Community Comments, CC1: 'Comment on egusphere-2024-720', Owen Cooper, 02 May 2024

Major Comments:

1) As stated on the first line of the abstract, methane is an important ozone precursor, but this paper does not address the distribution and trends of methane. Why has methane been omitted? Methane should be addressed as studies have shown its impact on recent ozone increases (Zhang et al., 2016), and as shown in Chapter 6 of IPCC AR6 (Szopa et al., 2021), the only future scenario with an increasing tropospheric ozone burden is SSP3-7.0, which is driven by increasing methane. NOAA GML observations of methane (https://gml.noaa.gov/ccgg/trends_ch4/) show that methane concentrations in the atmosphere have increased sharply since 2005 (an 8% increase from 2005 to 2023).

Answer: While oxidation of methane and NMHCs was mentioned in the introductory statement in the article, it was not investigated in this study since we focused in this article more on the reactive species, NO2, CH2O, and CO. Methane is an important precursor and we also think future assessments should focus on its analysis to include methane and other precursors.

We have included the following paragraph in section 3.2 to address methane contribution to tropospheric ozone: While this paper focuses on ozone precursors with higher reactivity, we note that methane, with an assessed total atmospheric lifetime of 9.1 ± 0.9 years (Szopa et al., 2021), is also a crucial driver (Fiore et al., 2002; Isaksen et al., 2014), given its accelerated growing rate of 7.6 ± 2.7 nmol mol^{-1} yr^{-1} between 2010 and 2019 (Canadell et al., 2021), largely driven by anthropogenic activities (Szopa et al., 2021).

2) Lines 66-68 When summarizing global tropospheric ozone trends, the best reference is Section 2.2.5.3 in Chapter 2 of IPCC AR6 (Gulev et al., 2021). While observations in the Southern Hemisphere are limited compared to the northern hemisphere, the available in situ and satellite observations do indicate an increase of ozone since the late 20th century: "Observations in the SH are limited, but indicate average tropospheric column ozone increases of 2–12% (1–5 ppbv) per decade in the tropics (Figure 2.8c), and weak tropospheric column ozone increases (<5%, <1 ppbv per decade) at mid-latitudes (Cooper et al., 2020). Above Antarctica, mid-tropospheric ozone has increased since the late 20th century (Oltmans et al., 2013)."

Answer: We thank Owen for this comment and we have updated this sentence as follows: "…..radiative forcing of $(0.47^{+0.23}_{-0.23})$ W m^{-2}; Forster et al., 2021). Since the mid-1990s, free tropospheric ozone trends based on in situ measurement and satellite retrievals have increased with high confidence by 1-4 nmol mol^{-1} $decade^{-1}$ across the northern mid-latitudes and 1-5 nmol mol^{-1} $decade^{-1}$ within the tropics (Guleb et al., 2021). In the Southern Hemisphere, with more limited observation coverage compared with the Northern Hemisphere, the tropospheric column ozone shows an increase since the mid-1990s by less than 1 nmol mol^{-1} $decade^{-1}$ with *medium confidence* at southern mid-latitudes (Gulev et al., 2021, Cooper at al., 2020). Tropospheric O_3 short- and long-term..".

3) As stated in Section 5 of the 'Guidance note on best statistical for TOAR analyses': "One of the most critical components of statistical analysis is to acknowledge the uncertainty. Every estimation must be accompanied by a quantification of the associated uncertainty (or error bar), which is used to assess the reliability of the (trend) estimate and is considered to be as equally important as the estimate". According to the guidance note, all trends need to be reported with the 95% confidence intervals and p-values. Basically, a trend value without an uncertainty

estimate is meaningless. For example, on lines 463-466 model trends are compared to OMI/MLS trends. But because the model trends have no uncertainty range, the trend value is meaningless and no conclusions can be drawn from this comparison.

Answer: We added error bars to the model trends in Figure 11 and clarified the model trend discussion. All trends are now shown with their corresponding error.

4) Another important piece of advice from the 'Guidance note on best statistical for TOAR analyses' is that all TOAR analyses should abandon the use of the phases "statistically insignificant" or "statistically significant". Compelling arguments for this policy are provided by the highly influential paper by Wasserstein et al., 2019. The submitted paper has many instances of the phrases "statistically insignificant" or "statistically significant". These phrases need to be removed, and they can be replaced by statements from the authors regarding their confidence in the reported trend values. Advice is given in the Guidance Note, and this advice can also be applied to figures such as Figure 13.

Answer: We appreciate the advice, and we will certainly consider in future submissions.

5) Section 3.5 I found the section on LNOx to be too long and it lacks clear statements on lightning trends. While the section cited previous work that thunderstorm days have increased in some regions (and decreased in others), no number were given, so it's not clear by how much thunderstorm days have increased. In terms of flash rate, some regions showed increases and some showed decreases, but there was no summary statement that lets the reader know if lightning has clearly increased or decreased on the global scale. Line 714 states that lightning contributes to positive ozone radiative forcing, but it's not clear to me that this is really the case. In the UT ozone has a strong longwave radiative effect (i.e. it absorbs outgoing longwave radiation) and of course LNOx can affect ozone in the UT and therefore affect ozone's longwave

radiative effect. But are there any studies that have shown that LNOx impacts ozone's radiative forcing (as opposed to ozone's longwave radiative effect)? IPCC defines radiative forcing as the change in the Earth's radiative balance since 1750. If lightning is impacting radiative forcing then there must be conclusive evidence that lightning frequency has increased on the global scale. If there is no clear evidence for a global increase (or decrease) of lightning then the link to radiative forcing cannot be established.

Answer: Section 3.5 has been shortened and a table has been removed. Section 3.5.1 now has a rough estimate of the global trend in thunder days, and makes it clear that no long-term trend in global flash rates has been detected. All mentions of "radiative forcing" have been removed, and instead, the terminology, "effects by ozone on longwave radiation absorption" is now used.

6) lines 391-393 Regarding the number of ozone profiles required to accurately detect a trend, several studies over the years have shown that once-per-week sampling is often inadequate for accurate trend detection. A paper recently accepted for publication in the TOAR-II Community Special Issue (Chang et al., 2024) addresses this issue, and the paper's conclusions need to be considered when interpreting ozone trends based on sparse sampling.

Answer: We acknowledge this limitation, and we further clarified this aspect in the text including two references (Chang et al., 2020 and Chang et al., 2024). However, as mentioned by Chang et al (2024) only 3 European stations have achieved such high sampling frequency (Hohenpeissenberg, Germany; Payerne, Switzerland, and Uccle, Belgium). In the meantime, despite the lower sampling frequency at the rest of the global stations, ozonesonde observations continue to be the gold standard against which satellite data are validated. Furthermore, ozonesonde climatologies are critical to provide feedback to satellite observations for which they continue to be used with this long-term purpose. Thus, with proper acknowledgment of the frequency limitation, we believe that ozonesonde trends published in previously peer-reviewed studies are valuable information for the ozone community.

7) Line 362 Here it is claimed that the pandemic period led to increases in emissions and therefore an increase in the ozone rate of change, but no convincing references are provided to support this claim. The paper by Oleribe et al. 2021 has nothing to do with atmospheric chemistry, and the paper by Matandirotya et al., 2023 only looks at 3 cities in South Africa. This statement seems like speculation and it should be removed.

Answer: It is not mentioned in any part of the article that "the pandemic period led to increases in emissions and therefore an increase in the ozone rate of change"

We mentioned that the higher O_3 trends in the southern hemisphere is due to the lesser impact of the pandemic and we cited Oleribe et al. (2021) since they explained "why Sub-Saharan Africa Experienced a less severe COVID-19 Pandemic in 2020". Similarly, Matandirotya et al., 2023 assessed the NO_2 atmospheric air pollution over three cities in South Africa during 2020 COVID-19 pandemic, and both articles cover important regions in the southern hemisphere. However, we agree that these references do not provide concrete evidence as to why O_3 trends in the southern hemisphere were higher, therefore we remove this sentence as suggested.

8) Section 3.4.2 This section seems to only review ozone trends from previous studies by Wang et al., 2022 and by Christiansen et al., 2022. Does this paper actually calculate updated trends from available ozonesonde records? Section 2.2.2. in the Methods section lists ozonesondes as a data source, but I see no new data analysis.

Answer: Figure 9 was prepared by the authors for this paper, using trends from Wang et al 2022. As pointed out, this portion of the manuscript is based on a review of global ozonesonde trends calculated and published in previous studies (Wang et al., 2022 and by Christiansen et al., 2022). This is indicated in the caption of Figure 9. However, we added a sentence at the bottom of the corresponding methods section for further clarification.

Minor Comments:

line 66 The stated radiative forcing for ozone (0.34) is incorrect. As reported in Section 7.3.2.5 of Chapter 7 of IPCC AR6 (Forster et al., 2021), ozone has an assessed effective radiative forcing of 0.47 [0.24 to 0.70] W m–2.

Answer: The numbers we showed are correct for the global average Radiative Forcing (RF) of ($0.34^{+0.09}_{-0.06}$ W m^{-2}; IPCC, 2023) for clear sky conditions based on the *Ramaswamy et al., 2018 (IPCC AR6)*. Page 363 of the IPCC AR6 report states that "For total sky conditions, the range in globally and annual averaged tropospheric O3 forcing from all of these models is from 0.28 to 0.43 Wm−2,… The tropospheric O3 forcing constrained by the observational climatology is 0.32 Wm−2 for globally averaged, total sky conditions."

The numbers in the comment are for Effective Radiative Forcings (ERF), which include all tropospheric and land surface adjustments, particularly aerosol-cloud interactions to aerosol forcing, which is highly uncertain itself (Smith et al., 2020, https://doi.org/10.5194/acp-20-9591-2020). Both numbers are correct but reflect different things and are nevertheless very close (within the uncertainty range). However, since the reference Forster et al., 2021 is newer and include the latest increase of GHG as well as improved cloud parameterization for RF, we will update our number to that reference.

Line 76 More context needs to be given regarding the ozone increase of 40 ppb. Is this at the surface or in the free troposphere? Over which continent? If stating the extreme ozone increase, it would help to also provide the average ozone increase. A useful number is the approximate 45% increase in the tropospheric ozone burden. A recent paper published in ACP (Nussbaumer et al., 2023) is highly relevant to this submission and some discussion of their conclusions is warranted.

Answer: The sentence has been updated as follows: CMIP6 models simulate large increasing trends of surface concentrations of O_3 and $PM_{2.5}$ in East and South Asia with an annual mean increase of up to 40 ppb and 12 μgm^{-3}, respectively, over the historical periods (1850-2014; Turnock et al., 2020). The reference is useful, and we have cited it properly.

Line 350 When discussing the impact of COVID-19 on tropospheric ozone there are some key papers that should be cited: Steinbrecht et al., 2021; Chang et al., 2022; Putero et al., 2023

Answer: References are cited now.

Line 400 Very strong ozone trends above Japan since 2010 was not a major conclusion of Christiansen et al. (2022). They only show the higher ozone values after 2010 in the supplement, and they recommend that these data sets be treated with caution because the instruments changed from carbon-iodide to ECC after 2010; these time series have not been homogenized to correct for the change in instruments.

Answer: We updated the sentence to "For example, ozone in East Asia (Japan) has been increasing at a cautious rate of 3.5 to 5 ppbv/decade, particularly since 2010 (Christiansen et al., 2022)."