AUTHORS' RESPONSE TO THE REFEREE 1 COMMENTS

We thank Referee 1 for helpful comments regarding improving our manuscript. Below are point by point replies to the particular issues raised.

This paper deals with the recovery of Arctic ozone in a future climate under increasing greenhouse gas concentrations and declining inorganic chlorine. In contrast to earlier studies the authors use an ensemble mean of seven transient simulations to capture the interannual variability in Arctic ozone. The special focus is on the possibility of individual years with strong ozone depletion even after 2060 when halogen loading has become relatively low. I find the paper appropriate for publication in ACP after my minor suggestions have been considered and my questions have been clarified.

General comments:
 The authors need to be very careful with the references in the text pointing to the single figures. There are some mistakes which can confuse the reader.

 It need to be clearly stated that the years '2060' and '2063' are model years coming out of a not nudged simulation and are therefore relatively arbitrary. In some parts it sounds as if we get a really strong ozone loss in the future year 2063 and a really weak one in 2060. This needs to be clarified.

 The authors have included a lot of citations in their manuscript and compare their results with many of these studies. In some cases they need to be more specified. In my opinion some studies do not exactly show what is stated here.

 There are a lot of typos in the references. A cross check should be done before final publication.

These points are addressed below.

Specific comments:
 Page 4, line 27: How are orographic and non-orographic gravity waves parameterized? Please provide a reference.

We have added this information to the manuscript.

 Page 5, lines 15 - 17: I don't really understand why these six year bins are excluded. The supplement only shows which years are affected. Please provide some more information on this, here or in the supplement.

For these periods there was an error when the model was run and we do not have sensible data.

 Page 6, line 11: 'In all six reactions, a net loss of 2 odd oxygen molecules occurs per cycle.' → Do you mean 2 molecules ozone? But in cycle 3 there is only one. Please clarify this.

We have clarified this in the manuscript.

Each cycle leads to net loss of 2 odd oxygen molecules (O_x=O_3+$O(^3P)$+$O(^1D)$). As discussed in Lee et al. (2002), the concentrations of ozone are significantly larger than of atomic oxygen, in particular in the polar lower stratosphere in winter/spring (which is the region studied here), therefore, we assume that $[O_x] \approx [O_3]$ and, thus, $d[O_x]/dt \approx d[O_3]/dt$.

□ ***Page 6, line 24: 'averaged from 65°N to the pole.' → Why do you use exactly 65°N 90°N? Have you also tested other latitudes, for example 60°N - 90°N and does this change the results?***

The 65-90°N average was chosen as a compromise between capturing a proportionally large fraction of the polar vortex while minimising the proportion of the extra vortex region, and including the edges of the vortex where in early winter halogen activation will take place preferentially (as there is little sunlight at higher latitudes).

□ ***Page 7, line 6: see comment above***

As above.

□ ***Page 10, lines 17 - 19: '...in agreement with Langematz et al. (2014).' → This statement should be specified. Do you compare with Figure 2a from Langematz et al. (2014)? From this figure I see a significant trend at 100 hPa, which is not the same as in your study. Moreover, you have to note that the time ranges are not identical.***

We have changed the text. ("In comparison, Langematz et al. (2014) found a statistically significant cooling trend in early winter over 1960-2100 throughout the polar stratosphere.")

□ ***Page 11, line 26: 'This is in broad agreement with the findings in Langematz et al. (2014).' → Where do you get this from? The focus in their study is on the vortex duration and not on the zonal wind trend. You need to be more specific with your comment.***

We have changed the text ("Langematz et al. (2014) analysed the timing of the formation of the NH polar vortex and found a statistically significant trend towards earlier vortex formation. It is possible that the strengthening of the stratospheric zonal wind in autumn/early winter in our ensembles could be related to a similar effect. ").

□ ***Page 12, line 3: Maybe you can call Section 3.3 'Case studies of exceptionally low and high ozone events' as you show results from both - low and high - and not only from low ozone events. This should be changed also in the Introduction (page 4, lines 5 - 7).***

We have changed the text to "Case study of exceptionally low and average ozone events".

□ ***Page 12, Section 3.3.: As you use free running, and no nudged model simulations, you won't expect that your 'model' years resemble 'real' years. Please make sure that the 'years' 2060 and 2063 are 'model' years. Do you really need this numbers? Maybe you can skip them and refer to low and high ozone events.***

We have added an explanatory sentence, and we also now refer to 'model years'.

□ ***Be very careful with the references on the figures:***

- ***Page 12, lines 25 - 26: ...(see Fig. 7(b) and 8 (b)).***
- ***Page 12, line 30: (Fig. 8(b)) and not 8(a)!!!***
- ***Page 12, line 31: (Fig. 8(b)) and not 8(a)!!!***
- ***Page 13, line 1: (Fig. 8(b)) and not 8(a)!!!***
- ***Page 13, line 26: (Fig. 9(c)) and not 9(b)!!!***

Thank you for spotting this, we have corrected the text.

 Page 15, line 31: '...account for ~20%...' → This is a very crude estimate. Please be more specific.

We believe that this level of accuracy is adequate. Given the limitations of the diagnostics used, it is more appropriate to give an order of magnitude estimate than a precise (but not necessarily accurate) number.

Technical corrections:
 Page 2, lines 9 - 10: reference for 'Montreal Protocol on Substances that Deplete the Ozone Layer'

We have clarified that this is an international treaty.

 Page 3, line 19: ... volume of PSCs (VPSC) → the abbreviation 'PSCs' has been introduced in line 9

We have corrected the text.

 Page 4, line 1: Stratosphere -troposphere Processes And their Role in Climate (SPARC)

We have corrected the text.

 Page 4, line 2: Please provide a reference for CCMI.

We have added the requested reference.

 Page 4, line 16: ... the recent SPARC Report on the Lifetimes of ... (SPARC, 2013;...) → Be careful that this is in line with the citation on page 21, line 14f.

We have corrected the text.

 Page 4, line 24: The dot at the end of the sentence is missing.

We have corrected the text.

 Page 5, line 13: You may introduce an abbreviation for 'sea-ice concentrations' here and use it on page 8, lines 18 and 27.

We have added the abbreviation.

 Page 5, line 17: ...long periods are excluded...

It is the 'total' that 'is' excluded.

□ ***Page 5, line 23: ... and a more minor ClO + O(3P) cycle... → You should include (Cycle 3, reference) as before.***

We have added this.

□ ***Page 11, lines 3 - 4: The references should be sorted by year.***

We have corrected the text.

□ ***Page 11, lines 29 - 30: ...(see also Langematz et al., 2014).***

We have corrected the text.

□ ***Page 12, line 12: ... higher than in model year 2063.***

We have corrected the text.

□ ***Page 12, line 15: use the abbreviation 'BDC', as introduced before***

We have corrected the text.

□ ***Page 13, line 17: ... ClO concentrations in 2063 compared to 2060 (Fig. 9(c)). → The figure shows a difference and not the concentrations in 2063.***

We have corrected the text. Also, we have added a figure showing the evolution of ClO at 21.5 km in the two case study years to the supplement.

□ ***Page 15, line 26: ...'exemplified by a case study in 2063.' → Either you include 'model year' here, or you skip the year. In the Conclusions I would prefer to skip the years and use 'low and high ozone events instead.***

As suggested, we no longer use “2063” and “2060” in Sect. 4 (i.e. Conclusions).

□ ***Page 15, line 32: '...in year 2063 and a year from the same period ... ' → Better: '...between this year and a year from the same period ...'***

Changed to “between this low ozone year and a year from the same period with near average springtime ozone”’

□ ***Page 17, line 4: ... Steil, B.; and Tian, W....***

We have corrected the text.

Page 17, line 5: The dot is missing at the end of the reference.

We have corrected the text.

Page 17, line 19: Drdla, K., and Müller, R.:...

We have corrected the text.

Page 19, line 10: ...Oberländer, S., ...

We have corrected the text.

Page 21, line 30: Tilmes, S., Müller, R., ...

We have corrected the text.

Page 22, line 5: ... and Müller, R.: ...

We have corrected the text.

Page 24, line 4: ... 11-year running average, respectively.

We have corrected the text.

Page 24, line 5:... 2060 and 2063, respectively, described in Sect. 3.3.

We have corrected the text.

Page 27, line 2: ... 11-year running average, respectively.

We have corrected the text.

Page 27, line 6: 'As in Figure 4, ...' → I would prefer an independent figure caption for Figure 6, as the only agreements with Figure 4 are the pressure levels and the meaning of the points and bars.

We have added an independent caption for former Fig. 6 (now Fig. 7).

REFERENCES

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Lee, A. M., Jones, R. L., Kilbane-Dawe, I., and Pyle, J. A.: Diagnosing ozone loss in the extratropical lower stratosphere, J. Geophys. Res.-Atmos., 107, NO. D11, 4110, doi:10.1029/2001jd000538, 2002.

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) is 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 $ClONO_2$+HCl reaction is not included on liquid aerosols. The HOCl+HCl, $ClONO_2$+H_2O and N_2O5+H_2O 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 N_2O_5+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 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 ~27×10^6 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 that 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 is 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.

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AUTHORS' RESPONSE TO THE REFEREE 3 COMMENTS

We thank Referee 3 for helpful comments regarding improving our manuscript. Below are point by point replies to the particular issues raised.

This article is based on an interesting study of the projection of future Arctic ozone using ensemble simulation from the UM-UKCA chemistry climate model. While other studies have been performed on this subject (e.g. WMO, 2011; Langematz et al., 2014), the originality of the study lies in the use of ensemble simulation, which allows the authors to estimate the intrinsic variability of the stratosphere, together with the impact of ozone depleting substances decrease and climate change on Arctic ozone. The paper is well written and informative for the projection of future Arctic ozone and I recommend publication in ACP, provided that important comments for improvement are taken into account.

Main comments

The main focus of the study is on the respective contribution of chemistry and dynamics on future Arctic ozone. In that respect diagnostics have been set up in order to evaluate the importance of chlorine chemistry in future ozone loss and the authors argue that halogen chemistry can still play a substantial role after mid-century. Since this result is rather intriguing, it deserves more attention in the article. A whole section is dedicated to the case study of winter 2063 but it is somewhat descriptive and does not demonstrate fully that the chemical loss is linked to halogen chemistry. For example, is the observed loss coherent with the known relationship between Cly levels, chlorine activation and PSC volume (e.g. Rex et al., 2004)?

An analysis of potential Vpsc vs. halogen induced ozone loss has been added to the updated manuscript (Sect. 3.2.3), where we also compare the model results to the study by Rex et al. (2004, 2006). As illustrated by the red star in Fig. 5 in the manuscript, the relationship between potential Vpsc and halogen loss simulated in the case study model year 2063 compares well with the fit to the ensemble data for that period. Also, we have added Fig. S3 shown below to the supplementary material to illustrate the evolution of Arctic mean ClO, Cl_2O_2, HCl and $ClONO_2$ for the two model case study years.

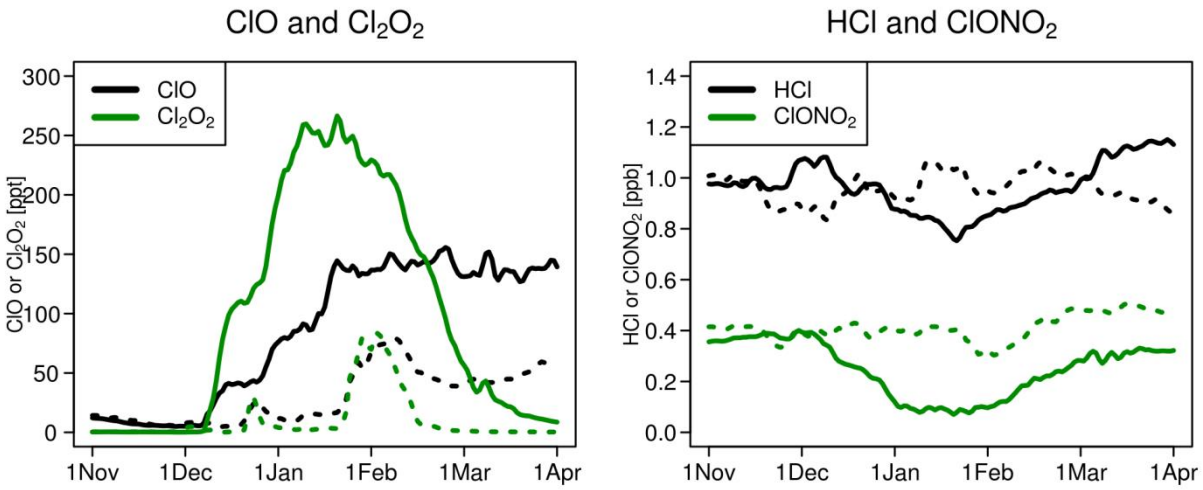

Figure S3. Timeseries of 65-90°N daily mean ClO and Cl_2O_2 [ppt] (left) and HCl and $ClONO_2$ [ppb] (right) at 21.5 km for the case study model years 2063 (solid lines) and 2060 (dashed lines).

What is the role of nitrogen chemistry that can sometimes be important in the Arctic mid-stratosphere as shown in Kuttipurath et al., 2010?

We have estimated the 65-90°N cumulative (1.Nov-30.Mar) ozone loss in the lower atmosphere (1-25 km) due to the $NO_2 + O(^3P)$ reaction to be ~2DU/4DU in the model years 2063/2060, respectively.

While we agree that all ozone loss cycles (HO_x, NO_x… etc) are important for the evolution of ozone (we now emphasize this in later part of Sect. 3.3), especially outside of the polar vortex and/or in the mid-/upper stratosphere, the focus of our study is on ozone losses due to halogen chemistry in the lower stratosphere. To avoid confusion we have removed the 65-90°N average chemical ozone loss diagnostic from the manuscript, leaving only the vortex-average quantity.

A quantification of PSC volume and a figure similar to Figure 2 but showing observations in order to demonstrate the skills of the model to simulate halogen chemistry would be useful.

An analysis of potential Vpsc vs. halogen induced ozone loss in the model has been added to the updated manuscript (Sect. 3.2.3), where we have made a comparison to the studies of Rex et al. (2004; 2006). While a comparison of the modelled total ozone column with observations was presented in Fig. 1(a), we have now added additional material (Sect. 2.1) that discusses present day ozone/ClO from a similar version of the model with 'nudged' meteorology and from observations (see the general authors' response). For the future period, it is of course impossible to compare our model results to observations.

Since the Arctic ozone loss is computed over the 65-90◦ N latitude range, it encompasses some loss from non-vortex air. This issue is acknowledged by the authors but would need some quantification.

The estimated vortex average halogen induced ozone losses for the two case study years 2063/2060 have been added to the updated manuscript (last paragraph in Sect. 3.3). See also the general authors' response.

From Figure 1, it seems that the interannual variability of Arctic ozone from the ensemble simulation is larger than the natural variability as seen from the observations. Can the authors comment on that and provide some statistics on this issue?

We do not agree. The variability in the observed ozone column appears comparable to or even somewhat larger than in our model.

In addition a more substantial description in section 2 of the skills of the UM-UKCA model in terms of polar ozone simulation is needed: e.g is there a cold bias of the polar stratosphere? How the strength and duration of the Northern vortex compare with observations, . . .?

A comparison of Northern hemisphere climatological stratospheric zonal wind and temperatures with reanalysis data has been added to the revised manuscript (Sect. 2.2). We have also added a comparison of present day polar ozone/ClO for a "nudged" meteorology version of UMUKCA with satellite observations.

See also the general authors' response.

Temperature trends: No mention is made of the evolution of the occurrence of sudden warmings in the ensemble simulation. It is thus difficult to distinguish radiatively induced with dynamically induced temperature trends. This issue should be addressed.

While we acknowledge that changes in the frequency of sudden stratospheric warmings (SSWs) may be important for determining polar temperature trends, a quantitative distinction between the dynamically and radiatively-induced temperature trends is beyond the scope of the study. In addition, data at sufficiently high temporal resolution (i.e. daily) required to calculate SSW occurrences are only available for one ensemble member, and this is unlikely to be adequate for diagnosing statistically robust changes in SSW frequency.
We have now changed the sentence 'The ensemble shows a radiatively-driven cooling trend…' in the abstract to 'The ensemble shows a significant cooling trend…'.

Minor comments
P5 l24: it is not clear how the 11-year solar cycle is simulated over the 21st century.

Solar cycle variability for the future period (after 2009) is included as in earlier periods but with a repeating sinusoidal 11-year cycle with an amplitude derived from observed cycle 23 (see Jones et al., 2011; Gray et al., 2013). We have added this information to the revised manuscript.

P7 l21: the products of the reaction are wrong: it should be Cl + O2.

Thank you for spotting this, but note that this has already been corrected after the initial quick referees' reviews and prior to the publication in ACPD.

P6 l4: This sentence is not so clear. The computational efficiency relates to the use of the diagnostics for evaluating halogen induced ozone loss.

We have improved the language by replacing 'computational efficiency' with 'computational ease'. Also, since we now include additional vortex-average halogen induced ozone loss diagnostics (see above and the general authors' response), we have added "…(except for the vortex-averaged quantities reported in Sect. 3.3, where daily means are used)…" to the sentence.

P9 l21: what is the contribution of slowing of gaz phase ozone loss cycles compared to changes in stratospheric transport in the earlier recovery of Arctic ozone?

Referee 2 also questions this point. We have removed this sentence as it is clearly confuses the reader.

P11 l4-13: In line with my major comments, the causes of the drops of Arctic ozone in late century, and the comparison with Langematz et al. (2014) study should be better substantiated.

We have now added more quantitative material, relating, e.g., potential Vpsc to halogen induced ozone loss; plus evolution of ClO_x and chlorine reservoirs for the model years 2063/2060 (see Figure S3).

P12 l10-18: a chemical loss of 40 DU is similar to the current Arctic ozone losses, while chlorine levels in 2061-2080 will be lower by more than a factor of 2. What PSC volume is necessary for such extreme loss?

The potential Vpsc calculated for the model case study year 2063 is now shown as the red star in Fig. 5 of the revised manuscript, and some text referencing to this is in Sect. 3.3.

P16 l13: what is the justification for the PV value to define the vortex?

This is a simple and fairy arbitrary choice based on a rough estimate of the maximum PV gradient. We are simply trying to be illustrative here. A similar approach has been used in other studies e.g. Müller et al (2005).

Figure 5: Case study years (2060 and 2063) should be highlighted in the figure.

We have added these as points to the figure (now Fig. 6).

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Authors' general response

We thank the three anonymous Reviewers for their helpful comments regarding improving our manuscript. We reply to the individual comments in the separate responses. In addition, Reviewers 2 and 3 raised a couple of common questions. These are in turn addressed below.

1. How well does the UMUKCA model simulate the present day Arctic ozone loss?

This will depend both on how well the model's chemistry scheme performs and how well the model reproduces the Arctic meteorology. These two factors are addressed below.

The performance of the UMUKCA CheS+ chemistry scheme can be evaluated using a similar model version but with a so-called specified dynamics set-up, i.e. in which model's temperature and winds are nudged towards meteorological reanalysis data. A comparison between the Arctic/Antarctic mean (65-90°N/S) total ozone columns in this nudged CCMI REFC1 CheS+(SD) integration[1] shows a good correlation between the simulated and observed ozone (Bodeker et al. 2005; Müller et al., 2008) (see Fig. S1). In the Antarctic, there is some overestimation of the ozone column in the model by up to ~15-30 DU (see Fig. S1).

For the cold March 2011, the model shows a positive bias in the zonal mean monthly mean ozone levels in the Arctic lower stratosphere (30-100 hPa) of up to ~0.9 ppb (~41%; for the 70-90°N mean, 50 hPa) compared with MIPAS satellite data (Fisher et al., 2008, Fig. R1). The positive ozone bias is commensurate with a negative bias in the zonal mean monthly mean ClO levels at this altitude (not shown). Note that there is some uncertainty in the observed zonal mean monthly mean ClO, in part due to sparse temporal sampling (twice a day) of the MIPAS instrument.

In general, all models will show some biases with respect to observations. Importantly, as in many studies, we compare the model for the present day and future periods in an internally consistent way; therefore, any biases will become less relevant for our study.

Regarding the NAT PSC formation and removal, as described in Chipperfield et al. (1999), the NAT PSC formation follows the equilibrium expression from Hanson and Mauersberger (1988). Although potentially imperfect, the use of equilibrium NAT scheme is a common technique employed in CCMs (see e.g. Morgenstern et al., 2010). The denitrification scheme assumes a relatively slow NAT sedimentation velocity of ~40 m/day for pure NAT PSCs, and a much faster NAT sedimentation velocity of ~1540 m/day in the presence of ice; the latter assumes coating of NAT PSCs onto ice particles.

Parts of the above information have been added to Sect. 2.1 of the revised manuscript. Figure S1 has also been added to Supplementary Information.

[1]We thank Paul Telford for preparing and running the nudged UMUKCA CCMI REFC1 CheS+(SD) integration discussed above (see also the updated Manuscript for full list of acknowledgements).

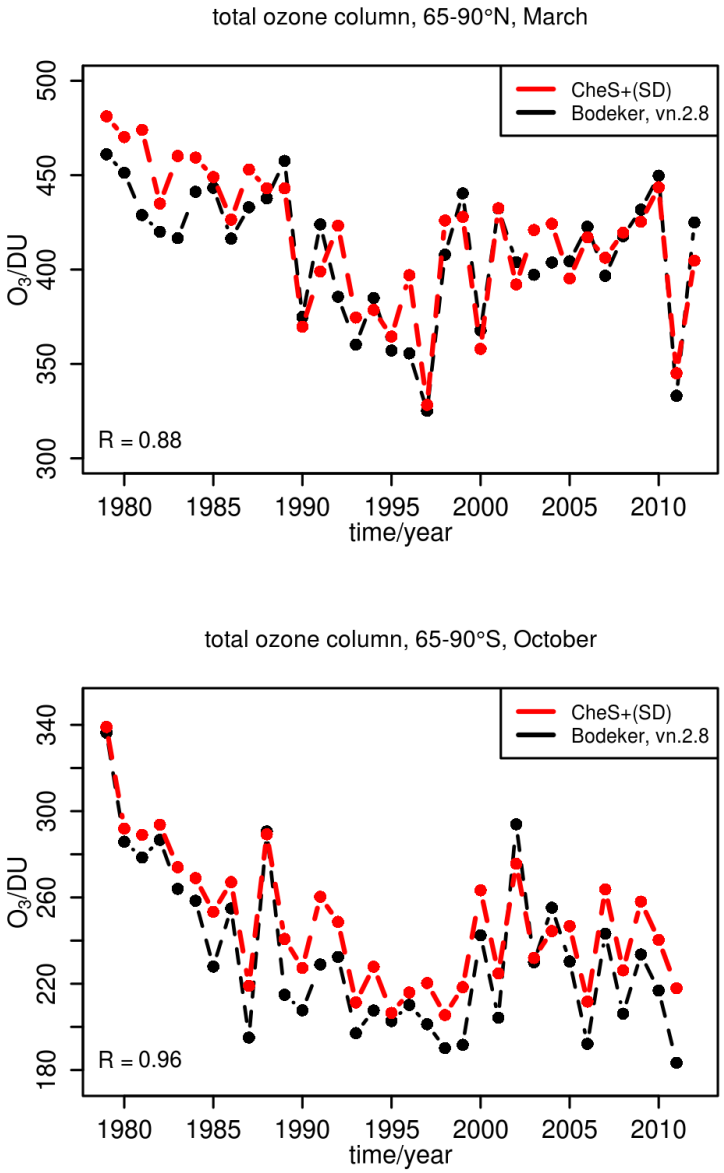

Figure S1. The evolution of 1979-2012 65-90°N March (top) and 1979-2011 65-90°S October (bottom) total ozone column [DU] in the nudged UMUKCA CCMI REFC1 CheS+(SD) integration (red) and observations (black, Bodeker total ozone column dataset: Bodeker et al., 2005; Müller et al., 2008).

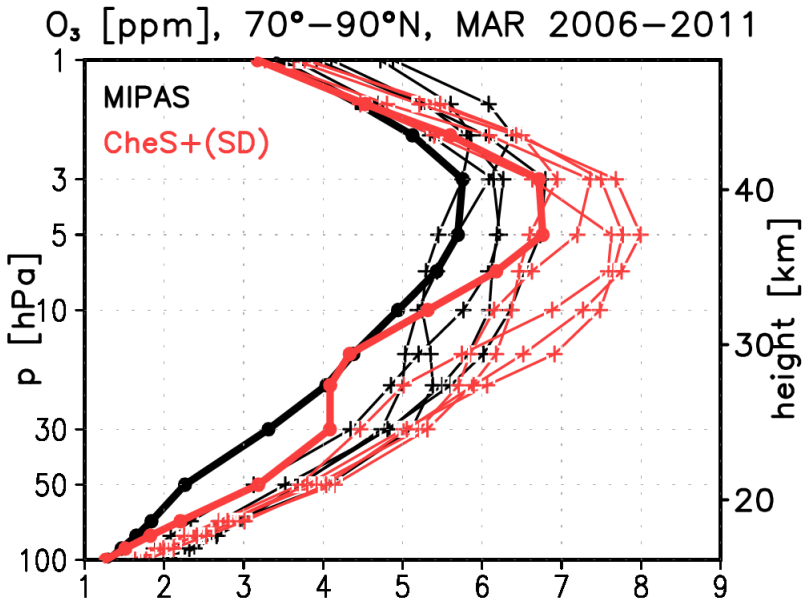

Figure R1. Monthly mean 70-90°N March ozone [ppm] mixing ratios in individual years from 2006 to 2011. Red lines are for the nudged UMUKCA CCMI REFC1 CheS+(SD) simulation and black lines are for MIPAS data (Fisher et al., 2008). Thick lines highlight the year 2011.

Beside chemistry, winter/springtime Arctic ozone levels are also strongly controlled by meteorology; any biases in temperature and winds will therefore impact on the simulated ozone. The UMUKCA REFC2 ensemble integrations described in the manuscript show a somewhat weaker and warmer present day Arctic stratospheric vortex in early/mid-winter, with a slight zonal wind bias of up to ~6 ms^{-1} in the mid-latitude lower/mid- stratosphere in March (see Fig. R2 and R3 of this response).

This information has been included in Sect. 2.2 of the updated manuscript.

As discussed in Sect. 3.2.3 of the updated manuscript, the model captures qualitatively the diagnosed relationship that higher Vpsc is associated with higher halogen induced ozone losses (Rex et al., 2004, 2006). The modelled correlation is R≈0.8 for the individual 20 year periods in the 21st century, demonstrating the coupling between chemistry and meteorology.

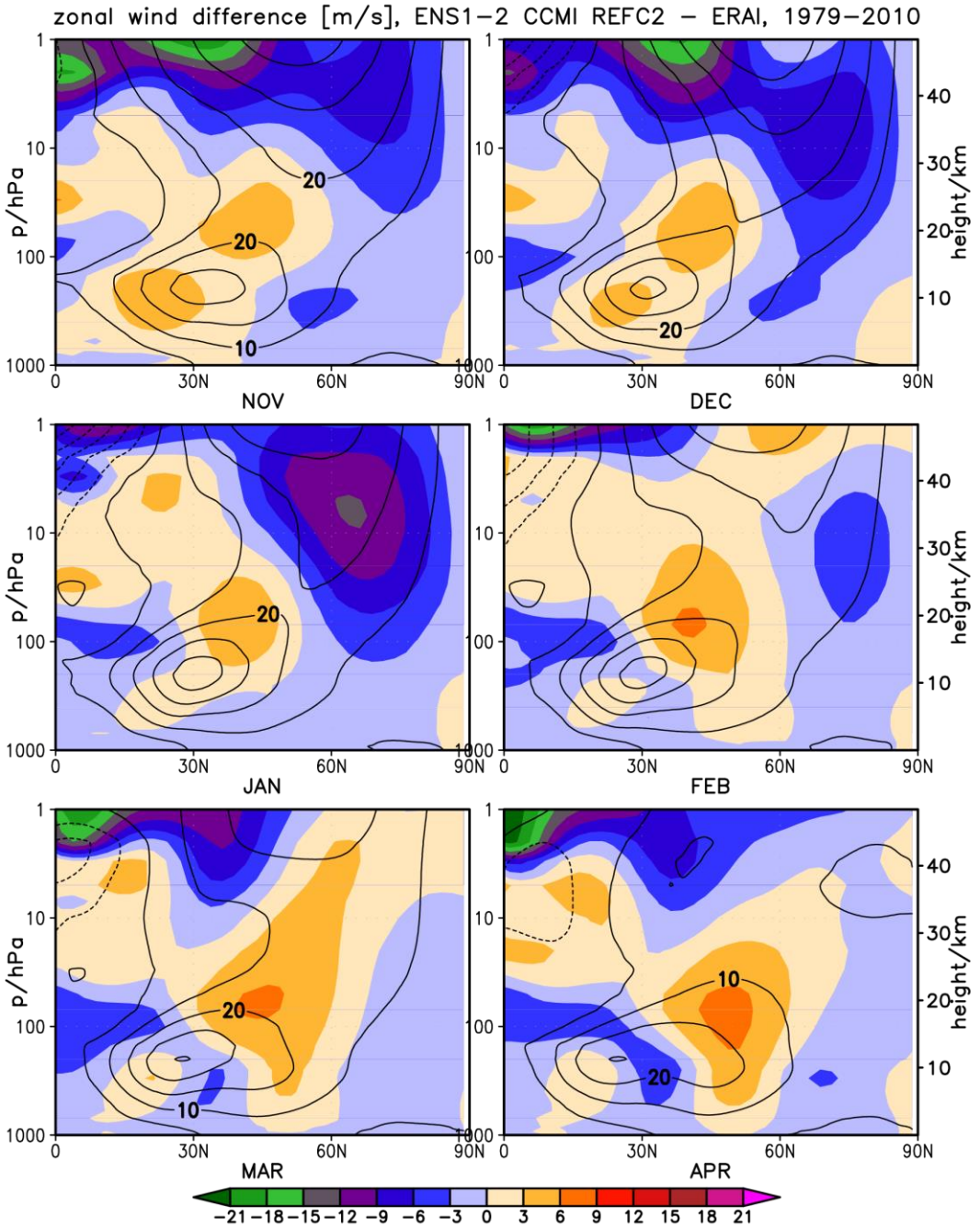

Figure R2. Contours: November to April monthly mean zonal mean zonal wind [m/s] climatology over 1979-2010 in the mean of ensemble members 1 and 2. Shading: the difference [m/s] between the model and the ERAI reanalysis (Dee et al., 2011).

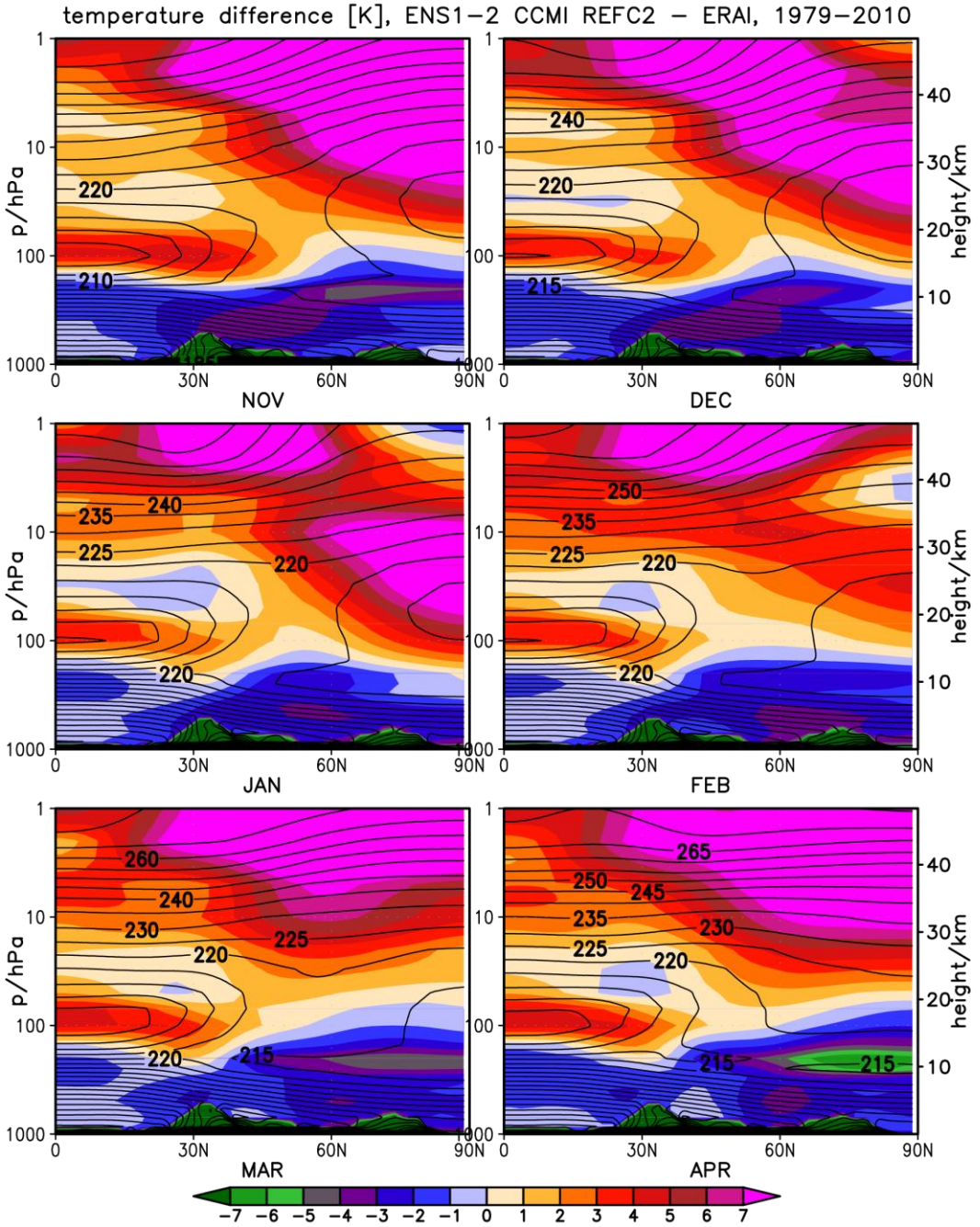

Figure R3. As in Fig. R2 but for temperature [K].

2. Chemical losses inside and outside polar vortex.

As stated in the manuscript, the model's passive ozone tracer does not participate in any chemical processing, and not just that due to halogen chemistry. This includes gas phase chemistry that is particularly important outside the polar vortex, as well as heterogeneous and gas phase chemistry inside and at the edge of the polar vortex. To avoid confusion with the interpretation of the passive ozone tracer diagnostic, we have replaced the Arctic mean passive ozone tracer with the vortex-averaged tracer in Fig. 8(a) of the revised manuscript. In addition, we have removed the last two sentences of Sect. 2.3 and the polar cap quantity from the last but one paragraph of Sect. 3.3. Lastly, we have reformulated the passive ozone paragraph in Sect. 3.3., including a cautionary sentence regarding the interpretation of the difference between the passive and full ozone column as resulting of a complex balance between chemical loss and production cycles as well as their interaction with transport throughout the winter.

In comparison, the halogen induced ozone loss diagnostic (Sect. 2.3) inside the polar vortex (calculated using various definitions of the polar vortex edge) lower atmosphere shows that the losses for model years 2063/2060 are mostly similar or even somewhat higher than those calculated for the Arctic mean (See Table 1 as well as Fig. R4-R5 below). This supports the use and interpretation of the Arctic mean diagnostic for analysing the long-term trends and variability in the halogen induced ozone loss for the full ensemble (Sect. 3.2.1).

The vortex-averaged halogen induced ozone losses for the two case study years have now been added to the manuscript (Sect. 3.3). We stress that we use a simple definition of the polar vortex edge, based on a rough estimate of a characteristic PV value for the maximum PV gradient.

CUMULATIVE HALOGEN-INDUCED OZONE LOSS [DU]

	65-90°N	$PV_{850K} \geq 600$ PVU	$PV_{450K} \geq 30$ PVU	$PV_{450K} \geq 35$ PVU
2063	39	44	46	43
2060	18	23	23	6

Table 1. Cumulative halogen induced ozone loss (1 Nov-30 Mar, 1-25 km) for the two case study years 2063 and 2060, calculated for the 65-90°N mean and a number of polar vortex edge definitions.

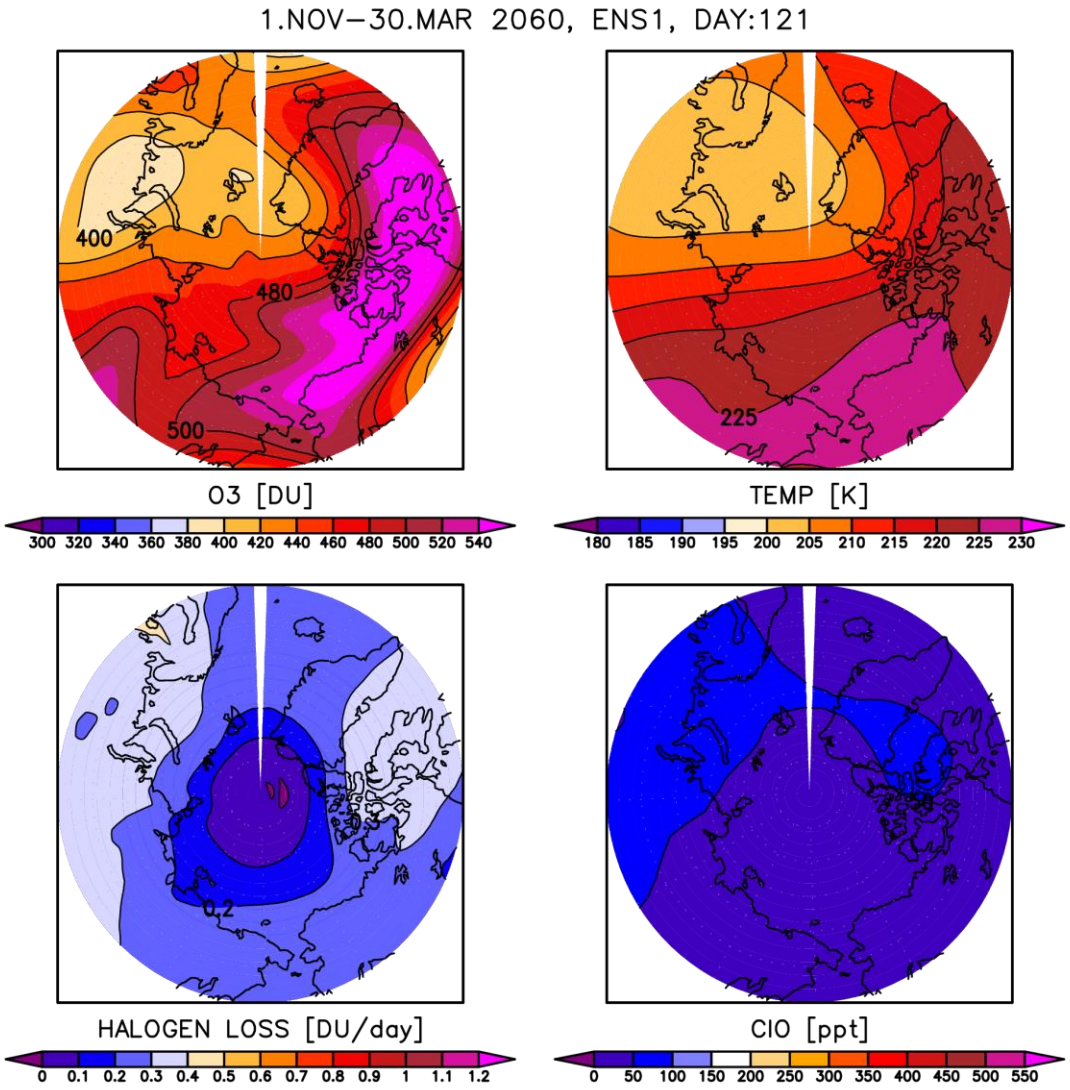

Figure R4. Daily mean total ozone column [DU] (top left), temperature at 21.5 km [K] (top right), halogen induced ozone loss in the 1-25 km layer [DU/day] (bottom left) and ClO [ppt] (bottom right) at 21.5 km simulated on 1 March in the case study year 2060.

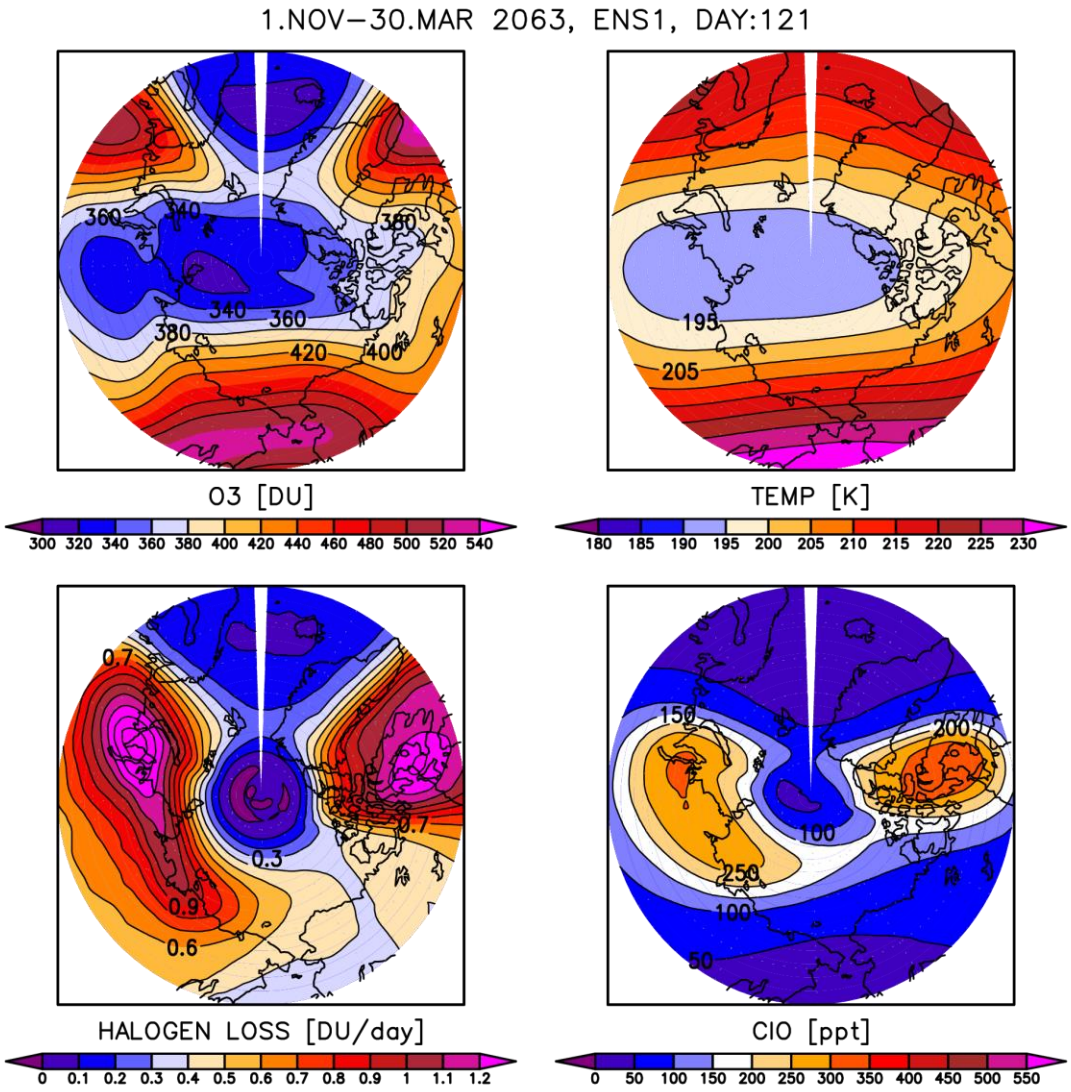

Figure R5. As in Fig. R4 but for the case study model year 2063.

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Future Arctic ozone recovery: the importance of chemistry and dynamics

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Abstract. Future trends in Arctic springtime total column ozone, and its chemical and dynamical drivers, are assessed using a 7 member ensemble from the Met Office Unified Model with United Kingdom Chemistry and Aerosols (UM-UKCA) simulating the period 1960-2100. The Arctic mean March total column ozone increases throughout the 21st century at a rate of ~11.5 DU decade^{-1}, and is projected to return to the 1980 level in the late 2030s. However, the integrations show that even past 2060 springtime Arctic ozone can episodically drop by ~50-100 DU below the corresponding long-term ensemble mean for that period, reaching values characteristic of the ~~to~~ near present day average level~~values~~. Consistent with the global decline in inorganic chlorine (Cl_y) over the century, the estimated mean halogen induced chemical ozone loss in the Arctic lower atmosphere in spring decreases by around a factor of two between 1981-2000 and 2061-2080. However, in the presence of a cold and strong polar vortex elevated halogen induced ozone losses well above the corresponding long-term mean continue to occur in the simulations into the second part of the century. The ensemble shows a significant ~~radiatively-driven~~ cooling trend ~~modelled~~ in the Arctic winter mid- and upper stratosphere, but there is less ~~consistency~~ confidence in the projected temperature trends~~across the seven ensemble members~~ in the lower stratosphere (100-50 hPa). This is partly due to an increase in downwelling over the Arctic polar cap in winter, which increases transport of ozone into the polar region as well as drives adiabatic warming that partly offsets the radiatively-driven stratospheric cooling. However, individual ~~years~~ winters characterised by significantly suppressed downwelling, reduced transport and anomalously low temperatures continue to occur in~~to~~ the future. We conclude that despite the ~~future~~ projected long-term recovery of Arctic ozone, the large interannual dynamical variability is expected to continue in the future, thereby facilitating episodic reductions in springtime ozone columns. 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.

1 Introduction

There was a period of rapid growth in the use of chlorofluorocarbons starting from the 1960s ~~onwards;~~ it is now well understood that ~~led to the suggestion that~~ these compounds, essentially inert in the troposphere, ~~could~~ undergo photo-degradation in the stratosphere and that their breakdown products ~~could~~ initiate depletion of the stratospheric ozone layer (Molina and Rowland, 1974). In 1985, Farman et al. reported a significant springtime depletion of ozone in Antarctica. Statistical analyses of long-term ground-based and satellite data sets subsequently demonstrated widespread, albeit smaller, downward trends at many locations (see, e.g., the Report of the International Ozone Trends Panel, WMO, 1988). These discoveries prompted a massive international research effort, which ~~soon~~ later confirmed the key role of halogen species in driving both the polar (Anderson et. al., 1989) and mid-latitude ozone losses (Hadjinicolaou et. al. 1997). Based on this scientific evidence, an international treaty, the Montreal Protocol on Substances that Deplete the Ozone Layer, came into force in 1987, and subsequent amendments and adjustments have significantly strengthened the regulatory control. Accordingly the stratospheric abundances of both chlorine and bromine peaked around the turn of the century and are now falling slowly, consistent with the long atmospheric lifetimes of some of the main ozone depleting substances (ODS) (SPARC, 2013; WMO, 2014).

While ~~initially~~ the major research questions were initially around the processes leading to ozone loss, the agenda has now shifted to the question of how and by when ozone levels in the stratosphere will return to earlier, historical values. However, it has been clear for some time (see Hofmann and Pyle, Ch. 12, WMO/UNEP, 1999) that ozone will not simply follow a path to recovery that is symmetric about the peak halogen loading; continuing increases in greenhouse gas (GHG) abundances mean that a future low-halogen stratosphere will not be the same as in the past.

Recent assessments of the state of the ozone layer (WMO, 2011, 2014; see also Eyring et al., 2010) paint a consistent picture of possible trajectories of ozone recovery based on integrations with chemistry-climate models (CCMs). First, because of the cooling of the stratosphere by GHGs (mainly CO_2), recovery of ozone to a particular level will occur before the corresponding recovery of halogen levels. Secondly, mid-latitude recovery in either hemisphere will occur before polar recovery. Recovery in the Arctic is expected by about 2030, but with a large inter-model range (see, e.g., Fig. 3.11 of WMO, 2011). Modelled recovery ~~in the~~of Antarctic springtime ozone occurs~~is~~ on average about 25 years later.

R~~Arctic r~~ecovery of Arctic ozone is particularly interesting and is the focus of this study. While similar chemical processes operate in the Arctic as in the Antarctic, where substantial springtime loss occurs every year, meteorological conditions in the Arctic are generally less favourable to cause extreme ozone depletion (Solomon et al., 2007, 2014). In particular, the Arctic stratospheric polar vortex in wintertime is, on average, more dynamically disturbed, resulting in higher temperatures and greater transport of ozone into the polar regions, and consequently the Arctic is generally not subject to the very large springtime ozone losses observed in the Antarctic (Tilmes et. al., 2006, Solomon et. al., 2007, 2014). However, when the winter/spring Arctic lower stratosphere is cold for a long period, substantial ozone depletion is to be expected (WMO, 2014). Occurrences of significant chemical depletion have been reported for a number of Arctic winters in the last two decades (e.g.

Goutail et al., 1999; Harris et al., 2002; Rex et al., 2002; Tilmes et al., 2004; Rex et al. 2006; Kuttippurath et al., 2010), and these have the potential to significantly impact on ecosystems and human populations. Indeed, the reduction of Arctic column ozone observed in late winter and early spring of 2011 was as large as that observed in the Antarctic~~a~~ (e.g. Manney et al., 2011). (Note however that Arctic column ozone is generally much higher than in the Antarctic, so that the late March values in 2011 were about twice those seen typically in springtime in the south, despite a comparable level of depletion).

The evolution of Arctic ozone will depend not only on the future trends in GHGs and ODSs but also on the behaviour of the stratospheric polar vortex, which is highly variable from year-to-year. In general, chemical and dynamical drivers of polar ozone depletion are likely to be related on interannual timescales (Tegtmeier et al., 2008). A strong and cold polar vortex favours low ozone. Transport of ozone to the high latitudes is much reduced and ~~low~~ cold temperatures in the lower stratosphere promote the formation of polar stratospheric clouds (PSCs) and ~~the~~ subsequent chlorine activation, thereby enhancing chemical ozone loss. In addition to PSCs, chlorine activation can also occur on cold sulphate aerosols (e.g. Hanson et al., 1994; Drdla and Müller, 2012; Wegner et al., 2012; Solomon et al., 2015). ~~It has been estimated that these two factors have contributed in roughly equal measure to recent variations in late winter/early spring total ozone (Rex et al. 2004, Tegtmeier et al., 2008).~~ In contrast, a warmer, more disturbed polar vortex should ~~experience~~ be accompanied by much smaller chemical destruction of ozone and enhanced transport from lower latitudes. The large interannual variability seen in Arctic late winter/early spring total column ozone results from complex interactions between these chemical and dynamical processes. It has been estimated that these two factors have contributed in roughly equal measure to recent variations in late winter/early spring total ozone (Rex et al. 2004, Tegtmeier et al., 2008). ~~In addition to PSCs, chlorine activation can also occur on cold sulphate aerosols (e.g. Hanson et al., 1994; Drdla and Müller, 2012; Wegner et al., 2012; Solomon et al., 2015).~~

There has been keen debate about the future evolution of conditions in the Arctic winter stratosphere. The stratosphere is expected to cool in the global mean under increased GHG concentrations (e.g. Fels et al., 1980), but the evolution of the highly variable Arctic lower stratosphere cannot be predicted with confidence. Rex et al. (2004) have reported a strong linear correlation between observed wintertime chemical ozone loss and a measure of the volume of ~~polar stratospheric clouds~~ (~~PSCs~~; (V_{PSC}). It has been suggested that long-term temperature changes in the Arctic polar lower stratosphere may have contributed to the coldest Arctic winters becoming significantly colder in recent decades (Rex et al., 2004; Rex et al., 2006; Ivy et al., 2014), reflecting the conditions that lead to larger V_{PSC} and facilitate greater chemical ozone losses. In contrast, Rieder and Polvani (2013) showed that increases in V_{PSC} since 1979, as estimated from multiple reanalysis datasets, are not highly statistically significant. While the future evolution of the Arctic polar vortex is the subject of keen debate, it is clear that any changes in the future will ~~certainly~~ have a ~~controlling~~ substantial influence on the Arctic ozone column.

Langematz et al. (2014) have studied the future evolution of Arctic ozone and temperature using a CCM. They found that rising GHG concentrations lead to a cooling of the Arctic lower stratosphere in early winter, but that there were no significant temperature changes in late winter or spring. They did not find a long-term downward trend in minimum Arctic

temperatures, nor any extension of the vortex break-up date into later spring. Consistent with numerous other studies (e.g. WMO, 2011, 2014), they found that Arctic ozone is expected to increase in the latter part of the century.

~~The~~ Langematz et al. (2014) ~~study~~ examined two transient 21st century integrations (with and without GHG changes) and a number of timeslice experiments with different boundary conditions. Our focus on Arctic ozone is similar to theirs, but we employ a different approach. We examine the evolution of Arctic springtime ozone in the UM-UKCA CCM (Morgenstern et al., 2009) ~~in~~ using an ensemble of 7 transient simulations carried out as part of the World Climate Research Programme (WCRP) Stratospher~~e~~ic~~,~~ troposphere~~ic~~ Processes and their Role in Climate (SPARC) chemistry-climate model initiative (CCMI, Eyring et al., 2013). This ensemble of simulations enables us to explore the year-to-year variability in Arctic polar ozone and its relation to the long-term trends in chemical and dynamical drivers over the 21st century. Section 2 gives information on the model and simulations performed. Section 3.1 describes the temporal evolution of Arctic springtime total column ozone. Section 3.2 examines the chemical, radiative and dynamical drivers of Arctic ozone during the 21st century. Section 3.3 highlights the importance of these drivers in a case study for ~~an~~ individual low and average ~~-~~ozone events simulated in the second half of the 21st century. Finally, Section 4 summarises the key results.

2 Model, experiment and methods

2.1 The Model

We use the UM-UKCA CCM that is built around the Met Office Unified Model (MetUM) in the HadGEM3-A configuration (Hewitt et al., 2011) at MetUM version 7.3. The model uses a horizontal resolution of 2.5° latitude by 3.75° longitude, with 60 vertical levels up to 84 km. We use the extended Chemistry of the Stratosphere (CheS+) chemistry scheme, which is an expansion of Morgenstern et al. (2009) where CFC-11, CFC-12, CFC-113, HCFC-22, Halon-1211, Halon-1301, CH_3Br, CH_3Cl, CCl_4, CH_2Br_2, and $CHBr_3$ are considered explicitly, resulting in an additional 17 bimolecular and 9 photolytic reactions. The chemical tracers O_3, CH_4, N_2O, CFC-11, CFC-12, CFC-113, and HCFC-22, are all interactive with the radiation scheme. The model and chemistry scheme were used for the recent SPARC Report on the Lifetimes of Stratospheric Ozone-Depleting Substances, Their Replacements, and Related Species (SPARC, 2013; Chipperfield et al., 2014).

As in Morgenstern et al. (2009), heterogeneous reactions on PSCs as well as formation and removal of ~~(~~nitric acid trihydrate~~;~~ (NAT) PSCs follow Chipperfield (1999), with the formation and removal of ice PSCs included in the hydrological cycle. Regarding the NAT PSCs, as described in Chipperfield et al. (1999), the NAT PSC formation follows the equilibrium expression from Hanson and Mauersberger (1988). The denitrification scheme assumes a relatively slow NAT sedimentation velocity of ~40 m/day for pure NAT PSCs, and a much faster NAT sedimentation velocity of ~1540 m/day in the presence of ice; the latter assumes coating of NAT PSCs onto ice particles. Stratospheric aerosols are prescribed for heterogeneous reactions, as well as in the photolysis and radiation schemes. Note that unlike on PSCs, the heterogeneous reactions between $ClONO_2$ and HCl as well as between N_2O_5 and HCl on sulphate aerosols are not included in the scheme. As heterogeneous

reactions on liquid aerosols can be important for the springtime ozone (see e.g. Solomon et al., 2015), the model heterogeneous chemistry scheme does not represent an exhaustive and fully realistic treatment of heterogeneous processes. However, using a similar model version Keeble et al. (2014) obtained a reasonable representation of the springtime Antarctic ozone hole, with the majority of the simulated Antarctic ozone depletion in the model driven by the heterogeneous reactions on the surfaces of NAT and ice PSCs. ~~This model was used for the recent SPARC Assessment of Lifetimes (SPARC, 2013; Chipperfield et al., 2014).~~

The 11-year solar cycle variability is included consistently in both the radiation and photolysis schemes. In the radiation scheme, we use the method employed in HadGEM1 (Stott et al., 2006) and HadGEM2-ES models (Jones et al., 2011)~~, using T~~total solar irradiance (TSI) data are those recommended in the fifth Coupled Model Intercomparison Project (CMIP5) (Wang et al., 2005; Lean, 2009), processed to force the mean of the 1700-2004 period to be 1365 W/m^2 (Jones et al., 2011)~~, from Solanki and Krivova (2003)~~. A~~and a~~ fit to spectral data from Lean (1995) is used to account for a change in partitioning of solar radiation into wavelength bins. In the Fast-JX photolysis scheme (Telford et al., 2013), ~~whilst the long-term evolution of TSI follows that of Solanki and Krivova (2003),~~ the change in partitioning of solar irradiance into wavelength bins is accounted for by scaling the bins according to the CMIP5 spectral solar irradiance data ~~as recommended for models in the fifth Coupled Model Intercomparison Project (CMIP5)~~ (Wang et al., 2005; Lean, 2009) for the years 1981 and 1986, and the long-term evolution of the processed TSI timeseries. Solar cycle variability for the future period (after 2009) is included as in earlier periods but with a repeating sinusoidal 11-year cycle with an amplitude derived from observed cycle 23 (see Jones et al., 2011; Gray et al., 2013).

The stratospheric climatology and variability, including that in the Arctic region, have been evaluated in similar stratosphere-resolving versions of the MetUM (Osprey et al., 2010; Hardiman et al., 2010) and UM-UKCA (Morgenstern et. al. 2009). The model includes parameterized orographic and non-orographic gravity wave drag (Scaife et al., 2002; Webster et al., 2003), and simulates an internally generated quasi-biennial oscillation (QBO) (Scaife et al, 2002). As shown by Morgenstern et al. (2009), the model reproduces the observed anti-correlation between polar ozone and jet strength in March, which is one indicator of the coupling between chemistry and meteorology discussed above.

A comparison between the Arctic/Antarctic mean (65-90°N/S) total ozone columns in a similar model version as used here, but with a specified dynamics set-up, i.e. in which model's temperature and wind fields are nudged towards meteorological reanalysis data, shows a good correlation between simulated and observed polar column ozone over the recent past (R=0.88 and R=0.96 for the Arctic and Antarctic mean, respectively, see Supplementary Material Fig. S1). In the Antarctic, there is some overestimation of the ozone column in the model by up to ~15-30 DU (see Fig. S1). For the cold March 2011, the model shows a positive bias in the zonal mean monthly mean ozone levels in the Arctic lower stratosphere (30-100 hPa) of up to ~0.9 ppb (~41%; for the 70-90°N mean, 50 hPa) compared with MIPAS satellite data (Fisher et al., 2008, not shown). The positive ozone bias is commensurate with a negative bias in the zonal mean monthly mean ClO levels at this altitude. Note that there is some uncertainty in the observed zonal mean monthly mean ClO, in part due to sparse temporal sampling

(twice a day) of the MIPAS instrument. In general, all models will show some biases with respect to observations. Importantly, as in many studies, we compare the model for the present day and future periods in an internally consistent way; therefore, any biases will become less relevant for our study.

The mean age of air in the tropical lower stratosphere, as well as in the northern high-latitudes, is at the young end of observationally-derived values, but its gradient between the northern mid-latitudes and the tropics is in a fair agreement with observations and other models (SPARC, 2013; Chipperfield et al., 2014). This indicates a significant improvement in its circulation as compared with the older atmospheric model version presented in Morgenstern et al. (2009) and in SPARC Report No.5 (SPARC, 2010).

2.2 The CCMI REFC2 Experiment

We follow the experimental design of the CCMI REF-C2 experiment (Eyring et al., 2013). Lower boundary conditions are used for CH_4, N_2O, H_2, CFC-11, CFC-12, CFC-113, HCFC-22, Halon-1211, Halon-1301, CH_3Br, CH_3Cl, CCl_4, CH_2Br_2, $CHBr_3$, and CH_3CCl_3, with values for ODSs specified from WMO (2011) and future GHG~~s~~ abundances are specified ~~as for~~according to the representative concentration pathway RCP6.0 (Fujino et al., 2006; Hijioka et al., 2008; Taylor et al., 2012). The prescribed s~~S~~tratospheric aerosols are as given by ~~prescribed for both heterogeneous and photolytic reactions, as well as in the radiation scheme, using~~ the SPARC climatology (SPARC, 2006). ~~However, unlike on PSCs, the heterogeneous reaction between $ClONO_2$ and HCl on sulphate aerosols is not included in the scheme. Consistently, using a similar model version Keeble et al. (2014) showed that the majority of the ozone depletion modelled in the Antarctic during spring can be attributed to the heterogeneous reactions on the surfaces of NAT and ice PSCs. The chemical tracers O_3, CH_4, N_2O, CFC-11, CFC-12, CFC-113, and HCFC-22, are all interactive with the radiation scheme.~~ Sea-surface temperatures (SSTs) and sea-ice concentrations (SICs) are taken from one ensemble member (r2i1p1) of the HadGEM2-ES RCP6.0 ensemble (MOHC, 2011; Jones et al., 2011). We perform two integrations from 1960-2099 and five over a shorter period from November 1980 to December 2080; the latter five were initiated from different initial conditions taken from a supporting perpetual year 1980 integration. As a result of a data issue, a total of five 6-year-long periods is excluded from the analysis of the ensemble (see Supplementary Information for more details). When compared against the ERA-Interim reanalysis (Dee et al., 2011), the integrations show a somewhat weaker and warmer present day Arctic stratospheric vortex in early/mid-winter, with a slight zonal wind bias of up to ~6 ms^{-1} in the mid-latitude lower/mid- stratosphere in March (not shown).

2.3 Diagnostics of chemical loss

We use two diagnostics to estimate the chemical ozone loss in the Arctic region. Firstly, we estimate the ozone loss due to halogen reactions directly from reaction fluxes. We use the diagnostic framework of Lee et al. (2002) in which the rate of odd oxygen (O_x=O_3+$O(^3P)$+$O(^1D)$) destruction is estimated for different catalytic cycles by determining the rates of their rate-limiting steps. The most important halogen-catalysed ozone loss cycles in the polar lower stratosphere are the ClO dimer

cycle (Cycle 1, Molina and Molina, 1987), ClO+BrO cycle (Cycle 2, Yung et al., 1980; McElroy et al., 1986) and thea more minor ClO+O(^{3}P) cycle (Cycle 3, Stolarski and Cicerone, 1974):

$$ClO + ClO + M \rightarrow Cl_2O_2 + M$$
$$\mathbf{Cl_2O_2 + h\nu \rightarrow Cl + ClOO}$$
$$ClOO + M \rightarrow Cl + O_2 + M$$
$$2*(Cl + O_3 \rightarrow ClO + O_2)$$
(NET: $2*O_3 \rightarrow 3*O_2$; (*Cycle 1*)

$$\mathbf{BrO + ClO \rightarrow Br + ClOO} \qquad \mathbf{BrO + ClO \rightarrow BrCl + O_2}$$
$$ClOO + M \rightarrow Cl + O_2 + M \qquad BrCl + h\nu \rightarrow Br + Cl$$
$$Br + O_3 \rightarrow BrO + O_2$$
$$Cl + O_3 \rightarrow ClO + O_2$$
(NET: $2*O_3 \rightarrow 3*O_2$; (*Cycle 2*)

$$\mathbf{ClO + O(^3P) \rightarrow Cl + O_2}$$
$$Cl + O_3 \rightarrow ClO + O_2$$
(NET: $O_3 + O \rightarrow 2*O_2$; (*Cycle 3*)

The rate-limiting step in each cycle is highlighted in bold font. Three further halogen cycles of lesser importance in the winter polar lower stratosphere (the BrO + O(^{3}P) cycle analogous to Cycle 3, and a pair of ClO + HO_2 and BrO + HO_2 cycles) are also included. In all six reactions, a net loss of 2 odd oxygen molecules occurs per cycle. Assuming that $[O_x] \approx [O_3]$ and, thus, $d[O_x]/dt \approx d[O_3]/dt$, we then calculate the rate of ozone loss from each cycle and integrate in time (from 1 November to 30 March) and altitude (from the surface to 25 km), resulting in an estimate of the cumulative ozone loss at each grid point due to these halogen reactions in the polar lower stratosphere. Whilst the rates (i.e. fluxes) of ozone loss are calculated at each grid point and every UM-UKCA timestep, we use the zonal mean monthly-mean diagnostics for computational ease (except for vortex-average quantities reported in Sect. 3.3, where daily means are used). for computational efficiency. The cut-off altitude of 25 km was chosen so as to capture most of the region subject to halogen activation on PSC surfaces. Since no further separation is made between the ozone losses due to heterogeneous and homogenous chemistry, the diagnostic is not equivalent to the ozone losses initiated exclusively by the heterogeneous reactions on PSCs, as it was presented in e.g. Chipperfield and Jones (1999), Chipperfield et al. (2003) and Keeble et al. (2014).

It is important to understand the characteristics of this diagnostic when integrated over the winter. First, we have calculated losses in an Eulerian framework and so our values are not directly comparable to the Lagrangian estimates that have been

calculated during some polar winters (see Rex et al. (2002) and Harris et al. (2002) for detailed comparison). Second, our values are integrated over 24 hours and will necessarily be lower than instantaneous rates calculated, for example, from observed, daytime observations of ClO. Third, unless otherwise specified we present data that have been averaged from 65°N to the pole. Unlike in the Antarctic, the Arctic vortex is not usually centered on the pole; it is mobile and highly variable, so our diagnostic will include areas where polar halogen chemistry is not active. However, the strength of our diagnostic is that it allows us to compare different model winters against each other. By definition, greater values of the diagnosed loss will be found in those winters where halogen chemistry is more important.

An additional diagnostic for ozone loss used in Sect. 3.3 is a passive ozone tracer, similar to that implemented in Chipperfield and Jones (1999) and Chipperfield et al. (2003). This chemically-inert tracer is initialised to the modelled ozone concentrations on 1 November each year. Whilst undergoing no chemical production or loss, it is transported by the circulation until the end of March. The difference between the chemical ozone field and the passive ozone tracer at the end of each winter represents the change in ozone levels due to all chemical processes. This diagnostic was only included in one ensemble member, discussed ~~below~~in Sect. 3.3.

~~In the Arctic, use of the passive tracer diagnostic generally indicates higher ozone loss than calculated from the fluxes especially in dynamically disturbed winters, consistent with the discussion above. In contrast, in the Antarctic much more similar values are calculated.~~

3 Results

3.1 Long-term evolution of polar total column ozone and Cl_y

Figure 1(a) shows timeseries of March total column ozone averaged over 65-90°N (henceforth referred to as Arctic mean) for the 7 UM-UKCA ensemble members. An analogous plot for 65-90°S in October is shown in Fig. 1(b). The corresponding Arctic mean total inorganic chlorine level (Cl_y) at 20 km is shown in Fig. 1(c).

The results in Fig. 1 are consistent with the multimodel means reported in WMO (2011, 2014) and Eyring et al. (2010). There is a reduction in Arctic total column ozone from the 1960s to the late 1990s, with the March ensemble mean dropping by ~15% to ~400 DU. This is consistent with the modelled increase in Cl_y over this period. A similar long-term decline is modelled in the springtime Antarctic, where the ozone reduction is much larger in both absolute and percentage terms (125 DU change in October, i.e. ~38%). This decline in 20th century polar ozone agrees well with observations (red curve; Bodeker total ozone column data set, v2.8, Bodeker et al., 2005; Müller et al., 2008).

As a result of the Montreal Protocol and its subsequent amendments and adjustments, the increase in ~~atmospheric~~ stratospheric Cl_y abundances has ceased and values are projected to decrease over the 21st century, returning to their 1980 levels by about 2060 in both the Antarctic and Arctic (WMO, 2011). In the Antarctic, the average total ozone returns to 1980 levels over approximately the same period and therefore closely follows the evolution of Cl_y (not shown). In contrast, total column ozone in the Arctic returns to 1980 levels by the late 2030s, which is approximately 15-20 years earlier than the

return date for Cl_y (Fig. 1(c)). ~~This occurs because of the slowing of gas phase ozone loss cycles under greenhouse gas induced stratospheric cooling (Haigh and Pyle, 1982), and through changes in stratospheric transport (see WMO (2011, 2014), and references therein).~~ In the Antarctic, the polar vortex is stronger climatologically and the evolution of ozone is largely determined by changes in halogen chemistry, in agreement with Austin and Wilson (2006). In the Arctic, the return date in UM-UKCA is somewhat later than indicated by the multi-model mean from the Chemistry Climate Model Validation 2 (CCMVal-2) project (Eyring et al., 2010), but is within the range of individual model estimates (WMO, 2011).

The individual curves in Fig. 1(a) and 1(b) highlight that the interannual variability in springtime total column ozone is much larger in the Arctic than in the Antarctic, with modelled values ranging from 346-487 DU in the 1981-2000 period. This large variability remains throughout the 21st century in the presence of the gradual reduction in Cl_y and the long-term increase in mean Arctic ozone. The ensemble mean ozone increases by about 11.5 (±1.3, i.e. ±2 standard errors) DU decade^{-1} over the 2000-2080 period, with its 11-year running mean increasing from 408 DU in 2000 to 482 DU in 2074. The minimum monthly mean ozone levels ~~found~~ at a single location anywhere poleward of 65°N increase at a similar rate to the Arctic mean ozone column, albeit the 11-year running mean value of this quantity in 2000 is ~50 DU lower than the corresponding Arctic mean value. T~~Notably, t~~he persistence of the large interannual variability is associated with years with particularly low Arctic ozone throughout the 21st century.~~–~~ In the second half of the century, March column ozone episodically drops not only below the 1980 level, but also to values of ~410-415 DU, which are close to the long-term minimum of the 11-year running ensemble mean around the turn of the century. In addition, one ensemble member simulates 383 DU in March of 2063 (see green point in Fig. 1(a)), which is comparable to the average values routinely found under present day conditions.

Figure 2 shows probability density functions (PDFs) of Arctic mean March total ozone for five 20-year intervals from 1981 to 2080. As is also evident in Fig. 1(a), the means of the PDFs (coloured diamonds in Fig. 2) progressively increase over the 21st century, consistent with the gradual decline in stratospheric Cl_y (coloured points in Fig. 2) and the super recovery of ozone (Eyring et al., 2010). Note that the first two decades of the 21st century have, on average, lower ozone compared to 1981-2000 (~14 DU decrease in mode, Kolmogorov-Smirnov (KS) test p-value of 0.052), with a suggestion of a change in skewness of the distribution. This results from the rate of reduction in column ozone at the end of the 20th century being larger than the rate of the subsequent recovery, which begins near the turn of the century.

Notably, the distributions ~~characterising the~~of Arctic March column ozone are negatively skewed, which indicates the occurrence of ozone values well below the 20-year mean. Austin and Wilson (2006) reported an increase in the interannual variability of March Arctic column ozone in the future. Clearly, the precise characteristics of the PDFs in Fig. 2, such as their width and skewness, vary between the five periods shown. Although there could in principle be a forced trend in these characteristics with time (e.g. Rex et al., 2004; Austin and Wilson, 2006), such factors will also be influenced by internal variability and decadal variability in SSTs/SICs~~sea ice~~ and solar forcing, which are common to all of the simulations (see Sect. 2). With the exception of an increase in the mean and mode of ozone over the century, we do not find systematic changes in other characteristics of the PDFs, such as their width, with time ~~(not shown)~~.

The occurrence of low ozone episodes ~~continue~~ in~~to~~ the future, with values episodically dropping by ~50-100 DU below the 11-year running mean~~,~~; ~~this~~ is particularly clear in the PDF for the last two decades modelled (2061-2080). In a single transient integration, Langematz et al. (2014) found that springtime Arctic ozone did not generally drop below the 1970-82 mean beyond around 2060. In the UM-UKCA simulations, years in which the springtime Arctic ozone drops below the 1980 level (~450 DU, see Fig. 1(a)) continue until at least 2080. This contrasting result may be due to differences, likely dynamical, in the representation of the Arctic winter stratosphere and its variability between the CCMs used in the studies, differences in experimental set-up (e.g. GHGs, SSTs and SICs~~sea ice~~), or the fact that we have more fully sampled the internal variability of the climate system by using an ensemble of integrations.

3.2 Future Arctic variability and trends in the chemical and dynamical drivers of ozone

Polar ozone is influenced by a complex interplay between chemical and dynamical processes. Over the 21st century, there may be forced trends in factors that affect both sets of processes, including atmospheric abundances of key chemical~~ly relevant~~ species, stratospheric temperatures, as well as the large-scale circulation; all of these may contribute to the evolution of ozone during this century. The following section discusses the temporal evolution of some of the chemical and dynamical processes that affect Arctic ozone in the UM-UKCA ensemble.

3.2.1 Trends and variability in chemical drivers

An estimate of the contribution of halogen induced chemical loss to the Arctic mean March column ozone and its time dependence is shown in Fig. 3(a). The scatterplot shows the Arctic mean March total column ozone versus the cumulative winter-time (1 November to 30 March) chemical ozone loss due to the six ~~most important~~ halogen cycles of most importance in the polar lower stratosphere integrated from the surface to 25 km, calculated using the Eulerian diagnostics discussed in Sect. 2.3. Note that the diagnostic is henceforth referred to interchangeably as 'halogen induced ozone loss' and, in short, 'halogen loss'. We use the diagnostic to compare particular Arctic winters against each other. The diagnostic constitutes a useful tool for examining both the interannual and interdecadal variability in the halogen induced ozone loss in our model and its contribution to variability in ozone. Recall that the calculated loss should not be expected to match quantitatively Lagrangian calculations of the chemical ozone loss in particular Arctic winters (e.g. Harris et al., 2002; Rex et al., 2004; Tegtmeier et al., 2008).

There is a significant correlation between the March total column ozone and the cumulative halogen induced loss during winter (Pearson correlation coefficient of -0.81 for the full distributions of all points). Around half of the estimated halogen induced ozone loss can be attributed to the ClO+BrO cycle, with further contributions from the ClO dimer and the ClO+O(^{3}P) cycles; the remaining three cycles together contribute a background of ~5-7 DU that varies only very little on both short and long-term timescales (not shown). As expected, there is a gradual decline in the average halogen induced loss over the 21st century, with the mean diagnosed loss almost halving between 2001-2020 and 2061-2080 (36 DU and 20 DU,

respectively). This reflects the long-term decrease in the global Cl_y levels, which also decline by about a factor of two in that period (Fig. 1(c) and 2).

Despite the long-term decrease in the mean halogen induced ozone loss, even after 2060 the halogen loss in individual years can still be more than a factor of two higher (i.e. by ~20-30 DU) than the 11-year running mean for that period (Fig. 3(b)). For example, the largest chemical losses during 2061-2080 are greater than 40 DU; this illustrates the continued potential for enhanced chemical loss to occur in the Arctic in the presence of favourable dynamical conditions (i.e. a cold and strong vortex). So, although the maximum halogen induced ozone losses later in the century are lower, as expected, than found during the period between 1980 and 2040 (Fig. 3(a)), ~~the halogen induced losses~~these processes still make an important contribution to the overall ozone anomaly in these years.

Chipperfield and Jones (1999) estimated the contribution of ozone losses due to heterogeneous halogen chemistry to the interannual variability in Arctic springtime ozone by comparing the standard deviations of diagnosed chemical loss and March total ozone column. In our calculations, after removing the 11-year running ensemble mean from both the Arctic column ozone and ~~diagnosed~~ halogen induced ozone loss time series, we find that the standard deviation of the estimated halogen loss constitutes nearly 30% of the standard deviation of the modelled March total column ozone over the entire 1981-2080 period of the integration. So, while halogen losses clearly contribute to the springtime Arctic ozone, the dominant driver in the model that determines the interannual variability is dynamics, ~~in agreement with~~ as in the study of Chipperfied and Jones (1999).

3.2.2 Trends in Arctic stratospheric temperatures

There is considerable interest in whether there has been any trend in minimum temperatures in the Arctic winter lower stratosphere in recent decades. This could increase the amount of PSCs formed in winter as well as their persistence, ~~and~~ thus enhanc~~e~~ing chemical ozone loss; such an increase, if present, could have contributed to years with low springtime ozone observed in the recent past (see e.g. Rex et al., 2004; Rieder and Polvani, 2013; Ivy et al., 2014). The direct radiative impact of increasing CO_2 levels is to cool the stratosphere, with the greatest impact in the upper stratosphere (Fels et al., 1980). Temperatures in the Arctic winter stratosphere are also strongly influenced by dynamical processes, which can enhance or offset the radiatively driven cooling (e.g. Bell et. al., 2010; Butchart et al., 2010; Langematz et al., 2014). In addition to long-term changes, the interannual variability is high and can potentially compound the identification of trends on shorter timescales.

Figures 4(a-c) show simulated Arctic mean temperature trends [K decade^{-1}] at five pressure levels in the stratosphere for the period 1981-2080 in early (November-December), mid (January-February) and late winter/spring (March), respectively. Black points denote the trends calculated for individual~~each~~ ensemble members, with the trend for the ensemble mean shown in red along with ±2 standard errors. A statistically significant cooling trend is found in the mid and upper stratosphere (at and above 30 hPa) throughout the winter. This is most robust in early winter (Fig. 4(a)) and late winter/spring (Fig. 4(c)), when all ensemble members show trends of the same sign. The magnitude of the trend increases with decreasing pressure in

agreement with earlier studies (Fels et al., 1980; Bell et. al. 2010, Oberländer et al., 2013; Langematz et al., 2014). However, in the lower stratosphere (100-50 hPa) there is less ~~consistency~~ confidence in the projected temperature trends across the ensemble throughout the winter. In early winter, the ensemble mean shows a weak cooling in the Arctic lower stratosphere (-0.15 K decade^{-1} at 50 hPa; trend small and not statistically ~~in~~significant at 100 hPa). At least one ensemble member shows a near zero trend. In comparison, Langematz et al. (2014) found a statistically significant cooling trend in early winter over 1960-2100 throughout the polar stratosphere.~~, in agreement with Langematz et al. (2014). However, at least one member shows a near zero trend. Similar is true for late winter/spring, with~~ In late winter/early spring, no trend in temperature was found in our study at 100 hPa. At 50 hPa and above, as discussed above, the results suggest an overall cooling trend; however ~~Notably~~, a large spread of magnitudes can be seen across the ensemble, with individual 50 hPa March temperature trends ranging from ~0 to ~0.5 K decade^{-1}. In mid-winter, the intra-ensemble spread is even larger and no significant trend in the ensemble mean lower stratospheric temperature is found.

A similar analysis of future trends in the local minimum Arctic temperature anywhere poleward of 65°N (T_{min}) calculated from monthly mean data reveals largely qualitatively similar results to the Arctic mean quantities shown in Fig. 4 (not shown). The results support the need for ensemble studies to confidently detect ~~model~~ simulated temperature trends in the polar lower stratosphere.

3.2.3 Trends in V_{PSC}

Stratospheric H_2O and HNO_3 levels are projected to increase in the future, which is likely to enhance levels of PSCs (Langematz et al., 2014). Moreover, the formation of PSCs will be further strengthened in winters with colder polar temperatures. Figure 5 shows a scatterplot of November to March mean potential V_{PSC} versus the halogen induced ozone loss in the polar lower stratosphere, as discussed in Sect. 3.2.1. Potential V_{PSC} is defined as average daily mean volume of air (1. November to 30. March, 1-30 km) in which NAT PSCs are thermodynamically possible according to the NAT equilibrium scheme used in the model. There is a tendency in the model for higher potential V_{PSC} to be associated with higher halogen induced ozone losses (R≈0.8 for the individual 20 year periods in the 21st century), in broad agreement with the observed relationship (Rex et al., 2004, 2006).

For the 1993-2005 period (as studied in Rex et al., 2006), the gradient of the linear fit (R=0.81) is 1.03 (±0.17) DU/10^6 km^3, which is about half of the value reported by Rex et al. (2006). We caution against a detailed quantitative comparison because of: differences between the Eulerian and Langrangian halogen-induced ozone loss diagnostics (see Sect. 2.3); the use of 65-90ºN average quantities instead of vortex averages; differences in the altitudes over which chemical loss has been integrated; as well as differences in the definitions of V_{PSC}.

The amount of halogen induced ozone loss past ~2040 that occurs for a given potential V_{PSC} is reduced with respect to the earlier periods, consistent with the projected future reduction in Cl_y levels and the resulting lower chlorine levels available for activation in the future period. However, the period 2061-2080 is characterised by occurrences of particularly high potential V_{PSC}; this is consistent with the occurrence of a number of very cold Arctic winters in the simulations (see Sect.

3.2.4 for the discussion of variability in the residual circulation), and could also be related to the rising HNO_3 and H_2O levels (not shown, see also Langematz et al., 2014). Hence, despite the lower Cl_y in the future, higher potential V_{PSC} still increases the likelihood for chlorine activation. Even though dynamics and transport will dominate the interannual variability of Arctic ozone in the future, halogen chemistry will still have the potential to contribute to instances of low Arctic springtime total column ozone.

3.2.43 Trends in atmospheric circulation and transport

In contrast to the Antarctic region, where interannual and interdecadal variability is smaller and, thus, springtime ozone levels within the polar vortex are mostly determined by the amount of halogen-induced loss, springtime ozone in the Arctic is strongly influenced by variability in transport within the Brewer-Dobson circulation (BDC). The deep branch of the BDC, comprising of rising tropical air reaching the mid and upper stratosphere, moving poleward and descending at high latitudes, is most relevant for ozone and climate in the polar regions (Lin and Fu, 2013; Butchart et al., 2014).

Figure 65 shows timeseries of the Arctic mean $\overline{w^*}$ a(the vertical component of the residual mean meridional circulation in the Transformed Eulerian Mean framework, see Andrews et al., 1987) at 30 hPa in DJF (note positive values indicate upwelling). There is a mean increase in downwelling over the polar cap from ~1.8 mms^{-1} to ~2 mms^{-1} over the 1981-2080 period at a rate of ~0.015 (±0.007) mms^{-1} $decade^{-1}$. Such an enhancement of the BDC is found in most CCMs (Butchart et al., 2010; Weber et al. 2011; Hardimann et al., 2014; Oberländer et al., 2013; Lin and Fu, 2013; Hardimann et al., 2014). A strengthening of the circulation will increase transport of ozone into the high latitudes, thereby contributing to the increase in springtime ozone over the century (Fig. 1(a)). In addition, it will drive adiabatic heating that would tend to offset the radiative cooling from increasing CO_2. This in turn will impact on the Arctic temperature trends discussed in Sect. 3.2.2, in agreement with Butchart et al. (2010) and Langematz et al. (2014). These authors attributed thereported statistically insignificant mid-winter (DJF and JF mean, respectively) temperature trendsresponses in the Arctic lower stratosphere, which they postulated to arise due to the compensation effect between the radiative cooling and dynamically driven warming.

In addition to the long-term trend in wintertime $\overline{w^*}$ over the Arctic, there is also large interannual and interdecadal variability throughout the century. For example, we find a series of winters in the 2060s and 2070s in which the downwelling is anomalously weak. These coincide with years with an anomalously strong and cold polar vortex (not shown) and anomalously low Arctic column ozone in spring (Fig. 1(a)). Our model calculations therefore suggest that the dynamical conditions favouring low springtime ozone are expected to continue to occur in the future. There is also a period in the 2050s when most of the ensemble members show relatively enhanced downwelling over the Arctic (Fig. 65) and higher column ozone amounts (Fig. 1(a)). Investigating the detailed drivers of this interdecadal variability is beyond the scope of this study, however, the results highlight the importance of both long-term trends and interannual and interdecadal variability for the future evolution of Arctic ozone.

Manney et al. (2011) and Langematz et al. (2014) have stressed that the occurrence of large ozone depletion episodes in the Arctic depends not only on the strength of the vortex during mid-winter, but also on its persistence into spring. A relatively long-lived vortex extends the period in which temperatures fall below the PSC formation threshold, thereby allowing for continued halogen activation, substantial denitrification, as well as postponeding the resupply of ozone to the pole following the vortex break-up. It is therefore important to consider possible future changes in the strength as well as formation and break-up dates of the vortex.

Figure 76 shows linear trends in zonal mean zonal wind (u) at 610°N in (a) November/December and (b) March. In early winter, the ensemble mean trend shows a strengthening of the winds in the lower and mid-stratosphere of ~0.1-0.25 ms^{-1} $decade^{-1}$. This is in broad agreement with the findings of Langematz et al. (2014) analysed the timing of the formation of the NH polar vortex and found a statistically significant trend towards earlier vortex formation. It is possible that the strengthening of the stratospheric zonal wind in autumn/early winter in our ensembles could be related to a similar effect. HowerImportantly, there are also ensemble members that show a near-zero wind changes in the lower/mid stratosphere and/or a weakening of the westerlies above (10 hPa), highlighting the challenge of extracting robust trends in the presence of large dynamical variability. The ensemble spread is much larger in March (Fig. 76(b)) and, consequently, no statistically significant ensemble mean trends in the vortex strength can be found (see also Langematz et al., (2014)).

We find that the large interannual dynamical variability that characterises the winter-time Arctic stratosphere is expected to persist in the future. ThereforeIn consequence, individual years characterised by dynamical conditions facilitating low column ozone will continue to occur into the future. In the presence of declining stratospheric halogen levels, this dynamical variability will become increasingly important for determining interannual variability in Arctic spring total column ozone. However, as discussed in Sect. 3.2.1, halogen chemistry will also have a role to play. This role will reduce during the next several decades but will nevertheless remain important.

3.3 Case study of exceptionally low and average ozone events

The previous sections have examined the future behaviour of Arctic ozone in the UM-UMKCA ensemble, as well as its chemical and dynamical drivers in the UM-UKCA ensemble. To further illustrate the potential impact of these processes in individual winters we now present a case study from the ensemble of an anomalously low total ozone event that occurs many decades into the future. Although the mean ozone column in the Arctic region increases steadily throughout the 21st century (Fig. 1(a)), individual winters continue to occur in which Arctic mean ozone is up to ~50-100 DU lower than the corresponding long-term mean. The green point in Figure 1(a) highlights the model year 2063 modelled in one of the ensemble members. This particular year has an Arctic mean March ozone column of 383 DU, which is lower than the long-term minimum of the ensemble mean found in the late 1990s. We compare this to a model year with near average total ozone from the same period (2060, marked by orange point in Fig. 1(a)), whose March Arctic mean ozone column of ~489 DU is approximately 100 DU higher than in the model year 2063. These two case study model years are analysed below. Note that the years 2063/2060 are used here as a naming convention, rather than in reference to some specified years in the future.

Figure ~~7~~ 8 shows timeseries of the daily Arctic mean total column ozone during the two case study winters of the model years 2063 (solid red) and 2060 (solid black). Column ozone increases throughout autumn and winter in both years due to transport of relatively ozone rich air from the tropics by the ~~Brewer-Dobson circulation~~BDC (Strahan et al., 2013). Note that the Arctic mean encompasses not only the stratospheric polar vortex, but also some regions outside of it. For this reason, Fig. ~~7~~8(a) also shows the vortex averaged column ozone for~~in~~ the two model years ~~2063~~ for comparison (dashed ~~red~~lines), defined here by the geographic region where daily mean Ertel's potential vorticity on the 850 K potential temperature surface is greater than 6×10^{-4} m^2 s^{-1} K kg^{-1}. These show that s~~S~~imilar but somewhat smaller differences between the two years are found when~~if~~ the vortex-average column ozone is considered, compared with the Arctic mean. ~~(not shown).~~

The differences in column ozone between the two model years are smaller during late autumn and early winter than in subsequent months; this is in broad agreement with comparisons of similar years in observations (Strahan et al., 2013). Larger differences between the two years occur from late December onwards. In 2063, column ozone increases more slowly between mid-winter and spring, particularly in the Arctic mean. On 1 February, the Arctic mean column is 354 DU in 2063, which is 67 DU lower than in 2060. This is associated with an anomalously strong and cold polar vortex (see Fig. ~~7~~8(b) and ~~8~~9(b)), and relatively weak stratospheric wave driving in early winter (not shown). The strong polar vortex is associated with reduced downward transport of air inside the vortex. Consistently, the DJF mean 30hPa $\overline{w^*}$ is a mere -0.95 mms^{-1}, which is ~1.05 mms^{-1} less negative than the 11-year running mean for that period (Fig. ~~5~~6).

The differences in column ozone between the two model years are even larger in late winter and spring. In 2063, the polar vortex remains strong until early April (Fig. ~~8~~9(b~~a~~)), and total ozone reaches only ~390-395 DU by the end of March. The vortex average column ozone levels off and oscillates around ~360 DU until the beginning of the vortex break-up in mid-April (Fig. ~~8~~9(b~~a~~)). In contrast, in 2060 the polar vortex weakens substantially in mid-February and does not fully recover before the transition to summertime easterlies occurs (Fig. ~~8~~9(a~~b~~)). Consequently, the Arctic mean column ozone rises to ~490-500 DU by the end of March.

Figures ~~9~~10(a) and (b) show time-altitude cross-sections of the differences in Arctic mean ozone mixing ratio, and ozone concentrations, respectively, between the model years 2063 and 2060. Small differences in ozone of up to ~0.5-1 ppm and ~1×10^{-12} molecules cm^{-3} are already present in late autumn at ~25 km, which contribute to the differences in column amounts evident in Fig. ~~7~~8(a). However, larger differences appear in the mid and upper stratosphere from the end of December onwards. This is particularly evident in the mixing ratios, with up to ~2.5-3 ppm less ozone between 30 and 40 km in 2063 compared to 2060. However, owing to the exponential decay in pressure with altitude the deficit is less pronounced in terms of absolute ozone amounts (up to ~0.5×10^{-12} molecules cm^{-3}). This anomaly is predominantly dynamically driven and is transported downward over the course of the winter, with differences in March maximising at 25-30 km. Ozone abundances are also reduced in the lower stratosphere in 2063 throughout winter and spring. Even though the differences in ozone mixing ratios at these levels are smaller compared with those in the mid and upper stratosphere, the higher ambient pressure results in significant changes in absolute ozone amounts. These therefore make a substantial contribution to the difference in total ozone columns in Fig. ~~7~~8(a). In general, the deficits at all altitudes magnify from mid-

winter to early spring, with ~1-1.75×10^{-12} molecules cm^{-3} less ozone in the 10-30 km region in 2063 compared to 2060 (Fig. ~~9~~10(b)). The difference in partial ozone column up to 25 km contributes a relative decrease of ~70 DU by the end of March. These are the altitudes where halogen catalysed ozone destruction plays its most significant role, as evidenced by elevated ClO concentrations in 2063 (Fig. ~~9~~10(c)).

Figure ~~7~~8(b) shows timeseries of the Arctic mean temperature at 22.7 km, in the region of NAT PSC formation, in 2060 (black) and 2063 (red). In 2060, the temperatures during early and mid-winter fluctuate around 210 K, but do not drop below ~203 K in the Arctic mean. In contrast, the Arctic mean temperature in 2063 oscillates near or below 195 K during most of January and February. The minimum local daily mean temperatures in 2063 reach 181 K (c.f. 191 K in 2060). The low temperatures allow ~~Th~~the formation of PSCs (see red star in Fig. 5, Sect.3.2.3), which in turn permits heterogeneous chemical reactions that lead to the activation of chlorine from its reservoir species, HCl and $ClONO_2$, into reactive forms (ClO, see Fig. S3, Supplementary Information). In early winter in 2063, this activation takes place predominantly near the sunlit vortex edges, which could contribute to some ozone loss in the mid-latitudes (Pyle et al., 1994; Millard et al., 2002). From mid-January onwards, Arctic mean ClO levels in the lower stratosphere are ~125-150 ppt ~~near 22~~at 21.5 km (Fig. S3) and ~100-125 ppt at 19 km (not shown). This is up to ~75-125 ppt higher than in 2060 (Fig. ~~9~~10(~~c~~b)). While the Arctic mean values are relatively modest, local daily mean ClO levels in March reach up to ~300 ppt at 19 km in 2063 (c.f. ~80 ppt in 2060). The very low temperatures in the model year 2063 also lead to substantial denitrification such that the formation of PSCs, and halogen activation, from mid-winter is strongly reduced (not shown).

The dashed blue line in Fig. ~~7~~8(a) shows vortex-average column ozone amounts in 2063 for a passive stratospheric ozone tracer, which when compared with the full (chemistry and transport) vortex-average ozone field enables a quantification of the relative roles of transport and (all) chemical processes in determining column ozone (see Tegtmeier et al. (2008) for an alternative approach). This passive tracer is initialised to the ozone field on 1 November and undergoes no chemical production or loss, but is advected by the circulation. The full ozone field and the passive tracer for 2063 track each other quite well, particularly in early and mid-winter. By the end of March~~:~~ both are relatively low and~~very~~ different from the ozone calculated in 2060 ~~(~100 DU difference in the columns for the full ozone field between 2060 and 2063 in March)~~ and it seems, therefore, that a major reason for the low column ozone in 2063 ~~difference between the two years~~ is dynamical in origin. There is also a difference between the full and passive ozone columns in 2063, which increases ~~during the late winter/early spring~~with time, with the cumulative effect of all chemical processes in the preceding five months averaged over the polar vortex contributing ~~to~~ a ~~~~2450 DU ~~difference~~ decrease to the total ozone column by the end of March in that year. ~~to the respective Arctic mean ozone columns (~25 DU within the vortex). Recall that the passive tracer ignores all chemical processes (and not just polar halogen chemistry). Chemistry evidently also plays a non negligible role in 2063.~~ This shows that while dynamical processes are the dominant factor contributing to the low column ozone in 2063, chemical processes can not be neglected. We caution, however, against over-interpretation of this value as it is the result of a complex balance between different chemical loss and production cycles throughout the depth of the atmosphere as well as their interaction with transport throughout the winter.

Using our other diagnostic, i.e. ~~In comparison,~~ the cumulative (1. November to 30. March) halogen induced ozone loss~~es~~ in the lower atmosphere (up to 25 km) ~~from 1 November to 30 March due to~~, we find that the 6 main halogen cycles (see Sect. 2.3) result in~~are ~~39~~40~~ DU ozone loss in 2063 compared to ~~18~~20~~ DU in 2060 (with very similar values of 44 DU and 23 DU, respectively, if we take vortex average). By this measure, the difference in estimated halogen losses between the two model years (~20 DU) ~~can therefore account for~~ is equivalent to about 20% of the full difference (~100 DU, see above) in the end of March Arctic mean total column ozone ~~at the end of March~~ between the two years. ~~The estimate of ~40 DU halogen loss in 2063 is in good agreement with the chemical loss of ~50 DU derived from the comparison of the passive ozone tracer and the chemical ozone field.~~ Whilst the two diagnostics (i.e. chemical ozone loss derived from the passive ozone tracer and the halogen-induced ozone loss in the lower atmosphere) ~~methods~~ are not equivalent, both indicate that dynamical processes are likely to be a dominant driver of future interannual variability in Arctic ozone. However, as illustrated by the later case, loss of ozone through halogen chemistry remains a~~n~~ smaller but nonetheless important contributor to the modelled Arctic ozone variability even in the second half of the 21st century.

4. Conclusions

On its own, the projected decline in stratospheric Cl_y over this century is expected to lead to the recovery of stratospheric ozone to pre-1980s values. However, a complex balance of chemical and dynamical processes will determine the precise evolution of Arctic ozone. For example, some studies have suggested that the Arctic lower stratosphere has become colder, with an increase in the incidence of temperatures low enough for the formation of PSCs (Rex et al., 2004, 2006). This could be related to the recent observation of particular years with exceptionally low Arctic springtime total column ozone (e.g. Manney et al., 2011). If there is indeed a trend, a key question is whether it is a forced response to increased greenhouse gas concentrations. However, other studies (e.g. Rieder and Polvani, 2013) have argued that any observed changes in the Arctic stratosphere are consistent with natural variability. How the Arctic stratosphere and Arctic springtime ozone will evolve in the future therefore remains a key research question.

In this study, future trends in Arctic springtime total column ozone are assessed using a 7 member ensemble from the UM-UKCA CCMI REFC2 integrations. The Arctic mean March total column ozone increases throughout the 21st century in the simulations at a rate of ~11.5 DU decade^{-1}, and is projected to return to the 1980 level in the late 2030s. This is consistent with the long-term reduction in Cl_y levels~~ and~~ GHG-induced stratospheric cooling as well as strengthened large scale meridional circulation, and is in overall agreement with previous modelling studies (Eyring at al., 2010; WMO, 2011, 2014). Importantly, the integrations indicate that the high interannual variability that characterises the Arctic ~~region~~ stratosphere today will persist in the future. Even beyond 2060 ozone can episodically drop ~50-100 DU below the corresponding long-term mean to near present-day values. These events coincide with an anomalously cold and strong polar vortex and reduced downwelling over the polar cap. These dynamical conditions are associated with reduced transport and mixing of ozone into the Arctic region and enhanced formation of PSCs which promotes activation of chlorine from its reservoir forms.

With regard to long-term trends, the ensemble mean shows a statistically significant cooling throughout most of the polar stratosphere in early winter, in agreement with the findings of Langematz et al. (2014). However, ~~an~~ individual ensemble member can show ~~a~~ near-zero temperature trends~~response~~ in the lower stratosphere, highlighting the need for ensemble studies for confident detection of trends in the polar lower stratosphere. During mid-winter, the inter-ensemble spread is larger and a significant cooling trend in the ensemble mean is only identifiable at pressures less than 30 hPa. In March, while all the ensemble members indicate cooling of the mid and upper stratosphere, the inter-ensemble spread is large in the lowermost stratosphere at 100 hPa and no ensemble mean temperature trend can be found.

The results indicate a strengthening of the deep branch of the Brewer Dobson circulation in boreal winter, which is manifested by an increase in downwelling over the Arctic from December to February of ~0.015 (±0.007) mms^{-1} $decade^{-1}$ (at 30 hPa); in agreement with previous modelling studies (Butchart et al., 2010; Weber et al., 2011; Oberländer et al., 2013; Lin and Fu, 2013; Hardimann et al., 2014). Such an increase in the large-scale circulation will increase transport of ozone into the high latitudes, thereby contributing to the long-term increase in the spring Arctic ozone. In addition, it will also drive adiabatic warming in the Arctic stratosphere, thereby offsetting the radiatively-driven cooling from increasing CO_2. This compensation may contribute to the lack of statistically significant temperature trends in the lower stratosphere in mid-winter, in agreement with the results of Langematz et al. (2014). The simulations also show a small strengthening of the zonal mean zonal winds in the extratropical lower and mid stratosphere in early winter of ~0.1-0.25 ms^{-1} $decade^{-1}$, but no significant trends in ~~spring~~March.

Consistent with the decline in Cl_y over the century, the estimated halogen induced chemical ozone loss in the lower atmosphere decreases by around a factor of two between 1981-2000 and 2061-2080. This indicates a reduced role of halogen chemistry as a driver of springtime Arctic ozone and its variability. However, there are still individual winters ~~in the~~ later in the century~~period~~ when dynamical conditions are favourable for the occurrences of elevated halogen losses. We conclude that despite the future reduction in levels of stratospheric halogens, chemical processes will play a smaller but non-negligible role in determining ~~the occurrence of low column~~ Arctic springtime column ozone throughout the century.

These points are exemplified by a case study of a low ozone year simulated in the 206~~3~~0s. Even though the ensemble mean March Arctic Cl_y in the lower stratosphere has dropped by ~50% with respect to the year 2000, the ~~corresponding~~ column ozone in this year is ~100 DU lower than the 11-year running mean and is close to the long-term minimum modelled near the turn of the century. This particular modelled winter is characterised by an anomalously strong and cold polar vortex that persists largely undisturbed until mid-April. Significantly suppressed transport reduces ozone supply throughout the winter. In addition, the anomalously low temperatures facilitate PSC formation and subsequent chlorine activation. The difference in the estimated halogen losses between this low ozone year and a year from the same period with near average springtime ozone is equivalent to ~~account for~~ ~20% of the difference ~~between~~ in~~the~~ March total ozone column between the two years ~~in 2063 and a year from the same period with near average springtime ozone~~.

To conclude, our integrations suggest that the long-term recovery of Arctic ozone, driven by the future reduction in stratospheric Cl_y, GHG-induced cooling, and the increased strength of the large-scale stratospheric meridional circulation,

means that the likelihood for individual years with springtime Arctic ozone depletion as severe as that observed in 2011 (e.g. Manney et al., 2011) will decrease in the future. However, the large interannual variability that characterises the Arctic polar vortex is expected to continue and this is likely to facilitate significant deviations of March ozone columns from long-term background values. Whilst our results suggest that dynamical processes will continue to play an important role for determining Arctic springtime ozone in the future, halogen chemistry will remain a smaller but non-negligible contributor for many decades to come.

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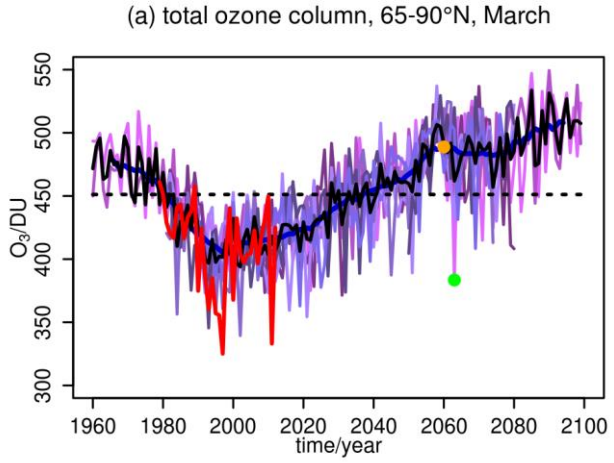
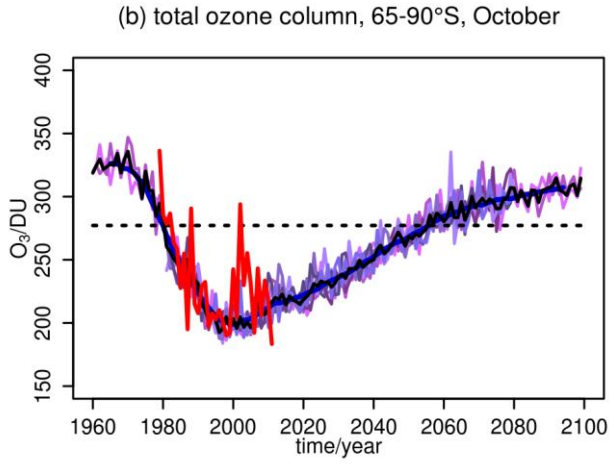

(a) total ozone column, 65-90°N, March
O3/DU
300
350
400
450
500
550
1960
1980
2000
2020
2040
2060
2080
2100
time/year


(b) total ozone column, 65-90°S, October
O3/DU
150
200
250
300
350
400
1960
1980
2000
2020
2040
2060
2080
2100
time/year

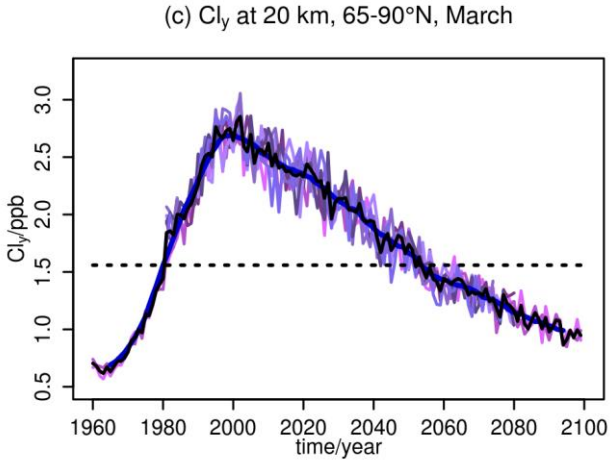

(c) Cly at 20 km, 65-90°N, March
Cly/ppb
0.5
1.0
1.5
2.0
2.5
3.0
1960
1980
2000
2020
2040
2060
2080
2100
time/year

Figure 1. Timeseries of monthly mean polar averaged (65-90°) total column ozone [DU] from 1960-2100 for (a) the Arctic in March, and (b) the Antarctic in October. (c) As in (a) for the March total inorganic chlorine loading (Cl_y) at 20 km [ppb]. The red lines in (a) and (b) denote observations from the Bodeker total ozone column dataset (Bodeker et al. (2005); Müller et al. (2008)). Thick black and blue lines denote the ensemble mean and its 11-year running average, respectively. Orange and green points in (a) denote the two case study years of 2060 and 2063, respectively, described in Sect. 3.3.

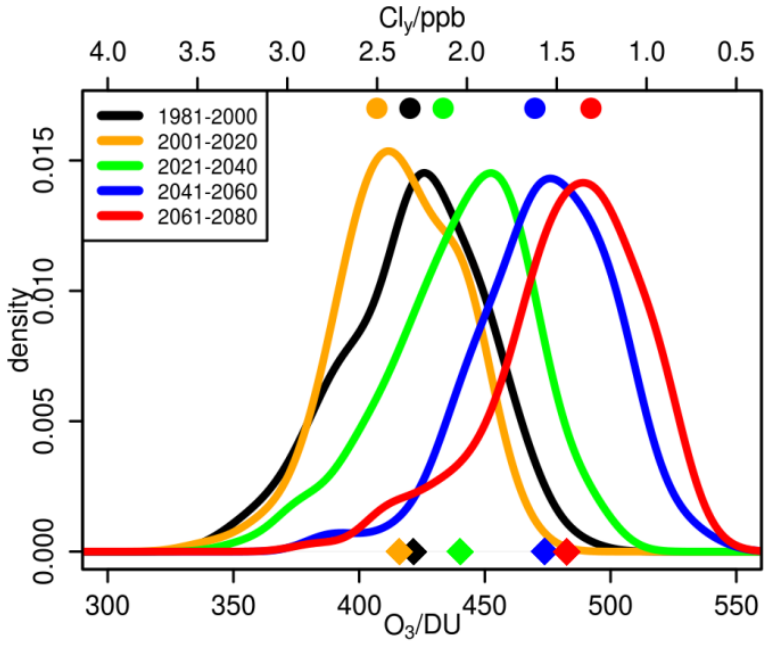


Figure 2. Probability density functions (PDFs) for 20-year intervals taken from the period 1981 to 2080 for Arctic mean total ozone column [DU] in March. Each PDF contains data from 7 ensemble members. Coloured diamonds indicate the 20-year means of the total ozone columns and coloured points the corresponding means of Arctic mean Cl_y [ppb] at 20 km.

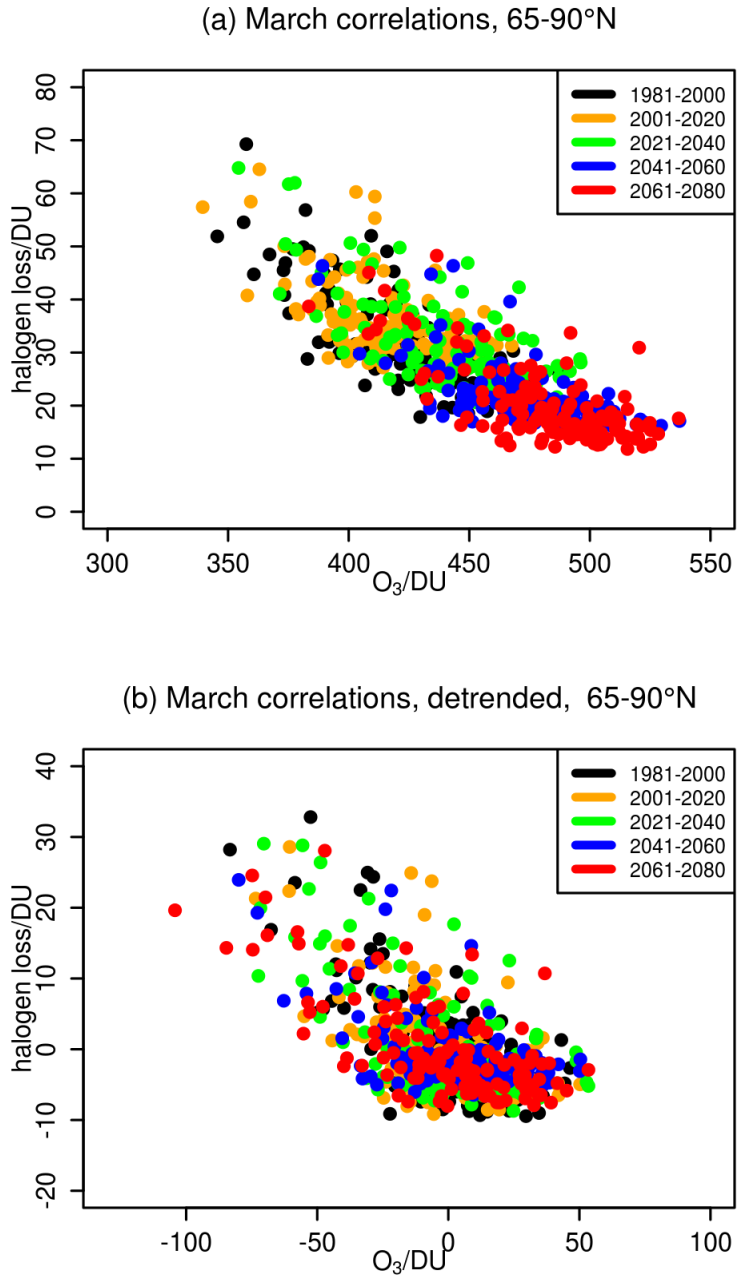


Figure 3. (a) Scatterplot of Arctic mean March total column ozone [DU] versus cumulative (1 November to 30 March) halogen induced total ozone loss [DU] (see Sect. 2.3) summed from the surface to 25 km. Each point shows a single winter and the colours denote the same 20 year intervals as in Fig. 2. (b) As in (a) but for deviations of column ozone and halogen losses from their corresponding 11-year running ensemble mean.

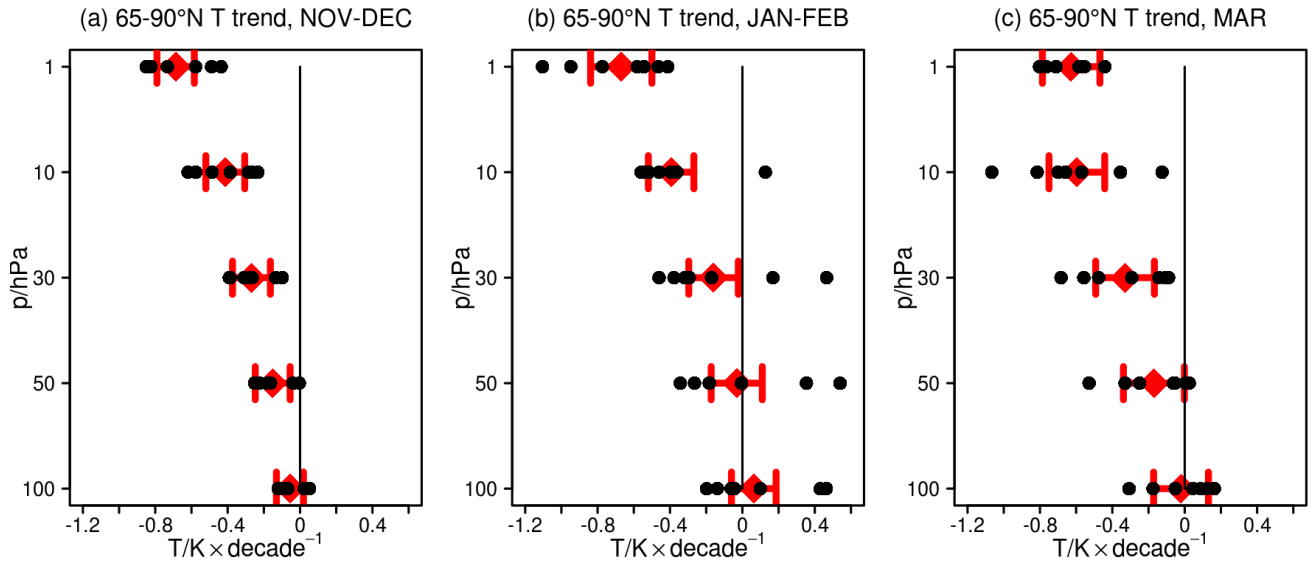


Figure 4. Linear trends in Arctic mean temperature [K decade^{-1}] at selected stratospheric pressure levels in (a) November/December, (b) January/February and (c) March calculated for each ensemble member individually (black points) for the period 1981-2080. Red diamonds and whiskers show the corresponding trends (±2 standard errors) calculated for the ensemble mean ~~±2 standard errors~~.

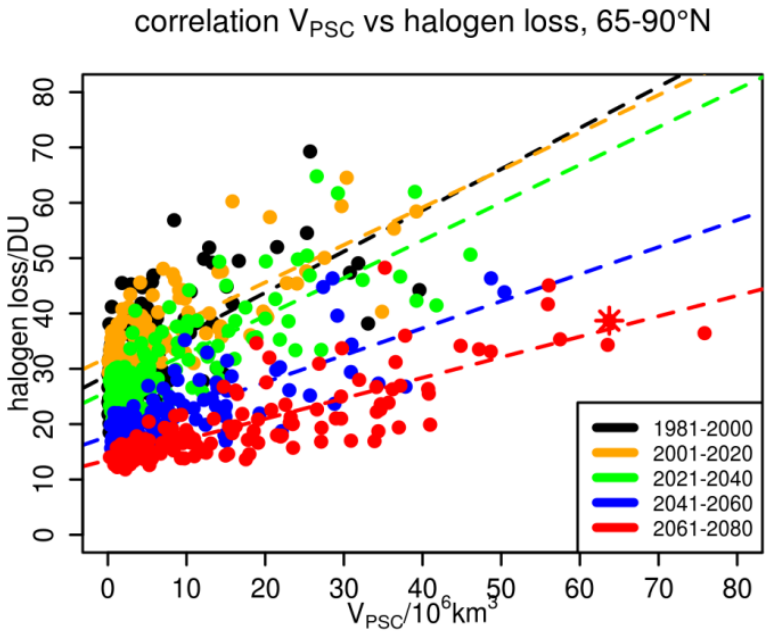


Figure 5. Scatterplot for November to March mean potential V_{PSC} [$\times 10^6$ km^3] against halogen induced ozone loss [DU] over 1-25 km integrated over the same months. Colours denote the same 20 year intervals as in Fig. 2. Dashed lines indicate linear fits for the individual 20 year periods. The model year 2063 analysed in Sect. 3.3 is highlighted with a red star.

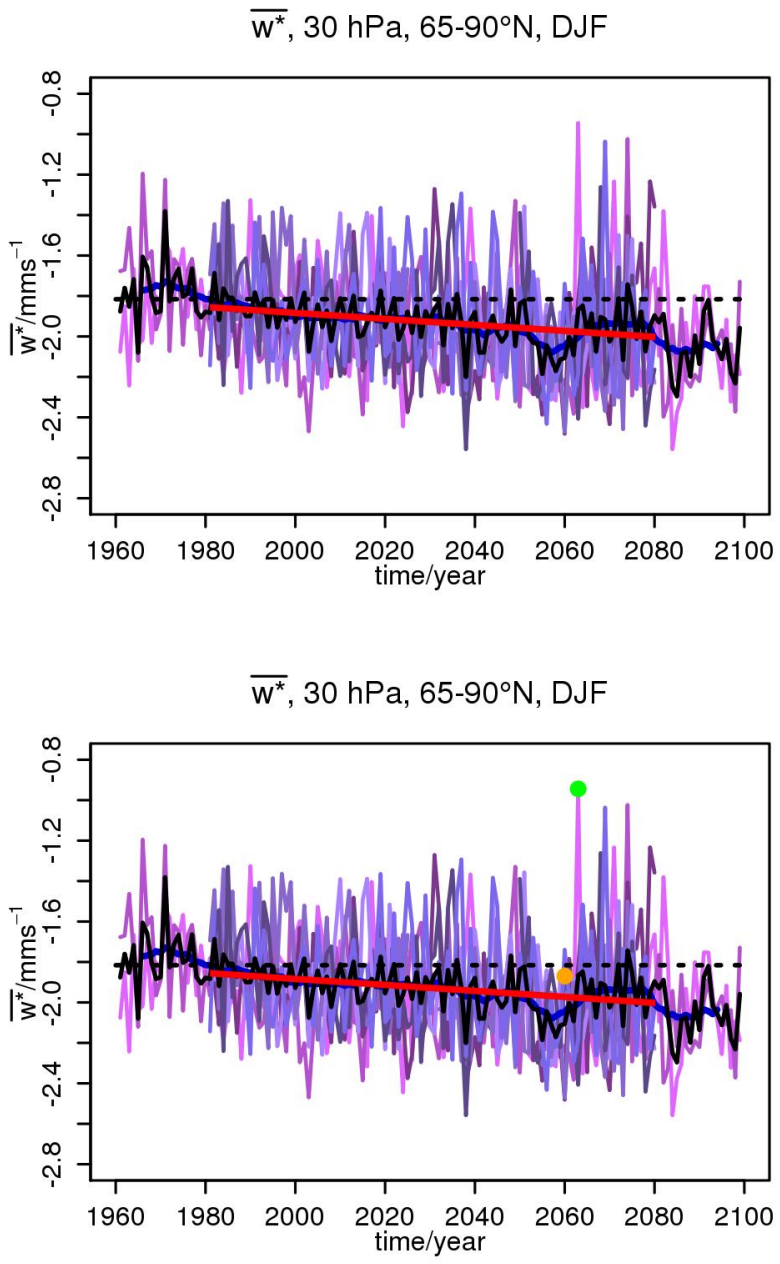


Figure ~~5~~6. Timeseries of DJF mean residual vertical velocity, $\overline{w^*}$ [mms^{-1}], over 65-90°N at 30 hPa from 1960-2100. Thick black and blue lines denote the ensemble mean and its 11-year running average, respectively. Thick red line shows the linear trend calculated for the ensemble mean over the 1981-2080 period. Orange and green points denote the two case study model years 2060 and 2063, respectively, discussed in Sect. 3.3.

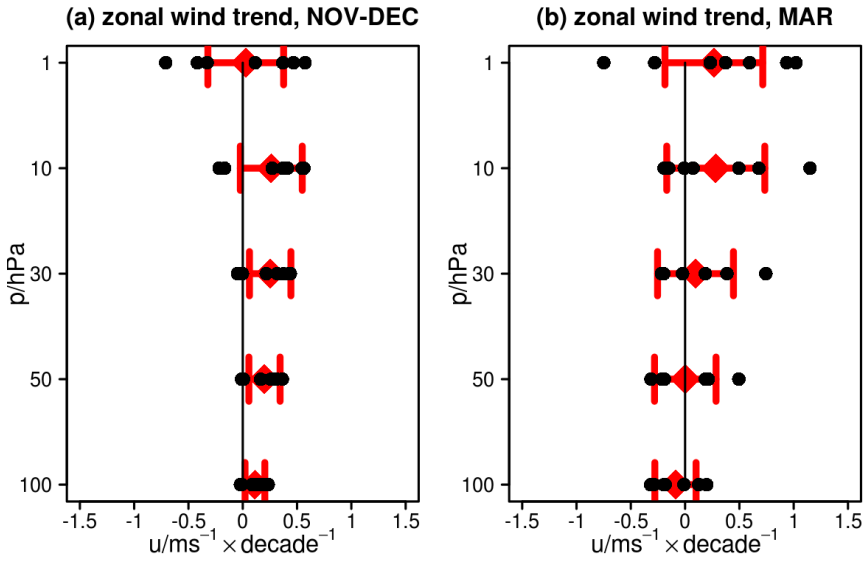
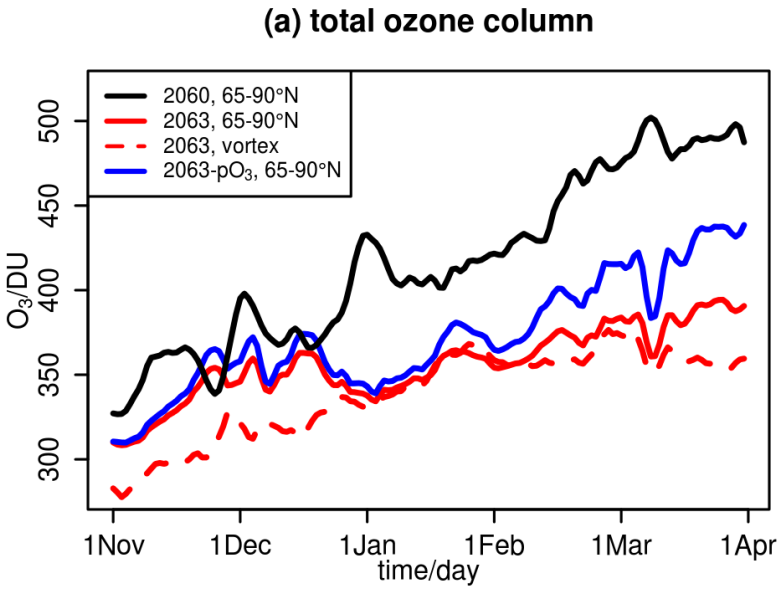
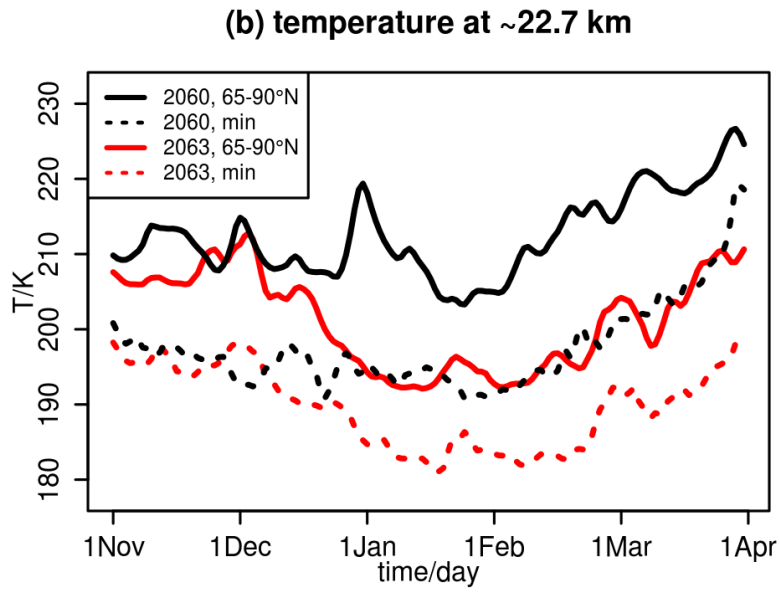
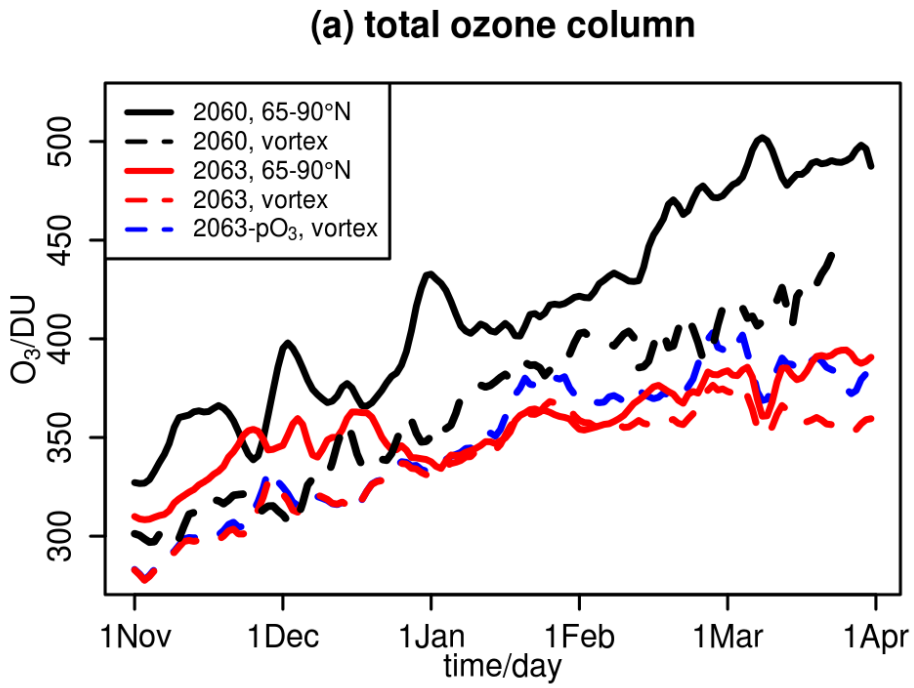


Figure ~~6~~7. ~~As in Fig. 4, but for the linear trends in zonal mean zonal wind [ms^{-1} decade^{-1}] at 60°N in (a) November/December and (b) March.~~Linear trends in zonal mean zonal wind [ms^{-1} decade^{-1}] at 61°N at selected stratospheric pressure levels in (a) November/December and (b) March calculated for each ensemble member individually (black points) for the period 1981-2080. Red diamonds and whiskers show the corresponding trends (±2 standard errors) calculated for the ensemble mean.


(a) total ozone column
2060, 65-90°N
2063, 65-90°N
2063, vortex
2063-pO3, 65-90°N
O3/DU
300
350
400
450
500
1Nov
1Dec
1Jan
1Feb
1Mar
1Apr
time/day


(b) temperature at ~22.7 km
2060, 65-90°N
2060, min
2063, 65-90°N
2063, min
T/K
180
190
200
210
220
230
1Nov
1Dec
1Jan
1Feb
1Mar
1Apr
time/day


(a) total ozone column
2060, 65-90°N
2060, vortex
2063, 65-90°N
2063, vortex
2063-pO3, vortex
O3/DU
300
350
400
450
500
1Nov
1Dec
1Jan
1Feb
1Mar
1Apr
time/day

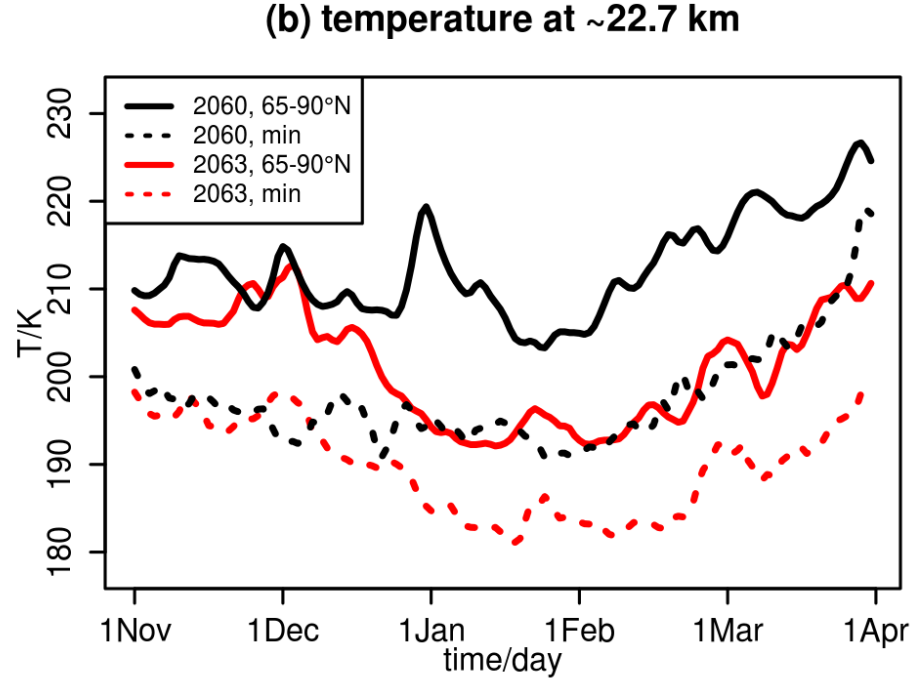

(b) temperature at ~22.7 km
2060, 65-90°N
2060, min
2063, 65-90°N
2063, min
T/K
180
190
200
210
220
230
1Nov
1Dec
1Jan
1Feb
1Mar
1Apr
time/day

Figure ~~7~~8. Timeseries of Northern Hemisphere (a) daily total column ozone [DU] during the winters 2060 (black) and 2063 (red), and (b) temperature [K] at 22.7 km. Solid lines show the Arctic mean (65-90°N), dashed ~~red~~ lines in (a) show~~s~~ polar vortex average~~s for 2063~~ (see text for details) and dotted lines in (b) show minimum daily mean temperatures found anywhere poleward of 65°N. The blue dashed~~solid~~ line in (a) shows the evolution of the vortex-averaged~~Arctic mean~~ passive ozone tracer in 2063.

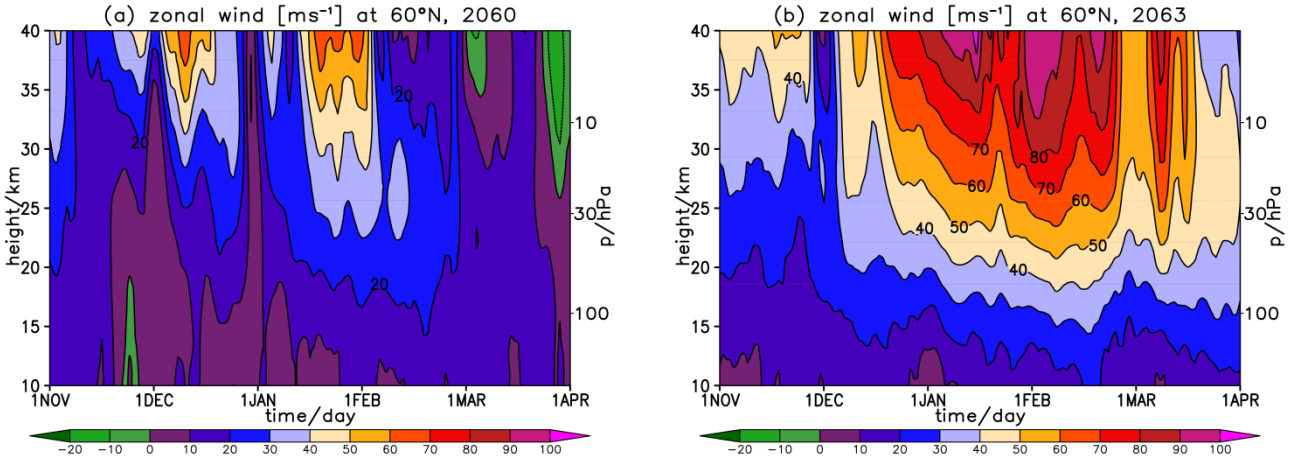


Figure ~~8~~9. Time-altitude cross-sections of the daily Arctic (60°N) zonal mean zonal wind [ms^{-1}] in (a) 2060 and (b) 2063.

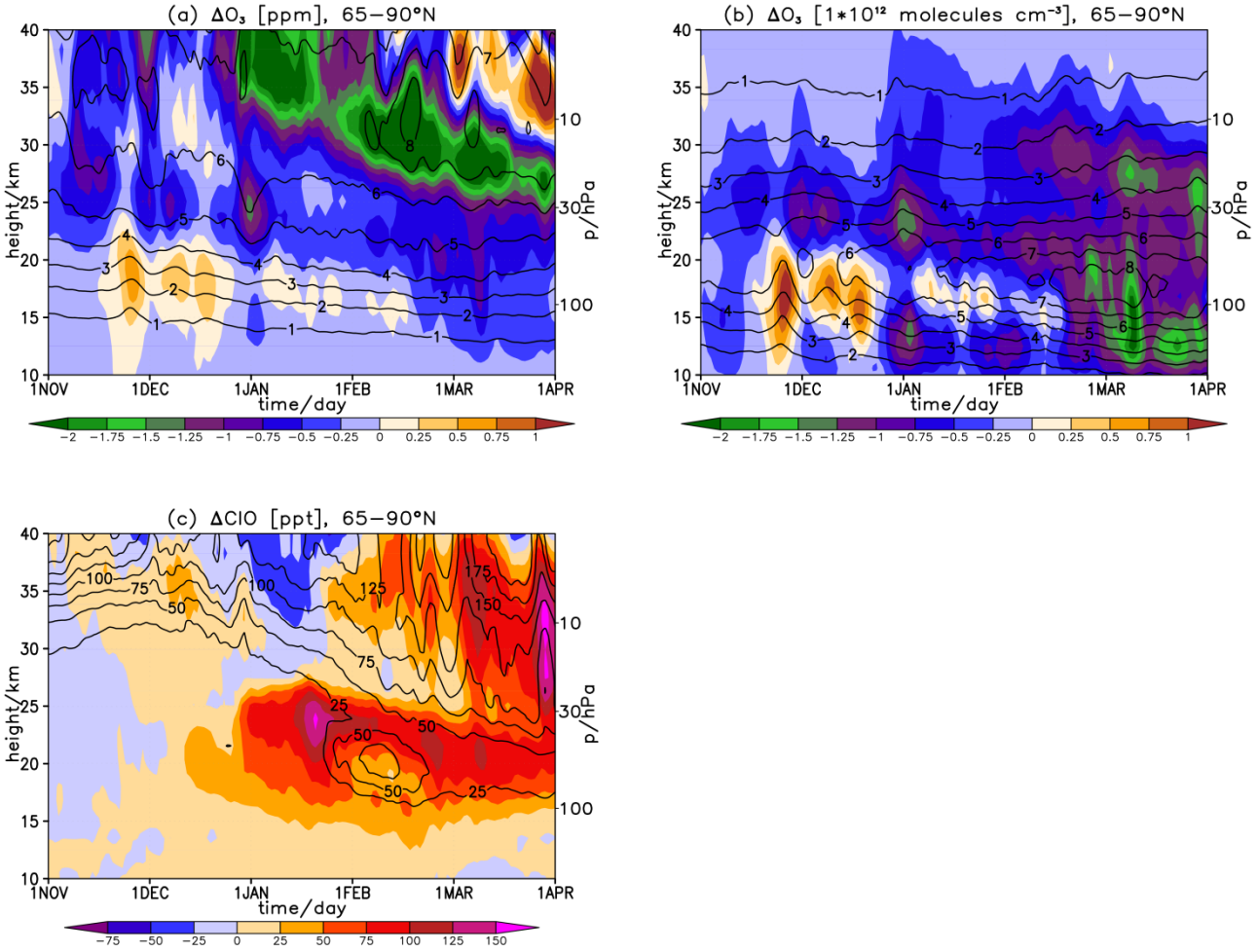


Figure ~~9~~10. Colours: Time-altitude cross-sections of the Arctic mean differences between the years 2063 and 2060 in (a) ozone mixing ratios [ppm], (b) ozone number density [10^{12} molecules cm^{-3}] and (c) ClO mixing ratio [ppt]. The solid contours show the year 2060 for reference.

As noted in Sect. 2.2~~,~~ of the manuscript, our UMUKCA REFC2 ensemble of integrations consists of 2 full 1960-2099 integrations (ENS1-2) and 5 shorter runs ~~from~~ covering November 1980 to December 2080 (ENS3-7). For technical reasons, data from 5 six-year-long intervals ~~in total~~ were excluded from the analysis, in particular:

- July 2025 – June 2031 in member number 5 (ENS5)
- April 2074 – March 2080 in member number 5 (ENS5)
- April 1996 – March 2002 in member number 6 (ENS6)
- April 2043 – March 2049 in member number 6 (ENS6)
- August 1982 – July 1988 in member number 7 (ENS7)

An example of the resulting timeseries is shown in Fig. S2~~1~~ for 65-90ºN March total ozone column.

Supplementary Figures

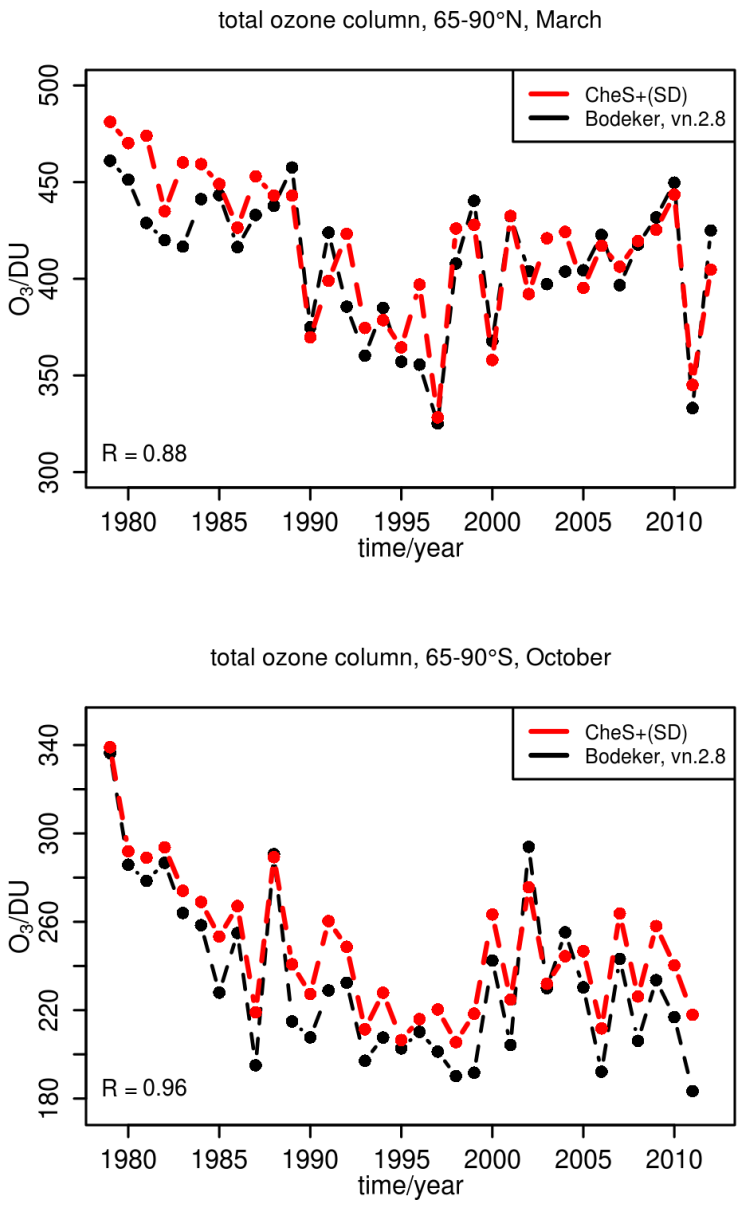


Figure S1. The evolution of total ozone column [DU] over 1979-2012 for 65-90°N March (top) and over 1979-2011 for 65-90°S October (bottom) in the nudged UMUKCA CCMI REFC1 CheS+(SD) integration (red) and observations (black, Bodeker total ozone column dataset: Bodeker et al., 2005; Müller et al., 2008). See Sect. 2.1 for details.

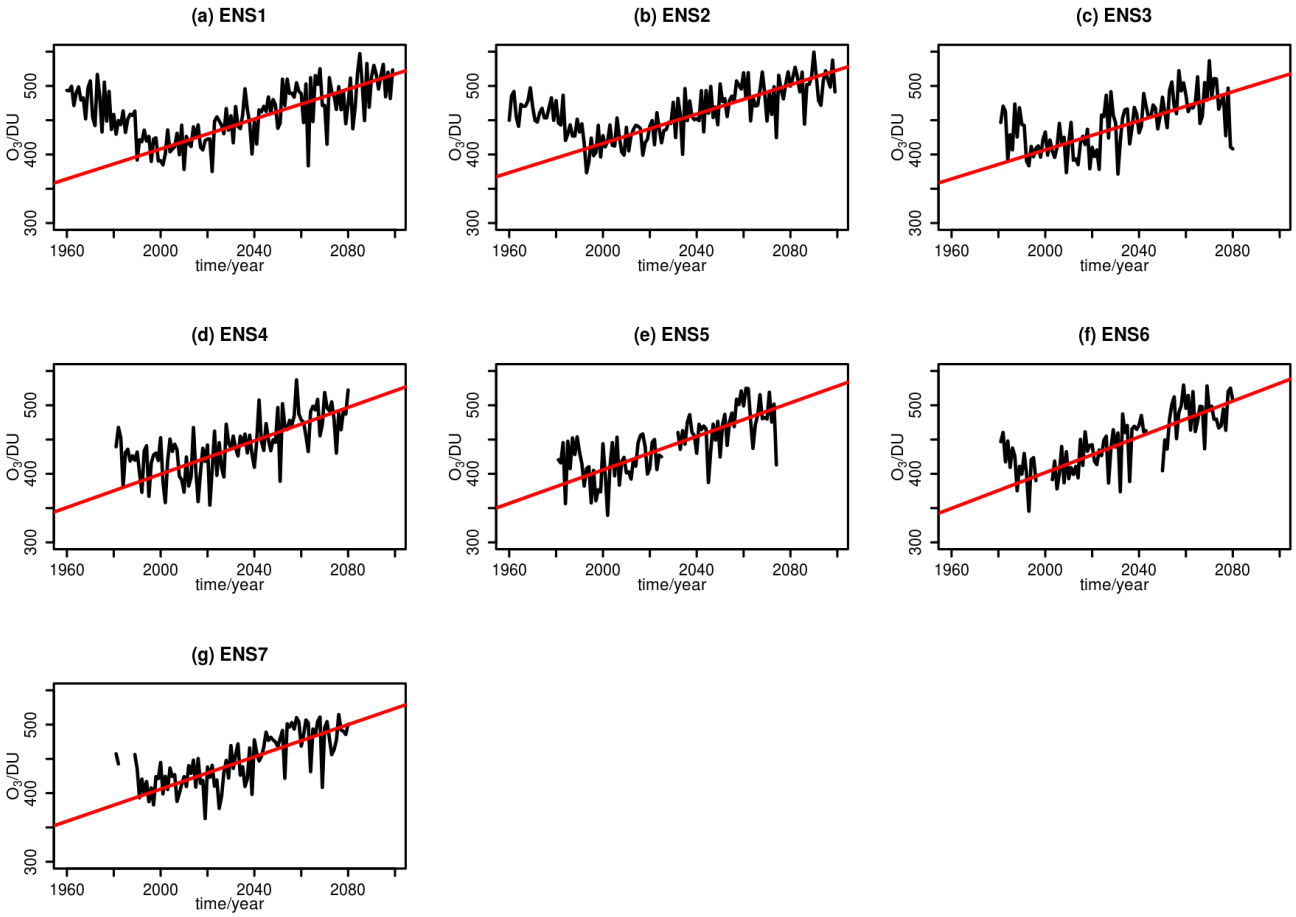


Figure S2~~1~~.~~:~~ (a-g) Timeseries of 65-90°N March total column ozone ~~columns~~ [DU] for individual ensemble members as labelled (black). Red lines show the corresponding ~~long-term~~ linear trends over the 2000-2080 period.

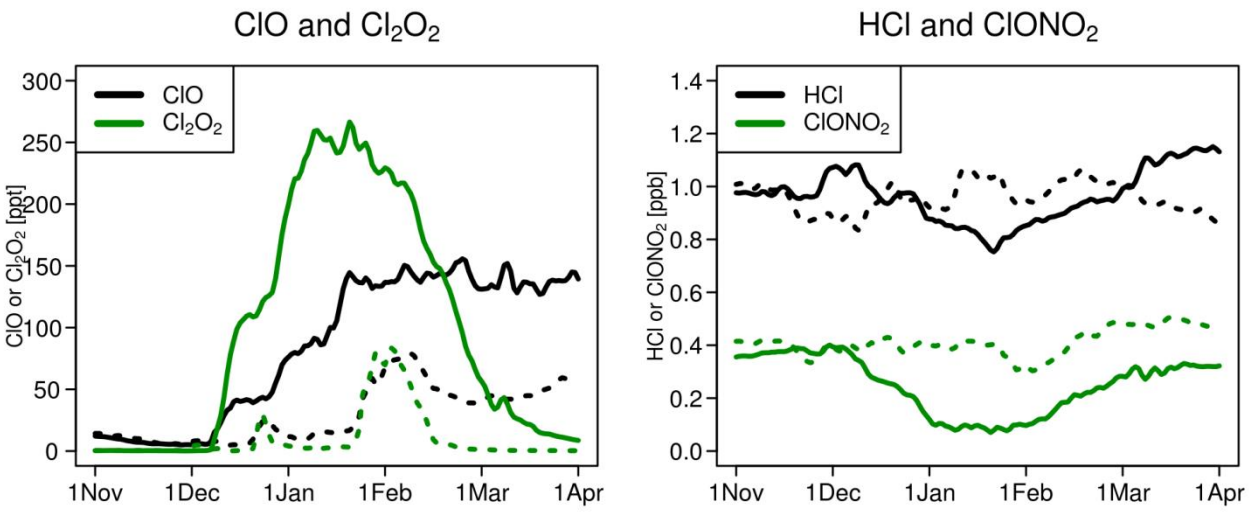


Figure S3. Timeseries of 65-90°N daily mean ClO and Cl_2O_2 [ppt] (left) and HCl and $ClONO_2$ [ppb] (right) at 21.5 km for the model case study years 2063 (solid lines) and 2060 (dashed lines).