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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 #2

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General assessment

This manuscript investigates the role of stratosphere-troposphere exchange of ozone on the tropospheric ozone budget and its drivers – climate change and declining ODS concentrations – over the 21st century. In general, the evaluations are straightforward and the manuscript is well written. While I see some shortcomings in the experimental setup of the simulations to answer the questions posed in a clean way (see major and minor comments in the following), the study offers some new and interesting insights, which warrant publication.

Major comments

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Discussion paper

A major issue of this study results from trying to answer question 5 – How is the ratio of stratospheric ozone in the troposphere changing in the future? The problem lies in how the authors have set up their simulations to answer the questions 1-5 posed, and that they do not allow for a clean separation of the different factors (ozone precursor, ODS, CH4, GHG effects) influencing stratosphere-troposphere exchange of ozone. As such the quantification of the relative contributions of the climate and ODS drivers seems somewhat unsatisfying, since the ratio of stratospheric to total ozone in the troposphere depends also on the mixture of the other contributing factors driving ozone in the simulations. While the study still is insightful, the shortcomings (contributions of CH4 and ozone precursor induced ozone production) need to be discussed more thoroughly.

If I understand right, Figure 8 is meant to quantify the relative contributions of ODS-decline and GHG-increases to the stratospheric ozone contribution to tropospheric ozone increases. However, I think the discussion of this Figure is not very logical. On P20 you focus first on the discussion of the relative contributions, which doesn't answer the question you pose at the beginning of the paragraph and in my eyes puts the wrong emphasis on these results. Per definition or design of your simulations, it is a given that the stratospheric contribution in the ODS-only simulation will be the main effect on tropospheric ozone levels (since nothing else is changing), so nothing surprising. The smallish regional differences can be explained by inconsistencies in boundary condition settings or internal variability of the climate system. Hence, the absolute numbers and their relative contributions to the ozone changes in the full simulation are of much more relevance and should be discussed first.

List of comments

Abstract: I feel that the abstract could be much improved by summarising more succinctly and more clearly the main conclusions of the manuscript. At the moment they read like a conclusions section with too much (and nevertheless not enough) detail to me. In particular L29-30 are unclear to me, since you do not explain through which

mechanism GHGs lead to increased tropospheric ozone loss.

Abstract L31: The notation of 'stratospheric column ozone in the troposphere' is confusing to me and seems too close to the notion of the stratospheric column ozone we usually refer to (that resides in the stratosphere). Could you say 'the column-integrated O3s in the troposphere' instead, or anything similar?

P8L178ff: Appenzeller et al (1996) did only address the mass flux, not the ozone flux with their approach. It is important to note that the approach you follow is that of Hegglin and Shepherd (2009), which has to be seen as an extension of Appenzeller et al. Please add this reference to reflect this.

P4L77 Usually, scientists refer to 'idealized model simulations' where simplified models are used and not full-blown chemistry-climate models that hopefully are at least somewhat realistic. Suggest rephrasing here and further down (L104) as well.

P6L188 correct to '. . .change in the future.'

P7L150 Using only 5 years of spin-up seem somewhat short to me given that you would need to bring the stratosphere (with transport times around 5 years in the upper stratosphere) into an updated state. Did you make any tests to see whether the model has no remaining drifts?

P10L209-215 The explanation of this alternative method of estimating STE is unclear to me. Did these references really use the loss of O3s and not O3 to infer STE as a residual from O3 production/loss? What additional information would this yield compared to looking at O3s change as you do here?

P10L223ff and P11L232 It would have been more convincing to compare the model results here to actual measurements in these figures to test the realism of the transition region and stratospheric transport in EMAC. However, I realize that this may be beyond the scope of this paper and do hence not request you to do so. However, it is difficult to say what you learn from a comparison with assimilated MOPPITT data as shown

in Barre et al (2013), since these data do not have the required vertical resolution to resolve the transition region. I suggest to instead compare to the ACE-FTS derived correlations in Hegglin et al. (2009) who conveniently show the CO-O3 correlations for the 30-60N latitude band in DJF (and other seasons, see their figure 7) as you have chosen in your figure. Here you see (in contrast to the Tian et al (2010) paper) that the CO-O3 correlations has a strong seasonality and latitudinal dependency. Judging by eye in the apple-to-apple comparison when using the Hegglin et al FIgure, I would say EMAC is resolving the transition very well, not just reasonably well with CO values at O3=0.1, 0.5, 1,5 ppmv of around 90, 30, 17 ppbv, respectively.

Hegglin, M. I., C. D. Boone, G. L. Manney, K. A. Walker, A global view of the extra-tropical tropopause transition layer from Atmospheric Chemistry Experiment Fourier Transform Spectrometer O3, H2O, and CO, J. Geophys. Res., 114, D00B11, doi:10.1029/2008JD009984, 2009.

P11L228 The reference to Pan et al 2007 seems missing.

P12L259 IPCC (2013) has a newer compilation/assessment of STE ozone fluxes derived from different methodologies, please update to the range indicated there.

P13L276-8 It seems to me that the ozone influx from the stratosphere is larger in spring than in summer according to your results in Figure 2.

P14L280 correct to '. . .in the SH.'

P14L282 again, I would prefer here '. . .the column-aggregated stratospheric ozone in the troposphere. . .'

P14L311 Please update this statement with respect to the IPCC 2013 results.

Tables 2 and 3: Please provide statistical uncertainties for your trend estimates.

P16L358 See also major comment above. It seems crucial to highlight already in the methodology section that precursor emissions are not evolving in the GHG- and ODS-

only simulations. Or did I miss this point? Can you provide an argument/estimate of the effect changes in precursor emissions could have on your results? This seems a major inconsistency in the design of your study with a potentially large effect on the amounts of ozone with stratospheric-origin in the troposphere that should be more thoroughly discussed also in the results and conclusion section. Not only are ozone precursor emissions potentially affecting the lifetime of O3s in the troposphere, but models predict that they had a major effect on lowermost stratospheric ozone concentrations as well, which will likely influence your derived ozone fluxes through non-linear chemical reactions.

P17L372-5 Reading your manuscript, I found this to be a very interesting and puzzling result, which you explain further down in more detail. However, to better envision what is going on I would appreciate to see how -dM/dt changes over time in particular given that this is the second major term in determining the ozone flux into the troposphere. I hence suggest adding a figure that quantifies the LMS mass changes over the 21st century.

P19L424-6 I do not see that O3s is transported further down in the NH than in the SH in Figure 7, rather it seems the opposite. Also, the explanation seems not really an explanation since the chemical lifetime is equally long in the SH winter than in the NH winter. Please check.

P20L447 correct to '. . . a similarly strong . . .'

Figure 8 caption: Please indicate that the numbers you show are changes in ozone and not absolute amounts and do not just refer to Figure 6. Using delta O3 in the figure titles would achieve the same result.

Figure 9 caption L1008: did you mean 'inter-annual'?

P21L484 Sentence seems incomplete.

P21L386-8 Another factor that needs to be discussed are tropospheric ozone precursor

emissions and their effect on the tropospheric ozone burden in the RCP6.0 simulation, since this will be another confounding factor when discussing relative changes in O3s contributions to total tropospheric ozone.

P22L504 correct to '. . . will consist of 46% ozone from. . . '

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