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: 186-189) 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].

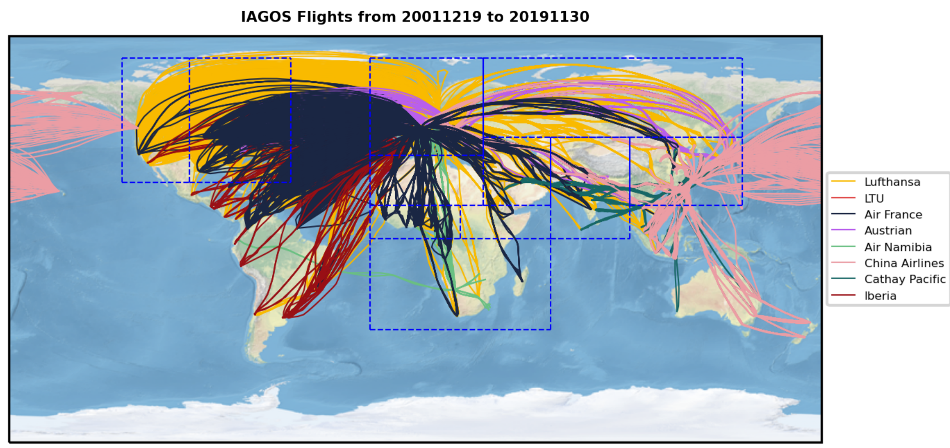

Figure 1: Trajectories of every IAGOS flights

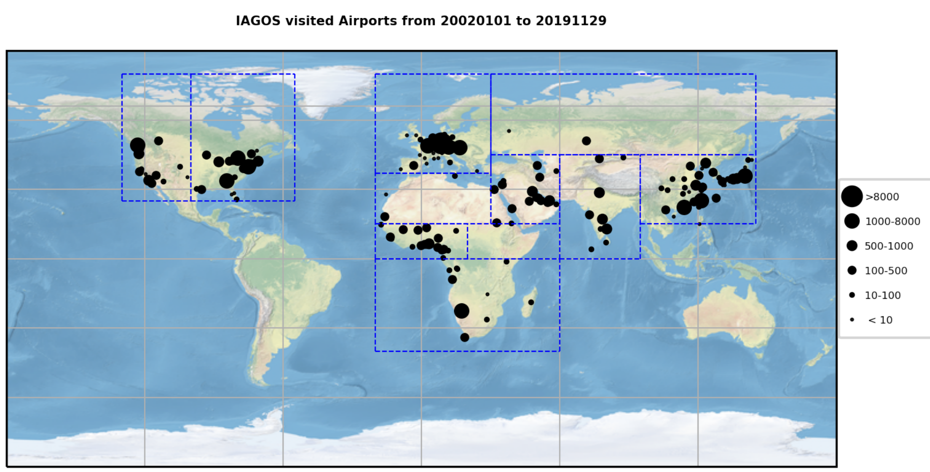

Figure 2: Map of the visited airports by IAGOS aircrafts

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 (244-250)

- 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 (275-280)

- 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 (345-348/356-358).

- 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 (379-391).

- 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 (428-435)
- 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 (471-476)

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 (225-235)

- 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 (251-255)

- 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 (257-274)

- 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 (323-333)

- 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 (364-378).

- 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 (399-406)

- 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 (416-426)

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.

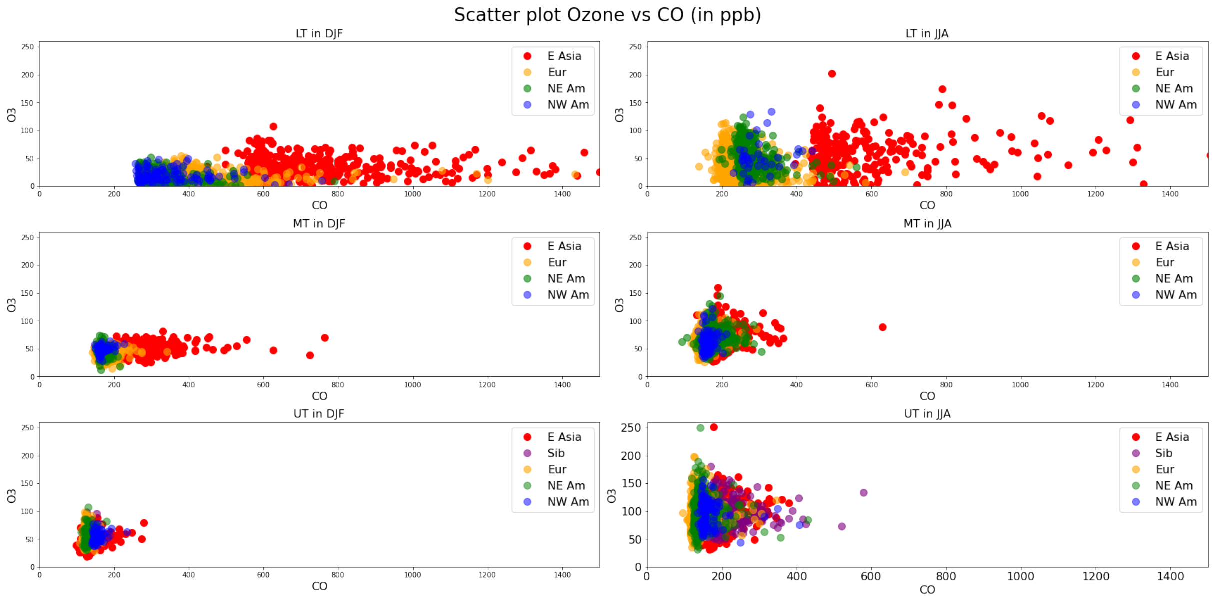

Figure 3: Scatter plot Ozone vs CO in the observed anomalies

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 303)

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 321). 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 397):

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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