Review response 2

June 2024

The paper presents a seasonal analysis of CO pollution plumes (anomalies) sampled by IAGOS commercial aircraft over different regions of the world for the period 2002-2019. Modeled footprints and global emission inventories for CO anthropogenic and biomass burning are used to simulate contributions to CO along each flight track and attribute the observed anomalies to emissions by type and by source region. I assume this study was made possible thanks to a lot of careful work and continued support for the IAGOS program but the paper does not give much details about this although it provides several references for previous analyses of the data. The authors use footprints and emission inventories "semi quantitatively" for emission attribution, I assume previous work has shown this is a reliable approach. The paper clearly presents graphic summaries for the CO anomaly analysis by region and the text further describes how seasonality in some atmospheric transport processes and emissions can explain the results. The discussion of ozone levels in the anomaly plumes is mostly descriptive by region. It seems that in only a few cases do CO anomalies correspond to ozone anomalies. Is there another paper that looks more holistically at those ozone anomalies and the processes behind them? It would be nice to help the reader understand the significance (and maybe limitations) of your analysis and findings for ozone. The importance of the IAGOS dataset and this work may be made stronger with a more organized argumentation in the introduction. Some of the text there is repetitive and some general statements are not backed up by references. The conclusion mostly summarizes the findings but could maybe also be more explicit about future work and why continuing and potentially expanding those measurements, adding other tracers... is important for the next decades.

We thank the reviewer for his/her important comments that will help improve this paper. We respond below to each specific point.

A few references on ozone have been added in order to better discuss our results in light of the literature:

- Yang et al. [2019]
- Chang et al. [2017].
- Lu et al. [2018]
- Gaudel et al. [2018]
- Cohen et al. [2018]

- Nowak et al. [2004]
- Hudman et al. [2004]

There are two important references of the ITCT 2k2 campaign over the northern pacific ocean and focusing on the chemical characteristics of pollution plumes from east asia. [Nowak et al., 2004, Hudman et al., 2004].

To clarify, ozone is not the focus of this paper as it is a first step before using a global CTM. Here, only levels of ozone are presented, since only CO and O3 measurements are available at this scale. We can not do a complete analysis of the processes behind each value but we plan to make a follow up analysis on the processes leading to ozone in pollution plume with a chemistry transport model. A paragraph on those perspectives have been added to the conclusions.

A paragraph on the Ozone limitation and perspectives has been added to the conclusions (lines 600-605): These O_3 values give information on its possible production in polluted plumes. However, without the measurements of additional chemical compounds (like VOCs and NOx for example) it is difficult to draw robust conclusions. To go further into the analysis on the O_3 in pollution plumes, information on more chemical compounds is required. The current perspective is to carry a similar study with a Chemistry Transport Model in order to get further information on the provenance of O_3 values but also on the amount of O_3 productions in polluted plumes, especially in regions with elevated values of O_3 like Siberia and the Middle East.

An other paragraph on the perspective about the importance of the IAGOS measurements as well as future perspective has been added to the conclusions (lines 590-594):

We have presented a detailed analysis of the characteristics of high carbon monoxide plumes and their associated ozone anomalies in different regions of the world. It is important for the IAGOS infrastructure to continue those measurements and to expand the regions sampled by the research infrastructure in order to provide these diagnostics in additional regions. This is particularly important in the tropics, where anthropogenic emissions are increasing and impact on the O_3 trend globally [Zhang et al., 2016]. Increased number and sampling frequency of measurements of NOx and aerosols by IAGOS will be available and valuable for future analysis focusing on O_3 photochemical production or air quality.

1 High level comments:

Are the findings new?

Yes, in terms of (i) synthesis study with dense and global data sets (ii) allowing a robust statistical analysis (iii) It is one of the only study focused on the extreme pollution anomalies around the world.

What are some key implications ?

- We thus provide diagnostics robust enough to further allow any "smart-evaluation" analysis

of global model pursuing the goal of having the good mixing ratios for the good reasons (CO in particular here).

- We show the significant impact of anthropogenic emissions and in particular of certain key regions on the CO anomalies in the MT and UT.
- We show that the most extreme anomalies are almost always related to biomass burning emissions.
- Ozone on average shows higher values in CO plumes and can even reach very high values under certain conditions.

Why are trends or interannual variability not explored? I think I may be able to guess but you may want to be explicit about it in the paper, ie. if the dataset year to year spatiotemporal coverage does not allow for this type of analysis.

As guessed, this is beyond the scope of the paper because the spatiotemporal coverage does not allow for this type of analysis. Some regions are sampled regularly for only a few years as it can be seen in the figure B1 in the appendix. We focus here on the "ID" of CO anomalies (i.e. where do they come from ? which transport pathway ? how is ozone inside such anomalies ? ...).

Be explicit about the nature of the IAGOS dataset for people less familiar with this work: Mention they are measurements on commercial aircraft, in the introduction and mention IAGOS in the conclusion too.

We thank the reviewer for the comment, as advised we added the following sentences in blue to the introduction and conclusion of the revised manuscript.

- IAGOS (In-service Aircraft for a Global Observing System; http://www.iagos.org) is a European research infrastructure using commercial aircraft in order to measure the atmosphere composition. Lines (86-88)
- IAGOS is a research infrastructure which uses commercial aircraft to measure atmospheric composition. Lines (488-489)

The consistency of the data calibration and the data quality throughout the period and across instruments is assumed but it may be nice to include a couple of sentences on that.

The following sentence has been modified in the methods section (lines 109-111).
The consistency between the MOZAIC, IAGOS and CARIBIC datasets as well as the internal consistency of the CO and O3 measurements since 1994 have been tested [Nédélec et al., 2015, Blot et al., 2021].

Further define the CO anomalies: how many consecutive datapoints above the q95 threshold are needed to become an anomaly plume?

The definition of the CO anomalies can be found in the methods section (lines 194-195) :
The CO anomalies are defined as CO values exceeding the threshold for three consecutive measurements (i.e. a distance of approximately 3 km during cruise phase).

I found a few typos or small corrections. Another thorough reading would be great to make sure all of these are taken care of. For example, fix a few inconsistencies throughout the article about how you refer to your regions.

Thank you for your comment. A thorough proofreading has been made by one of the co-authors, native english speaker.

2 Detailed comments:

2.1 Abstract:

First sentence should be clear the analysis is done for large regions of the globe.

In-situ measurements from IAGOS are used to characterise extreme values of carbon monoxide (CO) in large regions of the globe. (line 1-2)

You cover some of the findings for some region but results for India are not mentioned, even though they have their own section.

L14: Indian CO anomalies have drastically different characteristics depending on the season as the wet and dry phases of the monsoon have an important impact on the transport of the pollutant in this regions.

The much higher CO in anomalies over E Asia may be nice to mention here too.

L9: The largest values of CO are found in Eastern Asia in the lower and middle troposphere.

2.2 Introduction:

References would be great for statements on model limitations to reproduce or predict extreme weather events and extreme pollution events

Modified (lines 21-23): Extreme weather can sometimes be incorrectly reproduced and predicted by the global and regional models (e.g. Shastri et al. [2017], Lavaysse et al. [2019]). Extreme pollution events can also be difficult to predict, as they can be explained by multiple factors such as abnormal weather conditions and/or unusually intense emissions (either from anthropogenic or natural sources, or both).

Not clear about the impact of extreme pollution events on climate, maybe expand on what you mean with climate here and add references.

Deleted

Pollution is often referring to conditions in the boundary layer. What does it mean for the troposphere?

Pollution is often from the BL as it is emitted at the surface. After emissions it can however be exported out of the BL.

The text in the introduction makes it sound like this paper/study can be used to improve model simulations of extreme pollution plumes. How would this be done?

Probably not directly, but the purpose of this study was to better understand the origins of the CO and the main characteristics of the pollution and CO anomalies. Global model can use the diagnostics given here to verify that the model's pollution patterns have similar characteristics and have the right mixing ratios for the right reasons.

L 27-30: "This compound In the troposphere, ozone is photochemically produced from NOx and VOC (Volatile Organic Compounds)/ or CO (Seinfeld and Pandis, 2008). Hence, a good estimation of its chemical precursors as well as better understanding of the processes leading to their distributions at global scale is of prime importance."

Done line 32.

L 44: Owen et al., 2006 should be Cooper et al., 2006 (Owen is the firstname and Cooper is the lastname of the author).

The first author is R. C. Owen, Owen Cooper is second author.

L 72-75: "We present here a quasi-global overview over almost 20 years of extreme CO mixing ratios and their associated O3 values, as seen by IAGOS. The goal of this paper is to characterise

the seasonal, regional and vertical CO mixing ratios anomalies for different regions over the globe for almost 20 years as seen by IAGOS along with the simultaneously recorded O3 between 2002 and 2019." These two sentences say the same thing. Please remove repetition.

Done line 90

L 101-109: Is the last paragraph on the model needed in the measurement data set section 2.1? It is mostly repeated in section 2.2. Similarly the first paragraph in 2.2 repeats some of what is in section 2.1. Please revise to focus on what belongs in each section.

Done line 120.

2.3 Methods:

L 116: "The Bbiomass Bburning emission inventory used in this version..." remove uppercase letters from biomass burning and check if this should be singular, or plural.

Done line 130.

Section 2.2 : In the model, you only look at direct emissions of CO not CO chemically produced?

Exactly, we mostly focus on the emitted CO in the study as it is harder to account for CO production without a chemistry transport model.

Figure 1 may have better contrast for the Americas if the oceans were kept white. Could the legend be placed outside of the map to not cover part of it and you can make it a little bigger too? Are the acronyms for the GFED regions defined somewhere in your paper? Especially as you refer to boreal emissions several times, I assume you refer to emissions from BONA and BOAS.

Thank you for the comments, the figure has been fixed and a table of the acronyms has been added to the appendix (see table 1 below).

L 165: "At higher altitude, the samples are less influenced by local emissions..."

Done: L191.

Figure 2: There are two blue lines, so the CO measurements one would need to be referred to as dark blue. There are horizontal and vertical dashed lines. Are the vertical ones needed? Clarify

Acronym	Full name
BONA	Boreal North America
TENA	TEmperate North AMerica
CEAM	CEntral AMerica
NHSA	North Hemisphere South America
SHSA	South Hemisphere South America
EURO	Europe
MIDE	MIDdle East
NHAF	Northern Hemisphere AFrica
SHAF	South Hemisphere AFrica
BOAS	BOreal ASia
CEAS	CEntral Asia
SEAS	South East Asia
EQAS	Equatorial Asia
AUST	AUSTralia

Table 1: Table GFED acronym:

you refer to the horizontal dashed line for the 95th percentile for the CO for that region/season; you could give the value for q95 in the caption. What altitudes did the measurements in the Figure cover? What happens during the data gaps seen in the Figure?

Caption corrected

Table 1: Specify this is for CO and for different seasons in the caption. Put the unit (ppb) in the caption, not the table itself. Explain what "no data" means. Do you need to show results for seasons you will not discuss.

Caption corrected

L 172-173: "SOFT-IO is then used as a qualitative tool to assign a source type to each of the detected anomalies. This diagnostic is only applied if the contributions modelled by SOFT-IO are above a detection threshold defined as 5 ppb." You use 5 ppb for all altitude bins? Does it matter?

Yes but this criteria is almost only important for the UT layers where concentration and SOFT-IO contributions are low. Sauvage et al. [2017] showed that the detection frequencies of CO plume were decreasing at higher altitudes.

2.4 Results:

Figure 3: You would need to define Low BB in the caption.

Done

Legend is covering the a) in the first plot.

Corrected

Can you comment on the high mean for DJF and JJA CO anomalies in E Asia, plots a and b? What anthropogenic sources contribute the most here?

A paragraph on the high values of CO in East Asia has been added in the revised manuscript (lines 245-242) in blue below. At this altitude, the highest values of CO are found in Eastern Asia during both seasons. The anomalies can even reach a mixing ratio over 700 ppb in DJF. Those extremely high values are due to the important emissions from local anthropogenic sources and especially from the industrial and residential sectors [Qu et al., 2022].

Figure 5: "At this altitude 24 anomalies over out of the 5341 observed..." The unattributed anomalies in grey are very hard to see. Maybe that text could be in the main text not the Figure caption.

For this altitude layer, it is true but we want to keep it the same for every figure.

L 217-218: "BB contributions comes in the vast majority from Boreal America and Asia." plural

Done.

L 219-220: " In JJA, the plumes attributed to BB emissions are the most intense" plural

Sentence removed

Figure 6: remove volume from "volume mixing ratio". You are reporting dry air mole fractions here, is this true?

Done.

L 227: keep Figure 7 (and Figure 3) singular to avoid confusion. The figures have 4 subplots.

Done.

L 258-260: "In a lot of regions most of the emissions from BB are from the two boreal regions (Boreal America and Boreal Asia), which is probably due to the higher emissions height of those fires increasing the probability of the emitted CO to reach the UT."

Modified (lines 320-322): Most of the BB contributions are from the two boreal regions (Boreal America and Boreal Asia), which is probably due to the higher emissions height of those fires increasing the probability of the emitted CO reaching the UT [Dentener et al., 2006].

L 267: replace WNam with NWAm. Also simplify by splitting this sentence into two. One is about anomalies attributed to CEAS emissions and the other sentence is about CO anthropogenic emissions (if I understand correctly).

Done.

L 302-305: About the 2015 fires in Eq Asia. Can you add references? For ex: https://www.pnas.org/doi/ful

Done line 383.

Also fix typo: "caracterized" should be "characterized"

Done

L 308: "The anomalies measured during the months MAM have similar characteristics than to the anomalies from DJF but this time..."

Sentence removed

L 321: replace "The Gulf of guinea" with "the Gulf of Guinea"

Done

L 326. Remove "Obviously". It is rarely used in scientific writing, to my knowledge.

Done.

L 328: "most of its detected anomalies are attributed to emissions from local fires."

Done line 421.

L 342: "Fig.13 and Fig.14 show..."

Done L433.

L 355 and 367: Replace "wild fires" with "wildfires" "Wildfires" was misused here and we replaced it with "fires" L 372-373: Use uppercase for G in gulf, "gulf of Guinea" appears twice in this sentence. Same for L 387.

The term "wildfires" has been replaced by "fires" in the revised manuscript.

2.5 Conclusion:

L 403: Fix regions acronyms to be consistent with earlier ones. "NWam, NEam and Weur" should be NW Am, NE Am and Eur, I presume.

Done line 504.

L 417-418: Fix repetition in the sentence " This transport of pollution to Northeast Siberia is partly due to the East Asian monsoon, which transports air masses from Southeast Asia to Northeast Siberia."

Ok line 531.

L 438: fix typo: " the emissions (both atnthropogenic and BB)"

Done.

L 452 "observed with a thresholds defined as the 75th or 99th", singular for threshold.

Done line 586.

Fix the end of the conclusion: Remove the paragraph L 458-461 as it is repeated with an improved sentence for the ozone piece in the last paragraph.

Done.

Remove "obviously".

Done.

Be more specific. What specifically would you want to further study in those high CO plumes and therefore what measurements or "data" would you need?

The perspective paragraph of the conclusion has been updated in the revised manuscript (lines 605-616):

These O_3 values give information on its possible production in polluted plumes. However, without the measurements of additional chemical compounds (like VOCs and NOx for example) it is difficult to draw robust conclusions. To go further into the analysis on the O_3 in pollution plumes, information on more chemical compounds are required. The current perspective is to carry a similar study with a Chemistry Transport Model in order to get further information on the provenance of O_3 values but also on the amount of O_3 productions in polluted plumes, especially in regions with elevated values of O_3 like Siberia and the Middle East.

We have presented a detailed analysis of the characteristics of high carbon monoxide plumes and their associated ozone anomalies in different regions of the world. It is important for the IAGOS infrastructure to continue those measurements and to expand the regions sampled by the research infrastructure in order to provide these diagnostics in additional regions. This is particularly important in tropical regions, where anthropogenic emissions are increasing and impact on the O_3 trend globally [Zhang et al., 2016]. Increased number and sampling frequency of measurements of NOx and aerosols by IAGOS will be available and valuable for future analysis focusing on O_3 photochemical production or air quality.

2.6 Appendix

Figure A1: fix title and caption " Number of flights per regions" region should be singular

Done.

You have 3 supplementary figures A1, D1 and E1. I do not understand the A1,D1, E1 choice for naming those figures. Fig. B1 and Fig. C1 are showing up after the references so maybe make sure they are in order and the number 1 for A1, B1 etc seems unnecessary, unless it is how the journal asks for these supplementary figures to be labeled.

The Figures are now correctly placed before the references.

References

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