s’ with ‘found in the 1990s’). We do not discuss ‘the lowest value for March mean ozone in the ensemble’ but the lowest value of the ensemble mean.

• p 12, l 16: This effect could be reduced by considering equivalent latitude.

We agree with the reviewer, but we have chosen to show vortex-averages to illustrate this instead. This approach also reduces the contribution of extra-vortex air, although we note the relative simplicity of the vortex definition used (see below).

• p 12, l 17-19: Why did you choose 850 K to define the vortex? This is above the altitude where most halogen induced ozone loss occurs. Also how is the PV value defined (citation?)?

Although the 850K level is above the main region of halogen induced ozone loss, it is a common measure for defining the polar vortex. We also note that our study examines both dynamical and chemical processes. Therefore, the choice of only a single level as representative of a polar vortex edge is, by definition, imperfect. The threshold is based on a rough estimate of where the maximum PV gradient occurs at this level. (See also figures R4-R5 in the general response that show stereographic maps of key quantities over the polar cap).

• p 12, l 20: I suggest showing the vortex average data.

As suggested by the reviewer, we have added the vortex-average total column ozone data for 2060 to Fig. 7a. (now Fig. 8a in revised manuscript)

• p 13, l 9: this is an important point that should also be brought across clearly in the abstract.

It was not clear to us which point on P13 L9 the reviewer was referring to. Since we do not give specific details of the model case study years in the abstract, we do not feel it would be fitting to add in a specific detail of this kind to the abstract. However, we emphasise that the increasing importance of dynamical processes for Arctic springtime ozone in the future is stated at the end of the abstract: "Whilst our results suggest that the relative role of dynamical processes for determining Arctic springtime ozone will increase in the future, halogen chemistry will remain a smaller but non-negligible contributor for many decades to come."

• p 13, l 18: as stated before, heterogeneous reactivity in general should be more important than NAT formation in particular. Also, is there any formation of ice particles in the model for the year 2063?

As discussed in Sect. 2.1 of the updated manuscript, the heterogeneous reactivity in the NH polar regions in these experiments is likely to be dominated by NAT PSCs.

The November-March mean volume of ice (1-25 km) in the model year 2063 is $\sim 27 \times 10^6 \text{ km}^3$.

• p 14, l 2-4: it is interesting to note that only part of the effect of the anomaly has its origin in processes in the polar vortex. Doesn't this mean that in the model only part of the chemistry driven effect is caused by halogen chemistry? Again I suggest to bring this message more clearly across the the abstract.

These issues are addressed in general authors' response (see also below).

We note that the chemical ozone loss diagnostic derived from the passive ozone tracer results from a complex balance between chemical loss and production cycles as well as their interaction with transport throughout the winter.

To avoid confusion, we now state it clearly in the text; we have also removed the 65-90°N passive ozone tracer diagnostic from the text (and replaced with vortex-average quantity in the former Fig. 7a (now 8a)), as well as reformulated parts of the last two paragraphs of Sect. 3.3. Lastly, the vortex-average halogen induced ozone losses for the two case study years have been added to the manuscript (Sect. 3.3).

• p 14, l 9: here you state that the halogen effect is 40 DU but above you state that the polar vortex effect is only 25 DU. Does this mean that in the model, a significant fraction of the 2063 anomaly is driven by halogen chemistry outside of the vortex? Is there chlorine activation outside of the vortex in the model? This discussion at this stage and the attribution of ozone loss to processes needs to be improved.

As above, these issues are addressed in the general authors' response.

In the original version of the manuscript we state that, indeed, halogen induced ozone loss in the 1-25 km layer is ~ 40 DU. The chemical loss, as derived from the passive ozone tracer, due to all cycles integrated from the surface to the top of the atmosphere, vortex-averaged, is ~ 25 DU. Note that the two diagnostics are not equivalent. The chemical ozone loss diagnostic derived from the passive ozone tracer results from a complex balance between chemical loss and production cycles as well as their interaction with transport. To avoid confusion, we now state it clearly in the text, as well as modify the manuscript as described above and in the general authors' response.

Note also that the vortex-average halogen induced ozone losses for the two case study years (now added to the manuscript, but see also the general authors' response), show mostly similar or somewhat higher values than the Arctic mean values.

• p 14, l 21: citation for 'other studies'

We have added the requested citation.

• p 15, l 31: it is not clear to me where the number of 20% is coming from. on p. 14, you report that the halogen induced loss in 2063 is twice that of 2060, which might be the first order information of interest here. However, to me the question is still open of how much the difference between 2060 and 2063 is a polar vortex effect and in how far it is influenced substantially by out of vortex processes.

The difference in the 65-90°N halogen induced ozone loss between the two model years is ~20 DU (we have added this to the manuscript). The difference in the total ozone column by the end of March between the two years is ~100 DU. Hence, the difference in the estimated halogen loss in the polar lower stratosphere equates with ~20% of the full ozone difference. We have now changed 'can therefore account for' to 'is equivalent to'.

Vortex-average halogen induced ozone losses for the two case study years have been added to the updated manuscript.

See also the general authors' response.

REFERENCES

Haigh, J. D., and Pyle, J. A.: Ozone perturbation experiments in a two-dimensional circulation model, Q. J. Roy. Meteor. Soc., 108, 551-574, 10.1002/qj.49710845705, 1982.

Keeble, J., Braesicke, P., Abraham, N. L., Roscoe, H. K., and Pyle, J. A.: The impact of polar stratospheric ozone loss on Southern Hemisphere stratospheric circulation and climate, Atmos. Chem. Phys., 14, 13705-13717, doi:10.5194/acp-14-13705-2014, 2014.

SPARC: SPARC Report on the Lifetimes of Stratospheric Ozone-Depleting Substances, Their Replacements, and Related Species, SPARC Report No. 6, 2013.

Morgenstern, O., et al., Review of the formulation of present-generation stratospheric chemistry-climate models and associated external forcings, J. Geophys. Res., 115, D00M02, doi:10.1029/2009JD013728, 2010.

Hanson, D. and Mauersberger, K.: Laboratory studies of the nitric acid trihydrate - Implications for the south polar stratosphere, Geophys. Res. Lett., 15, 855-858, 1988.