Author's response to community comments on "Drivers of change in Peak Season Surface Ozone Concentrations and Impacts on Human Health over the Historical Period (1850-2014)"

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We would like to thank the TOAR-II community for their helpful and constructive comments on the manuscript. Below we have responded to each comment in turn and made alterations to the manuscript where appropriate (shown enclosed in "*speech marks and italic font*" and any deletions from the manuscript shown with a strikethrough "~~example~~"). The referee comments are shown first in grey shading and the author's response is shown below in normal font.

This review is by Owen Cooper, TOAR Scientific Coordinator of the TOAR-II Community Special Issue. I, or a member of the TOAR-II Steering Committee, will post comments on all papers submitted to the TOAR-II Community Special Issue, which is an inter-journal special issue accommodating submissions to six Copernicus journals: ACP (lead journal), AMT, GMD, ESSD, ASCMO and BG. The primary purpose of these reviews is to identify any discrepancies across the TOAR-II submissions, and to allow the author teams time to address the discrepancies. Additional comments may be included with the reviews. While O. Cooper and members of the TOAR Steering Committee may post open comments on papers submitted to the TOAR-II Community Special Issue, they are not involved with the decision to accept or reject a paper for publication, which is entirely handled by the journal's editorial team.

Comments regarding TOAR-II guidelines:

TOAR-II has produced two guidance documents to help authors develop their manuscripts so that results can be consistently compared across the wide range of studies that will be written for the TOAR-II Community Special Issue. Both guidance documents can be found on the TOAR-II webpage: https://igacproject.org/activities/TOAR/TOAR-II

The TOAR-II Community Special Issue Guidelines: In the spirit of collaboration and to allow TOAR-II findings to be directly comparable across publications, the TOAR-II Steering Committee has issued this set of guidelines regarding style, units, plotting scales, regional and tropospheric column comparisons, and tropopause definitions.

The TOAR-II Recommendations for Statistical Analyses: The aim of this guidance note is to provide recommendations on best statistical practices and to ensure consistent communication of statistical analysis and associated uncertainty across TOAR publications. The scope includes approaches for reporting trends, a discussion of strengths and weaknesses of commonly used techniques, and calibrated language for the communication of uncertainty. Table 3 of the TOAR-II statistical guidelines provides calibrated language for describing trends and uncertainty, similar to the approach of IPCC, which allows trends to be discussed without having to use the problematic expression, "statistically significant".

General comments:

Lines 52-54

This discussion seems to be referring to the suggestion that surface ozone at northern mid-latitudes might have doubled from the 1950s to the year 2000, and that this rapid increase is not reproduced by the models. However, the idea that ozone doubled is now out of date. In the first phase of the Tropospheric Ozone Assessment Report, Tarasick et al. (2019) conducted the most comprehensive assessment of historical ozone observations, digging up old and forgotten datasets missed by other studies. Tarasick et al. (2019) did not find evidence for a doubling of surface ozone, and concluded that ozone increased by roughly 30-70% from the mid-20th Century until the early 21st Century. These findings were accepted by IPCC AR6 (Gulev et al., 2021), and the CMIP6 models generally agreed with this rate of increase (Griffiths et al., 2021).

Thanks for the comment on this. However, we specifically refer to the Tarasick et al., (2019) paper and the 30-70% increase in surface ozone over the second half of the 20^{th} Century and into the 21^{st} Century on lines 42 to 43 of the introduction section. We specifically mention that global model simulations tend to agree with this observed range, although might slightly overpredict it across some regions. We however do mention a doubling of surface ozone concentrations and we have reworded this in the abstract and results section to be line with previous studies.

We have also included an additional reference to the Tarasick et al., (2019) results on line 237 as follows:

"*In addition, this simulated historical change in global OSDMA8 is within the observed change of surface ozone across the northern Hemisphere of 30 to 70 % (approximately 10 to 16 ppb) from Tarasick et al., (2019).*"

Line 71

A more recent analysis of the ozone climate penalty is provided by Zanis et al., 2022.

We have Included a reference to the Zanis et al., (2022) paper in the sentence that ends on line 71.

Line 194

The cold bias in UKESM is mentioned here, but no explanation is given. Could this be related to higher aerosol loading in this model?

We have included an additional sentence on line 195 to comment on this as follows:

"*The cold temperature biases simulated by UKESM1-0-LL have been previously attributed to an excessive aerosol forcing, with recent changes to aerosol and cloud properties in an updated version (UKESM1-1-LL) showing an improved representation of historical surface temperatures (Zhang et al., 2021; Mulcahy et al., 2023).*"

Zhang, J., Furtado, K., Turnock, S. T., Mulcahy, J. P., Wilcox, L. J., Booth, B. B., Sexton, D., Wu, T., Zhang, F., and Liu, Q.: The role of anthropogenic aerosols in the anomalous cooling from 1960 to 1990 in the CMIP6 Earth system models, Atmos. Chem. Phys., 21, 18609–18627, https://doi.org/10.5194/acp-21-18609-2021, 2021.

Mulcahy, J. P., Jones, C. G., Rumbold, S. T., Kuhlbrodt, T., Dittus, A. J., Blockley, E. W., Yool, A., Walton, J., Hardacre, C., Andrews, T., Bodas-Salcedo, A., Stringer, M., de Mora, L., Harris, P., Hill, R., Kelley, D., Robertson, E., and Tang, Y.: UKESM1.1: development and evaluation of an updated configuration of the UK

Earth System Model, Geosci. Model Dev., 16, 1569–1600, https://doi.org/10.5194/gmd-16-1569-2023, 2023.

Line 236

I realize that the output is focusing on 10-year periods, but the year 1855 doesn't really coincide with 1850, as listed on line 227.

The population data is only available at the start of each decade in every 10-year period before 2000, which doesn't coincide with the middle of the 10-year mean period that we have used as output from the model. Therefore, we have picked the population at the start of each decadal time-period to be used as representative of the whole 10-year time period.

We have amended the sentences on line 226 to 229 of the manuscript as follows to mention this point:

"*An assessment of population exposure to ozone is also provided by calculating the number of people exposed to OSDMA8 concentrations above the TMREL in 3 different decadal time periods (1850-59, 1980-89 and 2005-14 ~~2010~~). Population data for these periods is obtained for the nearest available years (1850, 1980, 2010) from the Hyde (History database of the Global Environment) dataset (Klein Goldewijk et al., 2017).*"

Line 242

Here it is stated that ozone more than doubled in most regions from 1950 to 2010, but from my reading of Figure 3, the increases were not this great. As the plots are small, it is not easy to read the numbers exactly, but ozone in South Asia seems to increase from about 30 ppb to nearly 60 ppb, so for this region one could argue for a doubling, but not for the other regions. For example, it seems that ozone in Western Europe increased from 30 ppb to about 44 ppb (an increase of about 50%) from 1950 to 2010, with a similar increase in High-income North America. These 50% increases fall within the 30-70% increase reported by Tarasick et al. (2019), based on historical observation at northern mid-latitudes.

Thanks for the comment on this and line 242 to 245 has been amended to:

"*Across most regions OSDMA8 concentrations only show small increases up until about 1950. After this time period concentrations rapidly increase (~~more than doubling~~ by approximately 50%) to 2005-2014 values, driven by the large rapid changes in all anthropogenic precursors of ozone (CH4, NOx, CO, Non-CH4 VOCs), with the largest relative changes occurring in NOx emissions (Fig. 2).*"

Figure 3 shows that the highest ozone mixing ratios in 1850 occurred in Central Sub-Saharan Africa, and Figure 5 shows that the highest attributable fraction occurred in the same region. What drove ozone production in this region in 1850? Biomass burning?

Indeed, ozone production in 1850 is probably likely due to the presence of elevated biomass burning and natural emission sources of ozone precursors (NOx and VOCs) within the model in this time period over this part of Africa.

Figure 4

The concept of the ozone climate penalty indicates that ozone concentrations in high emission regions should increase (if emissions are held constant) due to the increase in heatwave conditions (across highly populated areas) that are more conducive to ozone formation. However, the histpiSST simulation shows that ozone exposure increased slightly when the climate was fixed at 1850. One would think that ozone exposure would decrease if the climate was fixed at the colder 1850 conditions (i.e. less heatwaves in highly populated areas). What is the explanation for the increase?

The comment is correct in that the climate change penalty in figure 4 (comparing the difference of histSST and histpiSST) shows a small reduction in global population exposure to ozone concentrations above the theoretical minimum risk exposure level (32.4ppb). The comparison in Figure 4 does not reflect the actual change in ozone concentrations across regions but rather shows if there is a change in exposure to elevated ozone concentrations which could be harmful to human health. For example, across regions like south and east Asia the regional ozone concentrations remain above 32.4 ppb in both histSST and histpiSST and as such there is no change in population exposure above 32.4 ppb. Figure 4 is meant to highlight if there has been a shift in the population exposure to ozone concentrations in exceedance of 32.4 ppb (and therefore elevated risk to human health) because of the changes in different ozone drivers. Figure 6 and Figure A4 of the manuscript show the actual change in surface ozone due to climate change and highlight the increase in ozone concentrations across continental areas, including populated regions of north America, Europe and Asia.

Line 285

Unites should be United

Changed

Line 290

You can't really say that there was no risk globally in 1850, as ozone values in Central Sub-Saharan Africa were above 32.4 ppb.

Thanks for the comment. A new sentence on line 290 has been included to reflect the elevated concentrations in Central sub-Saharan Africa and non-zero risk to human health in 1850.

"*The major exception to this is that AF values are approximately 5% in the 1850-59 period over the Central Sub-Saharan Africa region. This non-zero AF is due to the elevated pre-industrial OSDMA8 concentrations (Fig 3) arising mainly from biomass burning sources of ozone precursors over this region in this period, although the low population density in this region and time period would represent a low risk to health.*"

Line 355

Here it would be helpful to indicate the magnitude of the future temperature changes that produced the notable ozone climate penalty reported by Zanis et al. 2022.

The range of future temperature change in the study of Zanis (2.7 to 5.4°C by 2100) has now been included in line 355 as follows:

"*The magnitude of the changes in ozone due to 1K of warming over the historical period are consistent with studies analysing the change of ozone in response to future warming (Archibald et al., 2020c; Zanis et al., 2022), with the impact of climate change on surface ozone being less in this study due to the smaller level of historical warming compared to those projected for the future warming of 2.7 to 5.4K.*"

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