Review response 1

June 2024

Referee comment on "Seasonal, regional and vertical characteristics of high carbon monoxide plumes along with their associated ozone anomalies as seen by IAGOS between 2002 and 2019" by Lebourgeois et al. This manuscript provides a statistical analysis of extreme CO values of IAGOS database for different regions, seasons and vertical layers (lower/middle/high troposphere), both in terms of origin (using SOFT-IO software) and in terms of impact on O3 production. I found the manuscript to be well organised and clearly written. The methods used are scientifically sound and the figures chosen appropriately support the discussion and conclusions. The results provide an important overview of the CO plumes observed over 18 years of in situ measurements. However, I find that there could be more links to the main processes involved, as well as a fuller discussion of how the results align with recent literature, including key publications using the IAGOS datasets that are cited in the article (but not only). A discussion of how representative the in situ data used is of each major region should also be added, since I don't think that the data are randomly distributed within each region/layer.

We thank the reviewer for her/his comments that will help improving our study. We respond below to each specific point.

1 Introduction

I don't really see the relevance of the paragraph on satellite observations to this paper. However, a more precise description of previous results using CO and O3 observation datasets (in particular IAGOS) would be useful to better put the main results of this paper into perspective (in introduction and to discuss the results).

We thank the reviewer for this comment and as suggested, the following paragraphs in blue have been added in the introduction section (lines 71-83 in the revised manuscript):

Some studies have used the IAGOS database to study the characteristics of CO and O_3 values in the troposphere and lower stratosphere. This is the case for Cohen et al. [2018], which used this dataset to study the climatology and trends in O3 and CO in the UTLS. Petetin et al. [2018b], Lannuque et al. [2021], Tsivlidou et al. [2022] used IAGOS to study the characteristics of CO in different regions or altitude layers of the world. Tsivlidou et al. [2022] studied CO and O3 characteristics in the tropical regions. She highlighted the origins of the CO in the different regions of the tropics. She specifically showed the importance of the Anthropogenic emissions to explain the values of CO in the tropical troposphere. Lannuque et al. [2021] studied the meridional distribution of O3 and CO over Africa using IAGOS and the satellite IASI (Infrared Atmospheric Sounding Interferometer). They showed the importance of the ITCZ and the upper branch of the Hadley cell for the redistribution of the pollutants over Africa. The Pollutant emitted at the surface is transported by trade winds toward the ITCZ where it is transported to the UT and redistributed to higher latitude by the Hadley cell. Petetin et al. [2018b] studied the CO vertical profile over different airport clusters. They characterised their seasonal profile as well as the seasonality of the highest CO anomalies 95 and 99 percentile. They showed a strong seasonal variability of the most extreme anomalies in northern America which were due to BB emissions. He also looked at the origins of the CO responsible for the CO anomalies at the different airport clusters.

2 Methods

2.2: The description of the SOFT-IO software should include a paragraph on performance and uncertainties (in emission inventories and attribution using back trajectories). The important warning in § 122-127 could be written more clearly, and discussion of the performance in attribution in past studies could be helpful.

Thank you for this comment that will improve the manuscript. The performance and uncertainties of SOFT-IO are described in Sauvage et al. [2017], Tsivlidou et al. [2022]. In order to provide key information in this study, the following paragraph has been added in the revised version, lines 142-148 :

Sauvage et al. [2017] and Tsivlidou et al. [2022] made a thorough statistical evaluation of SOFT-IO. The model had a really good score in the detection frequency of the CO anomalies (above 93% on average). Detection frequency was at its maximum in the LT as most anomalies are from local emissions at this altitude. In the MT and UT the scores were lower but remained above 80% as the simulation of horizontal and vertical transport could suffer some errors. It is important to note that the study presented here aims at using SOFT-IO only as a qualitative tool to attribute a source type and a relative geographical origin to the emissions leading to the detected anomalies. SOFT-IO is a model which has already been used in several studies similar to the current study (e.g Petetin et al. [2018b], Cussac et al. [2020], Lannuque et al. [2021], Tsivlidou et al. [2022]).

2.3.1: The data is divided into large regions for analysis. It would be important to discuss the location of the data analysed within each region and for each layer. If I understand the data correctly, the profiles correspond to specific airports and the flight paths also follow specific routes. I think it would be important to better discuss the representativeness of the data set for each region/layer.

Thank you for the comment. As you advised, we added a map showing the position of the flight trajectories done by the IAGOS aircraft in each region. We have added the following paragraph (lines: 189-192) in the revised version :

IAGOS samples the lower and free troposphere during landing and take-off. Petetin et al. [2018a] showed that close to the surface, the IAGOS measurements are representative of urban areas and provide similar measurements to urban background stations. At higher altitudes, in the free troposphere, the samples are less influenced by local emissions and therefore are representative of regional background conditions following the flight tracks showed in Fig.B2 in the appendix.

A figure showing the flight tracks of every IAGOS flight has been added to the appendix. This corresponds to the cruise parts of the flights in the UT. Regarding LT and MT, data correspond to the vertical profiles part of the flight during take-off and landing over visited airports (the second map). Note that the average horizontal distance between airport surface and the 8 km altitude is about 300 km [Petetin et al., 2018a].

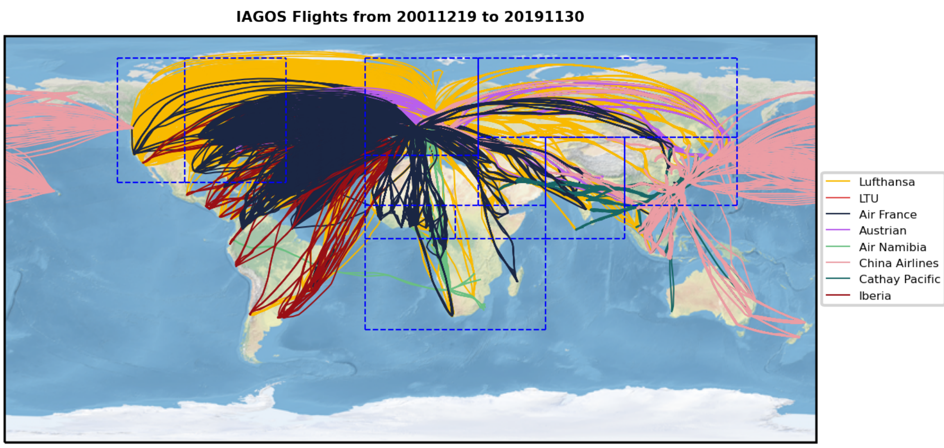

Figure 1: Trajectories of every IAGOS flights

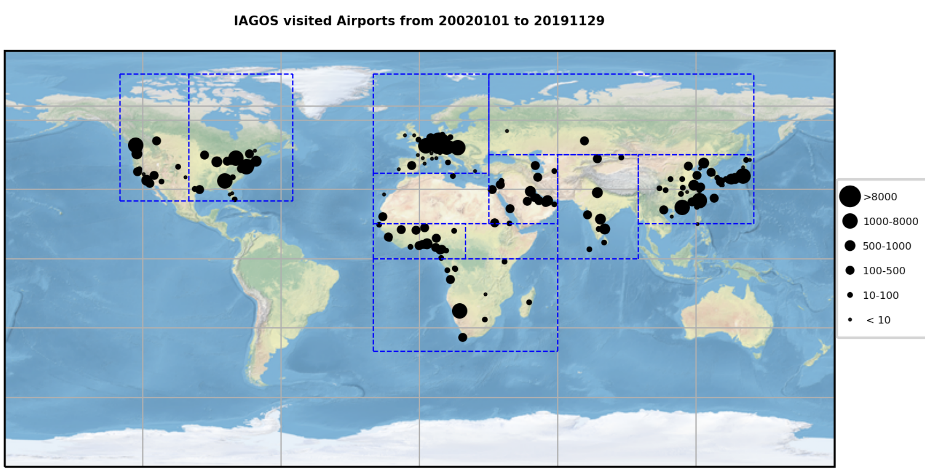

Figure 2: Map of the visited airports by IAGOS aircrafts

2.3.2: For the type of source attributed to the anomalies detected (l.175-180) , why did you choose to use the main characteristic and why not add the fractional contributions? Is it because *of too large uncertainties in attribution with SOFT-IO?*

As there are, by nature, anthropogenic emissions in almost every single plume, the objective of the methodology was to characterize if biomass burning plumes are dominant (i.e. >50%) or not (MIX, and of course ANT). That is the reason why we compile the main characteristics. The objective of this study is to see what is the dominant source in influencing CO anomalies.

3 Results

For all regions and layers, I was wondering what fraction of the detected anomalies are successfully attributed to a main source using SOFT-IO?

Thank you for the comment. As you advised in the methods part of the comment we have added a paragraph on the uncertainty of SOFT-IO from the previous study of Sauvage et al. [2017], Tsivlidou et al. [2022]. According to these studies on average 93% of the anomalies observed by IAGOS are also detected by SOFT-IO.

The seasonal variations in the LT are mainly attributed to variations in the local sources. Does that mean that seasonal variations in the background levels has little impact? Even if anomalies are selected, the final concentration is an enhancement above background, both for CO and for O3.

Yes, CO anomalies are selected and calculated over a regional and seasonal background. So, yes the seasonal variations are caused by variations in the local sources but also by changes in the background values (caused by various factors like chemical lifetime, higher emissions and less mixing). There is no selection of Ozone anomalies. The Ozone distributions are those sampled within the CO anomalies.

l. 207: The authors mention "a cycle of O3 destruction in CO-rich air masses": O3 is then lower than background in the corresponding area? It would be helpful to add some detail on the corresponding chemical processes (same comment for all regions). What could be the impact of other co-emitted compound such as aerosols? I understand that this is beyond of the scope of this paper but for each region, it would be helpful to have some reference to the literature on the subject.

Thank you for the comment. The following references have been added :

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

- Cohen et al. [2018]
- Nowak et al. [2004]
- Hudman et al. [2004]

The following paragraph regarding Ozone in the paper have been modified giving thus further clarification.

- In the LT in DJF our results are similar regardless of the region. We observe values of O_3 inside the CO anomalies close to the minima of the seasonal O_3 cycle. We can see that, in addition to the low photochemical activity linked to the boreal winter, we are seeing a cycle of O_3 destruction in the CO-rich fresh air masses. These low values of O_3 in polluted urban air mass are often characteristic with NO titration (e.g. Yang et al. [2019]).

  In JJA, the mean O_3 mixing ratios in the CO anomalies are closer to the median. However, there are strong regional variations showing the important local influence at this altitude. East Asia is a region with important O_3 values and a region having frequent high O_3 episodes [Chang et al., 2017b, Lu et al., 2018]. In this region anthropogenic CO anomalies are also associated with important O_3 values (20 ppb above the median). Lines (251-257)

- Fig.6 shows the mixing ratio of O_3 associated with high values of CO. In the MT there is almost no signal during the winter months (mixing ratio of O_3 inside CO anomalies is close or below the median) because of the relatively weak photochemical activity. In JJA, the O_3 mixing ratio within the CO anomalies is between the median and the 75th percentile of the total O_3 distribution, so the mixing ratio of O_3 in the CO plume is on average 5 to 10 ppb higher than the median values depending on the region. In East Asia, BB (and mixed) plumes are rare and mostly come from Boreal North Asia. O_3 values within those plumes are 20 ppb higher than the median and 10 ppb higher than the plumes from anthropogenic emissions. Lines (281-286)

- Previous studies already noticed the O_3 maximum over Siberia [Gaudel et al., 2018]. [Cohen et al., 2018] suggested that this maximum could be due to a higher stratospheric influence over the region.On average for the other regions, O_3 mixing ratios in CO anomalies are 13 ppb higher than their respective median and this difference can reach 21 ppb for the CO anomalies associated with Biomass Burning emissions. [...] Production or elevated values of O_3 during the transport of polluted plumes from East Asia have already been observed during the Intercontinental Transport and Chemical Transformation 2002 campaign (ITCT 2K2) [Nowak et al., 2004, Hudman et al., 2004], so similar processes could be at play here. Lines (350-356/365-367).

- The O_3 cycle shown here is similar to the cycle described in Lal et al. [2014] and obtained by a radiosonde, here the focus is on the O3 measured in the CO anomalies. In the LT, the minimum values of O_3 are reached during the summer monsoon in JJA. The low values can be explained by the increased marine influence during this period [Lawrence and Lelieveld, 2010]. At this altitude the O_3 values recorded simultaneously as the CO anomalies are low and show the low O_3 production in those plumes.

In the MT and UT, the maximum of the O_3 is reached during MAM, and the minimum is reached during DJF. In the UT, in DJF and MAM an important part of the CO anomalies come from northern African BB. These plumes are associated with higher values of O_3 (11 and 10 ppb above the median respectively for DJF and MAM). CO anomalies in JJA are caused by the local emission of anthropogenic CO rapidly transported to the UT by the important convective activity of the South Asian Summer Monsoon (SAMA). This rapid transport could explain that the associated values of O_3 are close to the median (65 ppb). In the post monsoon season (SON) BB anomalies from Equatorial Asia are added to the local anthropogenic anomalies. The values of O_3 in the BB plumes are low and close to the 25th percentile (44 ppb) which is explained by the lower background values of O_3 in Equatorial Asia compared to India [Cohen et al., 2018]. Lines (389-401).

- In the Middle East, O_3 values are among the highest in JJASO in the LT and MT. The summertime median is also higher than the median from East Asia (see Fig.13 and Fig.14) which is a region with identified extreme O_3 values [Chang et al., 2017a, Lu et al., 2018]. Li et al. [2001] suggested that the important tropospheric O_3 in Middle East were due to the constant import of pollution from different regions trapped in the upper level anticyclone and the strong subsidence associated to it cause an accumulation in the region. Here the CO anomalies detected are mostly caused by emissions from the Middle east rather than from long range transport. In the Middle East LT, values of O_3 inside CO anomalies attributed to anthropogenic emissions are lower than the 25th percentile, which is similar to the observation made on the northern hemisphere mid-latitudes. In the MT, the anthropogenic anomalies are close to the median during both seasons. Lines (440-447)

- Middle East shows the highest values of O_3 during JJASO. At this altitude layer CO anomalies are mostly from anthropogenic emissions originating from SEAS. Those anomalies show a 7 ppb enhancement compared with the median of 70 ppb. This is in agreement with a previous study from Li et al. [2001] showing elevated O_3 values in Middle East due to important import of anthropogenic pollution from polluted regions and very little from stratospheric intrusion. Middle East meteorological conditions are favourable for O_3 production [Duncan et al., 2008] as well as a constant important of pollutant Asian emissions [Stohl et al., 2002] as well as influx of NOx produced by lightning during the Asian monsoon Li et al. [2001]. Lines (482-488)

Reviewer is right that a complete and more in-depth analysis on the ozone distributions is beyond the scope of this paper. This is the first step of this statistical characterisation, with diagnostics on CO anomalies along with their ozone content. That gives material and diagnostics for additional studies with global models (CTM) to synthesize and integrate all the processes leading to ozone formation.

As mentioned in the general comments, it would be important to better discuss the results obtained in light of the literature, in terms of source contributions but also in terms of O3 enhancement in CO enriched air masses.

Thank you for the comment, we made important modification and added a lot of references in the results section of the revised manuscript in order to better discuss the result in light of the literature :

- Bergman et al. [2013]
- Chang et al. [2017a]
- Cohen et al. [2018]
- Cooper and Parrish [2004]
- Cooper et al. [2004]
- Dentener et al. [2006]
- Ding et al. [2009]
- Duncan et al. [2008]
- Field et al. [2016]
- Gaudel et al. [2018]
- Huang et al. [2012]
- Huntrieser and Schlager [2004]
- Jaffe et al. [1999]
- Kar et al. [2004]
- Lal et al. [2014]
- Lawrence and Lelieveld [2010]
- Lawrence [2004]
- Lelieveld et al. [2001]
- Li et al. [2002]
- Li et al. [2001]
- Liang et al. [2007]
- Lu et al. [2018]
- [Novelli et al., 1998]
- Pan et al. [2016]
- Sauvage et al. [2005]

- Stohl [2001]
- Stohl et al. [2002]
- Yang et al. [2019]

The following paragraphs in the revised manuscript have been modified :

- It is in agreement with the fact that inter-continental transport impacts mostly the Free Troposphere because of the stronger prevailing winds there. Long-range transport can also happen 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] than the typical west to east intercontinental transport which generally needs no more than a few days in the middle troposphere of the Northern Hemisphere [Jaffe et al., 1999, Liang et al., 2007].

  Most of the European pollution is exported via low altitude pathways, and can impact the concentration of CO into the LT of Eastern Asia North America and Northern Africa [Huntrieser and Schlager, 2004, Duncan et al., 2008, Li et al., 2002]. However those contributions in North America and East Asia are generally low compared with the mixing ratio of CO in the LT of those regions. Here, we are interested in the extreme values at the surface close to the major airports of the region (and therefore close to urbanised areas) so the low contributions from Europe are of minor importance but could have more impact in more remote parts of Asia. Lines (228-238)

- At higher altitudes, the measured CO is less influenced by the local conditions and emissions close to the sampled airport. This altitude layer is also more impacted by long-range transport as the strong westerly winds present in the Free troposphere (Middle and upper troposphere) allow a rapid transport of the polluted air-masses across the hemisphere [Jaffe et al., 1999, Stohl et al., 2002, Liang et al., 2007]. Lines (259-262)

- In this layer of the atmosphere, the local influence in the anthropogenic contributions (Fig.5.c) is still strong. Well known efficient processes for long-range transport of pollution are the Warm conveyor Belt (WCB) and frontal systems (e.g. [Cooper et al., 2004, Ding et al., 2009]) which can transport polluted surface air-masses to higher altitudes where important winds (e.g. jet stream at mid-latitude) can rapidly transport those air-masses to another continent. So, in general, there is important export of the pollutant from the regions at the western part of an ocean (start of the WCB) and the continent in the eastern part of the ocean will be the receptor (Europe and Western America) [Stohl et al., 2002, Huntrieser and Schlager, 2004, Cooper and Parrish, 2004]. This feature is well captured by SOFT-IO where we can see that an important part of the contribution in NW America is coming from Eastern Asia. It is also true for Europe where more than half of the contributions are coming from either North America or Asia. We can also see the lower contribution from long range transport in summer when the WCB is weaker [Cooper and Parrish, 2004]. East Asia is mostly impacted by its own pollution during the two seasons. The upwind continent is Europe which is not known for having efficient vertical transport processes and so being prone to important export of its pollution [Stohl, 2001]. East Asia on the contrary is one of the regions with the most efficient vertical transport [Stohl et al., 2002]. In JJA, BB contributions come mostly from Boreal America and Asia. Most of the time, the airports sampled by IAGOS are further south than most of the intense boreal fires. So, it is not surprising that little influence of the BB is detected in the LT. However, the influence from BB grows with altitude. In the MT, we observe an increased number of episodes attributed to either BB emissions or mixed sources in the MT of America and Europe in JJA (Fig.5.b). Fig.5.a shows that the plumes attributed to BB emissions are the most intense in JJA. Lines (263-280)

- The most polluted air-masses in the UT are often rapidly transported upward after their emission [Huang et al., 2012]. Among the emitting regions, Eastern Asia is one of the more prone to vertical uplift of its pollutants because of the important convective activity of the regions (WCB, east asian monsoon...) [Stohl et al., 2002] and the presence of the Tibetan plateau, which can play an important role by lofting polluted air mass into the upper part of the troposphere [Bergman et al., 2013, Pan et al., 2016]. Once in the UT, those air mass can be transported aroud the hemisphere, which can be seen by the anthropogenic contribution from SOFT-IO where CEAS alone accounts for at least 40% of the anthropogenic contribution in the different regions and even reaches 79% in NW Am. The total emissions of CEAS during this period account for about half of the northern hemisphere emissions.North America benefits also from important vertical uplift with the deep convection and mid-latitude cyclone starting in the regions [Cooper and Parrish, 2004]. North American emissions represent approximately 15% of the northern hemisphere emissions but only 14% of the modelled anthropogenic contributions in the different regions. The European region in contrast, is identified to have few vertical uplift pathways [Huntrieser and Schlager, 2004]. Lines (329-338)

- It is also the period of the winter monsoon in Southern Asia, this season is characterised by weak convective activity and Northern prevailing wind transporting pollution at low altitude toward the Indian ocean [Lelieveld et al., 2001, Lawrence and Lelieveld, 2010] and explaining the rather high values of CO in the LT and MT during this period and the low contribution from SEAS in the UT, at this altitude the anthropogenic CO anomalies receive an influence from CEAS and SEAS but also from NHAF. In JJA, it is the wet phase of the monsoon in India so the important convective activity and precipitation associated with this period [Kar et al., 2004] allow the rapid transport of the south-asian emission to the UT while preventing BB: almost all the CO anomalies are caused by anthropogenic emissions from India or the close proximity (SEAS and CEAS). In SON, the CO anomalies are at their maximum and are caused by anthropogenic emissions from SEAS and CEAS but also by BB emissions from EQAS. The BB anomalies are clearly the most intense during this season. It is interesting to note that in the vast majority the BB anomalies recorded by IAGOS during SON were from 2015. This year was hit by an important El Niño phenomenon characterised by especially intense fires over the Equatorial part of Asia [Field et al., 2016]. According to Kar et al. [2004], during this season in 2002 there was also important transport of CO from tropical fires. Lines (376-388).

- DJFM is the dry season in the Northern part of Africa, which causes high levels of CO from biomass burning emissions (see Fig.11.c). In the Gulf of Guinea, the maximum values of CO are reached during DJFM, which come from the important Biomass burning episode of the region during this season. It is also a region with a large population which explains the important anthropogenic contribution. In this region Lagos airport is the most visited by IAGOS flights and the accumulation of the pollutant observed in the LT during this season has already been characterised in Sauvage et al. [2005] and is caused by the Harmattan winds bringing rich CO air masses caused by the upwind fires. In JJASO, the southwesterly trade winds bring air-masses from the Atlantic ocean. These air-masses are cleaner with respect to anthropogenic pollution but can bring BB plumes from Southern Africa. Lines (408-416)

- The Middle East plumes have a high contribution from anthropogenic emissions in both seasons in the LT and the MT. The Middle East has been identified in previous studies as receiving the pollution of multiple regions [Li et al., 2001, Stohl et al., 2002, Duncan et al., 2008]. Europe is mostly exporting its pollution via low altitude pathways and we can see on Fig.11.c and Fig. 12.c that up to 20 % of the anthropogenic contributions can come from Europe. There are also contributions from Temperate North America and South and East Asia, but contrary to the European contributions these probably followed higher altitude pathways before sinking to the MT or LT [Li et al., 2001, Stohl et al., 2002]. We can also see important differences in the provenance of the anthropogenic contributions between DJFM and JJASO. In JJASO, we are seeing contributions mostly from the local regions (MIDE) similarly to the contributions in the LT. According to previous studies the Planetary Boundary Layer in this region can reach 4000 or 5000 meters in JJA [Gamo, 1996, Ntoumos et al., 2023]. So, this differences in the origins of the contributions between DJFM and JJASO may be caused by the higher PBL height in JJASO. Lines (426-436)

Although carefully conducted, the analysis reads a bit like a list. Perhaps a summary scatterplot of O3 versus CO could be used to get a more general view of the data set? A colour code could be used, for example, to differentiate regions / layers, etc.

The ozone distributions are only from the tropospheric branches because stratospheric air-masses are discarded and only the air masses with extreme values of CO are selected. We believe the ozone box plots are more informative (see figure 3 below). So, after exploration of this idea, we think that the suggested scatter plots are not meaningful for a summary.

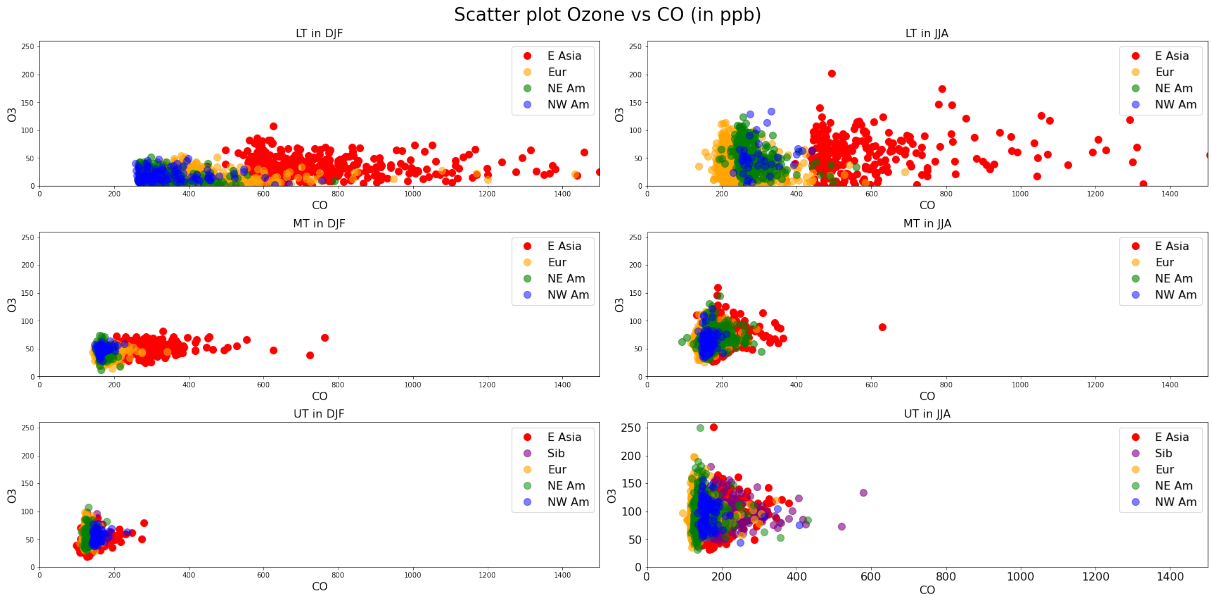

Figure 3: Scatter plot Ozone vs CO in the observed anomalies

4 Specific comments :

l. 51: Other O3 precursors have a long lifetime, CH4 for instance.

Corrected : "CO is one of the only with a long enough chemical lifetime"

l. 101: The paragraph on SOFT-IO should be included in the next section which is dedicated to the software.

Done Line 120.

l. 213: need reference for the larger number of convective events during summer.

Deleted.

l. 218: 'increased number of episodes. . . ' increased compared to what?

Sentence removed.

l. 244-245: again, increased compared to?

Corrected: "The mean mixing ratio of these episodes during the summer months also increased significantly compared to their winter values." (line 306)

Large increase in East Asia vs Siberia are attributed to quite different sources. What processes are at play? What transport pathways?

In East Asia the important summertime maximum is attributed to local emissions and the rapid vertical transport of the East Asian monsoon. In Siberia the maximum is both due to Boreal fires via pyroconvection [Nedelec et al., 2005] but also to anthropogenic emissions from East Asia transported by the east Asian monsoon.

Does that hold if not only the main features are kept, but the fractional contribution from different sources? Is the situation still that contrasted?

We tried both methods and no major differences were observed between the two methods.

l. 259-260: "which is probably due to the higher emission height..." Could this statement be checked? I agree that injection heights may be important here. What height is considered in SOFT-IO for source attributions?

Reference added [Dentener et al., 2006] (line 324). SOFT-IO uses emissions height given by GFAS (Rémy et al. [2017]). Those emissions heights are computed by the plume rise model from Paugam et al. [2015].

l. 315: Why is O3 particularly low in BB plumes for this region? Has this event (2015) been analysed in past publications (even using other methods)? In fact I also have the same question for other regions, such as African/ME BL, etc.

Those anomalies were from Tropical Asia and, according to Cohen et al 2018, it is a region with a lower O3 environment than India. Sentence added (line 400):

The values of O_3 in the BB plumes are low and close to the 25th percentile (44 ppb) which is explained by the lower background values of O_3 in Equatorial Asia compared to India [Cohen et al., 2018].

l. 367: Can BB be called 'wildfires' in this region? I would think there is significant contribution from agricultural burning as well. The use of the term 'wildfire' should probably be reviewed throughout the manuscript.

The term "wildfire" has been replace by "fire" in the revised manuscript.

l. 462: same phrases repeated.

Done

All figures for the statistical analyses: mention the total number of points in the subsets.

Good point, table added to the appendix (see table1 below).

Figures O3 (Fig 4, etc): What do colored dots represent? Average value? Why not show the full boxplot for each source? Not enough data?

		LT	FT	UT
NW Am	DJF	168	137	88
	JJA	66	87	133
NE Am	DJF	349	323	337
	JJA	409	589	1207
Eur	DJF	1192	1032	1180
	JJA	1701	1493	2186
Sib	DJF	no data	no data	181
	JJA	no data	no data	470
E Asia	DJF	480	944	1146
	JJA	415	711	937

		LT	FT	UT
India	DJF	150	164	414
	MAM	128	121	507
	JJA	155	141	890
	SON	123	155	417
North Af	DJFM	no data	no data	433
	JJASO	no data	no data	1285
Middle E	DJFM	404	275	338
	JJASO	432	330	1282
Gulf of G	DJFM	144	303	484
	JJASO	328	269	756
South Af	DJFM	79	148	367
	JJASO	49	179	713

Table 1: Number of observed anomalies for the different regions and seasons.

Yes the colored dot represents the average values. Yes, showing a boxplot different for each source would be challenging in some regions where not a lot of anomalies from a certain source are found.

References

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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 598-603): 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 604-609):

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 (496-497)

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 33.

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 244-246) 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 322-324): 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 387.

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 424.

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

Done L437.

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 508.

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 527.

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 581.

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 598-609):

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.

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Review Response 3

June 2024

1 Major Comments:

This manuscript provides a detailed analysis of the observed carbon monoxide mixing ratios above several regions sampled by the IAGOS program, and their associated emissions source regions. I find the analysis to be scientifically sound and the conclusions are consistent with the other papers submitted so far to the TOAR-II Community Special Issue. However, to increase this papers relevance to the TOAR-II effort I would like to recommend four areas for further analysis and discussion:

We thank the reviewer for her/his comments that will help improving our study. We respond below to each specific point.

1) There are some previous TOAR papers and other peer-reviewed studies that are relevant to this work and they should be cited (see papers referenced below).

Thank you, as advised the following citations have been added :

- Novelli et al. [1998]
- Kim et al. [2023]
- Gaudel et al. [2020]
- Gaudel et al. [2018]
- Lal et al. [2014]
- Lawrence and Lelieveld [2010]
- Lelieveld et al. [2001]
- Li et al. [2001]
- Nowak et al. [2004]
- Lu et al. [2018]

- Chang et al. [2017b]

2) This study touches on two important topics that have received little attention in the peer-reviewed literature, and the authors have an excellent opportunity to expand upon these topics. Specifically, these topics are the exceptionally high ozone mixing ratios in the UT above Siberia, and the high ozone levels in the lower, mid- and upper troposphere above the Middle East. Figure 5 (top left panel) of Gaudel et al. (2018) shows that the highest ozone values in the upper troposphere in June-July-August are found above Siberia. Figure 3c of Gaudel et al. (2020) shows that the ozone above the Middle East is even greater than ozone above China. These two regions have received relatively little attention in the peer-reviewed literature (an exception is Li et al., 2001). It would be helpful if these two regions can be given greater attention and highlighted in the abstract and conclusions as regions with exceptionally high ozone. Please elaborate on the potential contribution of biomass burning and anthropogenic emissions to the observed high ozone levels. The authors mention a potential contribution of the Asian summer monsoon to the high ozone levels above Siberia (i.e. the monsoon transports pollution from South and East Asia to Siberia). This is an excellent hypothesis and I think it should receive further attention.

Thank you for this comment that will improve the manuscript. As you noted, the two regions, Siberia and Middle-East present on average really high values of Ozone. As suggested these two regions will be highlighted and further discussed in the abstract, in the results section and in the conclusions (see below in blue).

- Among the studied regions, the troposphere above Middle-East and the UT of Siberia presented extremely high O3 values. lines (13-14)
- Previous studies already noticed the O_3 maximum over Siberia [Gaudel et al., 2018]. [Cohen et al., 2018] suggested that this maximum could be due to a higher stratospheric influence over the region. In the anthropogenic CO anomalies, the O_3 values are close to the background. However, as demonstrated in Fig.**??** a significant portion of polluted air masses are transported from the surface of East Asia to the UT of Siberia via the East Asian summer monsoon, which could potentially influence the production of O_3. On average for the other regions, O_3 mixing ratios in CO anomalies are 13 ppb higher than their respective median and this difference can reach 21 ppb for the CO anomalies associated with Biomass Burning emissions. Lines (350-356)
- In the Middle East, O_3 values are among the highest in JJASO in the LT and MT. The summertime median is also higher than the median from East Asia (see Fig.13 and Fig.14) which is a region with identified extreme O_3 values [Chang et al., 2017a, Lu et al., 2018]. Li et al. [2001] suggested that the important tropospheric O_3 in Middle East were due to the constant import of pollution from different regions trapped in the upper level anticyclone and the strong subsidence associated to it cause an accumulation in the region. Here the CO anomalies detected are mostly caused by emissions from the Middle east rather than from long range transport. In the Middle East LT, values of O_3 inside CO anomalies attributed to anthropogenic emissions are lower than the 25th percentile, which is similar to the observation

made on the northern hemisphere mid-latitudes. In the MT, the anthropogenic anomalies are close to the median during both seasons. Lines (440-447)

- In the UT O_3 values are maximum over Siberia. The O_3 measured within the BB anomalies are 15 ppb higher than the median but no elevated values are measured in the anthropogenic anomalies coming from Eastern Asia. Further investigations are needed to explain the extremely high values of O_3 measured in the UT of Siberia in JJA. Lines (530-534)

- In the lower and middle troposphere, the maximum O_3 values are found in Middle East. Previous studies assumed that the high O_3 in the regions were due to long range transport of polluted air masses followed by chemical production in the regions [Li et al., 2001, Duncan et al., 2008]. In the LT and MT most of the detected CO anomalies are from local anthropogenic emissions which either show low values of O_3 or values close to the median. In the UT, in JJASO CO anomalies are mostly from anthropogenic emissions from South East Asia. Those anomalies show enhanced values of O_3. Lines (573-578)

This paper is about CO anomalies and as such will focus on those two regions with this prism only, so the focus is made on the O_3 values inside the CO anomalies, but a further analysis on the high background of O_3 in these two regions is out of scope of the current paper and could be the subject of a future study.

3) Previous studies (Nowak et al., 2004, Figure 3; Cooper et al., 2002, Figure 8) have shown that scatter plots of ozone vs. CO are an effective way to highlight air pollution episodes and stratospheric intrusions (or air in the UTLS that is of stratospheric origin). These types of plots would be helpful for this study. For example, on line 277 the authors speculate that some of the high ozone values may be due to stratospheric influence. A scatter plot ozone vs CO could indicate instances of stratospheric intrusions.

This study focuses on the extreme values of CO and the stratospheric air masses are discarded in this study thanks to a filtering based on 2 PVU. Therefore, the high ozone values are not attributed to the stratosphere (or it is an outlier that should be further monitored as a case study). It can also be the (aged) stratospheric influence in the troposphere and we cannot see that with a scatter plot because we are only looking at the "high CO" branch of the scatter plot. So we believe that the suggested scatter plot is not meaningful for this analysis.

Objectives of the paper will be further clarified to avoid any "frustration" in the introduction, the following sentences have been added in blue below:

"Ozone values are presented as additional information. However, detailed analysis of the ozone values and climatology is outside the scope of the current paper."

4) Many of the study regions have well known trends in ozone and CO since 2000, but these trends are not addressed. The plots of average ozone and CO can therefore be misleading for certain regions. For example, during summertime, lower tropospheric ozone has decreased strongly in eastern North America since 2000, while it has increased in wintertime (see Figures 14 and 15 of Gaudel et al.; also see Chang et al., 2017). Lower tropospheric ozone has also increased *in East Asia (Lu et al., 2018; Kim et al., 2023). Globally, CO has decreased since the 1990s. For example, trends of global CO levels can be found at the bottom of this webpage maintained by the NOAA Global Monitoring Laboratory:* `https://gml.noaa.gov/ccgg/figures/` *(The figure called cotr_global.pdf shows a decrease of CO since the year 2000). Also, extreme CO air pollution events have decreased across the USA since 2000: https://www.epa.gov/air-trends/carbon-monoxide-trends Because ozone and CO have changed in many regions since the year 2000 it would be helpful to compare regions using data from the most recent years, when possible, rather than using 20-year averages.*

This analysis is not dedicated to trend analysis (already done by e.g. Cohen et al. [2018] regarding the IAGOS dataset). As the objective here is to characterise the "extremes of CO" over the entire IAGOS data set (to maximise the statistical robustness of the results), our strategy was to define the "extreme" as above the regional and seasonal Q95, assuming that the seasonal and regional differences (to be discussed) are "larger" than the global trend of CO. We believe our methodology is then of interest to detect and assess the variability of extreme events, occurring whatever the global trend. It thus provides a diagnostic picture of the origin (type of source and area) of the extreme CO that is important information to be further used in the model's evaluation (the right CO/O3 for the right reasons).

As advised, we also made the same analysis with only the last 10 years of the IAGOS datasets. The tables 1 and 2 below shows the 95th percentile computed with the full data period (2002 to 2019) to the one computed with the data from 2010 to 2019.

The differences between the two data sets can be seen in the figure below, which shows the results of the analysis conducted using the full data period and the figure using only the measurements from the last 10 years. This confirms that the trend of CO can be strong in some regions for the 95th percentile, however the differences between the two figures are not significant, and the origins and sources of the anomalies remain similar in regions with sufficient number of data. Consequently, our conclusions remain the same and it was decided to retain the full data period in order to have a larger dataset and to ensure greater statistical robustness. Indeed, it can be seen from figure B1 in the appendix that some regions are predominantly sampled in between 2002 to 2009. Removing this part of the data would result in a significant reduction in the number of measurements for those regions.

A paragraph regarding the trend and the sensitivity of our diagnostics to the period used in the analysis has been added to the conclusion (lines 587-592):

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 measurments. Several regions, showed a decreased 95th percentile in this datasets (see tables below). However, the origins and sources of the anomalies remain similar in regions with sufficient number of data. The conclusion the study remained largely unchanged for the CO anomalies of the last 10 years.

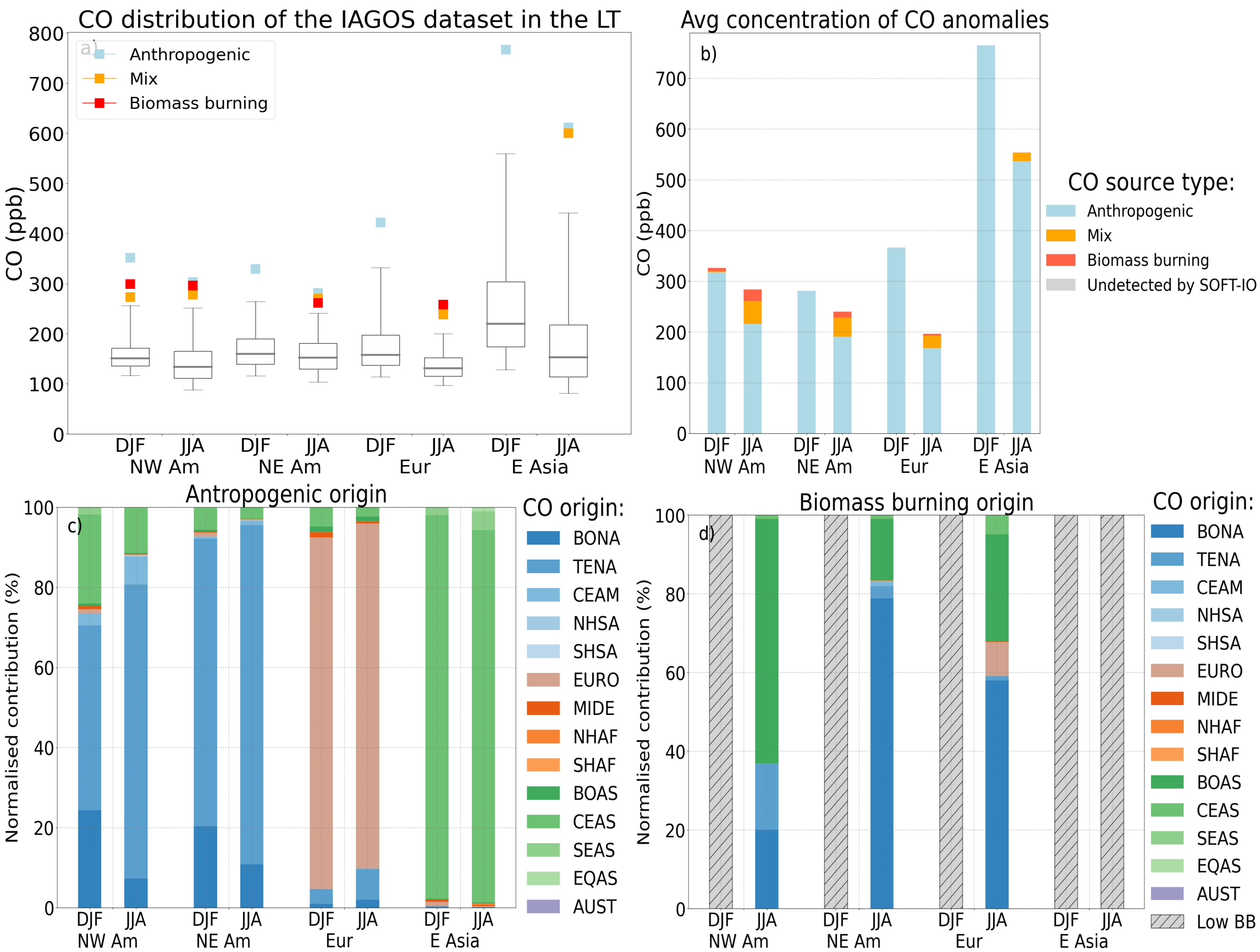

Figure 1: a) CO measured by IAGOS in the LT (below 2km). The box-plot represents the 5th, 25th, 50th, 75th and 95th percentiles of the CO distribution, while the coloured squares represent the mean values of CO inside the detected anomalies (each colour represents a type of CO anomaly attributed to a different source with SOFT-IO: red for biomass burning, blue for anthropogenic and orange for mix sources). b) Bar-plot showing the averaged mixing ratios of CO in all the detected anomalies (¿q95) in the LT in each region for JJA and DJF (given by the total height of the bar), and their proportion according to the different sources (blue for anthropogenic, red for biomass burning and orange for mix, the relative height of the coloured blocks represents the proportion of each type of anomaly). The proportion of plumes where no contribution is modelled by SOFT-IO are represented in grey (in this figure no anomalies are undetected by SOFT-IO over the 4804 observed). c) Regional origin (according to GFED regions, as in Fig. 1) of the anthropogenic contributions of the anomalies associated with mix and anthropogenic sources in the LT in NH extra-tropics (the hatched part cover region/season with not enough anomalies attributed to the mixed or anthropogenic categories) d) Same for the origin of the biomass burning contributions associated with mix and biomass burning anomalies. The Low BB patched (hatched grey patches) is applied if a regions has less than 3% of its plumes attributed to either mix or BB sources.

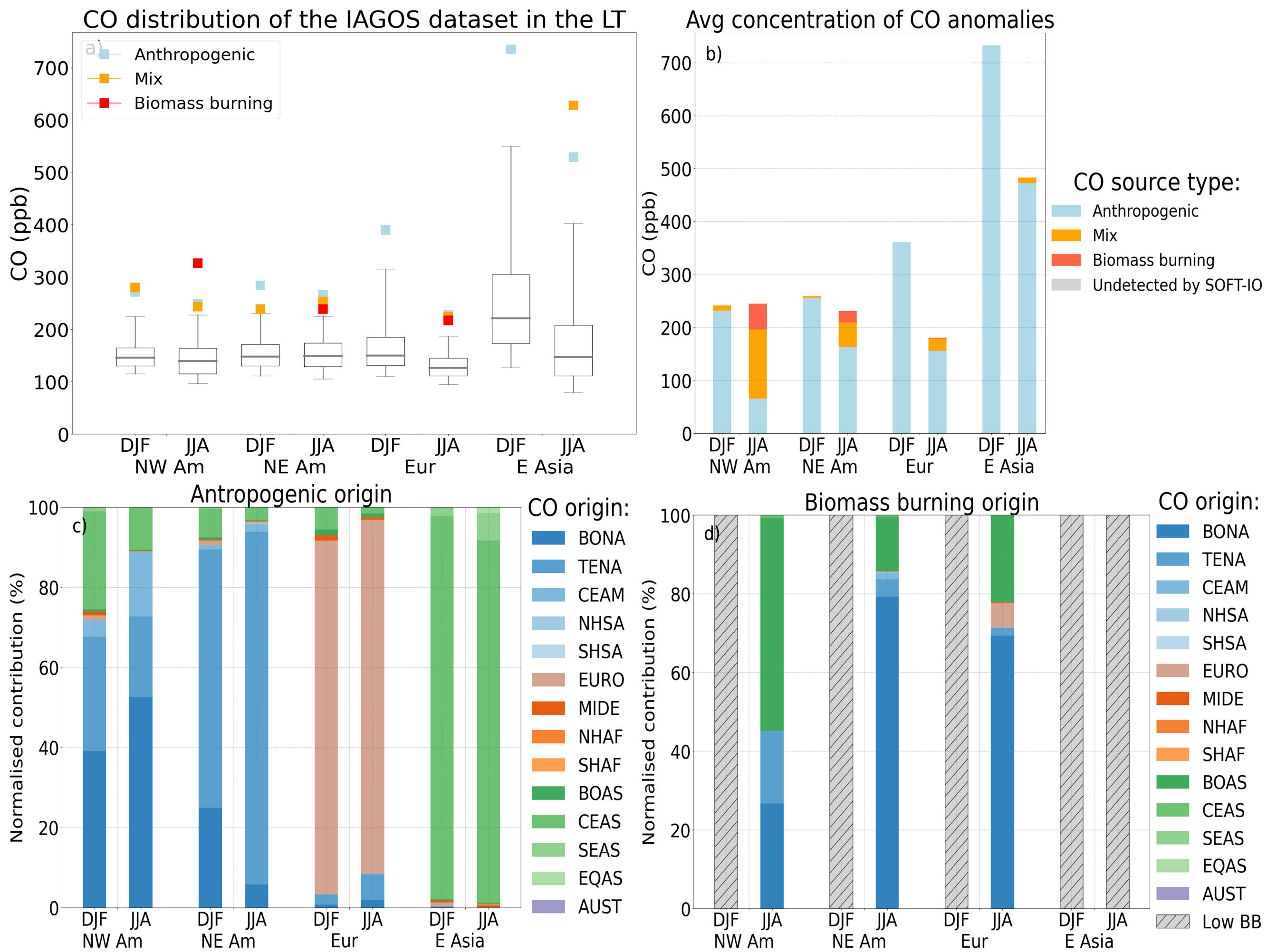

Figure 2: Same as figure with only the data from 2002 to 2019.

2 Minor Comments:

The paper contains many grammatical errors that should be corrected. A co-author with excellent English skills should carefully proofread the entire text.

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

Line 191 In addition to increased CO emissions in winter, there is also a well-known increase in CO lifetime in winter (due to less ozone and OH), which also explains the higher wintertime concentrations (Novelli et al., 1998).

Corrected.

		LT	FT	UT
NW Am	DJF	256	160	146
	JJA	251	149	145
NE Am	DJF	264	159	126
	JJA	241	156	132
Eur	DJF	332	158	126
	JJA	200	140	123
Sib	DJF	no data	no data	127
	JJA	no data	no data	181
E Asia	DJF	559	209	129
	JJA	441	173	162

		LT	FT	UT
NW Am	DJF	224	155	142
	JJA	227	168	140
NE Am	DJF	230	148	112
	JJA	225	156	126
Eur	DJF	315	150	117
	JJA	187	135	118
Sib	DJF	no data	no data	119
	JJA	no data	no data	168
E Asia	DJF	550	205	128
	JJA	403	160	153

Table 1: q95 values (in ppb) used as thresholds for the different regions using data from 2002 to 2019 on the left and using data from 2010 to 2019 on the right

		LT	FT	UT
India	DJF	424	157	132
	MAM	305	191	130
	JJA	267	134	131
	SON	470	150	150
North Af	DJFM	no data	no data	145
	JJASO	no data	no data	110
Middle E	DJFM	253	148	135
	JJASO	300	129	113
Gulf of G	DJFM	724	297	190
	JJASO	280	192	147
South Af	DJFM	219	132	172
	JJASO	400	245	197

		LT	FT	UT
India	DJF	399	155	131
	MAM	310	194	130
	JJA	237	132	132
	SON	468	140	155
North Af	DJFM	no data	no data	137
	JJASO	no data	no data	110
Middle E	DJFM	238	143	140
	JJASO	239	125	115
Gulf of G	DJFM	708	283	183
	JJASO	289	196	146
South Af	DJFM	252	165	162
	JJASO	457	263	195

Table 2: q95 values (in ppb) used as thresholds for the different regions using data from 2002 to 2019 on the left and using data from 2010 to 2019 on the right

Discussion of transport processes impacting India should reference previous work on this topic, e.g. Lal et al. 2014; Lawrence and Lelieveld, 2010; Lelieveld et al., 2001.

Thank you for the comment and as advised the paragraphs on India have been modified in the revised version.

- 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] and explaining the rather high values of CO in the LT and MT during this period and the low contribution from SEAS in the UT, at this altitude the anthropogenic CO anomalies receive an influence from CEAS and SEAS but also from NHAF. In JJA, it is the wet phase of the monsoon in India so the important convective activity and precipitation associated with this period [Kar et al., 2004] leads to rapid transport of the South-Asian emission to the UT while preventing BB: almost all the CO anomalies are caused by anthropogenic emissions from India or the close proximity (SEAS and CEAS). (Lines 376-383)

- The O_3 cycle shown here is similar to the cycle described in Lal et al. [2014] and obtained by a radiosonde, here the focus is on the O3 measured in the CO anomalies. In the LT, the minimum values of O_3 are reached during the summer monsoon in JJA. The low values can be explained by the increased marine influence during this period [Lawrence and Lelieveld, 2010]. At this altitude the O_3 values recorded simultaneously as the CO anomalies are low and show the low O_3 production in those plumes.

  In the MT and UT, the maximum of the O_3 is reached during MAM, and the minimum is reached during DJF. In the UT, in DJF and MAM an important part of the CO anomalies come from northern African BB. Those plumes are associated with higher values of O_3 (11 and 10 ppb above the median respectively for DJF and MAM). CO anomalies in JJA are caused by the local emission of anthropogenic CO rapidly transported to the UT by the important convective activity of the South Asian Summer Monsoon (SAMA). This rapid transport could explain that the associated values of O_3 are close to the median (65 ppb). In the post monsoon season (SON) BB anomalies from Equatorial Asia are added to the local anthropogenic anomalies. The values of O_3 in the BB plumes are low and close to the 25th percentile (44 ppb) which is explained by the lower background values of O_3 in Equatorial Asia compared to India [Cohen et al., 2018]. (Lines 389-401)

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