

February 2, 2025

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

Long-term satellite trends of European lower-tropospheric ozone from 1996 – 2017

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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.

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”.

Detailed comments:

Lines 59-73

The review of past ozone trend studies for Europe needs to be updated to include the most recent work, including a new TOAR-II study. While the papers by Oltmans et al. (2013) and Logan et al. (2012) are very good, they are now quite out of date, and aren't really relevant for current ozone trends.

An important consideration for trend detection is sample size. Since 1988 there have been quite a few studies that have examined the impact of ozonesonde sampling frequency on our ability to detect ozone trends in the free troposphere, as reviewed by the recent TOAR-II paper by Chang et al. (2024). Chang et al. (2024) also provide extensive analysis demonstrating that sparse temporal sampling by ozonesondes, especially once per week sampling, often fails to detect an ozone trend.

Estimates of free tropospheric ozone trends above Europe vary widely, based on the available ozonesonde time series. For example, free tropospheric trends (700-300 hPa) for the period 1994-2019 range from -1.56 ± 0.85 ppbv per decade (p -value <0.01) above Payerne, Switzerland, to $+2.26 \pm 1.04$ ppbv per decade (p -value <0.01) above De Bilt, The Netherlands (Chang et al., 2022). To improve our ability to detect trends, Chang et al. (2022) developed a method to merge all available IAGOS and ozonesonde time series above western Europe, and calculated a regional trend of 0.60 ± 0.20 ppbv per decade (p -value <0.01) for the tropospheric column (950-250 hPa) and a regional trend of 0.65 ± 0.19 ppbv per decade (p -value <0.01) for the free troposphere (700-300 hPa).

A new paper submitted to the TOAR-II Community Special Issue (Van Malderen et al., 2024) builds on the work of Chang et al. (2022), and shows clear ozone increases above Europe based on a merged dataset of ozonesonde, IAGOS, FTIR, Umkehr and lidar time series (stations are listed in Table 1, and additional time series information is provided in Table S1 in the supplement; Table 2 lists the data coverage, and Figure 3 shows the domain). Figure 12 and Table S2 (supplement) show that the 1995-2019 tropospheric column (surface to 8 km) regional ozone trend for Europe is 0.47 ± 0.20 ppbv per decade (p -value <0.01); the authors assign a confidence level of very high to this trend. Trends in the free troposphere (700-300 hPa) are even stronger: 1.25 ± 0.27 ppbv per decade (p -value <0.01); the authors assign a confidence level of very high to this trend.

The findings from Chang et al. (2022) and Van Malderen et al. (2024), along with the earlier findings of Gaudel et al. (2020), Christiansen et al. (2022) and Wang et al. (2022), all demonstrate that ozone has increased above Europe since the mid-1990s. It would be helpful if you can summarize this well documented ozone increase in the Introduction, and use it to frame the findings from your study.

Line 136

Here the manuscript introduces the ozonesondes that are used to evaluate the satellite products and the model, but I couldn't find any information on the actual time series that were chosen. To understand the ozonesonde analysis, and to judge if it supports the conclusions, the reader needs the following basic information: number of stations used, names of stations, coordinates of stations, sampling rate, instruments flown, time period analyzed, and finally, were the time series treated individually or merged? Van Malderen et al. (2024) is a very good reference for presenting this type of information. The authors mention that the ozonesondes are selected to match satellite overpass times of 10:00 and 13:30 LST. Presumably if sondes fall outside of the time window then they are discarded. What does this do to the sample sizes? Ozonesonde sampling rates are already too low for accurate trend detection, and if data are thrown out then the ability to accurately detect a trend diminishes even further.

In addition to using ozonesondes, why not also use the IAGOS profiles from Frankfurt? This is the only location in the world where ozone is profiled multiple times per day, and as a result, it is the only location in the world where we have high confidence that the monthly mean profile is accurate (we also

have high confidence in the monthly distribution, i.e. 5th to 95th percentiles). As a result, the ozone trend at this location is also highly accurate: $+1.16 \pm 0.77$ ppbv per decade (p-value<0.01) in the free troposphere for the period 1994-2019 (Chang et al., 2022).

Many of the authors on this paper are also authors on a paper recently published in the TOAR-II Community Special Issue that merges the GOME, SCIAMACHY and OMI products into a single tropospheric ozone product (Pope et al., 2023). Yet, as far as I can tell, the current paper makes no mention of the merged product, and treats the GOME, SCIAMACHY and OMI products separately. Why not include the merged product?

Line 359-361

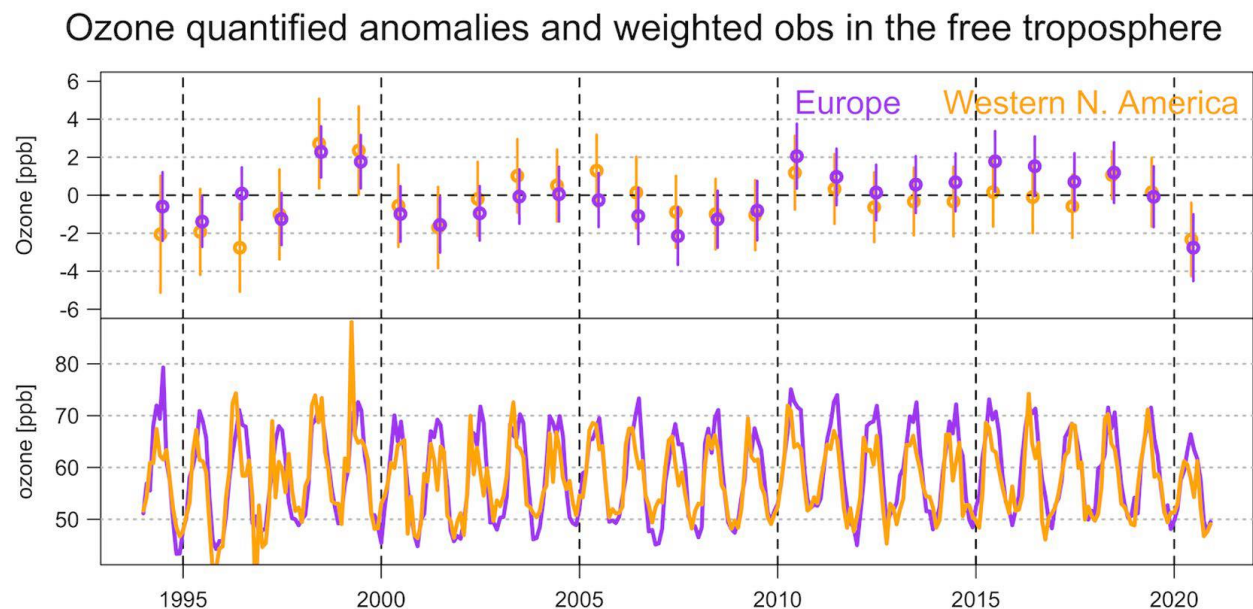
The paper concludes with this statement:

“As a result, it is difficult to detect a robust and consistent linear trend in European lower tropospheric O₃ between 1996 and 2017, which is masked by large inter-annual variability in the model and ozonesonde records and especially the UV sensor records.”

Regarding the in situ ozone trends, as described above, studies that use merged data sets with very large sample sizes are able to detect a positive trend above Europe for the period 1995-2019. The trend is there, but if the sample size is too small, the trend cannot be detected. As no details regarding your ozonesonde analysis were provided, I cannot judge why you were not able to detect an ozone trend, but my guess is that you are using time series with low sampling rates.

Figure 1

Your Figure 1 shows the month to month changes in ozone for the three satellite products from 1996 to 2017. Figure 9 from Chang et al. (2022) (pasted below) shows the monthly ozone values above Europe for the same time period, but based on merged IAGOS and ozonesonde data. It would be helpful to describe the variability of your satellite products in relation to these in situ observations.



From Chang et al. (2022): “Figure 9. Quantified annual ozone mean anomalies (with 2-sigma intervals) and uncertainty weighted time series in the free troposphere (700-300 hPa) above Europe and western North America.”

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