Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-64-RC1, 2018

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Future changes in the stratosphere-to-troposphere ozone mass flux and the contribution from climate change and ozone recovery" *by* Stefanie Meul et al.

Anonymous Referee #1

Received and published: 5 March 2018

(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

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Discussion paper

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.

(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 $\sim$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). More-

over, 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).

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.

(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.

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

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

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

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.

Page 11, line 244. Typo: "... Eqation".

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

Page 12, lines 252–259. Here it would be appropriate to include the STE budget term from the ACCMIP ensemble (i.e. 477 $\pm$ 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).

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).

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

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

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.

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)?

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).

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).

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).

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

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).

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

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

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.

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