

Response to Referee #2 comment on “Ground-based Tropospheric Ozone Measurements: Regional tropospheric ozone column trends from the TOAR-II/ HEGIFTOM homogenized datasets” by Van Malderen et al.

This paper presents a careful quantification of regional trends in tropospheric ozone from a range of ground-based measurements. The paper is well written, the analysis is thorough and the results are a valuable addition to the field.

My comments are minor. See below.

Thanks you for your minor comments. We believe we have answered and implemented those and thank you for your help in improving the manuscript.

General comments

I didn't catch why the chosen time periods run through 2021 for the TOST analysis but through 2022 for the HEGIFTOM analysis. I would suggest that the authors make it clear why a different date range was chosen for TOST vs HEGIFTOM.

The TOST dataset, developed by Liu et al., (2013a, b), has recently been improved and updated up to 2021 by Zang et al. (2024). After 2021, it is not yet available.

According to the TOAR-II Community Special Issue Guidelines (https://igacproject.org/sites/default/files/2023-04/TOAR-II_Community_Special_Issue_Guidelines_202304.pdf), 21st Century trends are defined for “time series beginning in the range 2000-2002 and ending in the range 2019-2021 (may include sites with data before 2000, but limit the analysis to 2000 and later)”. So, strictly speaking, we could have limited all the HEGIFTOM time series up to 2021 as well, but we preferred to add the extra year of data, if available, also for the calculating the trends from the individual site time series (accompanying paper Van Malderen et al., 2025).

At the end of Sect. 5.1, we added the sentence “It should be noted that all trends are calculated up to 2021, as the most updated version of the TOST dataset (Zang et al., 2024) is provided until the end of 2021.”

I did not follow the reasoning as to why different statistical approaches were applied to the TOST vs the HEGIFTOM datasets. It would be helpful to include some more explanation for the rationale for the choice of these particular approaches for application to these particular datasets.

We included a new Table 4 in the manuscript (see in our response to Referee #1 to find the new table) that summarizes the most important differences between the TOST and LMM approaches, not only in terms of the statistics. The choice of the statistical approach is tied to the characteristic of each dataset: TOST provides a gridded, trajectory mapped, monthly mean (tropospheric) ozone dataset, whereas the HEGIFTOM dataset is made of (partial) tropospheric ozone column time series at individual sites, from different techniques. So, to obtain regional trends from TOST, trends are estimated from merged data with QR, while with the LMM approach, synthesized (linear regression) trends from (well-correlated) individual time series within a region are calculated.

Specific comments

Abstract: “challenged by the diversity between satellite tropospheric ozone records and the sparse temporal and spatial sampling of ground-based measurements”.

I had to read this quite a few times before I could understand what this sentence was supposed to mean, particularly the phrase “challenged by the diversity”. I think that the authors are pointing to two different issues. One issue is that there are numerous satellite products for tropospheric ozone and that there are differences between those records that have not yet been well understood or accounted for. The other issue is that the “reference” ground-based datasets have

limitations in their spatial and temporal coverage. I think it would be much better to split this into two sentences to make the points clear.

We changed this sentence into “Quantifying long-term free-tropospheric ozone trends is essential for understanding the impact of human activities and climate change on atmospheric chemistry. However, this task is complicated by two key challenges: the differences among existing satellite-derived tropospheric ozone products, which are not yet fully understood or reconciled, and the limited temporal and spatial coverage of ground-based reference measurements.”

Line 87: “available satellite products disagreed on the sign of the trend”. This may be overly picky, but I would say that the analysis in the Gaudel et al. (2018) paper was not sufficient to determine whether there was truly disagreement or whether the differences between the trends presented were due to inherent characteristics of the different satellite datasets. I would suggest saying something like “early analysis of available satellite products did not provide a consistent picture of the sign of the trend”.

We implemented your suggestion.

Line 89 and Conclusions: Can you state at the end whether or not your conclusions are consistent with those from the IPCC AR6?

This is a very good point, and we made this comparison. We added the following bullet point to the Conclusions: “These findings are consistent with the conclusions of IPCC AR6. The HEGIFTOM regional trends in the free troposphere (FTOC, 700-300 hPa) and in the tropospheric column (TrOC, $p < 300$ hPa) for the period 1995-2019 (see Figure 12a and Tables S2 and S3), are very similar to the free tropospheric and tropospheric column trends assessed by IPCC AR6 (see Figure 2.8 in Gulev et al., 2021), which span a similar time period.”

Line 122: It would be helpful to state in this paragraph what the 5 different measurement techniques are, since this isn’t explained until later.

Done. Thank you for addressing this.

Section 2.2: Is it correct to say that there is no chemistry in TOST? Ozone is assumed to be constant along a given trajectory? Is that what “assigned along its forward and backward trajectory” means? Please clarify.

The answer is that TOST does not include photochemistry directly. However, TOST does account for chemistry to some extent. This is associated with the assumption that ozone concentrations in an air parcel remain constant along a 4-day trajectory (backward and forward) because the lifetime of ozone is generally longer than 4 days in the atmosphere. The forward or backward trajectory starts from the location of an ozonesonde at a given height for every 1 km from the surface to 26 km. The ozone mixing ratio value at that height is assigned to each grid cell along its 4-day forward and backward trajectories.

We included the sentence “As the lifetime of ozone is generally longer than 4 days in the atmosphere (Stevenson et al., 2006; Monks et al., 2015; Han et al., 2019; Prather and Zhu, 2024), it is conservative to assume that the ozone mixing ratio in an air parcel is constant along a given trajectory of 4 days, running either forward or backward from an ozonesonde profile.” And further, we completed a sentence (italic) with “Rather than simple linear or polynomial interpolation, the ozone measurement from the ozonesonde profile at the origin of a trajectory is assigned along its forward and backward trajectory paths, *starting from a location of the ozonesonde at a given height for every 1 km from the surface to 26 km. The ozone mixing ratio value at that height is assigned to each grid cell along its 4-day forward and backward trajectories.*”

Han, H., Liu, J., Yuan, H., Wang, T., Zhuang, B., and Zhang, X.: Foreign influences on tropospheric ozone over East Asia through global atmospheric transport, *Atmos. Chem. Phys.*, 19, 12495–12514, <https://doi.org/10.5194/acp-19-12495-2019>, 2019.

Monks, P. S., Archibald, A. T., Colette, A., Cooper, O., Coyle, M., Derwent, R., Fowler, D., Granier, C., Law, K. S., Mills, G. E., Stevenson, D. S., Tarasova, O., Thouret, V., von Schneidemesser, E., Sommariva, R., Wild, O., and Williams, M. L.: Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer, *Atmos. Chem. Phys.*, 15, 8889–8973, <https://doi.org/10.5194/acp-15-8889-2015>, 2015.

Prather, M. J. and Zhu, X.: Lifetimes and timescales of tropospheric ozone: Global metrics for climate change, human health, and crop/ecosystem research, *Elementa: Science of the Anthropocene*, 12, 1, <https://doi.org/10.1525/elementa.2023.00112>, 2024.

Stevenson, D. S., Dentener, F. J., Schultz, M. G., Ellingsen, K., Van Noije, T. P. C., Wild, O., Zeng, G., Amann, M., Atherton, C. S., Bell, N., Bergmann, D. J., Bey, I., Butler, T., Cofala, J., Collins, W. J., Derwent, R. G., Doherty, R. M., Drevet, J., Eskes, H. J., Fiore, A. M., Gauss, M., Hauglustaine, D. A., Horowitz, L. W., Isaksen, I. S. A., Krol, M. C., Lamarque, J. F., Lawrence, M. G., Montanaro, V., Müller, J. F., Pitari, G., Prather, M. J., Pyle, J. A., Rast, S., Rodriguez, J. M., Sanderson, M. G., Savage, N. H., Shindell, D. T., Strahan, S. E., Sudo, K. and Szopa, S.: Multimodel ensemble simulations of present-day and near-future tropospheric ozone. *J. Geophys. Res.* 111(D8). DOI: <https://doi.org/10.1029/2005JD006338>, 2006.

Caption for Figure 5: Figures show trends from 1995-2021 but statistics are based on median values over 1990-2021. Is this correct or should the date ranges be consistent?

In the caption of Figure 5, the 1990-2021 is a mistake. It should be 1995-2021. We are sorry for this. Thanks for pointing it out!

Line 543: There is a sentence that starts with “And”. Please check grammar.

We changed this:

“Alternatively, the impact of the FTIR and Umkehr time series, if available, on the TrOC trend, could be rather limited.”

Line 580: Consider adding a sentence or two that says something about possible reasons for free tropospheric increases that are not driven by precursor emissions/lower tropospheric increases. Changes in dynamics? Changes in OH availability?

We added: “While regional ozone trends can be influenced by interannual variability resulting from meteorological influences (e.g. ENSO) (Chandra et al., 1998; Oman et al., 2011, 2013; Ziemke et al., 2015; Lin et al., 2015, 2015, 2017; Lu et al., 2019; Xue et al., 2021; Jeong et al., 2023; Stauffer et al., 2024), model studies have consistently indicated that the hemispheric scale increase of ozone precursor emissions, especially in the tropics, is the dominant driver of positive ozone trends in the free troposphere of northern mid-latitudes (Verstraeten et al., 2015; Zhang et al., 2016, 2021; Fiore et al., 2022; Liu et al., 2022; Wang et al., 2022).”

Chandra, S., J. R. Ziemke, W. Min, and W. G. Read (1998), Effects of 1997–1998 El Niño on tropospheric ozone and water vapor, *Geophys. Res. Lett.*, 25, 3867–3870.

Fiore, Arlene M., Sarah E. Hancock, Jean-François Lamarque, Gustavo P. Correa, Kai-Lan Chang, Muye Ru, Owen R. Cooper, Audrey Gaudel, Lorenzo M. Polvani, Bastien Sauvage and Jerry R. Ziemke (2022), Understanding recent tropospheric ozone trends in the context of large internal variability: A new perspective from chemistry-climate model ensembles, *Environmental Research: Climate*, <https://doi.org/10.1088/2752-5295/ac9cc2>

Jeong, Y., Kim, S.-W., Kim, J., Shin, D., Kim, J., Park, J.-H., & An, S.-I. (2023). Influence of ENSO on tropospheric ozone variability in East Asia. *Journal of Geophysical Research: Atmospheres*, 128, e2023JD038604. <https://doi.org/10.1029/2023JD038604>

Lin, M., Horowitz, L.W., Oltmans, S.J., Fiore, A.M. and Fan, S., 2014. Tropospheric ozone trends at Mauna Loa Observatory tied to decadal climate variability. *Nature Geoscience*, 7(2), pp.136-143

Lin, M., Fiore, A.M., Horowitz, L.W., Langford, A.O., Oltmans, S.J., Tarasick, D. and Rieder, H.E., 2015. Climate variability modulates western US ozone air quality in spring via deep stratospheric intrusions. *Nature communications*, 6(1), p.7105.

- Lin, M., et al. (2017), US surface ozone trends and extremes from 1980 to 2014: quantifying the roles of rising Asian emissions, domestic controls, wildfires, and climate, *Atmos. Chem. Phys.*, 17, 2943–2970, 2017, www.atmos-chem-phys.net/17/2943/2017/doi:10.5194/acp-17-2943-2017
- Liu, J., Strode, S. A., Liang, Q., Oman, L. D., Colarco, P. R., Fleming, E. L., et al. (2022). Change in tropospheric ozone in the recent decades and its contribution to global total ozone. *Journal of Geophysical Research: Atmospheres*, 127, e2022JD037170. <https://doi.org/10.1029/2022JD037170>
- Lu X, Zhang L, Zhao Y, et al. Surface and tropospheric ozone trends in the Southern Hemisphere since 1990: possible linkages to poleward expansion of the Hadley Circulation. *Sci Bull* 2019; 64:400–9.
- Oman, L. D., J. R. Ziemke, A. R. Douglass, D. W. Waugh, C. Lang, J. M. Rodriguez, J. E. Nielsen (2011), The response of tropical tropospheric ozone to ENSO, *Geophys. Res. Lett.*, 38, doi:10.1029/2011GL047865
- Oman et al. (2013), The ozone response to ENSO in Aura satellite measurements and a chemistry-climate simulation, *JOURNAL OF GEOPHYSICAL RESEARCH: ATMOSPHERES*, VOL. 118, 965–976, doi:10.1029/2012JD018546, 2013
- Wang, H., Lu, X., Jacob, D. J., Cooper, O. R., Chang, K.-L., Li, K., Gao, M., Liu, Y., Sheng, B., Wu, K., Wu, T., Zhang, J., Sauvage, B., Nédélec, P., Blot, R., and Fan, S. (2022a), Global tropospheric ozone trends, attributions, and radiative impacts in 1995–2017: an integrated analysis using aircraft (IAGOS) observations, ozonesonde, and multi-decadal chemical model simulations, *Atmos. Chem. Phys.*, 22, 13753–13782, <https://doi.org/10.5194/acp-22-13753-2022>
- Xue, L., Ding, A., Cooper, O., Huang, X., Wang, W., Zhou, D., Wu, Z., McClure-Begley, A., Petropavlovskikh, I., Andreae, M.O. and Fu, C., 2021. ENSO and Southeast Asian biomass burning modulate subtropical trans-Pacific ozone transport. *National Science Review*, 8(6), p.nwaa132.
- Zhang, Y., West, J. J., Emmons, L. K., Flemming, J., Jonson, J. E., Lund, M. T., et al. (2021). Contributions of World Regions to the Global Tropospheric Ozone Burden Change from 1980 to 2010. *Geophysical Research Letters*, 48, e2020GL089184. <https://doi.org/10.1029/2020GL089184>
- Ziemke et al. (2015), Tropospheric ozone variability in the tropics from ENSO to MJO and shorter timescales, *Atmos. Chem. Phys.*, 15, 8037–8049, www.atmos-chem-phys.net/15/8037/2015/

Caption for Figure 13: Please state what the gray points represent. I see that this is stated in the text, but it should also be made clear in the figure caption.

It was already there: “In grey: the individual site trend estimates *from Van Malderen et al. (2025)*, with different symbols for the different techniques”, but we added, in italic, the reference to the HEGIFTOM individual site trends paper.