Reply to Anonymous Referee #2

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.

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

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.

The major goal of our study was to investigate the impacts of future changes in both GHG increases and ODS decline between 2000 and 2100 on STE and tropospheric ozone, both for the combined effect and the separate GHG and ODS effects. The additional effect of changes in tropospheric ozone production by changes in ozone precursors was implicitly included in the simulations: In the REF2100 simulation all ozone precursors were adapted to 2100; in the GHG2100 simulation the CH_4 GHG precursor was increased to 2100 values. With this model setup the goals of our study could be addressed. To clarify the inclusion of CH4 and precursors text has been added in Sections 2 and 4.

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.

We are sorry but are not able to understand completely the referee's comment. In Fig. 9 (old Fig. 8) the left column shows from top to bottom the absolute change in the tropospheric ozone column, the absolute change in the tropospheric column of stratospheric ozone, and how much of tropospheric ozone column change is due by stratospheric ozone (in %), all that for the total forcing

by GHG and ODS changes. The middle column shows the same quantities only for GHG increase; the right column for ODS decline. We agree that in the GHG and ODS cases, change patterns are only due to the implied forcing change (and nothing else). And it is true that in the ODS run, the relative contribution from the stratosphere is very high because there is no CH4 increase in the troposphere (hence no changed ozone production) and the effects of dynamical changes are minor (although the stratospheric contribution is not 100% everywhere). But the benefit of this figure – in our opinion - is that by comparing the change patterns of the individual forcings with those by the total forcing allows us to derive which forcing affects which region to which extent.

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.

The abstract has been rewritten and hopefully clarified.

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?

We have changed the notation.

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.

Reference Hegglin and Shepherd (2009) has been added to the text and the method clarified.

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.

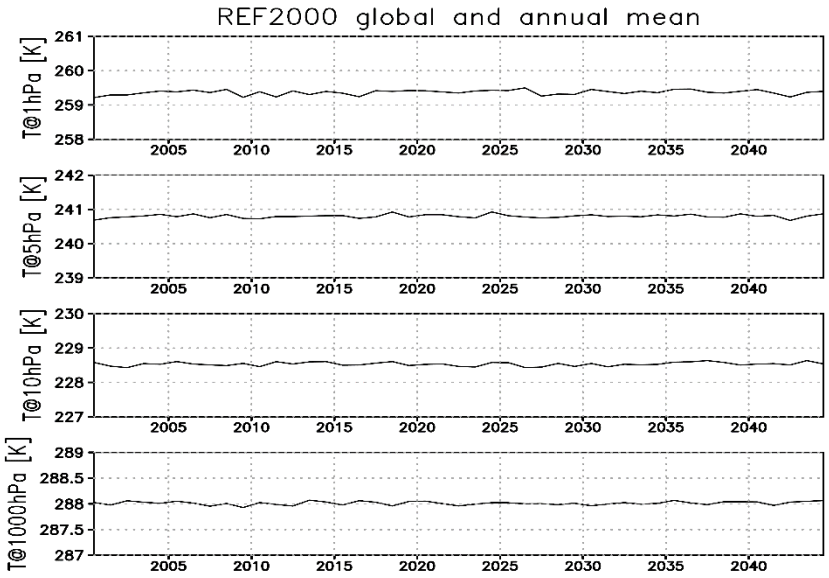
'idealized' not really needed here and removed at both places

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

done

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?

Yes, we checked that there were no drifts in the analysis periods of the model runs. The figure shows as an example global annual mean temperature of the REF2000 run (the analysis period extends from model year 2005 to model year 2045; all model years are run with year 2000 conditions).



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?

Yes, they use the loss of the diagnostic O3s tracer. However, since this is not relevant here we have removed this part.

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.

The reference to the MOPPITT data has been removed and a comparison to ACE-FTS in Hegglin et al. (2009) included in the text instead. ‘reasonably’ has been removed.

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

Reference has been added.

P11L228 The reference to Pan et al 2007 seems missing.

Pan et al. (2007) has been added in the list of references.

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

STE ozone fluxes from Table 8.1 in IPCC (2013) are used for comparison now. The section has been rewritten accordingly.

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.

This is true for the SH, but in the NH the largest influx occurs in early summer. Text has been clarified.

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

done

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

Has been added.

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

The reference to IPCC (2001) has been removed.

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

The uncertainty ranges were included in Table 2 and 3. It has been added that all changes are significant on the 95% confidence level.

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 ODSC4 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.

The reference simulation for the year 2100 includes changes in tropospheric precursor emissions, hence future changes in tropospheric ozone production (in addition to changes in STE). In addition, the GHG2100 simulation considering the effect of increased GHG concentrations includes increases in CH_4, i.e. the ozone precursor with the strongest trend in the RCP8.5 scenario, and consequently changes in tropospheric ozone production.

We have clarified this now by adding text in Sections 2 and 4.

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.

A new Figure 7 has been added showing dM/dt in the same format as Fig. 5. The text has been expanded to explain the seasonal variation of the seasonal breathing term for the total, GHG and ODS forcings.

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.

The text states: “In the SH, the abundance of stratospheric ozone increases throughout the troposphere down to the surface. More O3s seems to be transported further down than in the NH,…”. “Here” has been included to avoid misunderstanding.

P20L447 correct to ‘. . . a similarly strong . . .’

Done

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.

Done

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

Yes; changed.

P21L484 Sentence seems incomplete.

We removed the sentence.

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.

The role of tropospheric ozone precursors in the RCP6.0 and RCP8.5 simulations on the relative changes in O3s contributions in the troposphere had already been discussed in the manuscript by the following sentences:

“*Thus, the increase in the contribution of O3s in the future is slightly smaller in the RCP8.5 scenario than in the RCP6.0 scenario, despite the larger increase in OMF shown in Figure 2. Here, the different evolution of tropospheric ozone production in the two GHG scenarios plays a crucial role*.”

P22L504 correct to ‘. . . will consist of 46% ozone from. . . ’

Done