Review response

August 2024

1 General Comments:

Punctuation is confusing in many instances, mainly due to missing or misplaced commas and missing spaces.
Upper and lower case writing is often confusing and sometimes seems random, e.g. for region names, biomass burning, ...
Usage of the word "important" does not make sense in most instances. Please check all occurrences.

Thank you for the comment. A thorough proofreading has been made and the word "important" have been deleted or replaced in the manuscript.

The conclusions section has become rather long. I suggest to try to make it more concise and more clearly work out some key points rather than repeating all parts of the data discussion. The newly added paragraph on the data analysis being repeated with a shorter time series should be moved somewhere else. The same holds for the sensitivity test with the 75 or 99 percentiles.

Thank you for your comment, the conclusion has been changed and shortened. The paragraphs on the sensitivity test for the different time periods and different thresholds has been moved in the result section under a new subsection called "Discussion on sensitivity test analysis".

You can find below the new section "Discussion on sensitivity test analysis" (lines 485-496) :

Our study is based on extreme CO mixing ratios, which we have defined as observations above the regional and seasonal 95th percentile. In order to ensure the robustness of the results with respect to this parameter, we performed a sensitivity test to check whether any major changes in the features could be observed with a threshold defined as the 75th or 99th percentile. Overall, the same characteristics were observed with just a few differences. In the northern hemisphere, increasing the threshold causes a slight increase in the proportion of fire-related plumes (diagnosed as BB or MIX), which is not surprising as we have seen that these plumes are the most intense most of the time.

Our study focused on CO anomalies measured between 2002 to 2019, but important trends in CO and O_3 in the atmosphere have been observed in several of the studied regions (e.g. Novelli et al. [1998], Kim et al. [2023], Gaudel et al. [2020]). So, we performed the same analysis with only the last 10 years of the IAGOS measurements. Several regions, showed a decreased 95th percentile in this datasets (see tables **??** and **??** in the appendix). However, the origins and sources of the anomalies remain similar in regions with sufficient number of data. The conclusion of the study remained largely unchanged for the CO anomalies of the last 10 years.

You can find below the shortened conclusion (lines 498-544) :

IAGOS is a research infrastructure which uses commercial aircraft to measure atmospheric composition. In total, over the 18 years of measurements, more than 43,000 equipped IAGOS flights were made. This study is based on the in-situ IAGOS CO anomalies defined as the observations above the 95th percentile of each individual altitude range (LT, MT, or UT), region and season. In addition, SOFT-IO allows us to give a diagnosis on the main type of source as well as the region of emission responsible for the detected CO anomalies. SOFT-IO is based on FLEXPART retro-plumes initiated at each IAGOS measurement point. The back trajectory ensembles are then coupled to two CO emissions inventories : GFAS for the biomass burning and CEDS for anthropogenic emissions. The conclusions below relate only to CO anomalies (values above the 95th percentile of the region/season/altitude).

In the northern mid-latitudes, anthropogenic emissions peak in winter, and biomass burning emissions peak in summer. In the LT, the anomalies are strongly influenced by the higher local emissions during the winter months as well as the weak convection and low photochemical activity, which participate in the accumulation of the CO during this season. O_3 values in the CO anomalies of this season are between 8 and 23 pbb lower than the median in the different regions. In summer, at this altitude, there is significant regional variations which probably highlight the local environment more or less prone to O_3 production/destruction.

In the MT, the high CO plumes over NW and NE America, Europe in JJA are mainly due to boreal fire emissions. Those fires originate either from boreal Asia or boreal America. High CO plumes from anthropogenic origins still account for a significant proportion of the anomalies, but unlike the LT, the origins of these emissions are split between a local and a long-range influence. East Asia continues to be dominated by anthropogenic pollution throughout the year and to show the higher CO levels observed at this altitude in the northern hemisphere. O_3 in the MT in JJA, shows higher values than normal in the CO anomalies (from 7 to 9 ppb higher than the median).

In the UT, in northern mid-latitudes, anomalies caused by BB emissions remain less frequent than those from anthropogenic emissions, but they are the most intense during the boreal fire season (JJA). East asian emissions strongly influence the anomalies observed in the different regions of the northern hemisphere. The O_3 mixing ratios associated with CO anomalies are regularly higher than normal and O_3 reaches its maximum over Siberia. So, the exported CO plumes from Siberia will cause high anomalies of O_3 in the regions with lower O_3 environment.

The LT and MT of the Indian subcontinent are mostly influenced by the local anthropogenic emissions. In the UT however, the pollution pattern is dominated by the phases of the monsoon. In DJF and MAM, a significant contribution is coming from anthropogenic and BB emissions from Northern Africa. In JJA, strong convective activity favours the export of local pollution to the UT and most plumes are therefore attributed to local anthropogenic emissions. In SON, the UT above India can be strongly affected by BB emissions from in the maritime continent.

CO anomalies in the African troposphere follow a different regime. Fires are much more frequent and are responsible for a large proportion of the CO anomalies, even in the lower layers of the troposphere. The CO anomalies observed throughout the troposphere over Africa are deeply influenced by transition between the wet and dry seasons and the ITCZ shift which changes the transport patterns of the emitted CO. A part of the CO can also come from southern Asian emissions where it has been transported upward by the South Asian monsoon and trapped in the South Asian monsoon anticyclone before being transported westwards and influence the northern part of Africa and the Middle East. In the LT and MT, the maximum O_3 values are found above Middle East. Previous studies assumed that the high O_3 in the regions were due to long range transport of polluted airmasses followed by chemical production in the regions [Li et al., 2001, Duncan et al., 2008]. In the African regions, O_3 levels are maximum during their respective dry season and the CO anomalies associated with BB emissions show enhanced values of O_3 (22 ppb and 28 ppb higher than the median respectively for the Gulf of Guinea and Southern Africa in the MT).

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 [**?**]. 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 Specific comments:

L6 "maximum are" does not make sense.

Corrected: In the Northern Hemisphere, CO reaches its maximum values in DJF in the lower troposphere, which can be attributed to elevated anthropogenic emissions and reduced convective activity during the season.

L10 "the the"

Corrected.

L11 What does previous refer to?

Corrected : The middle troposphere is a combination of the characteristics of the LT and the UT, with contributions from both local emissions and long-range transport.

L16 regions should be singular

Corrected.

L15/17 "drastically" is a somwhat strong an unscientific choice of wording here.

Deleted/Changed to "strongly".

L26 "their" seems to link peak values and climate impact to the models which does not make sense.

Corrected :"their climate impact" deleted.

L35 "is" should likely be "are"?

Corrected.

L51 belt -¿ belts

Corrected.

L58 What do you meen by "one of the only O3 precursors"? Is it one of several ozone precursors or the only one?

Corrected : "CO is a precursor to O_3 with a chemical lifetime long enough to reach the UT".

L72 which - who ?

Corrected.

L146 SOFT-IO should be upper case

Corrected.

L153 Using a cut-off value of 2 PVU for the UT will not completely remove stratospherically influenced air masses from the data set. There may be air masses with elevated ozone left, reflecting stratospheric influence which might at high altitude cause a bias in the background data. Since *ozone is not the focus of this study, this might be neglegible, but should be taken into account when studying ozone further (see also work by L. Millán et al. within the OCTAVE project). there could very well be a CO anomaly in the UTLS or even the lowermost stratsophere as was for example prominently seen during the extreme 2019-2020 Australian bushfire season.*

There is a statement regarding the possible contamination of the anomalies by stratospheric airmasses (lines 336-339) in the results section.

L162 line -¿ lines

Corrected.

L192 showed -¿ shown

Corrected.

L197 Please give a range of values here to give the reader an idea what the statistical relevance is without the need to consult the appendix.

Corrected : Depending on the altitude layer between 49 and 2186 anomalies have been observed per region/season/altitude layer (table A4).

L198 Is it relevant that the selection is repeated for each flight? The ditribution of datapoint across pint is to some extent random but should not matter for the analysis outcome and result shoule remain the same even if the attribution of data to a certain flight is not considered.

The Methodology is repeated for each flight (with the same thresholds for each region/saison/altitude layer), but if you think that this sentence is ambiguous, I suggest to remove it.

L219 Why is this statement in a single paragraph?

Changed position of the statement (line 221) : IAGOS observation are in the LT similar to urban background stations [Petetin et al., 2018]. So as expected, anthropogenic contributions have a strong local influence in this layer (Fig. 3.c). For example, anthropogenic contributions are almost entirely from local sources in NW America, NE America and Europe in the LT.

L229-232 This sentence is very complicate and difficult to read. "than the typical west to east (...) transport" does not make sense.

Changed to :

Inter-continental transport generally needs no more than a few days in the in the middle troposphere of the northern hemisphere because of the stronger prevailing winds there [Jaffe et al., 1999, Liang et al., 2007]. Polluted airmasses can also be transported for long distances at lower altitudes, or sink in the Boundary layer (BL) after being transported at higher altitudes, but it generally requires a few additional days [Stohl et al., 2002]. (lines 223-227)

L233 into -¿ in

Corrected.

L244 What are you referring to by "this altitude"? The LT?

Changed to "In the LT".

L257 How can O3 values be "important"?

Corrected into "elevated O3 values".

L263 double occurrence of "CO distribution"

Corrected.

L265 (...) are Warm Conveyor Belts (WCB) (...)

Corrected.

L266/267/274 How can winds or export of pollutants be "important"?

Corrected to strong/significant/significant.

L273/274 Check grammar.

Changed to : The upwind continent is Europe and there is no efficient vertical transport pathways over this continent. Therefore, it is not prone to export its pollution. By contrast, East Asia is one of the regions with the most efficient vertical transport [Stohl et al., 2002]. (lines 267-269)

L352 "portion (...) is"

Corrected.

L364/380/386/387/463/465 "important"?

The word "important" has been replaced by "significant/strong/high" or deleted when not necessary.

L376-380 This is a very long sentence.

Changed to : It is also the period of the winter monsoon in Southern Asia. This season is characterised by week convective activity and northern prevailing wind transporting pollution at low altitude toward the Indian ocean [Lelieveld et al., 2001, Lawrence and Lelieveld, 2010]. Consequently, explaining the relatively high values of CO in the LT and MT during this period (Fig.D1 and D2 in the appendix) as well as the low contribution from SEAS in the UT. In the UT, the anthropogenic CO anomalies receive an influence from CEAS and SEAS but also from NHAF. (lines 368-372)

L389 What do you mean by "and obtained by a radiosonde". Data in Lal et 2014?

Deleted.

L468/469 Abbreviations ASM and AMA seem not to be used afterwards, no need to define them.

Corrected.

L473 ratio - ratios

Corrected.

L475 extreme what?

Sentence changed : The signal observed in the climatologies studied by Lannuque et al. [2021] and in the CO anomalies studied here show similarity.

L497 Text says 19 years earlier.

Corrected.

L504 these - the?

Deleted.

L506 What do you mean by "median on average" - the average of the medians of all regions?

Sentence deleted.

L520 Which regions does "of the regions" refer to?

Sentence deleted.

L530/537 important

Changed to "significant" or deleted .

L 532 "no no"

Corrected.

L571 What do you mean by "must be respected"?

Corrected to : "is necessary".

Tables and Figures

Some of the figure titles are confusing, single panel figures, such as Fig 1, 2, 4 ..., do not need titles, but all the explanation should be in the caption.

Title deleted for each single panel figure.

Punctuation of captions is inconsistent, please use "." in the end.

Corrected.

Figure 2: what do you mean by "on the day/year" in the caption?

Replaced by "the 7h of June 2013".

Figure 3 (and analogous figures): Numbers in ppb are not concentrations but mole fractions or mixing ratios; please correct title of panel b

Table A1: upper/lower case in full name is not logic. TENA is explained with an upper case E in TEmperate but BONA is not expanded analogously.

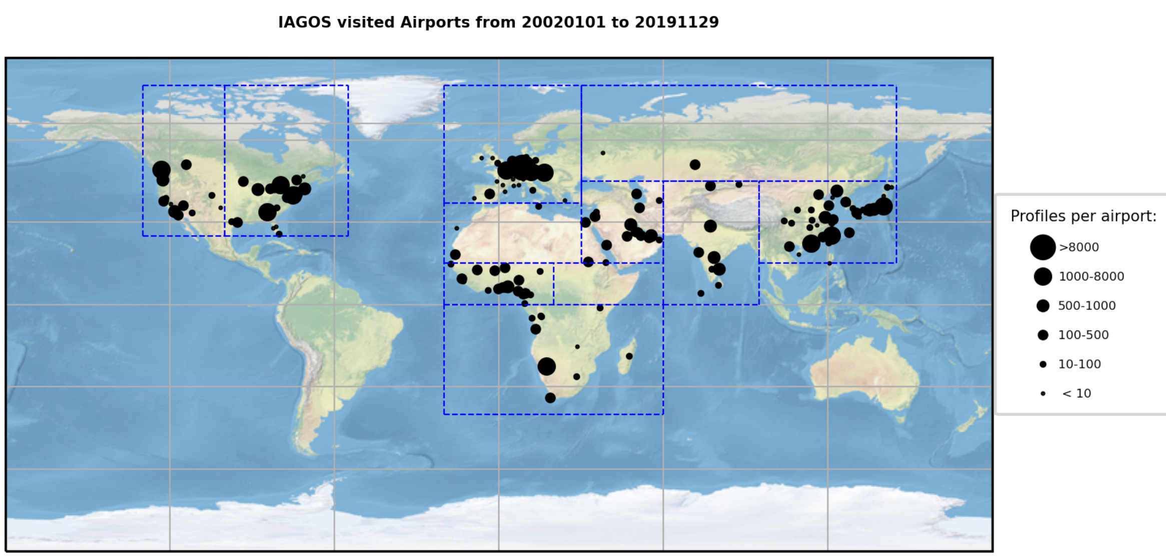
Corrected.

Figure B2: Singular/plural in caption

Corrected: "Trajectories of every IAGOS flight".

Figure B3: Please expand caption and fully explain what is shown. Does the symbol size stand for the number of profiles near one airport? Please add a label.

Corrected see figure below.



Number of profiles recorded per visited airport by IAGOS aircrafts.

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