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Comment on acp-2022-686

Anonymous Referee #2

Referee comment on "Tropical tropospheric ozone and carbon monoxide distributions: characteristics, origins, and control factors, as seen by IAGOS and IASI" by Maria Tsivlidou et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-686-RC2, 2022

Tsivlidou et al. provide an analysis of the longitudinal and vertical distribution of O3 and CO in the tropical band using IAGOS and IASI datasets. They apply the SOFTIO software to the IAGOS dataset to attribute the sources of CO mixing ratios above background values in different regions of the tropics. They conclude that anthropogenic emissions drive most of the CO enhancements measured by IAGOS throughout the tropical band, with occasional contribution from biomass burning emissions. Overall, I think the paper is overly long and hard to follow because of the lack of a clear structure in the discussion. This is really unfortunate, as it really undermines the value of the results presented throughout the manuscript. In any case, I have several major concerns that will need to be addressed by the authors before the paper can be accepted for publication in ACP. Below, I also raise several more specific points that should also be answered to.

General comments:

- I find the paper extremely hard to follow. It is extremely long (41 pages excluding references!), and would really benefit from a clear structure in the way the results are discussed. I would recommend using subsections for each cluster so that the reader can know straight on what exactly is discussed (e.g., which cluster is discussed? LT, MT or UT? Which month? O3 or CO? Measured mixing ratios or SOFTIO contribution? etc.).

  In my opinion, the section 3.1 does not bring anything to the paper. Discussing IAGOS data as done in the following sections is already plenty enough for one paper. In addition, the section 3.1 reads as purely speculative, and raises questions that are actually answered later on in the manuscript. What is the point of that?

  They are a lot of acronyms throughout the paper, especially with regard to the source regions of CO. It makes sense to use those in the figures, but the authors should consider righting the name of contributing regions in full to avoid having to refer to

Figure 1 all the time. In addition, NH and SH are often used in the paper as referring to the northern tropics and southern topics. This is really confusing as NH and SH are classically used for the whole northern and southern hemispheres. I would recommend using different abbreviations (NT and ST?).

The authors separate the troposphere in 4 different layers (surface, LT, MT and UT) but, unless I missed something, never define those layers in the manuscript. It complicates the discussion, as these definitions drive the interpretation of the data. This should be clarified at the beginning of the manuscript.

- A crucial piece of information is missing from the paper in my opinion, that bears on how the data should be interpreted. The IAGOS flights used by the authors date back from 1994 and 2002 for O3 and CO, respectively. However, there is no information on how the IAGOS flights used in the current analysis are spread throughout this time period. For a given cluster, are there always the same number of flights per month throughout the time period? Do all clusters span the same time range, or are some timeseries shorter than others? I assume that all clusters are different on the time range with data availability. If so, how would this impact the comparison of clusters, and the source attribution? There is a number of flights provided for each cluster in Figures 4, 6,7 and 8, but it is really unclear from the captions of these figures what this number refers to. Is that the average number of flights per month for the entire time period considered here? Or is that the total number of flights for all months in the given time period? If so, how do you quantify the uncertainty due to the low number of flights in some places (e.g., Mumbai or AbuDhabi)?
- There is no discussion at any point of the uncertainty in the contribution of sources to CO using the SOFTIO software. Throughout the paper, relative or absolute contributions of anthropogenic and biomass burning emissions to measured CO are provided, but with no associated uncertainties. I would imagine that this uncertainty is quite high (stemming from the uncertainty in emission inventories, uncertainty in back trajectory computation, etc.) and needs to be quantified.

  In the same vein, it became really hard to me to trust the contributions calculated by SOFTIO given the significant gap between those numbers and the actual mixing rations actually measured by IAGOS. First, the authors should explain more explicitly how background CO is calculated. Is it one common value for all sites? Is there a vertical resolution of the background? Then, for most sites in this study SOFTIO only explains a (small) part of the calculated CO anomalies, leaving in some cases hundreds of ppb of CO unaccounted for. To me, this issue needs to be discussed in more details, since the whole paper relies on the assumption that SOFTIO can explain "95% of the CO anomalies" as stated by the authors. At the very least, I would like to see on all figures (the ones that report CO contributions from AN and BB sources like figures 4 and 5) the proportion or absolute amount of CO anomalies that is unaccounted for. If and when that amount is larger than the contributions actually accounted for by SOFTIO, the authors need to discuss how much confidence can be put in the interpretation of those results. In addition, the authors invoke throughout the paper that the shortcomings of SOFTIO are due to underestimated AN emission inventories. Why would that be the case, rather than underestimation of BB emission inventories for instance? Another thought would be that the missing CO (unaccounted for) could be within the uncertainties of the SOFTIO calculated contributions, but this is hard to conclude on in the present paper as uncertainties are not discussed. In any case, if there is so much uncertainty in the AN emission inventories in the first place, that would impact SOFTIO calculated contributions everywhere and throughout the tropospheric column, right? So again, uncertainties should be discussed.

- One of the main conclusions of the paper is that anthropogenic emissions contribute much more to CO anomalies measured by IAGOS compared to BB burning. Is that really surprising considering that the large majority of the sites studied here are megacities, with several million inhabitants, and therefore with overwhelming local AN emissions? Imagine IAGOS airports were located in the middle of fires, wouldn't you get the opposite results? It is not clear to me how you can generalize your findings to the whole tropical band.

Detailed comments:

l.35 Change to (e.g., Edwards et al. 2006) as this was not the first paper to use CO as a pollution tracer.

l.37-38 It would be good to include a quantification of the respective sources of CO here, and an adequate reference for this statement on CO being primarily emitted by anthropogenic emissions. I imagine you are referring to emission inventories? Please explicit.

l.41-42 Bourgeois et al. (2020) recently presented global-scale distribution of O3 in the remote troposphere based on aircraft observations. Probably a good idea to acknowledge that here.

Bourgeois, I., Peischl, J., Thompson, C. R., Aikin, K. C., Campos, T., Clark, H., Commane, R., Daube, B., Diskin, G. W., Elkins, J. W., Gao, R.-S., Gaudel, A., Hintsa, E. J., Johnson, B. J., Kivi, R., McKain, K., Moore, F. L., Parrish, D. D., Querel, R., Ray, E., Sánchez, R., Sweeney, C., Tarasick, D. W., Thompson, A. M., Thouret, V., Witte, J. C., Wofsy, S. C., and Ryerson, T. B.: Global-scale distribution of ozone in the remote troposphere from the ATom and HIPPO airborne field missions, Atmos. Chem. Phys., 20, 10611–10635, https://doi.org/10.5194/acp-20-10611-2020, 2020.

l.42 "inadequate" doesn't sound like the right word here. Maybe use "due to the paucity of observations". Also, please resolve the conflict: the troposphere above "developing countries in the tropics" is not "remote". The remote troposphere applies to air far from emission sources, i.e., far from land.

l.61-62 I disagree with this statement. Field campaigns in the tropics have provided invaluable insights on the both the atmospheric chemistry and dynamics of this region, back from the 80's. The spatial coverage of these campaigns, especially airborne, is much larger than that of ozonesondes for instance, which map out O3 columns at very specific locations. I think that the least you can do is acknowledge the value of these campaigns and also name them (CAST, ATTREX, SAFARI, the NASA GTE campaigns (PEM-TROPICS), etc.).

l.64 Why would ozonesondes under-represent the tropical upper troposphere? If anything, the altitude ceiling of ozonesondes is much higher than that of IAGOS or other airborne measurements.

l.69 This is a bit misleading. Yes, you have long time-series from IAGOS flights, but you have strong heterogeneity in how the flights are spread in time and space throughout these periods. If you are going to call out all the limitations of other research networks, then you should acknowledge that here.

l.70 Madras is the fourth biggest city in India. Definitely not remote. Per definition, IAGOS flies in and out of airports, so you expect most of their tropospheric profiles to be influenced by regional emissions. You cannot qualify IAGOS as a remote troposphere observation network.

l.59-74: These two paragraphs read as trying too hard to show that IAGOS is much better than the other research networks and field campaigns in the tropics. It may not be the intention of the authors, but I think that they should rather focus on showing that all infrastructures are complementary and the combination of all are necessary to fully understand the kind of research questions they are addressing.

l.83 All research should be novel. Please remove "for the first time"

l.98-99 The horizontal resolution is the same during the ascend and descend phases and during the cruise phases?

l.166 How do you define the lower part of the UT?

l.117 How much of the data is discarded by applying this filter?

l.122 Please state the typical upper limit here

l.125-137 An important missing information here is a table that shows the number of flights per month for each of the sites (or clusters) shown in Table 1, the time length of the time series, if there are flights for every months of every year, etc. Basically, the authors could expand Table 1 to include all the information that the readers need to assess the distribution of IAGOS flights in time and space.

l.144 Does “intermediate” mean between the seasons? If so, why not take months during peak seasons?

l.161 most regions, including the tropics?

l.162 most of the regions, including the tropics?

l.166 Please specify where the remote area was located. You are only considering the UT to calculate the CO background. That does not seem right too me, since you will use this value to calculate CO anomalies throughout the tropospheric column and there is a vertical gradient in CO mixing ratios due to stratospheric air mixing. You should use the median CO value over a remote area averaged across the tropospheric column. Also, please provide a range of the CO background values thus obtained.

l.204-209 I don’t understand these figures and the associated discussion. The discussion talks about NH and SH emissions of CO, but the figures do not show all of the NH and SH. The maps are truncated to a latitude band that seems randomly chosen and does not seem to correspond to the tropical band as defined by the authors early on in the manuscript.

l.210-214 Why speculate here on the sources of CO and O3 when you are actually answering this question later on in the manuscript using SOFTIO?

l.220-225 This is not true. In this section, you only speculate, based on coincident maps of AN and BB emissions and CO and O3 concentrations, on the reasons of the anomalies. You have not shown any causality (coincidence is not a sound scientific argument) nor have you excluded other potential sources as long-range transport or stratospheric air mixing. In addition, the wave one pattern is already well described in the literature. You should cite appropriate literature here with regard to this effect. In any case, you should change “are related” to “could be related to”.

l.243 How do you define the LT?

l.245 What does this 58% value refer to? An average across the four months for the two sites? Why not include Gulf of Guinea in that average? Is that an average for the LT only? How do you define the LT? Please be more precise.

l.247 “extend” is not the right word here

l.257 October is more polluted than January and April in Central Africa and similarly polluted as July in Sahel and Gulf of Guinea

l.260-261 But this is also true for the other months, right? Look at January for instance, where the mean CO ranges between 250 and 750 ppbv between 0-4km. For that month, SOFTIO only attributes 200 ppbv of CO anomaly, so it clearly misses a large fraction of the CO source for that month as well. On what do you base your statement that this is due to an “underestimation of NHAF AN emission”? The fact that IASI also sees elevated CO (as mentioned in the following sentence) is no proof that they are AN originated. Why not an underestimation of BB emissions? Could it not be an issue with how you defined your background CO in the first place? This clearly needs to be explained in more details.

l.273 69 ppb, not 70.

l.277-280 and l. 287-289 Here you discuss O3 sinks in the LT, which is redundant with the following paragraph (l.290-297). I suggest keeping the following paragraph and removing these sentences to improve the readability of the manuscript.

l.314-317 Again, how can you conclude that SOFTIO underestimate AN emission, but not BB emissions for instance? It looks like SOFTIO struggles to match the CO anomalies near the surface for most African sites. Can that only be explained by incorrect AN emission inventory? If that is the reason, how can we trust the rest of the AN vs BB attribution by SOFTIO in the rest of the tropospheric column?

l.351 Madras is not an “oceanic site”, it is a mega-city (fourth biggest city in India).

l.442-445 This is true for all sites in South America, and most sites in Asia and in the Arabic Peninsula (on top of all sites in Africa). So, if all AN emission inventories are wrong in most regions, as suggested by the authors (and why is the reason for that?) how can the authors still quantify AN contribution throughout the tropospheric column with so much certainty?

l.561 So is 100 ppb the background level of CO? Shouldn’t the background value be altitude dependent, with lower values in the UT?

l.571-573 How so? O3 could be formed by BB emissions with the CO maxima being still

due to AN emissions. That O3 and CO maxima aren't collocated doesn't mean that O3 production isn't due to BB emissions.

l.575 Isn't O3 always formed by photochemical processes?

l.579 I was not aware that lightning also produces CO. How can LiNOx be responsible for elevated CO in the MT?

l.581 the large majority of your flights are over megacities. I think it is fair to assume that low O3 in the BL are due to titration by NOx

l.582-583 This is not obvious to me. Looking at the various figures, it looks like BB emissions contribute a lot in January to most of the African clusters. In addition, the O3 maxima is closer to the MT than the LT (although it is not clear how the authors dined these two layers) whereas the CO maxima is at the surface. So, to me there is clearly a discrepancy with the text here.

l.593 I have a hard time believing that CO would be diluted by BB impacted air masses. Please explain in more details.

l.621-622 The authors should rephrase this sentence, as it seems to imply causality between enhanced CO and enhanced O3. Do they really mean that the elevated CO in the LT is responsible for the elevated O3?

Conclusion There shouldn't be any references in the conclusion.

Table 2 Please indicate the pressure range for CO and O3 in the table, as they are not the same for the two species. Please change NH and SH to NT (northern tropics) and ST, otherwise it gets confusing.

Figure 2 Please indicate that those are monthly mixing ratios averaged from 2008 to 2019.

Figure 4 Please explain in the caption why Sahel and Gulf of Guinea are not included in the figure (for BB vs AN contribution) but rather given in the appendix.

Figure 7 Please expand the axes sot that all of the data are shown in panels 3,4 and 5.

Figure S1 The latitude band is not the same for all panels. Please homogenize.