ACP-2019-239

Old title: **Implications of constant CFC-11 concentrations for the future ozone layer**

Revised title: **Possible implications of enhanced CFC-11 concentrations on ozone**

by Martin Dameris et al.

Reply to the referee #1 comments

We thank the referee for taking the time to review our paper. In our revised manuscript we have considered all remarks and statements, which were raised in this expert's report. The paper has undergone significant changes. A second sensitivity simulation has been performed to enhance the scientific statement of our investigation. The title of the paper has been changed accordingly.

Among other modifications, we now provide a much clearer explanation about what the objectives of this study are. Apart from this, it is our intention to make it clearer for what purpose this study should not be taken, i.e. that it is not a robust projection of future ozone changes under different conditions regarding CFC-11. In the meantime, our strategy regarding our CCM sensitivity simulations received some support: The statements given by Harris et al. (2019; SPARC newsletter, summary of the CFC-11 workshop in Vienna, March 2019) are very clear in the sense that more concrete and more realistic projections of future CFC-11 levels are currently not possible. Therefore our approach is justified, allowing a simplified ("worst case") assessment of ozone changes due to enhanced CFC-11. Our second sensitivity study has been performed to provide a clearer picture with respect to the question on how much the recent CFC-11 changes so far have already affected ozone and how important the consequences are.

In the following we give our answers regarding all individual points raised by the reviewer. The statements, comments and suggested corrections raised by the referee are printed in black and our comments are presented in blue. If possible, we are pointing to the corresponding revised text block or paragraph. The revised manuscript including all changes (highlighted) and a cleared version (without highlighted changes) are provided.

Answers to referee #1 and our explanations

General comments by reviewer #1:
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.

The first motivation to start our sensitivity simulations in the year 2002 is the statement given by Montzka et al. (2018) that there is "a gap in our understanding of CFC-11 sources and sinks since the early 2000s". As it is stated now clearer in our manuscript, our major goal is to create an upper limit

case (SEN-C2-fCFC11_2050), which should allow an assessment of maximum consequences. In addition, we have now included a second sensitivity simulation (SEN-C2-fCFC11_2020), which allows an estimation of maximum changes due to the enhanced CFC-11 emissions in the last 18 years (2002 – 2019).

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.

As already said in our first reply to the referee, we understand the concerns of the referee regarding the set-up of our sensitivity simulation. Here we would like to explain our decision to start our sensitivity simulations in year 2002:

Taking the CFC-11 mean surface mixing ratio of the year 2002 and a constant value until 2050 was also motivated by Figure 2a in Montzka et al. (2018) and by Figure ES-2 in WMO (2018). Here they showed stable emissions between 2002 and 2012, but also they clearly indicated an increase of the emissions after 2012 (until 2017). In the meantime, the updated CFC-11 emissions shown by Harris et al. (2019; their Figure 1) indicated a further increase in 2018. 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 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 the future evolution of CFC-11 emissions. This is now also stated in the manuscript (second part of the Introduction starting on page 2, line 27; and also see the second part of Section 2 on pages 4 and 5).

If we would follow the suggestion given by referee #1, we are convinced that most of our results will only change slightly quantitatively, but not qualitatively. The related investigation carried out here is focusing mostly on differences between the REF-C2 and SEN-C2-fCFC11_2050 simulations. To our understanding, starting the SEN-C2-fCFC11_2050 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 REF-C2 simulation) are given as follow:

- 2002: 258,3 ppt
- 2015: 227,6 ppt
- 2017: 221,8 ppt
- 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-C2 and SEN-C2-fCFC11_2050 simulation **in 2050**.

Following the suggestion by the referee this would mean: The alternative sensitivity simulation 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**. It turned out that this CFC-11 difference value is

similar to the one, which was calculated before (i.e. a difference of 131,1 ppt in 2050 between SEN-C2-fCFC11_2050 and REF-C2). Therefore the calculated and presented differences of the TCO 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 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.

In the revised version of the manuscript, we have pointed out more clearly that our model set-up can be taken as an upper limit estimate ("worst case") regarding the CFC-11 background condition. It is now mentioned and discussed in the Introduction and the Discussion and Conclusion sections.

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.

We very much hope that the explanations given before are convincing the referee. From our point of view such an alternative sensitivity study (as suggested by the reviewer, covering 2017-2065 with fixed concentrations after 2017) will not lead to qualitative changes in our results. Moreover, we have demonstrated (see also the next point of our reply to the referee's comment) that the required CFC-11 emissions for establishing a constant surface mixing ratio in our sensitivity simulation SEN-C2-fCFC11_2050 after 2002 (i.e. a mean of about 90 Gg/year) are significantly higher than observed, but that they are not exorbitantly higher than the observed emissions in the last years. Therefore our approach for an assessment of the upper limit of consequences is definitely justified.

In addition we have performed another sensitivity simulation (SEN-C2-fCFC11_2020), which allows a rough estimation of the (maximum) consequences related to the enhanced CFC-11 levels in the past 18 years (2002 – 2019) of our numerical exercise.

Specific comments by reviewer #1:
- 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.

The exact emissions, which are required to achieve the corresponding CFC-11 surface mixing ratio, have been calculated. The respective CFC-11 emission values are now presented and discussed in Section 2 (lower part of page 4 and page 5). The tracer nudging procedure applied in our CCM EMAC diagnoses the amount of CFC-11, which is necessary to adjust to the prescribed surface mixing ratio. The cumulative CFC-11 emissions in the REF-C2 simulation (from 2002 to 2050) result in about 400 Gg. In the sensitivity simulation SEN-C2-fCFC11_2050, where the CFC-11 mean surface mixing ratio is kept constant at 258.3×10^{-12} mol/mol after the year 2002, the CFC-11 emissions required to achieve the constant surface mixing ratio value in our model after year 2002 is about 90 Gg/year (e.g. for year 2003 it is 87 Gg/year; the emissions in our model simulation are slightly increasing with time). The cumulative CFC-11 emissions (from 2002 to 2050) result in about 4500 Gg (i.e. roughly 4100 Gg more than in REF-C2). The emission values derived from observations given by Montzka et al. (2018)

are about 65 Gg/year (mean) for 2002 to 2012 and 75 Gg/year from 2014 to 2016 (see also Rigby et al., 2019). The figures presented by Rigby et al. (2019) and Harris et al. (2019, their Figure 1) with respect to the temporal evolution of CFC-11 emissions indicate a further increase after 2016 (incl. the 2018 value). In our second sensitivity simulation SEN-C2-fCFC11_2020 the cumulative CFC-11 emissions (from 2002 to 2050) result in about 2100 Gg (i.e. roughly 1700 Gg more than in REF-C2).

In summary, based on these calculated emission values derived from our model simulations it can be demonstrated that the prescribed surface boundary conditions are quite reasonable for an upper limit assessment of possible implications of enhanced CFC-11 levels.

- 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 have prepared the desired information regarding the level of confidence of the long-term changes, i.e. the (multiple) linear regression and the corresponding uncertainties of the trend estimates (we use ±2 standard errors as rough equivalent of the 95% confidence interval). The corresponding values are presented in revised Section 3.2. We did not focus on the statistical significance of the results 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.

The CCM EMAC results with respect to REF-C2 are part of the Dhomse et al. (2019) paper. A short paragraph about the general quality of our EMAC results is provided at the end of the revised Introduction (page 3, line 25 and following), allowing a rating. Another classification of our model results is given in the final section (Discussions and Conclusion; page 11).

- 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 now added some 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). Two sentences have been added, see page 6, lines 24 to 26.

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

The percentage increase of ClO_x in the US and LS for both sensitivity simulations are now presented in Section 3.1 (page 5, line 30 and the following lines).

- 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?

Regarding this point, a short statement is presented in Section 3.3 (page 9, lines 5 to 8) of 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 sensitivity simulation SEN-C2-fCFC11_2050.

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

Of course! Sorry. It has been changed.

- 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.

All figures (now Figures 2, 4, 5, 6, and also the new Figure 7) are now only showing the running 5-year mean.

We think that the manuscript has been improved by considering the comments of this referee. We very much hope that the referee agrees that this work contains interesting results, which are helpful for the rating of consequences regarding the unexpected CFC-11 emission in recent years. From our point of view such a sensitivity study gives interesting results due to enhanced CFC-11 surface mixing ratios in recent and coming years, whereas other ODS are declining as expected. Beyond that, the detailed ozone budget analysis at the end of our study further provides some new and interesting results regarding compensation and buffering effects associated with the different production and loss cycles (see the discussion on page 10, bottom paragraph).

ACP-2019-239

Old title: **Implications of constant CFC-11 concentrations for the future ozone layer**

Revised title: **Possible implications of enhanced CFC-11 concentrations on ozone**

by Martin Dameris et al.

Reply to the referee #2 comments

We thank the referee for taking the time to read our manuscript and for the critical statements and comments. Although the reviewer has concerns about the scientific content and value of the first version of our manuscript, we have prepared a revised version containing significant changes, which are related to the points raised by this referee. Among others, another sensitivity simulation has been performed and the results are discussed. From our point of view the scientific content of our study has clearly improved and the general statement of our investigation is now much clearer. The title of the paper has been changed accordingly. We very much hope to convince the referee that this piece of work contains some interesting and new results, which should be published.

In the revised manuscript, we have now provided a much clearer explanation what the objectives of this study are. Apart from this it is our intention to make it clearer for what purpose this study should not be taken, i.e. that it is not a robust projection of future ozone changes under different conditions regarding CFC-11 (hence, to avoid confusion, also the title has been changed). In the meantime, our strategy regarding our CCM sensitivity simulations has received some support: The statements given by Harris et al. (2019; SPARC newsletter, summary of the CFC-11 workshop in Vienna, March 2019) are very clear in the sense that more concrete and more realistic projections of future CFC-11 levels are currently not possible. Therefore, our approach is justified, allowing a simplified ("worst case") assessment of ozone changes due to enhanced CFC-11. Our second sensitivity study, which is now included, has been performed to provide a clearer picture with respect to the question on how much the recent CFC-11 changes so far have already affected ozone and how important the consequences are. Again this estimate can be taken as an upper limit related to new emissions.

In the following we give our answers regarding all points raised by the reviewer. The statements and comments raised by the referee are printed in black and our replies are presented in blue. If possible, we are pointing to the corresponding revised text block or paragraph. The revised manuscript including all changes (highlighted) and a cleared version (without highlighted changes) are provided.

Answers to referee #2 and our explanations

Before we are commenting the expert's report on a point by point basis, we want to add a general explanation to set our work into context.

Our analysis presented here does not aim at specifically investigating the effect of the newly discovered emission and numerous possible "directly" related scenarios. The recent paper by Montzka et al. (2018) triggered our interest in investigating the effect of enhanced CFC-11concentrations. Our objective is to create an upper limit case (SEN-C2-fCFC11_2050), which should allow an assessment of maximum consequences. In particular we think that our numerical sensitivity study is definitely new and interesting because

- we create and analyze a specific case study where the surface CFC-11 mixing ratio is fixed for a specific time period whereas other ODS decline as expected;
- we provide (most likely) an upper limit for possible scenarios ("worst case"); this could be seen as "what should be avoided/what could happen as a worst case (e.g. if a lot of the new

production is actually stored in banks)" somewhat in the tradition of the Newman et al. (2009) paper;
- we investigate when and where ozone loss due to enhanced CFC-11 levels occurs;
- we perform a detailed ozone budget analysis determining for the first time in such a scenario compensation and buffering effects associated with different production and loss cycles;
- we show that even with these enhanced CFC-11 surface mixing ratio it takes time to see the effect on ozone (due to strong inter-annual variability); and
- we have performed another sensitivity simulation (SEN-C2-fCFC11_2020), which allows a rough estimation of the (maximum) consequences related to the enhanced CFC-11 levels in the past 18 years (2002 – 2019).

We very much hope that all these points are now much clearer described in the revised manuscript and that the message of our study is now more obvious.

Preface:

This paper details the effect of a very specific future scenario of CFC-11 on ozone recovery. It is a response to the recent measurements showing the CFC-11 values are not dropping as quickly as predicted by compliance to the Montreal Protocol and thus implying illegal emissions. It outlines this one simple scenario in sufficient detail and the paper is well written. However, it is severely lacking in answering the questions necessary to understand the effect of the newly discovered emissions (see below for details). Thus, I cannot recommend publication of this paper in anything like its present form. I suggest the authors rethink the scope of the problem and expand their study considerably.

The referee is right in saying that our model approach is simple. However, there is a big knowledge gap regarding CFC-11 emissions (see Harris et al., 2019). As said already, therefore our approach is justified, allowing a simplified ("worst case") assessment of ozone changes due to enhanced CFC-11. In the revised version of the manuscript, we have pointed out more clearly that our model set-up can be taken as an upper limit estimate. This is now mentioned and discussed in the Introduction and the Discussion and Conclusion sections.

We believe that on the basis of our model study we can answer relevant questions, which are related to the effects of the additional CFC-11 emissions (i.e. constant CFC-11 concentrations). From our point of view the sensitivity simulations (SEN-C2-fCFC11_2050 and SEN-C2-fCFC11_2020), which have been performed in addition to the reference simulation (REF-C2), are certainly suitable for an upper limit sensitivity study regarding CFC-11. Our investigation should not be considered as specific future scenario that we deem likely. It is not our aim to distill more specific scenarios. This study aims to assess the magnitude of possible ozone changes under constant CFC-11 surface mixing ratio. Since currently we do not have more detailed information about possible future CFC-11 levels, such a "simple" assessment is in our understanding justified. The goal of this paper is to answer questions, which are related to possible implications due to enhanced CFC-11 emissions, but we do not claim that this is the actual path we are on. Moreover, this paper was not intended to answer questions, which are focusing on the origin or sources of the additional CFC-11 emissions.

Furthermore, we do not aim at making a complete assessment, which currently makes no sense since the future evolution of CFC-11 emissions is uncertain. Our study rather aims to provide an insight into the reaction of the ozone layer with respect to enhanced CFC-11 levels in the coming years. (Further, once a new "most likely" emission scenario is agreed on we are willing to provide such a simulation. A "full" assessment would also mean that different CCMs performing the same scenario.)

Last but not least and already mentioned above, it is an interesting sensitivity study to keep CFC-11 constant, whereas other ODS are declining according to RCP scenarios; the results are partly new and

informative. To fully carve this out, we have for the first time performed a detailed ozone budget analysis for such sensitivity simulations.

General comments:

The recent paper of Montzka et al. 2018 makes a strong case that there are illegal emissions of CFC-11 presently occurring and that these have been occurring since 2012 and even perhaps earlier. This brings up many questions for future ozone recovery including (but not limited to):

With our model study, which is based on a comparison of two sensitivity simulations with a reference simulation using a chemistry-climate model (here EMAC), we would like to provide a qualitative and also a rough quantitative assessment of possible implications of enhanced CFC-11 levels. Among others, it should help to assess the order of magnitude of ozone changes, which can be expected due to enhanced CFC-11 emissions.

Have the emissions to date affected future ozone recovery;

To answer this question in more detail, we have performed a second sensitivity simulation (SEN-C2-fCFC11_2020) for the revised version of our manuscript. The results are presented and discussed, in particular in the context of Figure 7. It allows an estimation of maximum changes due to the enhanced CFC-11 emissions in the last 18 years (2002 – 2019). It is found that an immediate full implementation of the Montreal Protocol again (with a drop down to zero of the additional CFC-11 emissions after 2020) eventually leads back to the direction of the expected ozone conditions around the end of the 2040s, as they are calculated in our REF-C2 simulation. In the REF-C2 simulation the model-diagnosed CFC-11 emissions are nearly zero after about 2030, whereas in the SEN-C2-fCFC11_2020 simulations they are steadily decreasing from higher values in 2020 down to zero around year 2050 (see the last paragraph of Section 3.2, page 7).

How much more would continuation of the present emissions to various end dates affect ozone recovery;

Based on the results of our simulations, in particular based on SEN-C2-fCFC11_2050, we find a maximum delay of the closure of the ozone hole by about 20 years under these “extreme” conditions (see the paragraph at the end of the Discussions and Conclusion section). Our result is in line with other model estimates, which are mentioned by Harris et al. (2019). A first estimate presented in WMO (2018) showed that if total CFC-11 emissions were to continue at levels experienced from 2002–2016 (67 Gg/year), the return of mid-latitude and polar EESC to the 1980 value would be delayed by about 7 years and 20 years, respectively. In our SEN-C2-fCFC11_2050 simulation a mean annual CFC-11 emission of about 90 Gg is assumed (see the description in the second part of revised Section 2) and therefore our assessment can be used as upper limit estimation.

What if the emission increased, what would that do to future recovery;

We think that our SEN-C2-fCFC11_2050 simulation is acceptable as upper limit assessment of possible future consequences. Another more extreme scenario would certainly be possible, but from

our point of view such a study would be purely speculative. We know that the consequences are clearly depending on the future evolution of the CFC-11 emissions (see Harries et al., 2019). Instead, we have now performed the SEN-C2-fCFC11_2020 simulation and are discussing the results of it in the revised manuscript. Here we discuss the related model results, which are indicating that the consequences due to the enhanced CFC-11 emission in the last 18 years are not dramatic, if the international community would immediately react, i.e., if the CFC-11 (additional) emissions are stopped now (assuming that the banks have not considerably increased).

What if there are banks of CFC-11 (and perhaps CFC-12) associated with the illegal emission;

To answer this question is highly speculative because there are major uncertainties regarding other ODS, which are related to CFC-11 production (for instance CFC-12 and HCFC-22). As pointed out by Harris et al. (2019), there are "major uncertainties in quantifying the excess emissions to unreported production", which are due to "(i) a possible increase in the leakage from banks; and (ii) the influence of atmospheric variability". To our knowledge, so far there are no other obvious changes identified, which are related to the levels of other ODS than CFC-11. Nevertheless, it is clear that other additional emissions or influencing processes would have the potential to further impact the ozone layer. For our paper we have decided not to discuss the possible implications of other, enhanced ODS and other related changes.

The present paper does not address these questions in any detail. Instead it only addresses one simple scenario: if the mixing ratio of CFC-11 stays constant through 2050, what is the effect on ozone. This assumes that the emission rate of CFC-11 stays at a constant level slightly higher than any inferred emission estimated in Montzka et al. and that this emission stays constant for the next 3 decades.

We hope that based on our explanations given before most of the raised question by the reviewer are sufficiently answered. From our point of view all the mentioned points are now adequately considered and discussed in the revised version. Certainly many other scenarios seem to be possible and in principle they could be performed with our model (but this would be expensive regarding computational costs); the number of possible future CFC-11 scenarios is arbitrary high. And as it has been stated before, the range of uncertainties is very large, and therefore the discussion of results of the scenario simulations would contain a large part of speculation. From our point of view such a discussion would not be efficient.

The results presented in this study definitely help to rate the potential of enhanced CFC-11 levels. Certainly our investigation based on SEN-C2-fCFC11_2050 provides "only" a rough estimation of possible implications, but taking into account the large uncertainties regarding the circumstances with respect to the CFC-11 emissions itself and to the effects on other ODS, in our view such a simplified assumption of a constant surface mixing ratio is absolutely justified as a "worst case" dating back to the point when expected emissions and observations started to diverge (i.e. year 2002).

Ignoring that this scenario is unlikely to occur given the international response to this issue, the real problem with the paper is that so little of the problem space is explored. I see limited value in modeling one (unlikely) scenario in detail and ignoring all other possibilities.

Based on our extended explanations in the revised manuscript and in particular with respect to the presentation and discussion of the results of the additional sensitivity simulation, we hope to convince the referee that our study is useful and that the scientific value of our investigation is given. We definitely did not ignore other possibilities, but, as mentioned before, due to the fact of the large range of uncertainties, we think that such a simplified numerical approach should be allowed.

I can only assume this choice was made because it is easy to implement in their model and it only took one new run. Unfortunately, the only question answered is that if a larger emission than inferred in Montzka et al. is continued for three decades it will have a negative effect on ozone recovery.

For certain, our choice was definitely not made because it is the easiest way to implement such a sensitivity simulation (i.e. SEN-C2-fCFC11_2050) in our CCM EMAC. As previously said, we have a clear objective: provide an upper assessment of possible consequences due to enhanced CFC-11 levels; we try to investigate a worst case scenario. Our main interest is related to the question: "What must be avoided?" (to be similar to the paper by Newman et al. (2009) "World avoided" paper). However, we appreciate the remarks given by the referee, so this has eventually led to our second sensitivity simulation (SEN-C2-fCFC11_2020), which surely has improved our study.

This will surprise no one and in fact it can be predicted by computing the perturbation of EESC in 2050 by changing the CFC-11 mixing ratio between the ref value to the new value. This is a "back of the envelope" calculation.

We were very surprised by this statement of the referee, in particular in the light of the results presented in Section 3.3 (Stratospheric ozone budget). In this section, for the first time we have prepared and presented a detailed analysis of the model results with respect to the ozone production and loss cycles for such a scenario simulation, i.e. enhanced CFC-11 levels. There are some "surprises" (may be not only for us), for instance that the effects are smaller than expected due to some compensating effects by other ozone production and loss cycles. Our budget analysis based on REF-C2 and SEN-C2-fCFC11_2050 nicely shows (Figure 8) compensating effects for the global annual mean and in the upper stratosphere in the Southern Hemisphere polar region, whereas in the lower stratosphere in the Southern Hemisphere polar region the compensation does not occur. We show, which processes are buffering the additional ozone destruction through higher CFC-11 values. We show where (altitude) and at which time ozone is reduced compared to the REF-C2. Further, we show that the additional emissions need time to affect ozone. It would have taken years to register some deviation in the ozone columns due to the additional emissions. In addition, we have analyzed the effect of the temperature change on the chemistry (see discussion on page 9).

I expect much more of the problem space explored in a paper addressing the effect of illegal emissions of CFC-11 on ozone recovery and with a chemistry-climate model to use. As stated above, I recommend that the author team rethink the issues the Montzka et al. paper uncovered and take a real stab at helping answering them. It is necessary to frame the problem in terms of various possibilities for the emissions (and bank changes) and then from there predict mixing ratio scenarios and finally model time series. Only then can the reader understand the scope of the problem and the possible effects on future ozone.

A classification into different terms of various possibilities for the CFC-11 emissions and based on this predict mixing ratio scenarios is certainly beyond the scope of our study. Our study would take a different direction; it would raise completely different questions. We think that with this paper we are providing an important contribution to solve some of the problems of possible implications on enhanced CFC-11 levels. We hope that the reviewer can support our view and our scientific approach. In particular, we have rephrased the Introduction section and the Discussions and Conclusion section making the objectives of our numerical model study much clearer. A confrontation of the observed CFC-11 emissions (Montzka et al., 2018) with the emissions employed in our model simulations help to categorize our findings (see second part of Section 2). It is demonstrated that the corresponding CFC-11 emissions are reasonable in our model simulations, which are required to keep the surface mixing ratio constant. Therefore, the adopted simulations can be used as an upper estimate of possible consequences of enhanced CFC-11 emissions.

We very much hope that the referee can follow our argumentation line and that the reviewer agrees that the revised manuscript is acceptable for publication in ACP.

~~Implications~~ Possible implications of ~~constant~~ enhanced CFC-11 concentrations ~~for the future ozone layer~~on ozone

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Abstract. This ~~investigation~~ numerical model study is motivated by the observed global deviation from assumed emissions of chlorofluorocarbon-11 (CFC-11, $CFCl_3$) in recent years.~~results presented by~~ Montzka et al. (2018)~~. They~~ discussed a strong deviation of the assumed emissions of ~~chlorofluorocarbon-11 (~~CFC-11~~, $CFCl_3$)~~ in the past 15 years, which indicates a violation of the Montreal Protocol for the protection of the ozone layer. An investigation ~~Chemistry-Climate Model (CCM) study~~ is performed based on Chemistry-Climate Model (CCM) simulations~~,~~ that ~~investigating~~ analyze the consequences of ~~a constant~~an enhanced CFC-11 surface mixing ratio ~~for stratospheric ozone:~~. In comparison to a reference simulation (REF-C2), where a decrease of the CFC-11 surface mixing ratio of about 50% is assumed from the early 2000s to the middle of the century (i.e. mixing ratio in full compliance with the Montreal Protocol agreement), ~~a~~ two sensitivity simulations ~~(SEN-C2-fCFC11) is~~ are carried out: One simulation ~~where~~ in which after the year 2002 the CFC-11 surface mixing ratio is kept constant until 2050 (SEN-C2-fCFC11_2050); this allows a qualitative estimate of possible consequences of stable CFC-11 surface mixing ratio on the ozone layer ("worst case"). In the second sensitivity simulation, which is branched off from the first sensitivity simulation, it is assumed that starting in year 2020 the Montreal Protocol is fully implemented again, which leads to a delayed decrease of CFC-11 in this simulation (SEN-C2-fCFC11_2020) compared to the reference simulation; this enables a rough and most likely upper-limit assessment of how much the unexpected CFC-11 emissions to date have already affected ozone. In all three simulations climate evolves under the same greenhouse gas scenario (i.e. RCP 6.0) and all other ozone depleting substances are declining according to this scenario. Differences between ~~these~~ the reference (REF-C2) and the two sensitivity simulations (SEN-C2-fCFC11_2050 and SEN-C2-fCFC11_2020) ~~simulations~~ are ~~shown~~discussed. ~~These illustrate possible effects on stratospheric ozone.~~ In the SEN-C2-fCFC11_2050 simulation~~The~~ the total column ozone (TCO) in the 2040s (i.e. the years 2041-2050) is ~~in~~ particularly affected in both polar regions in winter and spring. ~~At the end of the 2040s m~~Maximum discrepancies of TCO are identified with reduced ozone values of up to around 30 Dobson Units in the Southern Hemisphere (SH) polar region during SH spring (in the order of ~~10~~15%) ~~in the SEN-C2-fCFC11 simulation~~. An analysis of the respective partial column ozone (PCO) for the stratosphere indicates that strongest ozone changes are calculated for the polar lower stratosphere, where they are mainly driven by the enhanced stratospheric chlorine content and associated heterogeneous chemical processes. Furthermore, it turns out that the calculated ozone changes, especially in the

upper stratosphere, are surprisingly small~~er than expected~~. For the first time in such a scenario we perform a complete ozone budget analysis regarding the production and loss cycles. The budget analysis shows that ~~In~~ in ~~this altitude region~~the upper stratosphere the additional ozone depletion due to the catalysis by reactive chlorine is compensated partly by other processes related to enhanced ozone production or reduced ozone loss, for instance from nitrous oxide (NO_x). Based on the analysis of the SEN-C2-fCFC11_2020 simulation it turned out that no major ozone changes can be expected after year 2050, which are related to the enhanced CFC-11 emissions in recent years.

1 Introduction

The estimation of the future evolution of the ozone layer is a central part of the UNEP/WMO Scientific Assessment of Ozone Depletion. For that reason Chemistry-Climate Models (CCM) are carrying out long-term simulations (for several decades). These models are performing comprehensive numerical simulations under well-defined boundary conditions which are prescribing possible future changes of ozone depleting substances (ODSs), particularly those related to changes of chlorofluorocarbon (CFC) concentrations. In recent years, model guidelines have been defined, to facilitate the inter-comparison of CCM results from different modelling groups world-wide. For instance, in 2012 the Chemistry-Climate Model Initiative (CCMI), under the umbrella of IGAC/SPARC, defined the boundary conditions for the next round of coordinated reference (REF) and sensitivity (SEN) simulations (Eyring et al., 2013). The boundary conditions for the CCM simulations consider not only the expected changes of ODSs according to the regulations of the Montreal Protocol and its amendments, but also the influence of different climate change scenarios. Here, the greenhouse gas concentrations for the Representative Concentration Pathways (RCPs) adopted by the IPCC for its 5th Assessment Report (AR5) in 2014 (van Vuuren et al., 2011) were recommended. The suggested reference simulation for the future (REF-C2) assumes full compliance with the Montreal Protocol, expecting more or less no further production of CFCs.

More recently the respective CCM results were presented and discussed in several scientific papers, for instance by ~~Dohmse~~ Dhomse et al. (2018). Among other things the question of ozone recovery was investigated~~,~~. Further, it was analyzed~~and~~ how the speed of recovery and the return date are affected by the expected decrease of ODSs and by climate change. The results of the CCM simulations were taken into account as the foundation for the latest WMO ozone report (WMO, ~~2019~~2018).

During the preparation phase of WMO (~~2019~~2018) a paper by Montzka et al. (2018) was published, indicating a clear deviation of the expected surface concentration of chlorofluorocarbon-11 (CFC-11, ~~(~~$CFCl_3$) in the past 15 years. Observational datasets discussed by Montzka et al. (2018; M18) showed that (1) in the last 10 years (until 2017) the decline of CFC-11 surface mixing ratios was obviously much slower than expected (see Figure 1a in M18); (2) the decline of CFC-11 surface mixing ratios was nearly constant from 2002 to 2012, whereas in the following years the decrease of CFC-11 surface mixing ratios decelerated (here until 2017; see Figure 1b in M18). The measurements indicated increased CFC-11

emissions after 2012 (see Figure 2a in M18; see also Figure ES-2 in WMO, ~~2019~~2018). Montzka and colleagues (M18) mentioned that these observations imply "a gap in our understanding of CFC-11 sources and sinks since the early 2000s".

Based on these findings a significant impact on the recovery of the ozone layer seems to be possible, in particular, if CFC-11 emissions do not decline as previously anticipated (e.g. Daniel and Velders et al., 2011; Carpenter and Reimann et al., 2014). Therefore the assumption of decreasing CFC-11 surface mixing ratio in the future by CCMI is partly questionable. Currently the future evolution of CFC-11 emissions ~~in the future~~ is ~~unclear~~uncertain (Harris et al., 2019).~~:~~ Therefore, for this numerical study we cannot decide, which future scenario is most likely and also the possible ranges of future CFC-11 emission changes are difficult to estimate. ~~Here, we~~Our approach is to employ an ~~simplified~~ upper limit ("worst case") scenario of ~~future~~ CFC-11 emissions in a sensitivity simulation (SEN-C2-fCFC11_2050) by imposing~~through~~ constant surface mixing ratios of CFC-11 from 2002 onwards (Figure 1). The sensitivity simulation covers the period from 2002 to 2050. This sensitivity simulation should not be considered as a specific future scenario that we deem likely. As said this model set-up can be taken as an extreme (upper limit) scenario regarding CFC-11 background conditions. The reason for starting this sensitivity simulation in the year 2002 is motivated by a statement of M18 that since the early 2000s there are obvious uncertainties with respect to the sources and sinks of CFC-11 (see end of previous paragraph). We note that stable CFC-11 emissions are not equal to stable CFC-11 surface concentrations. But due to the fact that the future evolution of CFC-11 emissions (and also the surface mixing ratio) is uncertain, such a simplified (crude) assumption of a constant surface mixing ratio is absolutely justified as a "worst case" dating back to the point when expected emissions and observations started to diverge. Although M18 hinted that the additional CFC-11 emissions might be released in eastern Asia (see also Rigby et al., 2019), we do not impose any specific regional features due to the long lifetime of CFC-11 (e.g. Rigby et al., 2013). In comparison with the reference simulation (REF-C2) the sensitivity simulation SEN-C2-fCFC11_2050 allows us to investigate the potential impact of previously unaccounted CFC-11 emissions. ~~The sensitivity simulation covers the period from 2002 to 2050.~~ It ~~allows~~ enables a rough estimation of the additional possible ozone loss under ~~unchanged~~ constant CFC-11 ~~emissions~~ surface mixing ratio in the coming years and how it may impact the timing of full recovery of the ozone layer. In addition, the second sensitivity simulation (SEN-C2-fCFC11_2020, see also Figure 1) is carried out, to allow for an estimate of ozone changes in case of full recognition and implementation of the Montreal Protocol again in the coming years. After an 18-year period of constant CFC-11 surface mixing ratio (in line with SEN-C2-fCFC11_2050 from 2002 to 2019), the CFC-11 surface mixing ratio is decreasing in the following years (in parallel with REF-C2) under the assumption that the recent additional CFC-11 emissions will drop down to zero starting in year 2020.

It is the aim of this study to show when and where ozone loss occurs due to additional influx of CFC-11 into the atmosphere while other ODSs decline as expected. Our scenario described in SEN-C2-fCFC11_2050 could be taken as "what should be avoided", somewhat in the tradition of the Newman et al. (2009) paper. Furthermore the SEN-C2-fCFC11_2020 provides an estimate of the impact of the temporary increase CFC-11 emissions in recent years on the ozone layer and its recovery. Our analysis presented here does not aim at specifically investigating the effects of the discovered CFC-11 emissions and

numerous "directly" related scenarios. Here we want to assess the impact of enhanced CFC-11 surface mixing ratio as a sensitivity study. The foundation for our numerical exercise is the REF-C2 simulation, which was performed with our CCM EMAC (see description in Section 2). The results of this reference simulation were one of our several contributions to the Dhomse et al. (2018) study and WMO (2018). The results of EMAC were checked against observation (for the past) in detail (e.g. Jöckel et al., 2016) and were also compared with results derived from other CCMs. No obvious weaknesses or significant deficiencies could be identified.

After a short description of the used CCM and the analyzed model simulations (REF-C2, SEN-C2-fCFC11_2050 and SEN-C2-fCFC11_2020~~SEN-C2-fCFC11~~) in the next section (Sec. 2), the CCM results are presented in Section 3. For the results, we focus on changes of total column ozone and partial stratospheric columns in specific geographical regions and seasons. To the best of our knowledge, a detailed ozone budget analysis for such sensitivity simulations is performed and presented for the first time. Finally, the discussion and conclusion ~~will be~~are presented at the end of this paper.

2 Description of the model and simulations

For this study, the CCM EMAC (abbreviation stands for "European Centre for Medium-Range Weather Forecasts – Hamburg (ECHAM)/Modular Earth Submodel System (MESSy) Atmospheric Chemistry model") is used in the version 2.52 and is operated at a resolution of T42L90MA corresponding to a quadratic Gaussian grid of approx. 2.8 by 2.8 degrees in latitude and longitude with 90 levels up to 0.01 hPa. More details were presented by Jöckel et al. (2016).

The joint IGAC/SPARC Chemistry-Climate Model Initiative (CCMI) proposed several reference and sensitivity simulations ~~among others~~ for CCM studies. The aim was to support upcoming ozone and climate assessment reports. In this connection an internally consistent simulation from the past into the future between 1960 and 2100 has been suggested (Eyring et al., 2013). This transient reference simulation, i.e. REF-C2, as used in this study, is forced by trace gas projections and prescribed sea surface temperatures (SSTs) and sea ice concentrations (SICs). The projection component of REF-C2 uses greenhouse gas concentrations (i.e. CO_2, CH_4, and N_2O) that follow the Intergovernmental Panel on Climate Change (IPCC) Coupled Model Intercomparison Project Phase 5 (CMIP5) Representative Concentration Pathways 6.0 (RCP 6.0) scenario (van Vuuren et al., 2011). Monthly mean global SST and SIC data, which were simulated by the climate model HadGEM2 with an interactive ocean (Hadley Centre Global Environment Model version 2, data used for the RCP6.0 scenario; see Jones et al., 2011), ~~are~~ were used as boundary conditions for the REF-C2 simulation.

For this study, in addition to the REF-C2 simulation (for more details see Jöckel et al., 2016; here it is the "RC2-base-04" reference simulation), ~~a~~ two specific sensitivity simulations (SEN-C2-fCFC11_2050 and SEN-C2-fCFC11_2020) ~~is~~ are designed to address the possible consequences of ~~constant~~ additional emissions of CFC-11, which ~~lead to constant CFC-11 surface mixing ratios and therefore~~ affect the chlorine content of the stratosphere after some years. In ~~both~~ all EMAC simulations mixing ratios of ODSs (CFCs: $CFCl_3$, CF_2Cl_2, CH_3CCl_3, CCl_4; HCFCs: CH_3Cl, CH_3Br; Halons: CF_2ClBr,

CF_3Br) in the lowest model layer are adapted by Newtonian relaxation to observed or projected surface mixing ratios (Kerkweg et al., 2006). The applied tracer nudging procedure diagnoses the emission flux of CFC-11, which is necessary to adjust to the prescribed surface mixing ratio. In the REF-C2 simulation the mean CFC-11 surface mixing ratio in the year 2002 is 258.3×10^{-12} mol/mol (see Table 5A-3 in Daniel and Velders et al., 2011) and it is significantly reduced by more than 50% (127.2×10^{-12} mol/mol) in the year 2050 (i.e. the mixing ratio of the baseline (A1) scenario; WMO, 2011). The 2050 value is projected under the assumption of full compliance with the Montreal Protocol. The cumulative CFC-11 emissions in the REF-C2 simulation (from 2002 to 2050) result in about 400 Gg. Apart from one point the sensitivity simulation (SEN-C2-fCFC11_2050) is identical to the reference simulation (REF-C2): ~~for simplicity,~~ in the sensitivity simulation the CFC-11 mean surface mixing ratio is kept constant at 258.3×10^{-12} mol/mol after the year 2002, whereas they decline in ~~the reference simulation~~REF-C2. The CFC-11 emissions required to achieve the constant surface mixing ratio value in our model after year 2002 in SEN-C2-fCFC11_2050 is about 90 Gg/year (e.g. for year 2003 it is 87 Gg/year; the emissions in our model simulation are slightly increasing with time, likely due to the small reduction of the CFC-11 lifetime; see for instance SPARC, 2013). The cumulative CFC-11 emissions (from 2002 to 2050) result in about 4500 Gg (i.e. roughly 4100 Gg more than in REF-C2). The emission values derived from observations given by Montzka et al. (2018) are about 65 Gg/year (mean) for 2002 to 2012 and 75 Gg/year from 2014 to 2016 (see also Rigby et al., 2019). The figures presented by Rigby et al. (2019) and Harris et al. (2019) with respect to the temporal evolution of CFC-11 emissions indicated a further increase after 2016. In the second sensitivity simulation SEN-C2-fCFC11_2020 after year 2020 full adherence of the Montreal Protocol is assumed. The starting point of this simulation is aligned with SEN-C2-fCFC11_2050 assuming the same constant CFC-11 surface mixing ratio between 2002 and 2019 (i.e. 258.3×10^{-12} mol/mol), but with decreasing mixing ratios after 2020 until 2050. The cumulative CFC-11 emissions (from 2002 to 2050) result in about 2100 Gg (i.e. roughly 1700 Gg more than in REF-C2). A schematic of our three model simulations is presented in Figure 1.

In ~~the~~ both sensitivity simulations we do not emphasize specific regions regarding outstanding changes of the CFC-11 surface mixing ratio, e.g. in eastern Asia (see discussion in the Introduction). The modified CFC-11 boundary condition in the SEN-C2-fCFC11_2050 and ~~–~~SEN-C2-fCFC11_2020 simulations should cause a change of the stratospheric chlorine loading after about 10-15 years (e.g. Engel et al., 2002). ~~The SEN-C2-fCFC11 simulation covers the period from 2002 to 2050.~~

3 Presentation of CCM results

3.1 Reactive chlorine

Based on the prescribed changes of the CFC-11 boundary conditions the stratospheric content of reactive chlorine compounds (ClO_x = Cl + ClO + OClO + 2ClOOCl +$2Cl_2$ + HOCl) is expected to change. In Figure ~~1~~ 2 the simulated change (i.e. SEN-C2-fCFC11_2050 and SEN-C2-fCFC11_2020~~SEN~~ minus REF-C2~~REF~~, respectively) of reactive chlorine mixing

ratios with time is shown for the lower stratospheric (LS, near 50 hPa) and the upper stratosphere (US, near 2 hPa). Because ~~these two~~all model simulations are ~~both~~ operated in a "free-running-mode" (i.e. not having the same meteorology), the year-to-year difference (~~grey~~thin red and blue) curves as shown in Figure ~~1~~ 2 (and also in other figures shown afterwards) ~~is~~ are indicating the possible range of inter-annual fluctuation. Obviously it takes about 10-15 years of time (as expected) before the ClO_x values of the ~~two EMAC~~REF-C2 and SEN-C2-fCFC11_2050 simulations clearly diverge from each other. At the end of the SEN-C2-fCFC11_2050 simulation (i.e. in 2050) the resulting absolute (mean) difference in EMAC arises to an approximately 6_x_10^{-12} mol/mol increase in the LS and about 50_x_10^{-12} mol/mol in the US compared to the REF-C2 simulation. This vertical dependence of the changes of chlorine mixing ratios is related to the reference ClO_x profile in EMAC, which displays a distinct maximum at about 2-3 hPa (not shown). It turns out that in the US (above 30 km) in the 2040s (i.e. the time period from 2041 to 2050) the amount of chemically active chlorine species (ClO_x) in the SEN-C2-fCFC11_2050 simulation is ~~roughly~~ on average about 17~~-21~~% larger than in the REF-C2 simulation~~.~~; in the LS (below 30 km) the respective amount of ClO_x is enhanced by about 30%. As can be expected, the respective ClO_x differences between the SEN-C2-fCFC11_2020 with respect to the REF-C2 simulation in the 2040s are clearly smaller, i.e. about 2x10^{-12} mol/mol in the LS (about 14%) and about 25x10^{-12} mol/mol in the US (about 9%).

3.2 Total and partial ozone columns

The impact of the enhanced atmospheric ClO_x content due to constant CFC-11 surface mixing ratio after 2002 on total column ozone (TCO) is shown in Figure ~~2~~3. It illustrates the differences ~~between the REF-C2 and the SEN-C2-fCFC11 simulation~~ of the mean annual cycle for the last decade ~~of the SEN-C2-fCFC11 simulation~~ depending on the geographical latitude between the REF-C2 and the SEN-C2-fCFC11_2050 simulation. The largest changes of TCO are found in both polar regions: in particular in the Northern Hemisphere during winter (December, January, February) and in the Southern Hemisphere in late winter (August) to early spring (September, October). In the SEN-C2-fCFC11_2050 simulation the TCO values are clearly reduced by up to about 30 Dobson Units (DU) (in the order of ~~10~~15% in the Southern Hemisphere) in comparison to the REF-C2 simulation. During other times of the year and in other latitudinal regions the identified TCO changes are much smaller. They are mostly below ± 5 DU.

A closer look at the near global mean (60° S – 60° N) temporal behavior of TCO (Figure ~~3~~4) indicates a recovery of the ozone layer in both simulations (upper part of the figure, for both simulations the solar cycle was removed, see figure caption). The results of the SEN-C2-fCFC11_2050 simulation are showing slightly smaller TCO values in comparison to REF-C2 and a slightly flatter slope of the linear trend line (this regression accounts for possible auto-correlation at lag 1, see e.g. the method described in Tiao et al., 1990). The linear regression based on the results of the REF-C2 simulation (here the black line presented in the upper part of the figure) indicates an increase of 1.7 DU/decade for the TCO (annual near global mean), whereas the linear regression based on the results of the SEN-C2-fCFC11_2050 (red line in the upper part of the figure) shows a reduced increase of 1.3 DU/decade. This finding is supported by the TCO difference (lower part of the

figure), which indicates a small reduction of the TCO (in SEN-C2-fCFC11_2050) of about up to ~~3~~ 2 DU (in the order of less than 1%) until 2050. The linear regression (again accounting for possible lag 1 auto-correlation) gives -0.5 DU/decade (± 0.25 DU/decade; given by two times the estimated standard error, which corresponds roughly to the 95% confidence interval and will be used throughout this paper as a measure of uncertainty). This effect can be rated as negligible in comparison with the expected annual fluctuations in this region. Therefore in the following we focus on the analyses of the polar regions, in particular on the Antarctic region in September. One reason for choosing the month September for further analyses of Antarctic ozone chemistry is that we found here the most obvious ozone changes. Another reason is that this month is less noisy compared to October (Solomon et al., 2016).

In the Southern Hemisphere polar region (70° S – 90° S) in September obvious ozone changes can be identified (Figure ~~4~~5). The mean differences of the TCO between the 2000s and the end of the simulation amount to about 20 DU, indicating that the mean September ozone values in the SEN-C2-fCFC11_2050 simulation are about 10% lower than in the REF-C2 simulation. The trend estimate gives -4.1 DU/decade (± 1.7 DU/decade). This trend estimate and the uncertainties have been obtained by multiple linear regression, which accounts for possible lag-1 auto-correlation and uses the difference of the temperature anomalies (at 100 hPa over 70° S – 90° S) from the REF and the SEN simulation as second independent variable besides the linear trend. It is found that much of the inter-annual variability can be explained by including the difference of the polar temperature anomalies at 100 hPa in the regression model. This agrees with Langematz et al. (2016), who have used temperature anomalies to regress polar TCO. The temporal evolution of TCO differences and the size of the ozone disturbance found in the Northern Hemisphere polar region in January have the same order of magnitude (not shown), but the signal is ~~very~~ more noisy because of the stronger dynamic variability.

Now we are looking in more detail into stratospheric partial columns of ozone (PCO), for the upper stratosphere (US, above about 30 km, i.e. the 10 hPa pressure level) and the lower stratosphere (LS, between 100 hPa and 10 hPa) ~~– i.e. columns below and above 10 hPa, respectively –~~ for the Antarctic region in September. Figure ~~5~~ 6 shows the PCO differences for the US (top part) and LS (bottom part) between the SEN-C2-fCFC11_2050 and the REF-C2 simulation. Both are showing the expected negative trend, indicating lower values at the end of the SEN-C2-fCFC11_2050 simulation in the late 2040s. Again the trends are obtained by the same regression model as for the TCO (see previous paragraph) but for the US the temperatures at 10 hPa have been used. The mean PCO changes for the US with about 2 DU are much smaller than those calculated for the LS (about 20 DU). The temporal evolution of the PCO differences in the LS show ~~almost~~ similar results as found for the TCO differences (Figure ~~4~~5): in the SEN-C2-fCFC11_2050 simulation the TCO is reduced by about 20 DU until the year 2050. The strongest signature of ozone change found in the polar LS points to the importance of heterogeneous chemical processes. Viewing the vertical profile of the differences of net-ozone production rates of ClO_x between the REF-C2 and the SEN-C2-fCFC11_2050 simulation in the 2040s clearly indicates an absolute minimum (i.e. less net-ozone production in SEN-C2-fCFC11_2050) at around 50 hPa and another relative minimum at about 1.5 hPa in September (Figure ~~6~~8, right part; see explanation in Section 3.3). Complementary, looking at the PCO changes in middle and lower latitude

regions (60° S – 60° N, not shown) the partial column differences clearly ~~show~~ indicate that the small ozone differences detected in the TCO (lower part of Figure ~~3~~4) are affected by ozone reductions of similar magnitudes in the US and the LS, displaying only small contributions to the TCO.

In Figure 7 we are confronting the calculated differences between the simulations SEN-C2-fCFC11_2050 and REF-C2 as presented in Figures 4 to 6 with the corresponding results derived from SEN-C2-fCFC11_2020 and REF-C2. In all four parts of Figure 7 it is obvious that an immediate reimplementation of the Montreal Protocol eventually leads back to the direction of the expected ozone conditions around the end of the 2040s, as they are calculated in our REF-C2 simulation. In the REF-C2 simulation the model-diagnosed CFC-11 emissions are nearly zero after about 2030, whereas in the SEN-C2-fCFC11_2020 simulations they are steadily decreasing from higher values in 2020 down to zero around year 2050. This is caused by the prescribed CFC-11 surface mixing ratio as indicated in Figure 1.

3.3 Stratospheric ozone budget

In the following, a ~~more~~ detailed analysis of individual ozone production and loss processes is carried out. This ozone budget analysis is used to investigate the role of separate chemical cycles and reactions, which are responsible for ozone production and loss in the stratosphere. For this analysis the MESSy tool StratO3Bud (cf. Meul et al., 2014, based on Jöckel et al., 2006) is employed. The respective reactions responsible for stratospheric ozone production (attributed to photolysis hυ, HO_2 and CH_3O_2) and loss (attributed to O_x, NO_x, HO_x, ClO_x and BrO_x) are described by Meul et al. (2014, see their Table 2). In Figure ~~6~~ 8 the results of this budget analysis are shown as changes of the ozone production rate between the SEN-C2-fCFC11_2050 and the REF-C2 simulation $\left(\Delta P^{prc}_{SEN-REF}\right)$ with respect to the total production rate in the REF-C2 simulation. The explicit formula for calculating the changes of the ozone production rate at a certain level is given as

$$\Delta P^{prc}_{SEN-REF}(lev) = \frac{\sum_{lat \in R} P^{prc}_{SEN}(lat, lev) - \sum_{lat \in R} P^{prc}_{REF}(lat, lev)}{\sum_{lev} \sum_{lat \in R} P^{tot}_{REF}(lat, lev)}$$

Here, P denotes the temporal mean and zonally summed ozone production rate (molecules/s). The subscript denotes the respective simulations and the "prc" superscript denotes, which process (hυ, HO_2, CH_3O_2, O_x, NO_x, HO_x, ClO_x, and BrO_x) is analyzed. Loss cycles are regarded as negative production rates. Further, the superscript "tot" denotes the sum of all positive production rates (namely of hυ, HO_2, CH_3O_2) and the summation goes over all latitudes, which lie in the respective latitudinal band R. Here we show profiles of $\Delta P_{SEN-REF}$ for the annual global mean and the Southern Hemisphere polar region (70°S – 90°S) during September 2041–2050. In the Antarctic spring season (Figure ~~6~~8, right part) obviously the enhanced content of reactive chlorine in the SEN-C2-fCFC11_2050 simulations is responsible for the intensified ozone loss in the LS (around 50 hPa) and US (around 1.5 hPa). In the LS also ozone loss through the BrO_x cycle is enhanced, probably related to the enhanced chlorine loading as ozone loss due to the reaction of BrO with ClO is attributed to the BrO_x cycle (cf. Meul et al., 2014). On the other hand, as a consequence of more available chlorine, other loss cycles or production processes

show a tendency to compensate the enhanced ozone destruction by chlorine. For instance the catalytic NO_x-cycle shows some balancing in the altitude region between about 19 km (50 hPa) and the stratopause (about 0.7 hPa). This means that in the 2040s the ozone depletion by NO_x is clearly reduced (i.e. a relative ozone production) in the SEN-C2-fCFC11_2050 compared to the REF-C2 simulation above about 40 hPa. The results with respect to global annual means of the ozone production and loss in the 2040s shown in Figure ~~6~~ 8 (left part) indicate that below about 50 hPa no obvious changes are detected. Above 20 km the ozone loss is strongly affected by reactive chlorine and again some compensation effects in the US due to other competing ozone loss cycles are clearly identified. ~~-~~The positive values with respect to the photolysis rates indicate a slight downward shift of the ozone layer (ozone maximum) to lower altitudes. Probably, this is due to enhanced ozone loss through chlorine at higher altitudes, which allows more UV radiation to reach lower altitudes, where this additional radiation in turn causes higher photolysis rates.

The analogous ozone budget analysis is carried out for changes of the ozone production rate between the SEN-C2-fCFC11_2020 and the REF-C2 simulation. As expected, the vertical dependence of the ozone production rates in the 2040s looks similar as in the analysis of the SEN-C2-fCFC11_2050, namely (not shown): (1) the global annual means of the ozone production and loss below about 20 km (50 hPa) do not indicate obvious changes. Higher up ozone loss is strongly affected by reactive chlorine and again some compensation effects in the US due to other competing ozone loss cycles are clearly identified, but all ozone production rate changes are about half as strong as found between the SEN-C2-fCFC11_2050 and the REF-C2 simulation (see Figure 8 left part). (2) In the Antarctic region during September in the 2040s the intensified ozone loss in the LS (around 50 hPa) through the ClO_x and BrO_x cycles are again obvious, but the ozone destruction rates are only one third of the magnitudes, which are found between the SEN-C2-fCFC11_2050 and the REF-C2 simulation (see Figure 8 right part).

Finally, to~~To~~ check the possible impact of temperature changes due to enhanced CFC-11 concentrations on ozone chemistry we ~~have~~ analyze~~d~~ the overall temperature trends in the US (near 1 hPa) and LS (near 50 hPa) and also the differences between REF-C2 and SEN-C2-fCFC11_2050. The global annual mean long-term temperature behavior in the REF-C2 simulation is indicating a cooling of about 1 K in the LS and of about 3 K in the US from the early 2000s until to the year 2050. The temperature difference between REF-C2 and SEN-C2-fCFC11_2050 amount to an additional cooling of about 0.3 K in the US in the SEN-C2-fCFC11_2050 simulation, whereas no obvious change in the long-term behavior can be identified in the LS (not shown). It is difficult to separate the individual contributions of the additional cooling in a coupled CCM simulation, i.e. radiative cooling by enhanced CFC-11 concentrations and by less ozone in the stratosphere caused by the enhanced chlorine loading without additional diagnostics. We assume that both processes will contribute to the calculated additional cooling in the SEN-C2-fCFC11_2050 simulation.

~~To take~~Taking a closer look to the Southern Hemisphere polar region in spring, the REF-C2 simulation is indicating a clear cooling trend of about 4 K in the US (near 1 hPa) until 2050, whereas no obvious trend can be identified in the LS (not shown). With respect to temperature differences of SEN-C2-fCFC11_2050 and REF-C2, the US does not show a clear

change, whereas in the LS the SEN-C2-fCFC11 simulation suggests some additional cooling from the early 2000s until 2050 by about 2 K. However, this difference is superposed by large inter-annual fluctuations.

With this in mind we can try to ~~value~~ evaluate the calculated ozone differences in the ~~SEN-C2-fCFC11~~sensitivity simulations in comparison to REF-C2. In the global mean US, on the one hand enhanced chlorine mixing ratios are leading to enhanced ozone depletion by the catalytic ozone destruction cycle; on the other hand the extra cooling is known to create a reduction of the ozone depletion rates by gas-phase chemistry (e.g. Haigh and Pyle, 1982). It turned out that the net effect here is slightly negative, i.e. indicate that ozone differences between REF-C2 and ~~SEN-C2-fCFC11~~the sensitivity simulations in the US are dominated by the enhanced chlorine content. In the global mean LS, where no clear cooling is ~~calculated~~simulated, the smaller ozone values are therefore mainly caused by the enhanced chlorine content.

Looking closer at the South polar region in spring, it is obvious that in the US again the enhanced chlorine content is mostly responsible for the slightly reduced PCO in the SEN-C2-fCFC11_2050 simulation (Figure ~~5~~6, upper part). In the LS, where heterogeneous chemical processes are the most important drivers of ozone changes, the enhanced chlorine mixing ratios intensify the ozone destruction. This leads to significantly reduced PCO over the time (Figure ~~5~~6, lower part), which eventually leads to the indicated (slight) extra cooling of the polar lower stratosphere in spring. A first analysis of Polar Stratospheric Cloud (PSC) statistics for the REF-C2 and SEN-C2-fCFC11_2050 simulation displayed, however, no considerable trend in the PSC surface area (not shown). Therefore we cannot identify any hint for enhanced chlorine activation.

4 Discussions and Conclusion

After the detection of an unexpected and persistent increase in global emissions of CFC-11 (Montzka et al., 2018; see also update in Harris et al., 2019) it is still unclear (i) how much these additional emissions have already affected stratospheric ozone, (ii) how ~~these~~ the CFC-11 emissions ~~will~~ could further develop in the next years, and (iii) how large the potential for a disturbance of the temporal evolution of ~~these emissions will affect the recovery of~~ the ozone layer is. The discussions during the International Symposium in Vienna (March 2019) on the unexpected increase of the CFC-11 emissions came to the conclusion that a major problem is to create a realistic assessment of future CFC-11 levels (Harris et al., 2019). There are many factors, which have significant uncertainties, for instance the role of bank emissions or a possible co-production of CFC-12 (CF_2Cl_2) with CFC-11. There was a general acceptance that higher CFC-11 emissions are creating enhanced ozone depletion, but so far the corresponding magnitudes of ozone disturbances are uncertain. This study aims to estimate the possible upper limit of ozone changes due to enhanced CFC-11 surface mixing ratios in recent and coming years, whereas other ODS are declining as expected. For that reason a simplified study based on CCM simulations is conducted first, to estimate roughly the implications of a constant mean CFC-11 surface mixing ratio for ozone depletion instead of reducing CFC-11 on longer time scales ~~in the next decades~~, and second, how strong the maximum ozone effect is due to the additional

CFC-11 emissions in recent years. To keep things as simple as possible we do not consider regional differences with respect to CFC-11 emissions in our sensitivity simulations. From our point of view considering regional differences would not have relevant effects on the presented results because of the long lifetime of CFC-11 (e.g. Rigby et al., 2013; Engel and Rigby et al., ~~2019~~2018)~~.~~, which leads to global mixing (Hoffmann et al., 2014). ~~This study aims to estimate the order of magnitude of ozone changes due to constant CFC-11 surface mixing ratios in the coming years. In our view such a~~Our simplified approach is assuming an extreme boundary condition (i.e. constant CFC-11 surface mixing ratios until year 2050 in SEN-C2-fCFC11_2050 and until year 2019 in SEN-C2-fCFC11_2020), which is justified since ~~there are many uncertainties with respect to the future evolution of CFC-11 emissions;~~ currently a more ~~precise~~ realistic approach with respect to future CFC-11 levels is ~~almost impossible~~not available (Harris et al., 2019). Therefore, the presented results should not at all be taken as a ~~solid~~ robust prediction of future conditions. ~~They should turn the view to the potential impact of unchanged CFC-11 surface mixing ratio on the ozone layer.~~

The presented results indicate that mostly the ozone layer over the Arctic and Antarctic in late winter and spring is ~~significantly~~ in particular affected by the prescribed CFC-11 surface mixing ratio change. In our case, at the end of the SEN-C2-fCFC11_2050 simulation the impact on TCO culminates in a maximum ozone decrease of up to 30 DU in both polar regions (Figure ~~2~~3). The calculated ozone changes at ~~middle~~ mid-latitude and tropical regions are surprisingly small (~~around~~ less than ±5 DU) and therefore are mostly not ~~statistically~~ significant in the sense that the range of variability is in the same order of magnitude. An estimate of possible ozone changes in the late 2040s based on the perturbation of "Equivalent Effective Stratospheric Chlorine" (EESC) may lead to similar results, but appropriate explanations are lacking. Therefore, for the first time we perform a detailed ozone budget analysis of such sensitivity simulations showing interesting results with regard to compensation and buffering effects associated with different production and loss cycles. It turns out from our ~~analyses~~ results that the strengthened ozone depletion by enhanced chlorine is partly compensated by other ozone depleting catalytic cycles (e.g. NO_x) and other molecules (e.g. HO_2). For the global mean picture, there is no big ~~TOC~~ TCO difference visible, as the effects of ozone production and loss processes are nearly cancelling. In the polar regions however, although there are also compensating effects, the signal is noteworthy in spring (e.g. about 20 DU for the Antarctic region). We identify where (altitude) and at which time the ozone amount is decreased in the SEN-C2-fCFC11_2050 simulation compared to REF-C2. The ozone response to CFC-11 changes looks quasi linear, but the processes in the background are obviously non-linear.

Finally, based on the results of our SEN-C2-fCFC11_2050 simulation we ~~present a simple~~roughly estimate ~~for~~ the possible shift of the closure of the ozone hole over Antarctica under the implied conditions of this sensitivity ~~due to the effect of constant CFC-11 surface mixing ratios after the year 2002~~. For this we are ~~using~~ looking at the temporal evolution of the total stratospheric ClO_x loading in the REF-C2 (started in the middle of the 20th century) and the SEN-C2-fCFC11_2050 simulation. In the REF-C2 simulation ClO_x values are strongly increasing from 1960 onwards and are highest at the end of the 1990s. Starting in the 2000s the ClO_x concentration is decreasing. The 1980 value of the REF-C2 simulation can be

regarded as a reference for chlorine conditions before the ozone hole appeared. This "pre-ozone hole" chlorine content is reached again around the year 2050 in ~~the~~ our REF-C2 simulation, which is some years earlier than the multi-model mean based on all REF-C2 simulations as calculated by Dhomse et al. (2018). By extrapolating the linear regression line of the ClO_x content (for ~~2017~~ 2002 to 2050) of the SEN-C2-fCFC11 2050 simulation (not shown) into the future, we estimate, that a pre-ozone hole, i.e. "1980", ClO_x loading is likely to be reached before 2070. Therefore we can roughly estimate a maximum delay of the closure of the ozone hole of somewhat less than about 20 years when keeping CFC-11 surface mixing ratio at a 2002 level vs. a decline of CFC-11 surface mixing ratio as it is reached through adherence of the Montreal Protocol. Considering that ~~Dohmse~~ Dhomse et al. (2018) determined the closure date for the ozone hole by the year 2060 ~~(the multi-model-mean for the analyzed REF-C2 simulations)~~ and that the one sigma standard deviation is in the range of about ±5 years (see also Figure 4.22 by Langematz and Tully et al., ~~2019~~2018), this indicates that the calculated effects of constant CFC-11 surface mixing ratio could have a non-negligible effect on the closure date of the ozone hole. This finding is in line with other model results mentioned by Harris et al. (2019) that the closure of the ozone hole and ozone recovery as a whole will be delayed depending on the CFC-11 emission levels. A first estimate presented in WMO (2018) showed that if total CFC-11 emissions were to continue at levels experienced from 2002–2016 (67 Gg/year), the return of mid-latitude and polar EESC to the 1980 value would be delayed by about 7 years and 20 years, respectively. For the Arctic region enhanced stratospheric chlorine content means that there is the possibility of stronger ozone depletion under specific dynamic conditions (i.e. a stable and cold polar vortex until March) for a slightly longer time period (e.g. Dameris and Godin-Beekmann et al., 2014).

The presented results do not show ~~very~~ dramatic consequences for the global mean ozone layer due to ~~the unchanged~~ enhanced CFC-11 surface mixing ratio for the next years, but indicate relevant changes in the polar regions in winter and spring. In the light of our results showing chemical feedback processes, which are diluting ~~their~~ the effects due to enhanced CFC-11 levels in parts, the compliance of the guidelines of the Montreal Protocol is absolutely necessary ~~and exceptions should not be allowed~~. Without a further strong regulation of the CFC-11 and other ODS emissions (e.g. Laube et al., 2014; Hossani et al., 2017), this could affect significantly the recovery of the ozone layer including the timing of the closure of the ozone hole – and this should be avoided!

Code and data availability. The Modular Earth Submodel System (MESSy) is continuously developed and applied by a consortium of institutions. The usage of MESSy and access to the source code is licensed to all affiliates of institutions, which are members of the MESSy Consortium. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information can be found on the MESSy Consortium Web-site (http://www.messy-interface.org).

Author contributions. Both sensitivity simulations were set-up and carried out by P.J. with support of M.D.; M.D. structured and composed the manuscript. The author team analyzed jointly the model data and compiled the results and all three authors contributed to the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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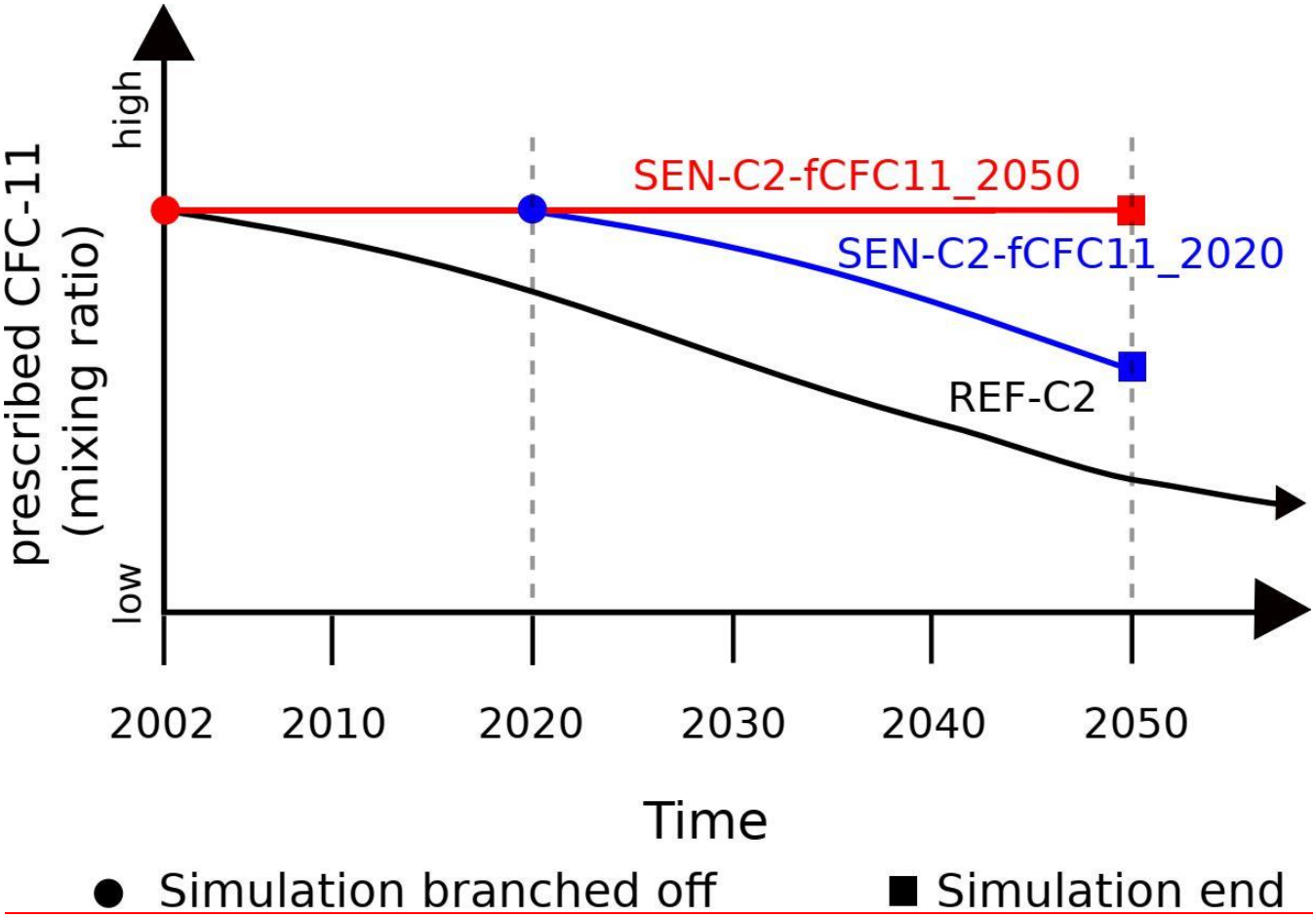

Figure 1: Schematic of the performed EMAC model simulations: A reference simulation (REF-C2) and two sensitivity simulations (SEN-C2-fCFC11_2020 and SEN-C2-fCFC11_2050) enabling an assessment of enhanced CFC-11 surface mixing ratios on the ozone layer.

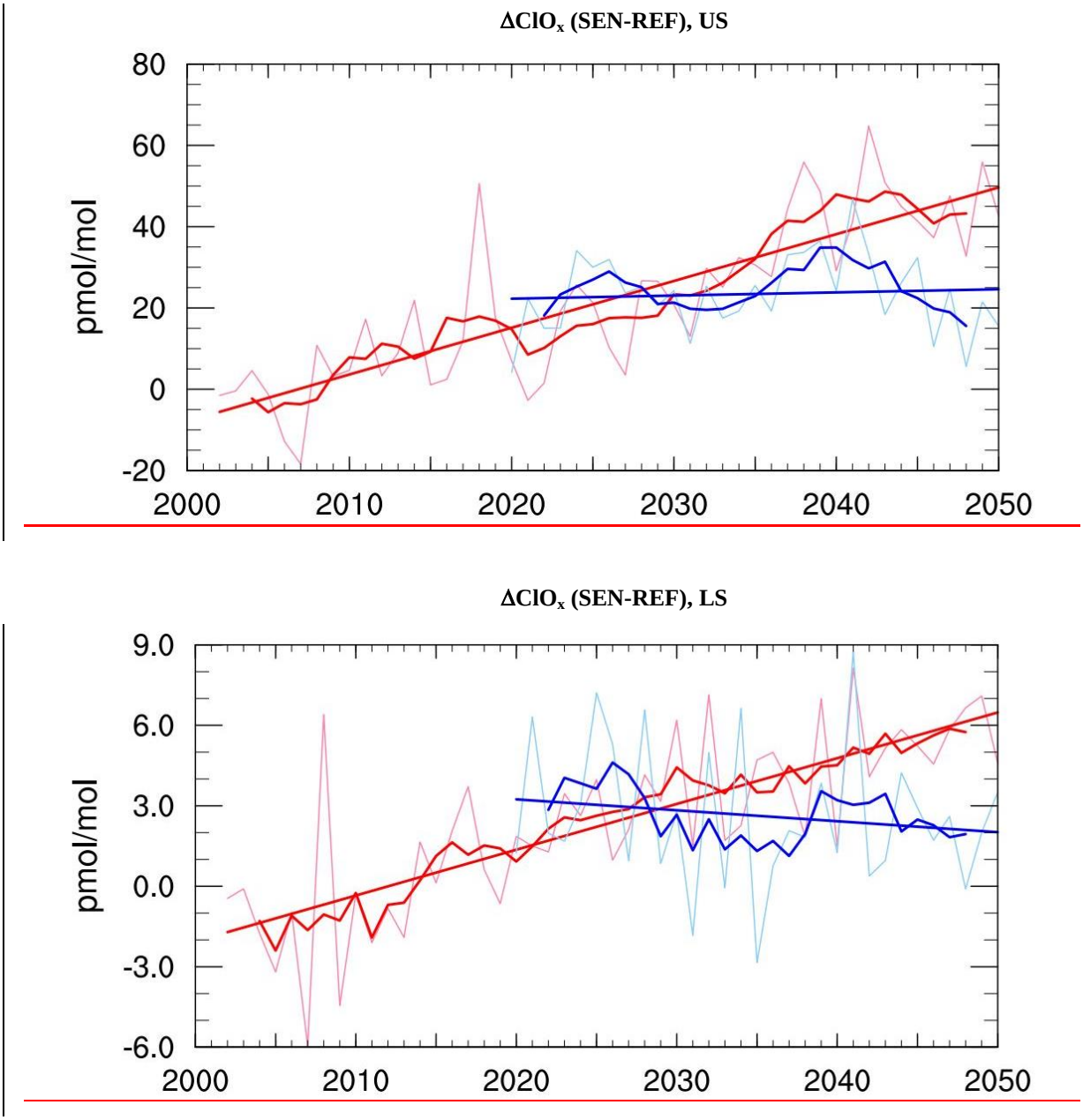

Figure 2: Temporal evolution of the annual global mean ClO_x mixing ratio differences (in mol/mol) at around 2 hPa (US, top) and 50 hPa (LS, bottom) between the SEN-C2-fCFC11_2050 and REF-C2 (in red) and between the SEN-C2-fCFC11_2020 and REF-C2 (in blue). The 11-year solar cycle (smoothed with a 1-2-1 filter) has been removed from both time series. The thicker curves in red and blue show the 5 year running means. The red and blue lines show the linear regression estimate of the unsmoothed time series.

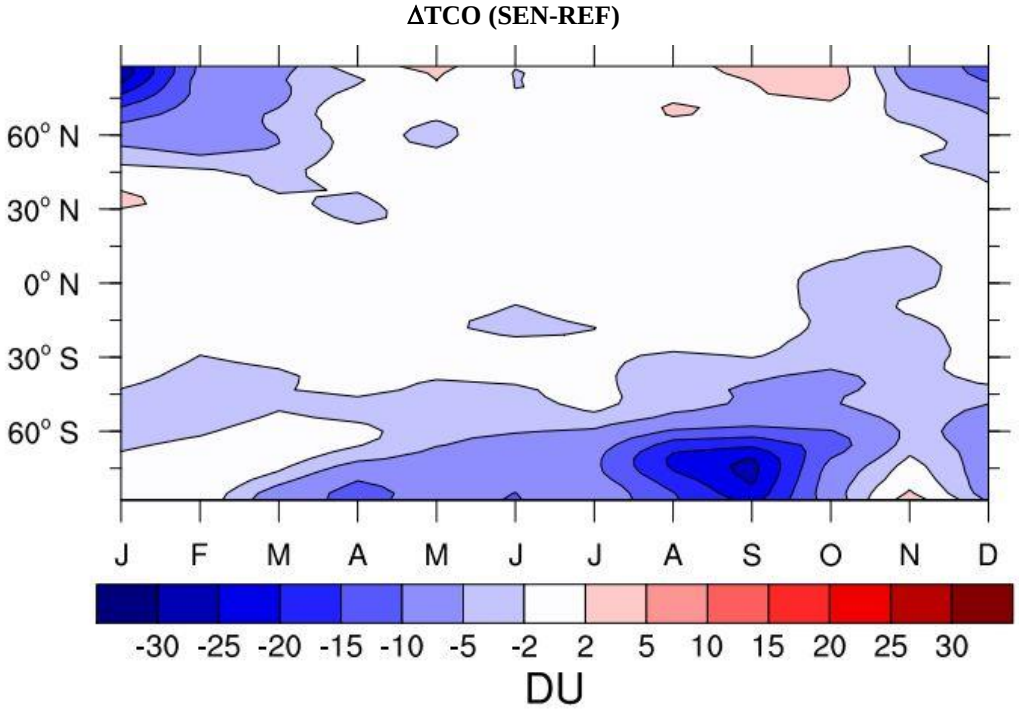

Figure 3: Mean annual cycle of total column ozone (TCO) differences (in Dobson Units, DU) between SEN-C2-fCFC11_2050 and REF-C2 for the 2040s (i.e. SEN minus REF).

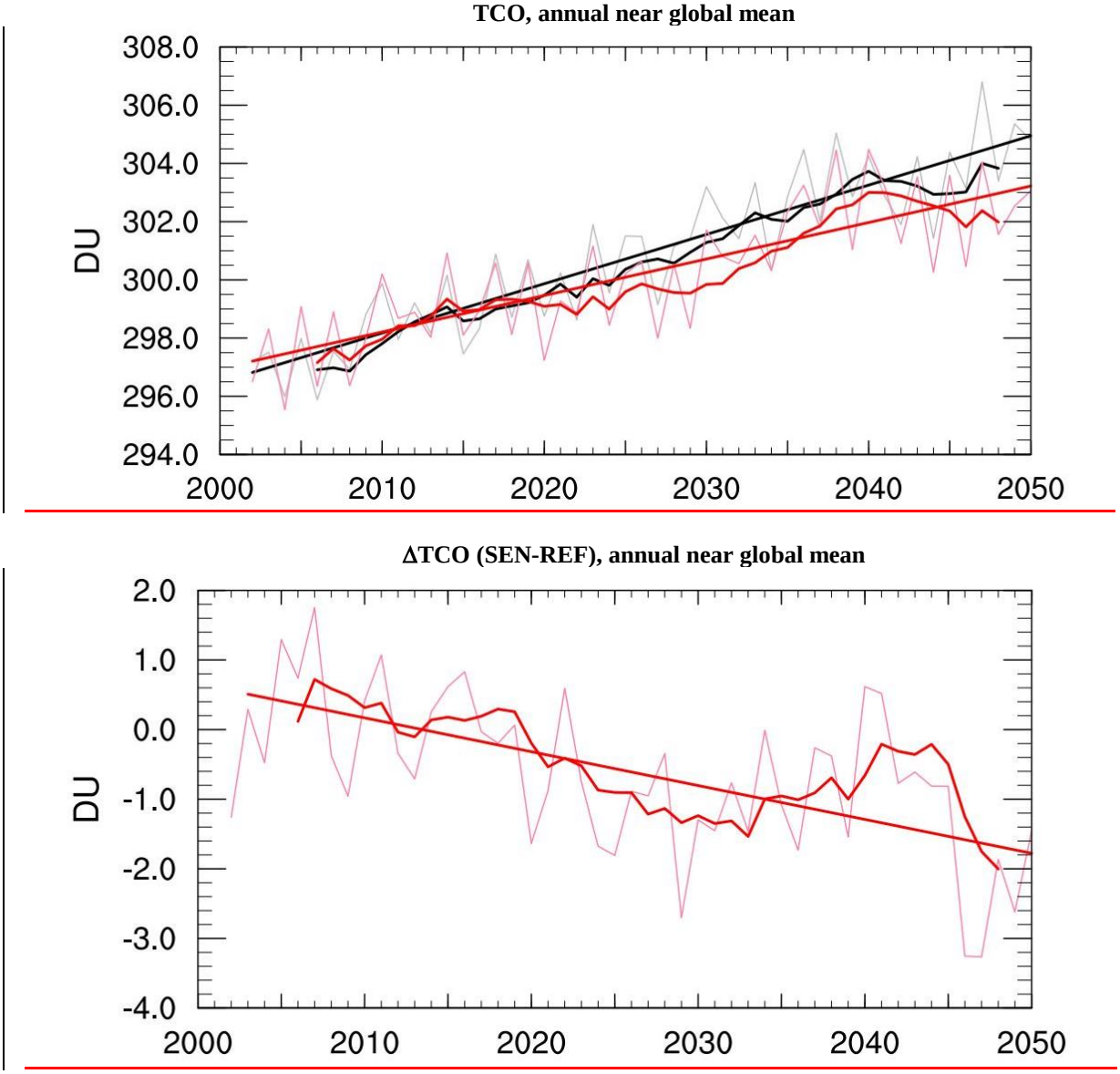

Figure 4: Top: Temporal evolution of total column ozone (TCO; in DU) for the annual near global mean (60° S – 60° N) in REF-C2 (black curves) and in SEN-C2-fCFC11_2050 (red curves). Bottom: TCO differences (in DU) between SEN-C2-fCFC11_2050 and REF-C2 (i.e. SEN minus REF). For the absolute TCO time series (top) the 11-year solar cycle (smoothed with a 1-2-1 filter) has been removed. Thicker curves show the 5-year running means, respectively. The corresponding lines (top and bottom) show the respective linear regression estimates based on the unsmoothed data.

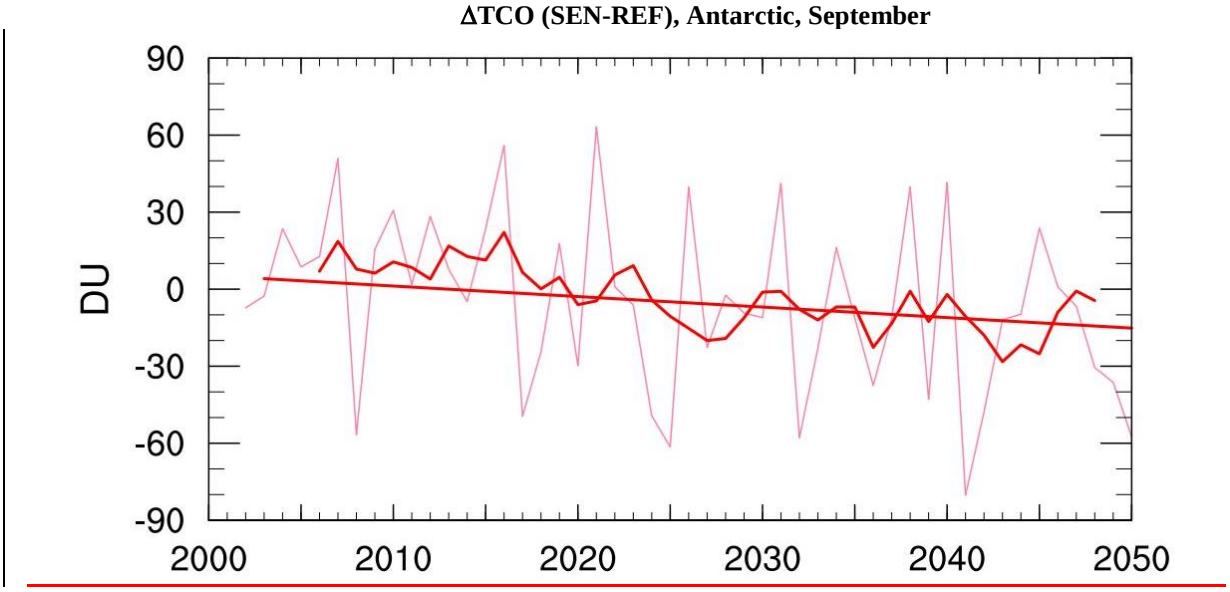

Figure 5: Temporal evolution of TCO differences (in DU) between SEN-C2-fCFC11_2050 and REF-C2 (i.e. SEN minus REF) for the Antarctic region (70° S – 90° S) in September. The thicker curve shows the 5-year running mean. The corresponding line shows the trend estimate between for the unsmoothed time series using a multiple linear regression – including differences of temperature anomalies as dependent variable - which accounts for possible autocorrelation with lag 1 (see text for details).

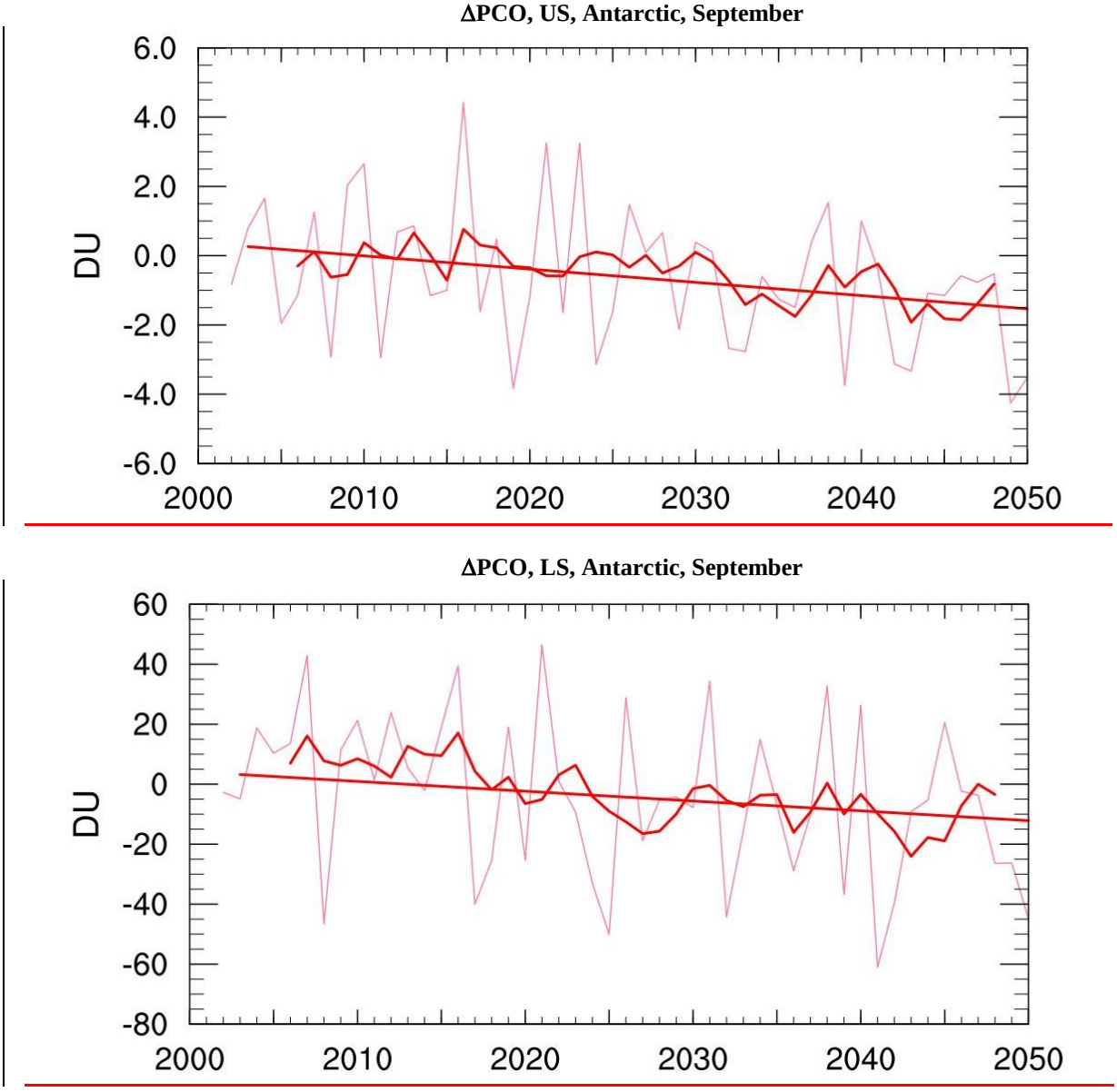

Figure 6: Temporal evolution of Partial Column Ozone (PCO) differences (in DU) between SEN-C2-fCFC11_2050 and REF-C2 (i.e. SEN minus REF) for the Antarctic region (70° S – 90° S) in September. Top: PCO are shown for the US (above 30 km); bottom: PCO are shown for the LS (below 30 km). Red thicker curves show the 5-year running means, respectively. The corresponding lines show the trend estimates for the unsmoothed time series using a multiple linear regression – including differences of temperature anomalies as dependent variable – which accounts for possible autocorrelation with lag 1 (see text for details).

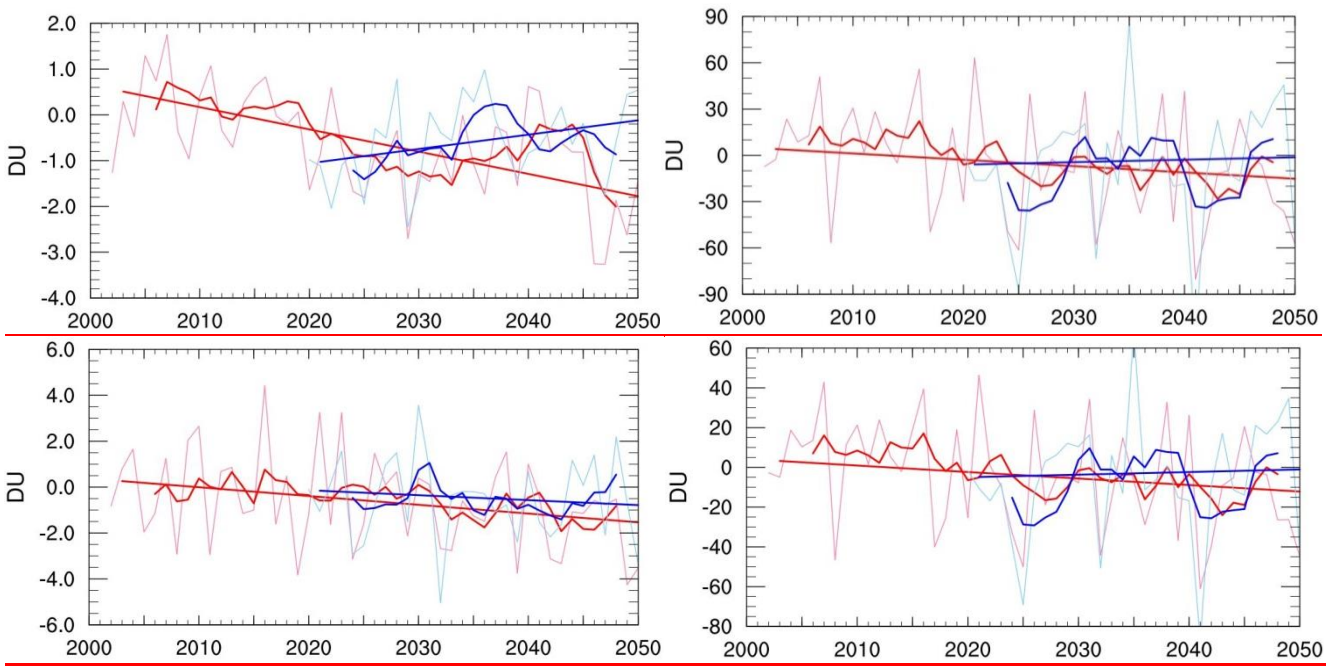

Figure 7: Different temporal evolution of column ozone differences (in DU) between the individual sensitivity simulations and the reference simulation (in DU): SEN-C2-fCFC11_2050 minus REF-C2 values are indicated in red and SEN-C2-fCFC11_2020 minus REF-C2 in blue. Top left: TCO for the near global mean (60° S – 60° N); top right: TCO for the Antarctic (70° S – 90° S); bottom left: PCO for the Antarctic (70° S – 90° S) in the US; bottom right: PCO for the Antarctic (70° S – 90° S) in the LS. The red and blue lines show the trend estimates for the unsmoothed time series using a multiple linear regression – including differences of temperature anomalies as dependent variable - which accounts for possible autocorrelation with lag 1 (see text for details).

Ozone production rate

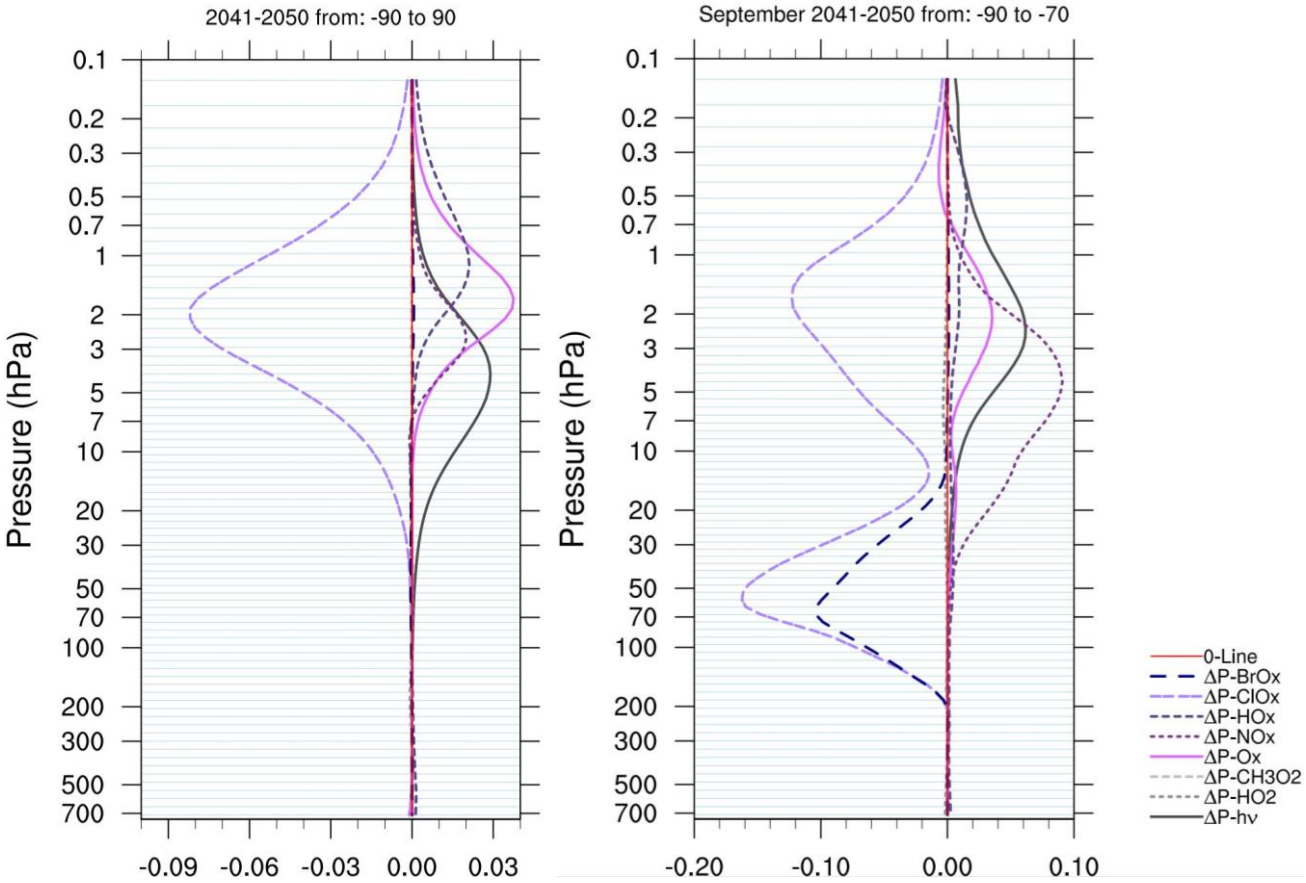

Figure 8: The relative change of ozone production rates (in %), which are normalized to the total column production (through photolysis hυ, HO_2 and CH_3O_2) in the REF-C2 simulation. For the individual ozone destruction cycles and molecules mean difference values are shown, which have been derived from the REF-C2 and the SEN-C2-fCFC11_2050 (i.e. SEN minus REF) simulation for the 2040s (from 2041 to 2050). Left: for the mean annual global mean profiles; right: for the South polar region (70° S – 90° S) in September. Negative values are indicating an intensified ozone loss or a decreased ozone production in the SEN-C2-fCFC11_2050 simulation, whereas higher values indicate more ozone production or less loss through a specific process. Thin horizontal lines indicate the nearest pressure levels to the model grid-boxes.