Comments by Owen R. Cooper (TOAR Scientific Coordinator of the Community Special Issue) on:

Tropospheric Ozone Precursors: Global and Regional Distributions, Trends and Variability

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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.

General Comments:

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, tropopause definitions and best statistical practices.

Guidance note on best statistical for TOAR 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".

<u>Recommendation</u>: Major revision

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).

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)."

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.

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.

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.

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.

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.

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.

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.

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.

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

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.

Figure 6
The color table is not aligned correctly with the ozone trend values. Weak positive trends of 0.0 to 0.5 DU per decade are shaded light blue instead of light orange. This makes negative trends appear to be far more prominent in the maps than they truly are. Please correct the color table.

Line 326
Are the small changes in tropopause height enough to have an impact on ozone trends? According to Figure 3 the tropopause height at northern mid-latitudes has increased at a rate of about 30 m per decade. Over the period 2004-2021 (the OMI/MLS record), this equals a total increase of the tropopause height of 51 meters. Assuming a typical tropopause height of 12,000 m, this means that the depth of the troposphere has increased by less than one half of one percent. I'm skeptical that this very small change has a meaningful impact on the ozone trend over such a short period.

Line 343
Here it is claimed that positive ozone trends are driven by oceanic emissions. Oceanic emissions of what? Please provide references that demonstrate that this is a known and valid explanation.

Figure 14
Please correct "United Utates"
Also, please remove the trend lines that extend beyond the bounds of the SCIAMACHY record.

Lines 355-357
"The trends during the time period (2005-2021) show a significant decline in O3 column trends in the northern hemisphere but a slightly increasing trend in the southern hemisphere."
This sentence is not phrased well. I think you are trying to make the point that when the years 2020-2021 are added to 2005-2019, then there is a change in the ozone rate of change. Please rephrase so that this point is clear.

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