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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Tropospheric ozone in CMIP6 Simulations" *by* Paul T. Griffiths et al.

Anonymous Referee #2

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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groups did not provide actual flux estimates of the contribution of stratospheric ozone, and instead relied on the outdated and flawed method of estimating the flux based on the residual of the P, L and D terms.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.

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. 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 network: https://www.esrl.noaa.gov/gmd/ccgg/trends_ch4/ Please also comment on the relative impact of methane and BVOCs on future ozone levels. 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.

2) To provide some background information for my comments in #2 above, here are the latest numbers on observed stratospheric ozone depletion:

Here is the primary link to: "Scientific Assessment of Ozone Depletion: 2018" https://www.esrl.noaa.gov/csd/assessments/ozone/2018/

Here is the link to: "Twenty Questions and Answers About the Ozone Layer: 2018 Update" https://www.esrl.noaa.gov/csd/assessments/ozone/2018/downloads/twentyquestions.pdf On page 46, Figure Q12-1 shows the decrease of ozone in the stratosphere by latitude; the observed reduction in the N. Hemisphere is easily less than 5%.

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

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Discussion paper

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.

Appenzeller, C., Holton, J. R., & Rosenlof, K. H. (1996). Seasonal variation of mass transport across the tropopause. Journal of Geophysical Research: Atmospheres, 101(D10), 15071-15078.

Holton, J. R., Douglass, A. R., Haynes, P. H., McIntyre, M. E., Rood, R. B., and Pfister, L. (1996) Stratosphere-troposphere exchange, Rev. Geophys., 33, 403–439.

Jaeglé, L. et al (2017), Multiyear composite view of ozone enhancements and stratosphere-to-troposphere transport in dry intrusions of northern hemisphere extratropical cyclones. Journal of Geophysical Research, 122. https://doi.org/10.1002/2017JD027656

Olsen, M. A., Douglass, A. R., & Kaplan, T. B. (2013). Variability of extratropical ozone stratosphere–troposphere exchange using microwave limb sounder observations. Journal of Geophysical Research: Atmospheres, 118, 1090–1099. https://doi.org/10.1029/2012JD018465

Yang, H. et al [2016], Quantifying isentropic stratosphere-troposphere exchange of ozone, J. Geophys. Res. Atmos., 121, 3372–3387; doi:10.1002/2015JD024180

Minor Comments:

Line 20 Need to add uncertainty estimate to ozone RF: 0.4 +/- 0.2 W/mˆ2

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.

Line 42 Here the authors state: "Multiple satellite products corroborated by the global ozonesonde network indicate a present-day (2010-2014) tropospheric ozone burden of 338$\pm$6 Tg in broad agreement with the current range of model estimates (Gaudel et al., 2018)." Where did the estimate of 338$\pm$6 Tg come from? All of the satellite estimates of the tropospheric ozone burden in Gaudel et al. are listed in their Table 5, but this number does not appear in the table. Did the authors take the 3 values (TOST, IASI-FORLI and IASI-SOFRID) from the 2010-2014 column and produce their own range? If so then they need to specify that it relies on just the IASI and TOST (ozonesonde) products.

Lines 85-104 There is some good discussion here regarding the impact of changes in the BDC on tropospheric ozone. The authors should also consider the following paper that is the first to establish a link between the expanding Hadley circulation and observed changes in tropospheric ozone across southern mid- and high latitudes.

Lu, X., Zhang, L., Zhao, Y., Jacob, D.J., Hu, Y., Hu, L., Gao, M., Liu, X., Petropavlovskikh, I., McClure-Begley, A. and Querel, R., 2019. Surface and tropospheric ozone trends in the Southern Hemisphere since 1990: possible linkages to poleward expansion of the Hadley Circulation. Science Bulletin, 64(6), pp.400-409. http://refhub.elsevier.com/S2095-9273(19)30104-5/h0040

Line 126-127 What is meant by "transported" vs. "non-transported" chemical tracers? Aren't all tracers transported by the model winds?

Line 123 Four models are described, but the basic information on grid resolution and number of vertical layers is only provided for the GISS model. Please provide this information for all four models.

Line 259 Here you should specify that these sites are remote, as there are some urban and rural sites (such as Hohenpeissenberg, Germany, and Whiteface Mountain, New York) that have data since the early or mid-1970s.

Lines 269 and 275 The ultimate source of the surface ozone data is the NOAA Global Modelling Division, and credit should not be given to the person who processed the data (instead mention colleagues who processed data in the acknowledgements). So that the reader can find these data, the following URL needs to be listed in the Data Availability Statement: ftp://aftp.cmdl.noaa.gov/data/ozwv/SurfaceOzone/

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.

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/

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 correction was applied. I don't see mention of these particular correction factors in TOAR-Observations, and they need to be described here.

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.

Line 293 data were accessed

Line 293-295 Here the authors state: "A total of 23,392 profiles using Carbon-Iodine (Komhyr, 1969), ECC (Komhyr, 1971), and Brewer-Mast (Brewer and Milford, 1960) sondes from 82 sites world-wide were aggregated over the period 2005-2014." The great majority of the ozone profiles are made using the modern ECC method, rather than the much older carbon-iodide and Brewer Mast methods. TOAR-Observations shows that there are some biases between these methods. Please provide some numbers to indicate the percent of profiles made with the more reliable ECC method.

line 301 There seems to be a word missing after southern hemispheric: "Note that the northern hemispheric overestimate and southern hemispheric seen at the surface. . ."

Line 311 It's an overstatement to say that satellites provide daily near-global ozone observations. Their orbits don't even provide daily coverage in the tropics, and they can't see through cloud. For global coverage you basically need to build a monthly composite.

Figure 8 The caption says there is a dark blue line in the figure, but not to my eye. I see light blue (CESM2-WACCM), regular blue (MMM) and gray (is this GFDL-ESM4??).

Line 398 I get an increase of 25%, not 20%, as follows: 100*(350-280)/280 = 25%

Line 463 It's not clear which latitude band of the SH you are referring to when you say that ozone destruction reaches a minimum around the year 2000. Are you talking about 40 degrees south? If so, Zhang et al. did not show a shift in emissions from the SH tropics, southward to the SH mid-latitudes. Their Figure 1 in their supplement shows a broad increase of emissions from the equator to 30 or 40 degrees south. In other words, there is not a decrease in the tropics that is balanced by an increase at mid-latitudes (i.e. a shift from one latitude band to another). The latitudinal shift in emissions in Zhang et al. occurred in the NH.

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

Line 511 Misspelled: UEKSM1

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.

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