Author Response to Anonymous Referee #1 Comments

We thank the reviewer for her/his helpful comments. Referee comments are given in black and author comments/actions in red.

"The paper by Williams et al. "Characterising the Seasonal and Geographical Variability of Tropospheric Ozone, Stratospheric Influence and Recent Changes" utilises satellite and ozonesonde observations and two chemistry-climate models to investigate the stratospheric influence on tropospheric ozone. The authors conclude that the influence of stratospheric on tropospheric ozone is larger than previously thought. The authors also assessed the tropospheric ozone over the periods of 1980-89 and 2001-2010, and find an overall significant increase in tropospheric ozone, and attribute 25-30% changes at the surface and 50-80% in the upper troposphere to the stratosphere-troposphere exchange. The paper is well written, and the analysis is generally thorough, but some clarifications and improvements are needed before the paper can be accepted for publication in ACP. Detailed comments are listed below."

Thank you for your positive assessment of our manuscript. We hope to have clarified the issues raised with details on changes given below.

"General comments:

A major concern is that this study includes only two CCMI model results, which reduces the robustness of the finding, especially in that "the influence of STE in the tropospheric ozone is larger than previously thought". Furthermore, using only simulations constrained with prescribed dynamics might obscure the changes due to dynamical feedbacks, especially when assessing long-term changes in ozone. Therefore, I suggest that the authors tone down the conclusion mentioned above, and instead focus on the uncertainty in the contribution of STE to the tropospheric ozone. The limitation of using prescribed dynamics CCM simulations should also be noted and discussed. A wider usage of CCMI models would address the first comment. As a minimum, the authors should give a reason for using only the two chosen models."

We acknowledge your concern that there are caveats with confining the analyses to only two models (such as the robustness of findings) but we argue that such detailed analyses would not be possible in the context of a wider range of CCMI models, at least not within the scope of this paper alone. However the main reason for our choice of these two models is the availability of the O_3S tracer which is defined similarly. Whilst some (but not all) of the other CCMs in CCMI have O_3S tracer simulations, they are not defined equivalently. We also acknowledge your point regarding the use of the specified dynamics simulations and agree that the above conclusion needs to be toned down accordingly, with greater emphasis on the uncertainty in the contribution of STE to tropospheric ozone. Whilst we are fully aware that the use of specified dynamic simulations can supress chemistry-climate feedbacks, the constraint on dynamics is critical for our quantification of historical changes due to the primary influence of transport and dynamics on the chemical distribution of ozone VMR. We agree that the limitations of using only two CCMs and specified dynamics simulations needs to be noted and discussed more widely, which we now address in the revised manuscript. In response to the first comment, we now make clear the reason for our choice of the two models.

“Regarding previous studies, I doubt that the paper by Lamarque et al. (1999) is still a very relevant reference that the authors focus their comparisons on, given that the approach used in Lamarque et al. (1999) was very simplistic compared to what can be achieved using more recent state-of-the-art CCMs. Also, there are a few more studies that have investigated the impact of STE on tropospheric ozone which the authors failed to cite, for example, Lelieveld and Dentener (2000), Hess et al. (2013), etc. Jos Lelieveld and Frank J. Dentener, What controls tropospheric ozone? JGR, 105, P3531-3551 2000.

P. G. Hess and R. Zbinden, Stratospheric impact on tropospheric ozone variability and trends: 1990–2009, ACP, 13, 649–674, 2013.

Therefore, a more thorough review of the recent literature would be desirable.”

Our explicit reference to Lamarque et al. (1999) relates to the similarity of their analyses to ours and we would argue that the comparison to their results highlights how our understanding of the influence of the stratosphere on tropospheric ozone has changed in the last twenty years as models have become increasingly more complex. However, we certainly agree with your point and thus also feel that other more recent publications on the matter (such as the ones suggested above) need mentioning, which will help for a more thorough review of the recent literature on the impact of STE on tropospheric ozone. This has now been implemented.

“Specific comments:

P2, L13: add “large” before “number”

Done.

“P4, L17: please add references here”

A reference has been added to support this point.

“P5, L5-L20: Can you describe the models’ characteristics in a more objective way here? Why do you choose these two models specifically (there are quite a few other models from CCMI that you could include)? Also describe the main differences between these two models.”

Please see paragraph for amendments to help describe such characteristics more objectively. A sentence has been added to justify the use of only the two models (based on the definition of the O_3S tracer which is similar in both models and is either absent or defined different in other CCMI models. A sentence has also been added highlighting the main differences between the two models which is logically structured in more detail in the following specific model sub-sections (2.1.1 and 2.1.2).

“P5, L28: Pease provide more details in chemical schemes used in EMAC.”

Please see following added sentence detailing the chemistry included in the MECCA chemistry submodel which goes into EMAC.

“P6, L4 & L21: Please provide more detailed information on how the O3S tracer is defined in terms of its chemical and dynamical nature in both models.”

Please see manuscript for additional added information on the chemical and dynamical constraints on defining the O3S tracer.

“P7, L19-L21: Please clarify if the AKs have or have not been applied to the modelled and ozonesonde data when you compare these two. It only makes sense to apply AKs when compare model/sonde data to the satellite data.”

No AKs have been applied for this comparison and rightly so as it only makes sense to apply AKs for satellite-model/satellite-sonde comparisons as you state. The sentence describes satellite-sonde validation efforts as reported by another study (Miles et al., 2015). Although this was cited further up, it has been cited again to avoid any confusion.

“P7, L27-L29: I don’t understand why “The 1000-450 hPa (0-5.5 km) OMI subcolumn data is considered a representative approximation of the full tropospheric ozone column amount, due to vertical smearing of information from above 450 hPa (_ 5.5 km).” is the case. Is it possible to show AKs?”

We remove this claim as the accuracy of this representativeness is strongly latitude and seasonally dependent and due to lack of supporting literature to support this statement.

“P9, L1-L4: Please add references here. There are a series of publications on JOSIE by Smit et al.

Smit, H. G. J., and D. Kley (1998), JOSIE: The 1996 WMO International intercomparison of ozonesondes under quasi flight conditions in the environmental simulation chamber at JuÂ´llich, WMO Global Atmosphere Watch report series, No. 130 (Technical Document No. 926). World Meteorological Organization, Geneva.

Smit, H. G. J., and W. Straeter (2004a), JOSIE-1998, Performance of ECC Ozone Sondes of SPC-6A and ENSCI-Z Type, WMO Global Atmosphere Watch report series, No. 157 (Technical Document No. 1218), World Meteorological Organization, Geneva.

Smit, H. G. J., and W. Straeter (2004b), JOSIE-2000, JuÂ´llich Ozone Sonde Inter-comparison Experiment 2000, The 2000 WMO international intercomparison of operating procedures for ECC-ozonesondes at the environmental simulation facility at JuÂ´llich, WMO Global Atmosphere Watch report series, No. 158 (Technical Document No. 1225), World Meteorological Organization, Geneva.”

Many thanks, these references have been added.

“P10, L19: Ozone is not at its minimum in SON in the SH, but maximum. Biomass burning emissions and STE usually dominate the seasonality of SH O3.”

We acknowledge this is the case already for the SH as a whole, but this sentence refers only to Antarctica and the Southern Ocean where the influence of stratospheric ozone depletion is ‘likely’ dominant.

"P11, L21-23: can you provide more details on the difference between these two NOx emission datasets?"

Further details added courtesy of the Hoesly et al. (2018) reference.

"P12, L1-L2: This seems slightly mis-leading on the function of the AKs. The purpose of applying the AVKs is to compare like with like."

We use Fig.2 (Table S1) to show and describe the importance of applying AKs and the effect it has on tropospheric ozone which we would argue is informative for an uninitiated reader to AKs and the necessity of its application for like for like model-satellite measurement comparisons. Sentenced revised slightly to avoid misleading the reader.

"P12, L4-L7: Similar features seem exist in both models; it seems more likely due to transport barrier than STE (which the STE maximises in winter)."

We agree with this statement and revise this rather speculative sentence to allude to this as a possible cause, as well as the magnitude of the stratospheric ozone hole which could explain the retention of this feature after applying AKs.

"P12, L7-L9: Does the difference in chemical schemes between the two models play a role here?"

There could be differences due to disparities in the implementation of emissions in the model schemes and different treatments (e.g. bulking of species in CMAM) but we cannot disentangle such influence apart from dynamics here in our evaluations. We now acknowledge this as having a possible influence in the biases in each model here and allude to the need for further investigation.

"P13, Fig 3: It is impossible to discern the RSD of ozonesonde data denoted as circles, due to a uniformed colour scale."

We have revised the colobar scale down from 0-20 % to 0-14 % to make clearer structure in the RSD plots. Ozonesonde RSD is now made more easily distinguishable (white outline) but note that the RSD for sondes is uniformly low with few exceptions. The reason for this unclear and would warrant further investigation.

"P14, Fig 4: The value of 100 ppbv O3 seems a bit too low for defining the tropopause. Using 100 ppbv O3 also deviates from the definition by Bethan et al. (1996) (cited in caption), which is based on the ozone gradient, defined as the minimum altitude where the vertical gradient of the O3 VMR is greater than 60 ppb/km, remains so for a further 200 m, and the O3 mixing ratio is greater than 80 ppbv, exceeding 110 ppbv immediately above the tropopause."

Thank you for spotting this error, we now remove the citation to Bethan et al. (1996) and simply state that we choose the 100 ppbv ozone isopleth as a rough approximation of the chemical tropopause height.

"P15, L16-L32: Please note the figures that you are discussing throughout this paragraph.

Also, the description/discussion in this paragraph can be simplified to focus on the main points."

All discussion in this paragraph refers to Fig. 4 and we now remind the reader at the start of this paragraph. We have revised and shortened this paragraph for easier reading and to make clearer the main points.

"P16, L1-L2: again, please can you refer to the figure(s) that you are discussing."

Done.

"P16, L27: Do you also apply AVKs to model data when compare them with ozonesonde data? If so, it is not necessary."

No we do not. We merely use the model-ozonesonde comparison to infer how the biases arise/change between OMI and the models as a result of applying AKs to the models in these comparisons (Fig. 1 and 2).

"P16, L28: do you mean "simplified" tropospheric chemistry scheme? It is unusual to use the word "conservative"."

Yes, word changed.

"P16, L29-L31: which comparison/figures you are talking about here? Please make it clear by referring to figures."

The additional smearing (increase in subcolumn ozone due to AK application) can be seen in Fig. 2 but the conclusion is made based on the model-ozonesonde comparison (Fig. 4) as should be clear from the previous few sentences. We however remind the reader that the influence of the AK can be seen in Fig. 2 to make this clearer.

"P16, L32-L33: It seems lacking context regarding "since vertical smearing of information is far more limited due to a higher tropopause.". Could you be specific? Where is the information regarding a higher tropopause?"

We base our statement on the higher climatological mean position of the tropopause in the tropics/sub-tropics compared with the extratropics which will result in less smearing of information from the stratosphere. Sentenced revised for clarity.

"P17: L1: "must induce" should be "must have induced""

Changed.

"P17: L3-L7: Showing the AKs might help with the discussion here."

We add in an example of the OMI monthly mean AKs for August 2007 (~ 47°S) to illustrate this point in to the supplement (new Fig. S2), which importantly shows that the 1000-450 hPa subcolumn is sensitive to influence from ~ 150-450 hPa pressure range. This indicates where the smearing of information can originate from. We refer the reader to this figure and emphasise this point in the manuscript.

"P21, L31: Please provide details on how you map the model data to ozonesonde measurements shown in Fig 7?"

Additional detail has been added. Ozonesonde profile measurements were aggregated for each month and for 10 degree latitude bands, which were then subsequently averaged over all 31 years (1980-2010) over all longitudes (zonal average). Similarly to Fig. 4, measurements were interpolated and averaged between ±20 hPa for each pressure level (350, 500 and 850 hPa).

"P23, L1: it is too general to say that "… are evident in the contemporary CCM simulation" while only two CCMs are used here."

Dropped the word "contemporary".

"P24, L11: What do you mean "even lower tropospheric ozone"?"

We refer to the lower troposphere but can see how such confusion may arise. The phrase "even lower" has been removed and we refer now to such influence extending down into the lower troposphere.

"P24, L20: What is the rationale for choosing these longitudes?"

Whilst the choice is a little arbitrary to illustrate that calculated changes are very much height-dependent, both the 30°W and 30°E transects intersect the small region of negative change over central Africa, which varies according to model and pressure level in terms of magnitude and location. The 30°W transect also intersects Greenland where there is significant model disparity at 350 hPa during MAM (Fig. 8a and 10a), although the cross-sectional view (Fig. 9a and 11a) shows such large differences to be confined only to the uppermost part of the domain near to the tropopause). We find this to be consistent with differences in tropopause height found by Hegglin et al. (2010) and our own finding that the lower branch of the BDC is stronger in CMAM. The 90°W transect (Fig. 9c and 11c) intersects the Himalayas where there are some interesting differences in the calculated changes on either side of this mountain range for both O_3 and O_3S and between each model. Overall the selected transects help the discussion in this section to understand the upper level of stratospheric contribution to changes in near-surface ozone (largest across the Eurasian continent) and the source of such features such as the negative trend region over Africa (not evident in O_3S). A shorter version of this rationale is now added here in the manuscript for clarity.

"P25, Fig 8: There are large areas in the SH that are denoted significant in CMAM (SON 500 hPa and SON surface plots), which are not reflected in the relevant discussion. Please check."

We now acknowledge this in the manuscript but do not discuss in depth as changes are small. The modest increase would appear to be related to long range transport from the SH subtropics and entrainment hemispherically by upper level winds especially. O_3S shows no such significant changes implying this increase is driven by changes in the troposphere.

“P27, L22: Please note which figure(s) you are discussing here? Is Fig 8?”

It is Fig. 8, yes. This has been added.

“P30, L10-L12: is “subtle shifts in the height of tropopause” shown anywhere?”

We do not show it anywhere but we do refer to Hegglin et al. (2010) earlier on in 4.1 (page 19, line 8) which finds that the CMAM tropopause is lower (some 30-50 hPa higher in pressure) than EMAC. This citation is now mentioned here also to support this statement.

“P34, L10-L12: What are the reference variables for these percentage changes?”

The percentage change values relate to the change values in ppbv we summarised earlier in 5.1 – e.g. The O_3 (Fig. 8) increase in the NH mid-latitudes during MAM/SON in the upper troposphere, being on the order of some 4-6 ppbv is seen to equate to a 1-3 ppbv increase in O_3S (Fig. 10) over the time period considered, hence we arrive at a ~ 25-50 % stratospheric contribution to the total change. We now mention here that such values are arrived at in conjunction with use of the tagged stratospheric (O_3S) tracer.

“P35, L2: Please specify re “some regions of the world".”

We now give examples where the increase is substantial and generally significant for both models in one or more seasons: W. Eurasia, E. North America, S. Pacific Ocean and the S. Indian Ocean.