Authors' response to reviewer comments on - "Tropospheric ozone in CMIP6 Simulations" by Paul T. Griffiths, Lee T. Murray et al.

This paper provides a current and necessary update to the global tropospheric ozone budget using 3 or 4 state-of-the-art models. The paper will be very useful to the research community, but it first needs a major revision to improve the analysis and discussion in three areas:

1) A major conclusion of Young et al. [2013] is that the projected increase of ozone during the 21st century under RCP8.5 would be almost entirely driven by the large assumed increase in methane. Methane is barely mentioned in this paper, and all focus is placed on BVOCs. It seems unlikely that methane has ceased to be a major factor, and the authors need to discuss the impact of methane on future ozone increases.

2) The paper emphasizes the impact of stratospheric ozone recovery on future ozone increases, but doesn't provide any clear analysis to support this claim. While stratospheric ozone decreases in the mid-latitudes of the southern hemisphere are in the range of 5-17%, the reduction of stratospheric ozone in the northern hemisphere is quite small, and is less than 5%. Given that the recovery in the Northern Hemisphere will only result in a small increase in the transport of stratospheric ozone into the troposphere, the authors need to provide separate estimates of the impact of ozone recovery on the ozone burden in the Northern and Southern Hemispheres.

3) The model groups did not provide actual flux estimates of the contribution of strato- spheric ozone, and instead relied on the outdated and flawed method of estimating the flux based on the residual of the P, L and D terms. Estimates of the stratospheric contribution to the tropospheric ozone budget need to be calculated using a flux-based approach.
I elaborate on these issues in my detailed comments below. Once these issues have been addressed the paper would be acceptable for publication in ACP.

We thank the reviewer for their extensive and valuable comments.

Major Comments:

1) Elaborating on comment #1 above, it would really help if the authors provided a description of ssp370, with a focus on projected methane concentrations. The paper provides no information on this scenario, other than a brief statement in the Conclusions that it is a "middle of the road" pathway. I had to perform a google search, which led me to this paper: O'Neill, B. C., Tebaldi, C., van Vuuren, D. P., Eyring, V., Friedlingstein, P., Hurtt, G., Knutti, R., Kriegler, E., Lamarque, J.-F., Lowe, J., Meehl, G. A., Moss, R., Riahi, K., and Sanderson, B. M.: The Scenario Model Intercomparison Project (ScenarioMIP) for CMIP6, Geosci. Model Dev., 9, 3461–3482, https://doi.org/10.5194/gmd-9-3461-2016, 2016.
I assume ssp370 must be SSP3-7.0 in O'Neil et al.? According to O'Neil et al. this is a medium to high end scenario with radiative forcing of 7.0 W m-2. This description doesn't really fit with the statement in the Conclusions that this is a "middle of the road" pathway.

Done. We have added a section describing the SSP-3.70 and corrected the description to 'regional rivalry' and added that the 7.0 Wm-2 is at the high end of the CMIP6 pathways. Reviewer 1 makes a similar point.

> As we saw from the ACCMIP results, the factor associated with RCP8.5 that caused ozone to increase over the 21st century was methane. I assume this would also play an important role in the current analysis, but the authors provide no information on the expected methane concentrations; they just say that it increases monotonically. Please provide a description of the expected methane concentrations in ppbv, with a comparison to the current rate of increase, as observed by the NOAA net-work: https://www.esrl.noaa.gov/gmd/ccgg/trends_ch4/

We have added a panel to Figure 1 showing the CH4 global mean concentrations specified to be used to drive the Historical and SSP370 experiments. The historical CH4 concentrations are from (Meinshausen et al., 2019)

> Please also comment on the relative impact of methane and BVOCs on future ozone levels.

We agree that this is an important point. AerChemMIP contains within it a series of experiments designed to attribute the effect of methane and VOCs for both historical as well as SSP3-7.0 scenario, such as histSST-piCH4 and ssp370SST-lowCH4, which address methane's role specifically, and through the piClim experiments which quantify the impact of BVOC [Collins et al., 2016]. The **relative** impact is difficult to attribute in the context of the CMIP/ScenarioMIP experiments analysed here therefore. For this paper, we excluded AerChemMIP experiments for reasons of multi-model data availability. We anticipate that many follow-on papers will look at this question in more detail and thank the reviewer for the suggestion.

> On line 403 the authors attribute the ozone increase in the late 21st century to BVOCs. But based on the results of Young et al. [2013] one would assume that methane would be more important. If this is no longer the case, then the authors need to bring BVOCs to the forefront and state very clearly that BVOCs are expected to make a greater contribution to increasing ozone than methane.

This is a good point, and we have made the manuscript clearer to emphasise that the increases in the ozone burden in the late 21st century, when NOx and CO stabilise, can be driven by many factors such as methane, climate effects or increase in stratosphere-to-troposphere transport, and added a reference to who discuss the role of methane.

> 2) To provide some background information for my comments in #2 above ["While stratospheric ozone decreases in the mid-latitudes of the southern hemisphere are in the range of 5-17%, the reduction of stratospheric ozone in the northern hemisphere is quite small, and is less than 5%. Given that the recovery in the Northern Hemisphere will only result in a small increase in the transport of stratospheric ozone into the troposphere, the authors need to provide separate estimates of the impact of ozone recovery on the ozone burden in the Northern and Southern Hemispheres."]

We have added a section to the supplementary material showing the data the reviewer requests. It is not possible, without interhemispheric ozone mass flux diagnostic output, to say more about the impact of stratospheric ozone recovery on the hemispheres separately.

3) Elaboration on comment #3 above. In the Conclusions (line 554) the authors state: "We find that STE fluxes are similar among the models" However, the authors provide no quantitative support for this statement because they did not actually calculate the flux of ozone from the stratosphere to the troposphere. Even though each of these state-of-the- art models has a fully coupled stratosphere-troposphere circulation, and even though other recent studies have directly calculated the ozone flux, this study relies on the old, and error-prone, method of simply inferring the flux based on the residual of P, L and D. There are errors associated with P, L and D, and therefore if you rely on these terms to infer the flux from the stratosphere it will reflect all of these errors. An excellent example is the residual term of UKESM1 in Figure 13. The inferred flux from the stratosphere drops to zero in the year 2000, which means that either there is complete ozone depletion in the stratosphere, or there is a complete collapse of the Brewer Dobson circulation. We know that neither of these scenarios is possible, and therefore this inferred flux from the stratosphere is nothing more than errors associated with P, L and D. This study needs to abandon the inference method of estimating STE and use a flux-based method that calculates the net ozone flux across the tropopause, or across the 380 theta isotherm. The 380 isotherm flux method is convenient because any stratospheric ozone that descends from the "overworld" across this layer will eventually enter the troposphere [Holton et al., 1996; Appenzeller et al., 1996]. While there is a delay of several weeks from the time the ozone crosses the 380 isotherm until it crosses the tropopause, it's fine to use this method to calculate an annual average flux. Recent paper that use this method are Jaegle et al., 2017; Olsen et al., 2013; and Yang et al., 2016.

We agree with the reviewer that the residual terms in the ozone budget calculated here, particularly for UKESM1, require clarification. We also agree that a dynamically calculated flux would be a valuable complement, but we note that the CMIP6 data request does not permit the analysis that the reviewer requests, as mass fluxes were not archived.

The use of the residual method is well-established [Stevenson, 2006; Young, 2013; Archibald, 2020], has been shown to agree with dynamical STE calculations [Griffiths et al., 2020] and to be valuable when the global ozone tendency is small [Wu, 2017]. Online mass flux methods are not without their own flaws; e.g., they are prone to double-counting due to rapid changes in tropopause altitude relative to downwelling. Mass flux across a fixed isotherm protects against this, but misses photochemistry in the extratropical lowermost stratosphere. We therefore could not agree wholly with the suggestion to abandon the residual method, nor its association with STE, particularly in its qualitative behaviour over time to emphasise the importance of the stratospheric ozone recovery to the determination of future tropospheric ozone budget and burden. We agree that comparison of the residual method to a dynamical calculation would be valuable, but unfortunately, the diagnostics do not exist.

Nevertheless, to follow up the reviewer's suggestion, we have, where available, sourced directly from the modelling centres the STE fluxes calculated dynamically. The figure below shows ozone transport w.r.t. the WMO tropopause (the CMIP6 PTP variable). It can be seen that the figure shows

good agreement between the magnitude of this term as calculated in the GFDL-ESM4 and UKESM1 models, particularly in the period around 1850, of 300-400 Tg per year, consistent with earlier model estimates, and with the residual in this period, but that GISS-E21-G model, with its consistently higher tropopause, is higher by about a factor of four. In the present day, GFDL-ESM4 and UKESM1 diverge, largely as the result of higher ozone depletion calculated in UKESM1.

The figure, derived from data outside the CMIP6 data request, shows the difficulty in using a meteorological (WMO) tropopause for these flux calculations. This is for two reasons: firstly, the altitude of the WMO tropopause, being a thermal tropopause is difficult to define precisely when there are large regions where the lapse rate is essentially constant, and model vertical resolution tends to be low. In these regions, however, the ozone gradients are high, and so the ozone flux across the tropopause is particularly sensitive to the location of the tropopause.

The use of o3prod, o3loss and dryo3 avoids some of these issues. Given that the bulk of ozone production and loss occurs in the lower troposphere (Archibald and Elshorbany, 2020) the budget terms are less sensitive to the precise location of the tropopause, but there is a significant production of ozone in the UT from lightning NOx. The precise size of this term is sensitive to tropopause definition.

We note that the o3prod and o3loss terms used to calculate the residual include the majority (>90%), but not the total, ozone production and loss channels, and are missing potentially important odd-oxygen loss pathways. The low STE term for UKESM1 may be partly a result of this and the strong ozone depletion simulated in UKESM1 (Keeble et al., 2020; Morgenstern et al., 2020; Skeie et al., 2020) as well as circulation changes (Morgenstern et al., 2020). This is also seen in the supplementary figure, in which the dynamical ozone transport declines to a lower value for UKESM1, compared to GFDL-ESM4, reflecting the greater decrease in lower stratospheric ozone in UKESM1 across the period 1850-2014. As the reviewer notes, changes to circulation may be important, and will be addressed in a subsequent study (Zeng et al., in prep).

We have expanded the discussion of the influence of the stratospheric impact to include the references the reviewer suggests, and to discuss the pros and cons of the two methods, noting the absence of data for dynamical calculations, including a reference to the supplementary figure, and highlighting the potential impact of the missing channels in the o3prod and o3loss terms. We will include the figure below in a supplementary section.
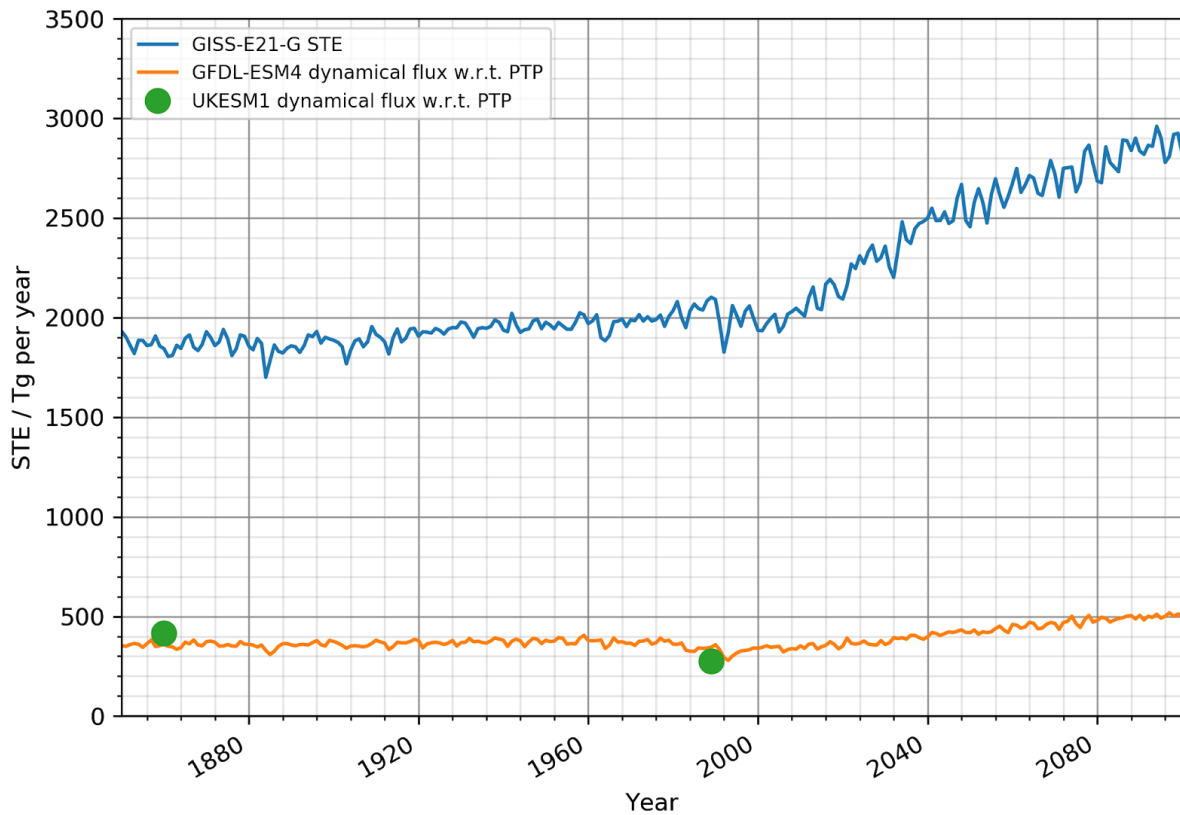
Line 20 Need to add uncertainty estimate to ozone RF: 0.4 +/- 0.2 W/m^2

DONE: uncertainty added.

Line 34 This statement needs to be reconsidered. Ozone's lifetime is very short and is mostly irrelevant to climate variability on interannual or decadal times scales (e.g. ENSO on a time scale of five years). The impact of climate variability is in relation to shifts in transport pathways and emissions. For example, in strong El Nino years there is increased biomass burning across Indonesia, which boosts ozone production in that region, while ozone decreases on the other side of the Pacific. This seesaw pattern has nothing to do with ozone lifetime and is a direct result of El Nino changing the distribution of ozone precursor emissions. Another way to think about it is in terms of isoprene, which only has a lifetime of a few hours. You can get large fluctuation in isoprene concentrations across the southeast USA just due to the impacts of the seasonal cycle and drought on emissions. You would get similar relative seasonal and interannual fluctuations if isoprene's lifetime was two weeks instead of a few hours.

We have amended the text to say that the ozone lifetime is sufficiently long to be transported over large distances, and that modes of climate variability may affect this transport.

Done. We have amended the manuscript to make clear that 338 +/-6 Tg refers to the available observations of the whole atmosphere burden derived from TOST and IASI data detailed in Gaudel et al. (2018) as the reviewer notes. This is necessarily a subset of the burdens derived from other satellite products.

Done. We have added a reference to this article.

Done. Amended to use the word 'species'

Done. We have added a table with these data.

Done. We have noted that these are 'remote' stations.

We have added this statement to the Data Availability statement. However, we note that the data used here is not the same as what is directly available from NOAA, as it has been corrected to account for changes in instrumentation over the historical record as described by Tarasick et al. (2019).

Figure 2. This is one of the most important figures in the paper, yet it is difficult to read because the panels are far, far too small. Please expand the figure so that it fills the width of the page.

Done. The PDF of the figure has been scaled to the page width.

Figure 4. This comparison should also include the NOAA site of American Samoa, in the marine boundary layer of the South Pacific (-14.2° S, 170.6° W, 42 m) which has continuous data from 1975 to 2015. The data are available here: ftp://aftp.cmdl.noaa.gov/data/ozwv/SurfaceOzone/

Done. We have added these data to the figure.

Lines 277-278 TOAR-Observations [Tarasick and Galbally et al., 2019] evaluated the historical ozone observations at South Pole (prior to 1974) and only included the 1961- 1963 data in their Table 5. The 1964-1966 and 1967-1973 data were not included, presumably because they were not considered to be as reliable. Here it seems that the 1964-1966 and 1967-1973 data were included, and that some type of correc- tion was applied. I don't see mention of these particular correction factors in TOAR- Observations, and they need to be described here.

Those data were not available at the time TOAR-Observations was written, but they have since been digitized by Sam Oltmans who provided them to Owen Cooper. The corrected Regener chemiluminescent automatic ozone analyser and the ECC analyser both show good agreement with the UV method (see TOAR-Observations, Table 2) and so do not warrant correction.

Line 290 This is the first time that Figure 3 is discussed, but it appears in the text after Figure 4. The numbers of these figures need to match their appearance in the text.

Done

Line 293 data were accessed

Done

Done. This statement has been changed. The reviewer is of course correct that the vast majority of these data are from ECC sondes; in fact all but one site (Hohenpeissenberg), which uses the Brewer-Mast sonde, some data from two Japanese sites, which switched to ECC after 2008, and a small amount of data from two Indian sites, which use a sonde similar to the Brewer-Mast. The Hohenpeissenberg data agree well with ECC data in recent decades, while the Japanese KC data measure a little low (2-6%)[TOAR-Observations]; the behaviour of the Indian sonde in recent decades is not well known.

Done. Have removed this sentence.

Done. Modified to 'high frequency'

Done. We now use CMIP6 standard colours for this plot. (https://github.com/IPCC-WG1/colormaps)

Amended

We agree with the reviewer that there is no shift in emissions from the SH tropics. Rather, we point out that the evolution of net chemical production in the background SH mid-latitudes reaches to its maximum in around 2000. We have rephrased this.

> Line 493 What does "shown in 18" mean? Figure 18?

Done. We apologise if this was not clear.

> Line 511 Misspelled: UEKSM1

Corrected. Thanks again.

> Figure 16 Why are these terms described as fluxes? Flux is the transport of mass across a unit area and will contain units of m-2, as shown by many examples here: https://en.wikipedia.org/wiki/Flux These terms are not fluxes and the y-axis label needs to be corrected. The caption of this figure also contains several typos.
> Line 538 It's not clear what is meant by "fluxes". Are you just talking about the deposition flux? The ozone production and loss terms should not be referred to as fluxes.

Done. We have adopted the term tendency to describe rates of change of quantities. "Fluxes" in Figures 13 and 15 have been replaced with "Tendency".

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