

Author Response to Anonymous Referee #2 Comments

Thank you for your comments. Referee comments are given in black and author comments/actions in red.

“The manuscript titled “Characterising the Seasonal and Geographical Variability of Tropospheric Ozone, Stratospheric Influence and Recent Changes” presents a very interesting analysis on the stratospheric influence on tropospheric ozone using two chemistry-climate models CMAM and EMAC, as part of the IGAC/SPARC CCM1 activity. The manuscript first shows that both models agree quite well with the observations from Satellite with the Ozone Monitoring Instrument (OMI) and from ozonesondes. Then the manuscript focuses on the models to study the variability of tropospheric ozone, stratospheric ozone and the stratospheric intrusions in order to assess how much stratospheric ozone impact tropospheric ozone. A statistically significant increase in tropospheric ozone is found across much of the world. The role of the stratosphere-troposphere exchange to such ozone changes ranges from 25-30% at the surface and 50-80% in the upper troposphere-lower stratosphere.

Although the manuscript is not so easy to read and follow, it is well structured; in particular, the summaries of each main section are very much appreciated.

I would suggest minor revisions, mainly clarifications, before the manuscript could be published.”

Thanks for your feedback. Hopefully implementation of the suggested minor revisions/clarifications will help to improve the legibility of the manuscript.

“General comments:

I found one general information missing about the models. It is the inferred stratospheric influx as mentioned in Young et al., 2013 (Table 2) for other CCM1 models. Could the authors add this information?

Young, P. J., Archibald, A. T., Bowman, K. W., Lamarque, J. F., Naik, V., Stevenson, D. S. et al.: Pre-industrial to end 21st century projections of tropospheric ozone from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP). Atmos. Chem. Phys., 13(4), 2063-2090, doi:10.5194/acp-13-2063-2013, 2013”.

This information is only from ACCMIP (a subset of CCM1 models) and therefore we summarise such information instead from table 8.1 from the IPCC WG1 AR5 report which includes the mean stratospheric influx from this subset of models, in addition to a range of other models and observational estimates. Sentence added in opening paragraph of section 2.

“Specific comments:

Line 1 p. 2: Could the authors give the period of time on which the change in ozone was calculated: 4-6 ppbv (5-10%)”.

This information should have been implicit as the time period was mentioned in the previous sentence (line 34-35, p. 1). Have omitted this detail here and given the period of time the change was calculated over at the end of the above sentence (line 2, p. 2) for clarity.

“Line 24 p.2: “background ozone” is used here, when I think it refers to “baseline ozone”. According to the Hemispheric Transport of Air Pollution 2010 Part A paper and Cooper et al.

(2014), “Baseline concentrations refer to observations made at a site when it is not influenced by recent, locally emitted or produced anthropogenic pollution. The term global or hemispheric background concentration is a model construct that estimates the atmospheric concentration of a pollutant due to natural sources only.

Cooper, O. R., Parrish, D. D., Ziemke, J., Cupeiro, M., Galbally, I. E., Gilge, S., ... & Oltmans, S. J. (2014). Global distribution and trends of tropospheric ozone: An observation-based review.

HTAP, T., 2010. Hemispheric Transport of Air Pollution 2010 Part A: Ozone And Particulate Matter, Air Pollution Studies No. 17”.

Thanks for pointing this out. We indeed misuse the term ‘background ozone’ as referring to the ‘baseline ozone’ when citing Cooper et al. (2014) and so have corrected this and added in the additional HTAP (2010) reference. Our reference to studies which refer to ‘background ozone’ remain, but we keep these citations separate from the two above.

“Line 12-14 p. 3: I am not sure to understand where “seasonal minimum’ comes from. According to Tang et al. (2016), the STE ozone flux in the Northern Hemisphere shows a maximum in late spring and early summer as well. Could the authors clarify the sentence?”

The phrase “seasonal minimum” on line 13 relates to the STE mass flux, not the STE ozone flux which we acknowledge has a seasonal maximum in late spring and early summer (in agreement with Yang et al. (2016)?) on line 12. Have revised this sentence, which hopefully clarifies this better and improves readability.

“Line 9 p. 5: [Typo] In “24, 6, 48 and 24 h”, “24” is written twice”.

This is an actual fact not a typo. These times refer to the nudging to temperature, vorticity, divergence, and surface pressure respectively. These are now listed following a colon and taken out of parentheses to avoid confusion.

“Line 26 p. 5: [Typo] Change “Langrangian” to “Lagrangian””

Typo corrected.

“Line 1 p. 9: Could the authors add references about the intercomparison campaigns between 1970 and 1990, for example Beekman et al. (1994). I would have the same comment for the “evidence that the ECC sondes have greater precision [...]”.

Beekmann, M., Ancellet, G., Megie, G., Smit, H., Kley, D., 1994b. Intercomparison campaign of vertical ozone profiles including electrochemical sondes of ecc and brewer-mast type and a ground based uv-differential absorption lidar. J. Atmos. Chem. 19, 259e288.”

References added, many thanks.

“Line 18 p. 9: The authors wrote, “A seasonal maximum in tropospheric ozone is evident in each hemisphere during spring, which is more pronounced in the Northern Hemisphere and extended in many regions through summer”. According to Figure 1a, the Northern Hemisphere shows a seasonal maximum in spring and summer. In spring the maximum is rather seen

above 80N. How confident are you on the retrieval of tropospheric ozone above 80N? Wouldn't be rather a stratospheric signal?

Could the authors add this particular polar region (>80N) where the spring maximum is seen?"

We make no change to the manuscript as the retrieval should not be trusted at these latitudes due to the influence of the OMI row-anomaly (rows on the 2-D detector which have become damaged or blocked by insulation blankets – mentioned on page 8, L6-9). We have instead extended the grey masking to cover this region in MAM/JJA.

"Line 21 p. 9: Use of the parenthesis: "northward (southward)". This is not really a good structure and the authors tend to overuse it through the manuscript. I would suggest writing it without the parenthesis. That would be better English and more fluent for the reader.

Sentence revised and we limit use of this structure elsewhere to only sentences where we deem appropriate and fluency is not compromised for the reader.

Figure 1 (p. 10): I would suggest to change the maximum limit of the colorbar. Tropospheric ozone (1000-450 hPa sub-column) barely reach 35 DU at a maximum. I would suggest to change 50 DU to 35 DU. The geographical variability of tropospheric ozone will then be easier to see.

Would the authors know what is happening above South Africa for JJA and SON? There is ozone values around 30 DU on the coast and above the ocean around but rather 20 DU on the continent, as there would be a continent/ocean barrier. It does not seem real."

We have revised this colour scale accordingly. We believe advection of precursor-rich air from the continent (due to biomass burning) and later formation of photochemically formed ozone to explain the higher values offshore, together with the reduced depth of the subcolumn over the relatively elevated South African mainland.

"Line 23 p. 12: Could the authors explain more, maybe with an equation, how they link the "interannual variability" and the "seasonal aggregates of the computed relative standard deviation (RSD) of the monthly mean O₃". It is not obvious. The interannual variability seems to be the variability year to year. Why would the authors study seasonal composites of RSD as a metric for the interannual variability?"

We have added an equation immediately below this sentence for clarity in how we calculate seasonal composites of RSD. The standard deviation is normalised with respect to the mean of each individual month over all years (2005-2010) to compute the monthly RSD which we then aggregate by calendar season. This metric therefore captures variability with respect to the monthly resolved seasonality over all years which we infer here as the interannual variability.

"Line 14 p. 17: Section 4. Could the authors explain more the difference between O₃S and O₃F? If there is any equation used, I would suggest adding them to the text. It is not so clear."

We now make this clear through revising this sentence and have also added the O₃F equation in line (in addition to the Fig. 5 caption).

“Line 18 p. 33: “RSD values of over 10%”. How does 10% compare with other values? It is not a clear evidence for the reader that it shows an influence of ENSO and QBO.”

According to Lee et al. (2010), a study that was originally cited in section 3.2, the QBO is estimated to induce anomalies of as much as 10-20 % in tropical tropospheric ozone, which would scale with the 10-20 % variability in RSD found in both OMI and the models. This is again referred to here to ensure the reader makes this connection but ‘ENSO’ is dropped as the study found that the influence from ENSO is likely much smaller (~ 3 %).

“Line 24 p. 33: “Taking this information into account”. To which information do the authors refer?”

This refers to the findings of section 3 (summarised in the paragraph above in section 6: conclusions). This is made clearer to the reader.

“Line 33 p. 33: The sentence started at this line and finishes line 2 p. 34, I think the sentence could be shortened.”

This sentence has been split in two.

“Line 34 p.33: “(no larger than 20%)”, I would suggest removing the parenthesis and writing “with biases no larger than 20%”.”

This has been revised.

“Line 13 p. 34: What does “high sensitivity to the tropopause” mean?”

This refers to the height of the tropopause being key to the calculated changes (due to the associated sharp vertical gradients in ozone). Changed to ‘known discrepancies in tropopause height’ as found in Hegglin et al. (2010).

“Figure S4 p. 6: [TYPO] in the caption change “CMAN” to “CMAM””

Thanks, typo corrected.