

## 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  $\text{Cl}_y$  levels, chlorine activation and PSC volume (e.g. Rex et al., 2004)?*

An analysis of potential  $V_{\text{psc}}$  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  $V_{\text{psc}}$  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  $\text{ClO}$ ,  $\text{Cl}_2\text{O}_2$ ,  $\text{HCl}$  and  $\text{ClONO}_2$  for the two model case study years.

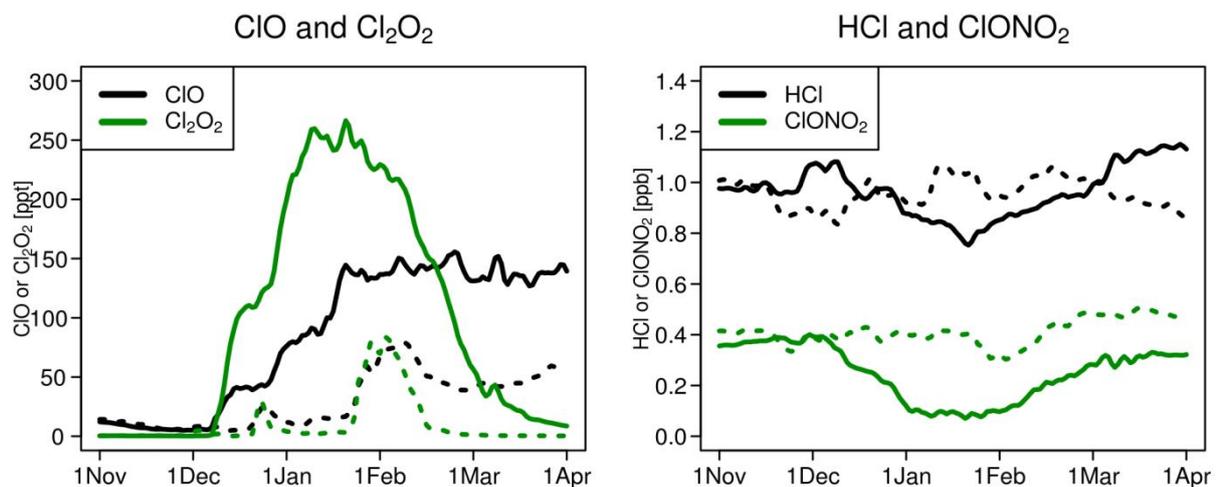


Figure S3. Timeseries of 65-90°N daily mean ClO and Cl<sub>2</sub>O<sub>2</sub> [ppt] (left) and HCl and ClONO<sub>2</sub> [ppb] (right) at 21.5 km for the case study model years 2063 (solid lines) and 2060 (dashed lines).

***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<sub>2</sub> + O(<sup>3</sup>P) reaction to be ~2DU/4DU in the model years 2063/2060, respectively.

While we agree that all ozone loss cycles (HO<sub>x</sub>, NO<sub>x</sub>... 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 V<sub>psc</sub> 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/CIO 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 14-13: 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<sub>x</sub> and chlorine reservoirs for the model years 2063/2060 (see Figure S3).

***P12 110-18: 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 113: what is the justification for the PV value to define the vortex?***

This is a simple and fairly 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).

#### **REFERENCES:**

Rex, M., Salawitch, R. J., von der Gathen, P., Harris, N. R. P., Chipperfield, M. P., and Naujokat, B.: Arctic ozone loss and climate change, *Geophys. Res. Lett.*, 31, L04116, doi:10.1029/2003gl018844, 2004.

Rex, M., Salawitch, R. J., Deckelmann, H., von der Gathen, P., Harris, N. R. P., Chipperfield, M. P., Naujokat, B., Reimer, E., Allaart, M., Andersen, S. B., Bevilacqua, R., Braathen, G. O., Claude, H., Davies, J., De Backer, H., Dier, H., Dorokhov, V., Fast, H., Gerding, M., Godin-Beekmann, S., Hoppel, K., Johnson, B., Kyro, E., Litynska, Z., Moore, D., Nakane, H., Parrondo, M. C., Risle, A. D., Skrivankova, P., Stubi, R., Viatte, P., Yushkov, V., and Zerefos, C.: Arctic winter 2005: Implications for stratospheric ozone loss and climate change, *Geophys. Res. Lett.*, 33, L23808, doi:10.1029/2006gl026731, 2006.

Gray, L. J., Scaife, A. A., Mitchell, D. M., Osprey, S., Ineson, S., Hardiman, S., Butchart, N., Knight, J., Sutton, R., and Kodera, K.: A lagged response to the 11 year solar cycle in observed winter Atlantic/European weather patterns, *J. Geophys. Res. Atmos.*, 118, 13,405–13,420, doi:10.1002/2013JD020062, 2013.

Jones, C. D., Hughes, J. K., Bellouin, N., Hardiman, S. C., Jones, G. S., Knight, J., Liddicoat, S., O'Connor, F. M., Andres, R. J., Bell, C., Boo, K. O., Bozzo, A., Butchart, N., Cadule, P., Corbin, K.

D., Doutriaux-Boucher, M., Friedlingstein, P., Gornall, J., Gray, L., Halloran, P. R., Hurtt, G., Ingram, W. J., Lamarque, J. F., Law, R. M., Meinshausen, M., Osprey, S., Palin, E. J., Chini, L. P., Raddatz, T., Sanderson, M. G., Sellar, A. A., Schurer, A., Valdes, P., Wood, N., Woodward, S., Yoshioka, M., and Zerroukat, M.: The HadGEM2-ES implementation of CMIP5 centennial simulations, *Geosci. Model Dev.*, 4, 543-570, doi:10.5194/gmd-4-543-2011, 2011

R. Müller, S. Tilmes, P. Konopka, J.-U. Grooß, H.-J. Jost: Impact of mixing and chemical change on ozone-tracer relations in the polar vortex, *Atmospheric Chemistry and Physics*, European Geosciences Union, 2005, 5 (11), pp.3139-3151