

Reply to Anonymous Referee #1

We would like to thank the referee for the time and the useful comments that helped to clarify important aspects of the manuscript. Our replies to the comments have been added in blue.

(1) General comments:

Ozone in the troposphere is important for the radiative budget (i.e. greenhouse gas, GHG), atmospheric chemistry (e.g. main source of hydroxyl radicals) and air quality (i.e. pollutant with negative consequences for the biosphere). Thus it is crucial to understand its evolution during the 21st century under different pathways (i.e. emission scenarios). Indeed, a key term influencing the abundance and distribution of tropospheric ozone is the stratosphere-troposphere exchange (STE). The study investigates future changes of STE following the RCP8.5 and RCP6.0 emission scenarios based on experiments of the EMAC chemistry-climate model. Projections based on chemistry-climate models are a valuable mean to probe ozone evolution associated with specific factors – i.e. such as ozone precursors, GHGs and ozone depleting substances (ODSs) – albeit significant disagreements compared to observations and between models are often found.

Overall, the study addresses relevant research questions with regard to the evolution of the STE, its drivers and the contribution of stratospheric-produced ozone to the tropospheric budget. The manuscript is technically well written. Specific comments are detailed below, which are intended to help the authors improve the paper. Briefly, I suggest more details about the “climates” (sea-surface temperatures and sea-ice concentrations, SSTs/SICs) imposed on the simulations to better interpret the comparison between the two emission scenarios considered, since they are from different models (e.g. different climate sensitivity) with consequences for ozone (e.g. chemistry and dynamics). Also, I suggest further acknowledgement to the various roles that methane plays on ozone chemistry – in the troposphere and the stratosphere – as well as its importance on the evolution of the STE. I understand in this set of simulations the effect of an enhanced Brewer-Dobson circulation and stratospheric cooling cannot be separated from the role of methane on ozone chemistry. The present study is recommended for publication after the specific and technical comments are addressed.

We thank the referee for addressing these two important aspects. We have addressed both in detail in our responses to the specific comments below, and adapted the manuscript accordingly.

(2) Specific comments:

a. The study explores how two different emission scenarios (RCP6.0 and RCP8.5) affect the stratospheric ozone mass flux into the troposphere. There is, however, an inconsistency between the SSTs/SICs used that have not been explored/detailed. In order to compare and better understand the differences in STE between these scenarios, it would be desirable to discuss how the underlying Earth System Models (ESMs) – MPI-ESM and HadGEM2-ESM – project climate. For example, the difference in global surface warming by the end of the century (2081–2099 relative to 1985–2005) between these ESMs may be as large as 1 K (higher for HadGEM2-ESM) under the RCP8.5 (see Table 3 in Friedlingstein et al., 2014 – doi.org/10.1175/JCLI-D-12-00579.1). Moreover, Fig. 11 in Jöckel et al. (2016 – doi.org/10.5194/gmd-9-1153-2016) shows that internally generated (EMAC-MPIOM) global

and annual mean SSTs for the same period but following the RCP6.0 scenario are 1 K lower compared to those projected by HadGEM2-ES. Also, the CMIP5 ensemble projects that annual mean surface air temperature anomalies for the same period under the RCP6.0 and RCP8.5 are of 2.2 ± 0.5 K and 3.7 ± 0.7 K respectively (i.e. mean difference of 1.5 K; see Table 12.2 in IPCC, 2013). Therefore, the difference between the SSTs/SICs imposed on the simulations under these emission scenarios may be relatively small, implying that ozone precursor emissions (i.e. methane) may account for a large fraction of the STE difference (i.e. rather than climate-induced).

We thank the referee for this important comment. We have addressed this issue in the revision by including the following text in Section 4:

“Note that the difference in OMF change by 2100 between the timeslice and transient simulations may not be due to the different GHG scenario alone. The HadGEM2-ES model which provided the SST/SIC distribution for the transient RCP6.0 simulation is known to have a higher climate sensitivity than the MPI-ESM which provided the SST/SICs for the EMAC timeslice runs (Andrews et al., 2012). This might lead to a somewhat stronger future SST increase, stratosphere-troposphere exchange and OMF in the transient RCP6.0 run than would arise using MPI-ESM SSTs/SICs. Hence, the differences in OMF by 2100 discussed here represent a lower boundary estimate of the expected OMF differences between the RCP6.0 and RCP8.5 scenarios.”

However, the following figure shows that the MPI-ESM RCP8.5 SSTs are clearly higher than the HadGEM2-ES RCP6.0 SSTs by the end of the century.

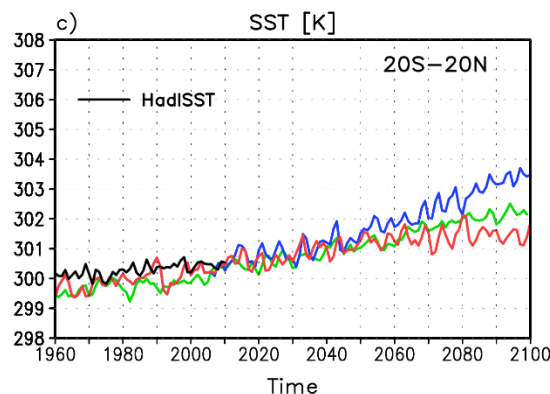


Figure 1: Annual mean SSTs from 1960 to 2100 averaged over 20°S–20°N for RCP4.5 (red) and RCP 8.5 (blue) GHG scenarios derived from MPI-ES, and for RCP6.0 derived from HadGEM2-ES (green). The RCP8.5 timeslice simulations in this study used 2095–2104 averaged SSTs from RCP8.5; the transient RCP6.0 used the transient RCP6.0 SSTs (green curve).

b. Methane is a GHG and plays various roles on ozone chemistry. Although, the authors already acknowledge the role of ozone precursors in the troposphere (i.e. methane; Page 5, lines 85–92; Page 20, lines 462–464), the effect on the stratosphere is not addressed (see below). Among other effects, it is an ozone precursor in the lower stratosphere (i.e. smog-like chemistry) and affects chlorine active/inactive partitioning, which is particularly important for polar regions under high concentrations of ODSs. An increase in methane abundances (e.g. boundary conditions) results in higher stratospheric ozone (e.g. Kawase et al., 2011 – doi.org/10.1029/2010GL046402; Revell et al., 2012 – doi.org/10.5194/acp-12-11309-2012), which will affect the stratospheric ozone mass flux into the troposphere. I guess in the GHG2100 simulation, methane is coupled to both, radiation and chemistry schemes. Therefore, the climate-induced and methane-related impacts on the STE cannot

be unambiguously separated (i.e. both included in the “GHG-induced changes”). I think this should be addressed more clearly in the manuscript and its main findings.

We included the following section to the introduction:

“Besides the temperature effect of the GHGs (mainly CO₂) on the ozone chemistry, the increasing abundances of CH₄ and N₂O also have an impact on the net production of stratospheric ozone: While higher N₂O concentrations are associated with an enhanced ozone loss in the stratosphere due to reactive nitrogen, a CH₄ increase not only causes a larger ozone loss in the lower and upper stratosphere but also leads to an increased ozone production in the lower stratosphere where it acts as an ozone precursor (e.g. Revell et al., 2012). In addition, CH₄ plays a role in polar ozone chemistry since changing CH₄ concentrations also alter the partitioning between halogen reservoir gases and activated halogen species (e.g. Nevison et al., 1999). The combined effect of the increasing GHG concentrations (including the interactions between the chemical cycles) on the net stratospheric ozone production is positive (e.g. Meul et al., 2014).”

We agree that in our simulations the climate-induced and the methane related impacts of GHG changes on STE cannot be separated, but this was not our intention but instead to distinguish between the GHG and ODS related effects on STE and tropospheric ozone change. A comment has been added in Section 2.1 for clarification.

(3) Technical comments:

Pages 1–2, lines 24–25. “. . . GHG effect on the STE change is due to circulation and stratospheric ozone changes. . .”. Does the latter refer to stratospheric cooling and/or methane-related chemistry? Please clarify.

The abstract has been rewritten to improve clarity.

Page 1–2, Abstract. Research question (4) refers to a comparison between the emission scenarios, yet I find no reference to this in the abstract.

The abstract has been rewritten to improve clarity.

Page 4, lines 77–79. Note Banerjee et al. (2016) in their “climate” simulations did not include methane (and N₂O) in the chemistry scheme.

A respective comment has been added to the text.

Page 6, lines 111–113. The sentence reads a bit confusing – i.e. 1960–1999 period is not RCP6.0.

The sentence has been rewritten.

Page 7, lines 150–152. I guess “well-mixed GHGs” are coupled to the radiation and chemistry schemes. Please clarify, this is important to interpret the findings.

This has been clarified in the text.

Page 11, line 244. Typo: “. . . Eqation”.

Corrected

Page 11, line 246. “. . . ozone flux into the NH is larger than into the SH. . .” Is this significant?

The difference between NH maximum and SH maximum is not significant so we removed this statement from the manuscript.

Page 12, lines 252–259. Here it would be appropriate to include the STE budget term from the ACCMIP ensemble (i.e. 477 ± 96 Tg/year; mean and sdev) that informed the last (AR5) IPCC assessment report (see Table 2 in Young et al., 2013; doi.org/10.5194/acp-13-2063-2013).

The paragraph has been rewritten including information from ACCENT, ACCMIP and IPCC (2013).

Page 12, lines 259–261. Note Olsen et al. (2002) presented an influx estimate for the NH midlatitudes (e.g. see their Table 1 for further estimates).

Thank you for this note. We now use the values from IPCC (2013) based on Young et al. (2013).

Page 13, line 289. Do you mean “O3s tropospheric columns. . .”?

Yes, ‘tropospheric’ added

Page 14, lines 302–303. “low(er). . . small(er)”?

Yes, corrected

Page 14, lines 309–311. I would also include where your model lies compared to the last IPCC report. As the authors commented, there is still a relatively large uncertainty on the STE term.

A broader discussion of the STE analyses from IPCC (2013) and model intercomparisons has been included in Section 3.2

Page 15, lines 325–326. Could the “. . . unintended neglect of minor chlorine source gases. . .” account partly for the difference on ozone mass flux trends compared to Hegglin and Shepherd (2009)?

Yes, the weaker ozone depletion due to the missing minor chlorine gases could contribute to the smaller negative trend. But we don’t have the possibility to quantify this effect, so decided not to discuss this in the text.

Page 15, line 346. I would explain a bit more what the “. . . more extreme GHG emission scenario” means (i.e. climate-induced and methane-related impacts).

‘climate change- and methane-related effects’ has been included in the text.

Page 16, lines 356–359. From Fig. 5 it seems that non-linearities may not have a significant impact on the STE, and that ODS-only and GHG-only largely account for the total change (i.e. largest changes in ozone precursors are for methane).

Unfortunately, we are not able to test the impact of non-linearities but we agree that the effect is probably small.

Page 17, lines 385–389; Page 19, lines 419–420; Page 20, lines 460–462; Page 23, lines 522–524. I think the effect of methane-related chemistry on the STE and ozone should be included (assuming methane is coupled to the radiation and chemistry schemes).

We have rewritten the statement:

“The increase of F_{in} (or $O_3 \times -\bar{w}^$) due to the GHG effect (Fig. 6d) is caused by an increase in the downwelling (i.e. $-\bar{w}^*$, positive for downwelling, Fig. 6e) of the BDC in the winter season with climate*

change (e.g., Sudo et al., 2003; Butchart et al., 2010; Oberländer et al., 2013) in combination with an ozone increase resulting from modified chemical production and loss rates in the stratosphere and enhanced meridional transport (Figure 6f)."

Page 18, lines 403–405. Please clarify this sentence.

We have added a new figure (Figure 7) showing the changes in the seasonal breathing term. We think that this make this section clearer.

Page 20, lines 456–457. This is a robust result from different CCMs and worth mentioning (Banerjee et al., 2017 – doi.org/10.5194/acp-2017-741; Iglesias-Suarez et al., 2017 – doi.org/10.5194/acp-2017-939).

Both manuscripts address the radiative forcing due to changes in ozone which is not a topic of this manuscript. Moreover, Iglesias-Suarez et al. (2017) is still under review. Therefore, the references have not been added.

Page 41 and 46, Figure 4 and 9. "TS RCP8.5" legend is a bit confusing since it includes year 2000 (i.e. REF2000).

We have modified the Figures. Now it is stated "TS obs+RCP8.5".

Pages 43–44, Figure 6 and 7. How is the 95 % confidence level calculated? Please explain.

The statistical significance of the future changes is tested by applying the Student's t-test. We have included this information in the text.

Page 45, Figure 8. It would be very helpful to have the annual and global mean values [DU] and [%] at the top of the figures.

Done