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Implications of constant CFC-11 concentrations for the future ozone layer

by Martin Dameris et al.

Reply to the referee #1 comments

Thank you to the referee for taking the time to review our manuscript. In the following we give our first answers regarding the points raised by the reviewer. The statements, comments and suggested corrections raised by the referees are printed in black and our comments are presented in blue. We will try to consider all of the raised points in the revised manuscript in an adequate manner.

Answers to referee #1

Thank you for your comments and suggestions to improve the manuscript.

General comments:
The paper by Dameris and colleagues investigates the potential impact of enhanced CFC-11 concentrations on future ozone by means of a coupled chemistry-climate model. A recent publication by Montzka et al. showed that atmospheric CFC-11 concentrations have not declined as expected from the Montreal Protocol. Motivated by this finding Dameris et al. conducted a sensitivity simulation with the CCM EMAC for the first half of the 21st century assuming constant CFC-11 levels after the year 2002. This simulation has been compared to a reference in which atmospheric CFC-11 develops in compliance with the Montreal Protocol. Although I like the idea of estimating the implications of increasing CFC-11 emissions on future ozone, I have major concerns related to the set-up of the performed sensitivity simulation. I totally agree with the authors that the future evolution of CFC-11 is not known and that therefore a simplified modelling approach may be justified, but assuming constant year 2002 concentrations was in my view the most unfortunate choice. We know from observations that atmospheric CFC-11 has further decreased since 2002, namely from about 258 ppt to about 230 ppt in 2017. That means that the sensitivity simulation assumes too high atmospheric CFC-11 for the time period 2002-2017. Thanks to its long lifetime this additional CFC-11 stays for a while in the atmosphere and makes a quantitative estimate of the recently discovered increased CFC-11 emissions for future ozone meaningless. I would have understood a sensitivity experiment that follows the observations until 2017 and assumes constant CFC-11 values afterwards.

We understand the concerns of the referee regarding the set-up of our sensitivity simulation, because the starting point of our simulation was the year 2002 instead of 2017. And yes, in these 15 years the mean surface CFC-11 mixing ratio decreased by 36,5 ppt (from 258,3 to 221,8 ppt) according to the prescribed scenario in the reference simulation. In the following we are trying to explain our approach hoping to convince the reviewer that her/his concerns are not justified.

The reason for starting the sensitivity simulation in the year 2002 was motivated by the statement of Montzka et al. that since the early 2000s there are obvious uncertainties with respect to the sources and sinks of CFC-11. Taking the CFC-11 mean surface mixing ratio of the year 2002 and a constant value until 2050 was primarily motivated by Figure 2a in Montzka et al. (2018) and also by Figure ES-2 in WMO (2019), because they showed stable emissions between 2002 and 2012, but also they clearly indicated an increase of the emissions after 2012 (until 2017). We are aware that stable emissions are not equal to stable concentrations. To work with a constant background value is of course a rough (crude) assessment of the future evolution of CFC-11. But since the future evolution of CFC-11

emissions (and also surface mixing ratio) is highly uncertain, such a simplified assumption of a constant surface mixing ratio is from our point of view certainly justified as an extreme scenario dating back to the point when expected emissions and observations started to diverge, and in particular because of the lack of more precise information about future evolution of CFC-11 emissions.

If we would follow the suggestion by ref #1, we are convinced that most of our results may only slightly change quantitatively, but not qualitatively. Our argument line is the following:

Our investigations are focusing on differences (!) between the REF and SEN simulations. To our understanding, starting the SEN simulation in the year 2017 (instead of 2002) and running it until 2065 (instead of 2050) would lead to similar results.

In Table 5A-3 (WMO, 2011) the mean surface CFC-11 mixing ratios (and used in our model) are given as follow:

- 2002: 258,3 ppt
- 2015: 227,6 ppt
- 2017: 221,8 ppt (not about 230 ppt as pointed out by ref #1)
- 2020: 213,0 ppt
- 2050: 127,2 ppt
- 2065: 94,4 ppt

In our presented work we have used the surface mixing ratio for the years 2002 and 2050, which means that we have reduced the CFC-11 surface value in this time period by about 50%, i.e. **131,1 ppt difference** between the REF and SEN simulation **in 2050**.

Following the suggestion by the referee this would mean: The alternative sensitivity simulation SEN would be started in 2017 (with a constant surface mixing ratio of 221,8 ppt, as calculated from Table 5A-3) and this simulation would be performed until the year 2065. This would result in a difference of the CFC-11 surface mixing ratio of **127,4 ppt in 2065** (SEN minus REF). It turned out that this CFC-11 difference value is similar to the one which was calculated in our work (difference of 131,1 ppt in 2050). Therefore the calculated and presented differences of the TOC and PCO changes (and also the results of the ozone budget changes) in our investigation should be very similar to the results of the alternatively suggested simulation by the referee. To our understanding the calculated ozone changes (SEN minus REF) are primarily affected by the prescribed CFC-11 differences (between REF and SEN) rather than from the CFC-11 background value, which is of course different (by 36,5 ppt at the surface) in 2002 and 2017.

From our point of view one could argue in our paper that our model set-up can be taken as an upper limit estimate (like an extreme scenario) regarding the CFC-11 background condition. We will prepare an appropriate formulation in the revised manuscript.

In my opinion this study requires additional efforts before becoming acceptable for publication in ACP. Either the authors perform a new sensitivity study with a more meaningful set-up (not necessarily the one outlined above, if there are better ideas), or they have at least to provide an estimate of the overestimated increase in stratospheric chlorine loading due to fixed CFC-11 levels between 2002 and 2017 and the subsequent ozone loss in their current sensitivity run.

In light of our statements from above, we refrain from performing a new sensitivity study (e.g. covering 2017-2065 with fixed concentrations after 2017). We hope that the presented arguments

regarding the similarity of the differences in the prescribed surface mixing ratios and related differences in stratospheric chlorine show that such an additional sensitivity is likely to influence the majority of our results only on a quantitative basis and not qualitatively. This is the case as we are focusing mainly on differences between the REF and the SEN which are mainly depending on the differences in the chlorine loading.

Specific comments:
- No matter which constant CFC-11 value the authors assume for their sensitivity, it would be interesting to estimate the emissions required to achieve or maintain these CFC-11 values. This would help to put the made assumptions into perspective, also with historic CFC-11 emissions, and to get an idea of how likely the chosen scenario is.

We can certainly estimate the exact emissions which are required to achieve or maintain our corresponding CFC-11 surface values. They will be presented and discussed in more detail in the revised manuscript.

A quick look on our model data output, the following value has been estimated:

The calculated CFC-11 emissions in the CCM EMAC (because of the prescribed surface mixing ratio), which are needed to keep the surface CFC-11 conditions stable on the value of 2002, turned out to be of the order of 87 Gg/a in the year 2003.

The emissions presented in Figure ES-2 (WMO, 2019; based on Figure 2a in Montzka et al., 2018) indicate mean CFC-11 emissions of 65 Gg/a, for the period from 2002 to 2012; for the years from 2014 to 2016 the CFC-11 emissions are around 75 Gg/a.

- In general I would appreciate to see some information on the statistical significance of the displayed differences between both model simulations.

Thank you for this suggestion. We will prepare and present some more information of the significance of our results in the revised manuscript! We did not focus on statistical significance as we know the general processes behind the additional ozone loss, i.e. we know that we put in additional chlorine which will change ozone abundances.

- As this study is based on one CCM only, it would be interesting to see a short discussion about the sensitivity of ozone recovery and return dates in EMAC to stratospheric chlorine compared to other CCMs, following the Dhomse et al. paper.

Will be considered in the revised manuscript!

- Fig. 4, 5, and 6 show Antarctic ozone chemistry for September. Usually October is shown for Antarctic ozone. I assume the authors chose September because Fig. 2 shows the largest difference between both model simulations in September. Some explanation would be helpful.

We will add additional explanations in the revised manuscript. Yes, we chose September because it shows the largest ozone changes. But also because September is less noisy compared to October (see e.g. Solomon et al., Science, 353, Issue 6296, 269-274, doi: 10.1126/science.aae0061, 2016).

- p4, l15-17: What is the percentage increase of ClOx in the LS?

Will be calculated and discussed in the revised manuscript.

- p6, l21-26: Do you attribute the discussed additional cooling in SEN-C2-fCFC11 to the additional CFC-11 or the changes in stratospheric ozone or both?

This will be explained and briefly discussed in the revised manuscript. It is difficult to determine the individual contributions of the additional cooling in a coupled CCM. We assume that both processes (i.e. radiative cooling by enhanced CFC-11 concentrations and by less ozone in the stratosphere caused by enhanced chlorine loading) will lead to the calculated additional cooling in the SEN simulation.

Technical corrections:
- p2, l9: Dohmse -> Dhomse
- p8, l15: Dohmse -> Dhomse

Of course! Sorry.

- Fig. 1, 3, 4, 5: I think one running mean, either 3 or 5 years, would be enough. Especially in Fig. 3 (top) the many different lines are rather confusing than helpful.

Thank you for the suggested correction regarding the figures. Will be changed!