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

An improved Trajectory-mapped Ozonesonde dataset for the Stratosphere and Troposphere (TOST): update, validation and applications

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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-II 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 practices 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".

Specific Comments:

A very important topic regarding detection of ozone trends in the troposphere is sampling frequency. Papers going back to the late 1980s have shown that low sampling frequencies (e.g. once per week ozone profiles) often fail to provide accurate monthly mean ozone values or reliable trends (Prinn, 1988; Logan, 1999; Cooper et al., 2010; Saunois et al., 2012; Chang et al., 2020; Chang et al., 2024). The modelling community is aware of this challenge (Lin et al., 2015; Barnes et al., 2016; Fiore et al., 2022) and they need long-term ozone observations with high sampling frequencies (greater than 3 times per week, if possible). The TOST product can help as it basically merges ozone observations on the regional scale, according to transport pathways, rather than through simple averaging across a pre-defined region. It would be very helpful to the modelling community if you could create a map that indicates the regions with the highest sampling frequencies, for example, areas with three or more observations per week, and regions with 5 or more observations per week.

The panels in Figure 9 are similar to my suggestion, but I'm not sure how to interpret these plots. For example, Figure 9e shows a dark green square over Hilo, Hawaii, which seems to indicate more than 180 samples for the month of January during 2000-2009. If I divide 180 by 10 years, then I get 18 ozone samples in a month, or a sampling frequency of more than 4 times per week. Sondes are only launched from Hilo once per week, so the other samples must be due to observations associated with the forward and backward trajectories. Given that Hilo is in the middle of the Pacific Ocean, it is probably more than 4 days of transport time from the nearest ozonesonde site, and therefore any trajectory in the 5x5 grid cell above Hilo must be associated with a Hilo ozonesonde. If this is the case, then the samples in the 5x5 grid cell are not independent. The algorithm must be counting the same observation several times while the trajectory slowly traverses the 5x5 grid cell.

Is there a way for you to determine the number of independent ozone values in a 5x5 grid cell? For example, can a forward or backward trajectory from a single ozonesonde only be counted once if it falls within a particular grid cell? If you can then make a plot showing the number of independent observations within a grid cell, then it is easier to relate TOST to a sampling strategy of 1, 3 or 5 profiles per week.

Lines 44-51

This introductory paragraph focuses on stratospheric ozone, while the topic of the TOAR-II Community Special Issue is tropospheric ozone. It's fine to discuss stratospheric ozone, as it impacts the troposphere, but a brief summary of the importance of tropospheric ozone is needed, especially when stating the importance of ozone for climate, as most of the radiative forcing is in the troposphere. See Chapters 2, 6 and 7 of IPCC AR6 (Gulev et al., 2021; Szopa et al., 2021; Forster et al., 2021).

When presenting the findings from the updated TOST product the focus is on the stratosphere and there is no analysis regarding tropospheric trends. The TOST product was used by the first phase of TOAR to show that ozone has increased in both hemispheres from 1998 to 2012 (Gaudel et al. 2018). TOST was also used by IPCC AR6, and the 1998-2012 positive ozone trends are consistent with the IAGOS trends over a slightly longer period (1994-2016), as shown in Figure 2.8 below (Szopa et al., 2021). It would be helpful to provide updated tropospheric ozone trends based on TOST-v2. It would also be helpful to show the extent of the negative ozone anomalies in 2020 caused by the COVID-19 economic downturn, as previously reported by Steinbrecht et al. 2021 and Putero et al., 2023 (published in the TOAR-II Community Special Issue).

Line 26

When saying the dataset has been updated to the most recent decade (1970s-2010s) it gives the impression that the final year in the dataset is 2019, but the final year is actually 2021. Please just list the full range of the dataset using the first and final years.

Lines 52-60

When reviewing the availability of long-term ozone profile records, please also mention lidar records. The lidar record at Observatoire de Haute Provence in southeastern France began in 1991; while the annual ozone anomalies from the lidar and the co-located ozonesondes differ due to sampling differences, both show a similar long-term ozone increase in the free troposphere, in the range of 1-3 ppbv/decade for 1991-2020 (Ancellet et al., 2022). Similarly, the JPL Table Mountain lidar north of Los Angeles shows an increase of 1 ppbv/decade for 2000-2023, as shown in the updated figure below (produced by Kai-Lan Chang using the method described by Chang et al., 2023). Since 2018 the Table Mountain lidar has a very high sampling frequency of 4-5 times per week. It also shows the decrease in ozone levels in 2020, associated with the COVID-19 pandemic.

Line 61

In addition to providing ozone retrievals for the stratosphere and troposphere, satellites also provide total column ozone retrievals.

Line 62

Satellite products can provide ozone retrievals for the lower, mid- and upper troposphere, with varying degrees of sensitivity, not just for the 6-10 km range (see section 3.3 of Gaudel et al., 2018)

Lines 68-72

A new area of global modelling involves the production of chemical reanalyses, which assimilate satellite data, to improve the quantification of tropospheric ozone, e.g. Miyazaki et al., 2020a,b; Colombi et al., 2021.

Line 130

The Data and Methods section needs to state how the tropopause is defined, as the product is provided in terms of both the troposphere and stratosphere. If a forward or backward trajectory begins in the troposphere and the final location of the trajectory particle, after 4 days, is above the tropopause, is this ozone observation categorized as being in the troposphere, or stratosphere?

Line 249-251

How were the IAGOS data averaged temporally? Into monthly means? What is the horizontal resolution? 5x5 degrees? How many airports were used? Did you use just the vertical profiles, or also the cruise level data? Do you have a data availability threshold? For example, do you require at least 4 profiles in a month to produce a monthly mean?

Line 540

The *Guidance note on best statistical practices for TOAR analyses* (described above) asks for all trends to be reported with 95% confidence intervals and p-values, and in units of ppbv decade⁻¹. In the submitted manuscript trends are only reported for the stratosphere and in units of ppbv year⁻¹. If ppbv year⁻¹ is the standard unit for reporting ozone trends in the stratosphere, then please retain this unit, otherwise please follow the TOAR guideline.

Figure S3

Why compare IAGOS and TOST over the range 50-150 ppb which includes tropospheric and stratospheric samples? If a monthly mean value for IAGOS observations is 100 ppb then it is very likely composed of both tropospheric samples (less than 100 ppb) and stratospheric samples (greater than 100 ppb). According to Figure S3d an IAGOS monthly mean of 100 ppb can correspond to a TOST value anywhere from 50 ppbv (mostly tropospheric samples) to 150 ppbv (mostly stratospheric samples). Clearly these two data sets are not sampling the same air masses and this is not an apples-to-apples comparison, so I don't see the value in these correlation plots.

Section 4.2

Every year, stratospheric ozone trends and variability are updated in the Global Climate chapter of the State of the Climate reports. The most recent edition (Dunn et al., 2023) provides an update through the end of 2022. In particular, Figure 2.64 compares several products and shows stratospheric ozone levels at 22 km for the latitude band 35N-60N, similar to your Figure 13. The SWOOSH product (Davis et al., 2016) is a combined satellite product, bias corrected against ozonesonde observations and provides global coverage. How does TOST compare to these other products, and does TOST provide any new information?



Curtain plot of the Table Mountain lidar record, 2000-2023 (data provided by Thierry Leblanc, JPL; method described by Chang et al., 2023). The decrease in 2020 has been attributed to the impact of the COVID-19 economic downturn (Steinbrecht et al., 2021).

(a) Surface and lower troposphere (b) Mid- and upper troposphere (c) Tropospheric column average TOST ozonesondes: 1998-2012 90 **o** low elevation IAGOS aircraft, mid-trop., 1994-2016 80 70 60 50 40 30 20 10 -10 -20 -30 -40 -50 ▲ IAGOS aircraft, upper trop., 1994–2013 ■ Hilo ozonesondes, mid-trop., 1994–2018 high elevation IAGOS aircraft Δ TOST Δ TOST Sat Latitude Sat3 Sat2 TOS' Sat1 TOST -60 <= 0.05 -70 < p <: -80 -90 -3-2-10123456 78 -3 -2 -1 0 1 2 3 4 5 6 7 8 -3 -2 -1 0 1 2 3 4 5 6 7 8 Ozone trend (ppbv per decade) Ozone trend (ppbv per decade) Ozone trend (ppbv per decade) Satellite products: Sat1 1979-2016 (TOMS, OMI/MLS)

Surface and tropospheric ozone trends

Figure 2.8 in IPCC, 2021:

Chapter 2. In: Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change doi: 10.1017/9781009157896.004

Sat2 1995-2015 (GOME, SCIAMACHY, OMI,

Sat3 1995-2015 (GOME, SCIAMACHY, GOME-II)

GOME-2A, GOME-2B)

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