Reviewer#1

The paper documents the methodology and results from the use of FLEXPART on the IAGOS dataset, with the goal of providing potential users with source attribution. The paper is well-written and provide a good description of the methodology. The application portion of the paper is more limited, focusing on a few examples and broad measures. Overall, I find the paper worthy of publication after consideration of the following points.

We would like to thank Reviewer#1 for her/his comments and suggestions that will improve our manuscript.
We clarified all the points raised by reviewer#1 and answered her/his different remarks in blue in this document.

Major point

While there is a wealth of information provided by all the parcels released along the flight track, the authors do not provide any information on the standard deviation (or any other statistical information) of the simulation perturbation. In particular, this seems to be of relevance to the discussion of Figure 11.

We provided statistical information in the submitted version through the percentiles information given in Fig 10b which are commented in Section 5.2.
In addition, as suggested by Rev#1, we have added in the revised version of the manuscript different statistical information.
SOFT-IO standard deviation has been added to Figure 11, as suggested by Rev#1, but also on Figs.#5 #6 #7 and #8 (see below for the modifications).
Additionally, we have also added standard deviation of the IAGOS vs SOFT-IO bias on Figure 10a, but not on Figs.12a and 13a for clarity reason.
The discussion related to the figures has been modified accordingly to take into account this new information on standard deviation in Section 5.2, as suggested by Rev.#1.

Minor points
- Line 162: It is not clear the vertical resolution is the most critical factor. Plenty of processes (as discussed in the paper) are not present in trajectories, or a choice of different parameters, could also be responsible for trajectory shortcomings.
We have modified line 162 in order to account Ref#1 remark:
“ Vertical resolution is one of the most critical factor for modeling such CO plumes with the best precision in terms of location and intensity (Eastham and Jacob, 2017)”

- Line 208: Why the ICARTT dataset? There are plenty of regional dataset that might have been of higher relevance than this one. It would be good to justify this choice
Ref#1 is true that there are plenty of regional dataset that could have been tested. The goal of using regional dataset in the paper is to evaluate the incidence of one of them respect to global emission inventories, not to evaluate the incidence of all regional dataset. We have chosen ICARTT because of improved results demonstrated in the representation of boreal biomass burning fires in some specific cases (Elguindi et al., 2010; Turquety et al., 2016). Boreal fires

can be associated with pyro-convection, generally poorly represented in global emissions inventories. As IAGOS has a quasi global coverage, global emission inventories are the first choice in the methodology. However ICARTT comparison showed that regional inventories could be used to obtain better results on limited case studies on CO observations related to extreme events such as pyro-convection, and suggests that other regional emission inventories could be then included in the future in SOFT-IO for specific case studies CO pollution.
We have added the following sentence lines 206-209:
"The aim is to test the ability of regional inventories in better representing simulated CO for specific case studies. The goal of using regional dataset in this paper is only to evaluate the incidence of one of them respect to global emission inventories, not to evaluate the incidence of all regional dataset. We have chosen ICARTT because of improved results demonstrated in the representation of boreal biomass burning fires in some specific cases (Turquety et al., 2016) as for example the one based on MOZAIC data by Elguindi et al., (2010). Global emission inventories are the first choice to interpret quasi global coverage of the CO IAGOS measurements. In the future we plan to include regional emission inventories for the study of specific events."

- Line 220: it seems that the CO lifetime is not part of this equation. This would be a serious issue since 20-day trajectories are considered. If used, what is the CO lifetime?
CO is considered as chemically passive tracer in the equation. Concentrations will only vary considering dispersion and mixing associated with dynamical processes along 20 days.
The only significant chemical sink of CO in the troposphere is OH attack. As stated in lines 80-81, CO has lifetime of months in the troposphere (Logan et al., 1981; Mauzerall et al., 1998), higher than the 20-day of backtrajectories. Folkins et al. (JGR 2006) calculated CO lifetime against OH attacks (their Fig. 11) between 20-25 and 80 days within the troposphere, confirming that trajectories lower than 20-25 days should be used to avoid chemistry issues in CO lifetime.

- Line 228: it is also important to recognize the CO tends to be mostly released during smoldering and so might not be as prevalent in pyrocumuli.
The following sentence has been added line 228:
"even if CO tends to be mostly released during smoldering"

- Line 286: it is not clear that it is always a straight linear decay with altitude.Â˘a How important is the definition of the background?
We agree that there is not always a straight linear decay of CO with altitude. However, as for most of the IAGOS vertical profiles CO is enhanced in the boundary layer (related to surface emissions), the calculation of the background by using the slope calculated in the free troposphere was the most accurate way to define the background.
This definition of the background could be in the future improved by using "climatological" CO vertical profiles. It will be only possible to use this with sufficient CO measurements above the different IAGOS airports, and this was not possible for the present study over 10 years of CO measurements, except for few exceptions (Frankfurt for instance). Note that the definition of the background does not enter in the SOFT-IO methodology neither in the final CO ancillary data included in the IAGOS database. The background is defined in the present study to extract CO anomalies in order to statistically evaluate the differences with the contribution in CO computed by SOFT-IO. Finally the CO background definition has a negligible incidence in the CO anomalies definition, as we focus on the anomalies higher than the percentile 75 (see Eq. 4 and 5 lines 303-304)

- Line 295: is there any assurance that the background from VP is consistent with UT where they connect? If not, is this an issue?
Two different methodologies are used to estimate the background in UT and VP, as we still do not have enough data over all airports to apply climatological background for VP. Background is not used to provide ancillary data of CO in the IAGOS database and its definition is quite subjective (see for instance Parrish et al., 2012, doi:10.5194/acp-12-11485-2012). We estimate a background in the submitted paper to evaluate SOFT-IO simulations respect to CO anomalies events.
This is neither an issue for the provision of CO ancillary data calculated with SOFT-IO in the IAGOS database, nor for the estimation of CO anomalies as we focus on events higher than percentile 75, as explained just above.

- Line 301: change “to consider” to “to be considered”
Done

- Line 366: it would be nice to show PV along the same track
PV has been added in dark green along flight track on Figs.6a and 8a (see below)

- Line 425: Figure needs an explanation of the color bar labels.
Explanation of the color bar levels has been added (see below)

- Line 465: change “less good” to “worse”
Change is done line 465

- Line 471: I think it would be quite illuminating to present an additional figure (within the text or in the supplement) with percentages instead of concentrations.

We have added additional figures of relative bias in supplement section (Figs S2a, S2b, S2c and S2d)

- Line 488: this might look quite different with percentages!
Figures with relative bias have been added in supplement (Fig S2a, S2b, S2c and S2d)

- Line 497: this seems like a very narrow explanation.Â˘a There are many things that could go wrong, not just pyro-cumulus.
Rev#1 is true. We have added the following sentence line 497:
“*, or these emission inventories are under estimated for such specific events*”

- Line 502: I think “sense” is better than “information”
Information has been replaced by sense

- Line 508: this seems like too many plots since very little discussion is attached to Them
Plots have been implemented over one page

- Line 513: as mentioned in my major point above, the question is but what is the range of the variability from the different parcels?Â˘a The only thing that this is showing is that the mean is within the observed standard deviation.

As mentioned previously, we have added standard deviation into the figure and discussed it in Section 5.2. We clearly see that the standard deviation of the model is within the standard deviations of the observations in the LT and in the UT, but not in the MT.

- Line 549: it is hard to get a sense of the change from the Taylor diagrams. If the authors want to keep them, it might be quite helpful to have arrows indicating the direction of the change.

We have added connection lines to help the reader interpreting the direction of change in the Taylor diagrams (see below)

- Line 555: this is actually incorrect. The anthropogenic emissions in MACCity originated from Lamarque et al. (ACP, 2010), except for the added seasonal cycle. Emissions were harmonized for year 2000 with the various scenarios (RCPs); therefore, any data post-2000 is actually the result of the scenario RCP8.5. The fact that they are fairly close is that they share many aspects (see paper above for more details).

"

Rev#1 is true. We have updated information concerning MACCity in our manuscript in order to consider this remark. The following sentences have been added:

"These results are not surprising as MACCity (Lamarque et al., 2010; Granier et al., 2011) is originated from various regional inventories (in addition to EDGAR), and expect to better represent..."

"*However as stated in Lamarque et al., (2010) both inventories share many aspects (for example over Latin and South America), and the differences between them...*"

Figures modifications requested by Rev#1:

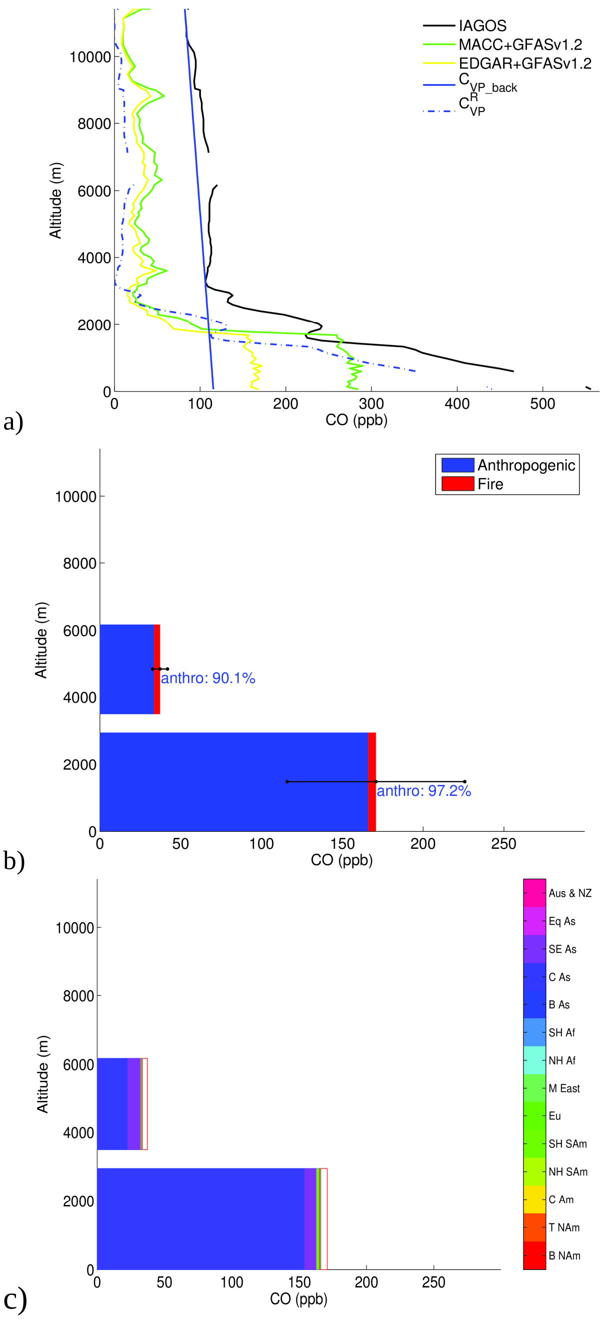


Figure 5: (a) Carbon monoxide profiles over Hong Kong during a MOZAIC-IAGOS flight landing on 22 October 2005. The black line indicates the observed CO profile while the blue line indicates the CO background deduced from the observations. Green and yellow lines indicate the simulated CO contributions using respectively MACCity and EDGARv4.2 for anthropogenic emissions, and using GFAS v1.2 for biomass burning emissions. Simulated CO is separated in (b) sources contribution (anthropogenic in blue, fires in red, standard deviation in black) and in (c) regional anthropogenic origins (14 regions defined for global emission inventory, http://www.globalfiredata.org/data.html, see Fig. S1; unshaded red square is for fire contribution), using MACCity and GFASv1.2.

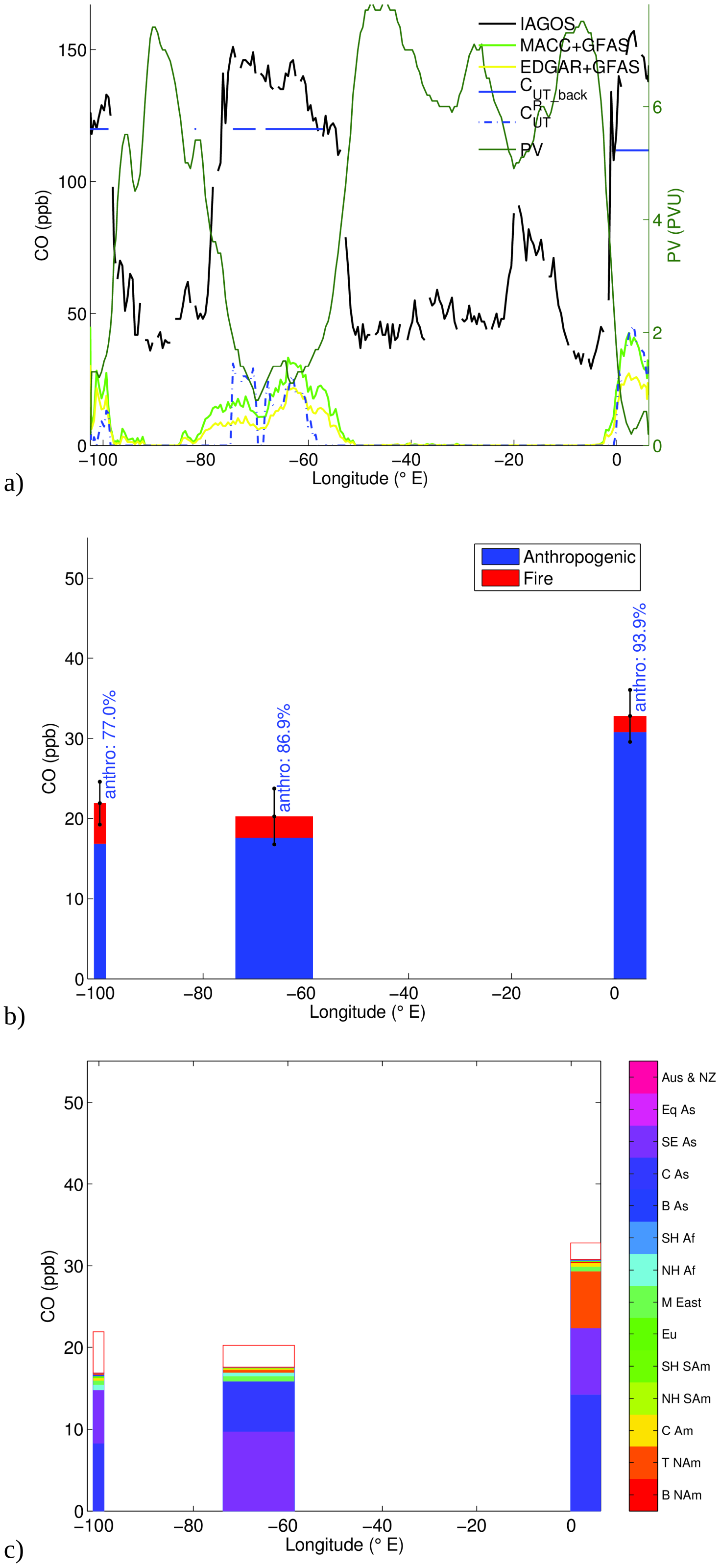


Figure 6: (a) Carbon monoxide zonal profile during the 10 March 2002 MOZAIC-IAGOS flight from Frankfurt to Denver. The black line indicates the observed CO while the blue line indicates CO seasonal background in the UT deduced from the IAGOS data set. Light green and yellow lines indicate the simulated contributions using respectively MACCity and EDGARv4.2 for anthropogenic emissions, and GFAS v1.0 for biomass burning emissions. Dark green represents potential vorticity (pvu) from ECMWF analyses. Simulated CO is separated in (b) sources contribution (anthropogenic in blue, fires in red, standard deviation in black) and in (c) regional anthropogenic origins (14 regions defined for global emission inventory, http://www.globalfiredata.org/data.html, see Fig. S1; unshaded red square is for fire contribution), using MACCity and GFASv1.0.

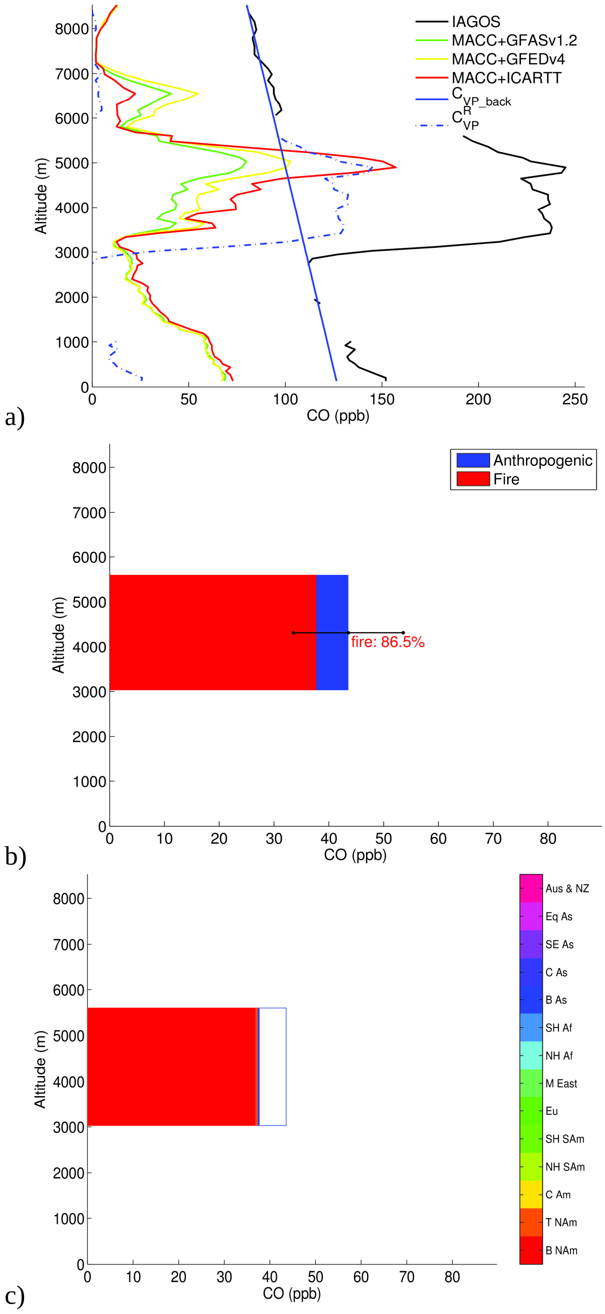


Figure 7 : (a) Carbon monoxide profiles over Paris during a MOZAIC-IAGOS flight landing on 22 July 2004. The black line indicates the observed CO profile and the blue line indicates CO background deduced from the observations. Green, yellow and red lines indicate the simulated contributions using respectively GFASv1.2, GFED4 and ICARTT for biomass burning emissions, with MACCity for anthropogenic emissions. Simulated CO is separated in (b) sources contribution (anthropogenic in blue, fires in red, standard deviation in black) and in (c) regional biomass burning origins (14 regions defined for global emission inventory, http://www.globalfiredata.org/data.html see Fig. S1; unshaded blue square is for anthropogenic contribution), using MACCity and GFASv1.2.

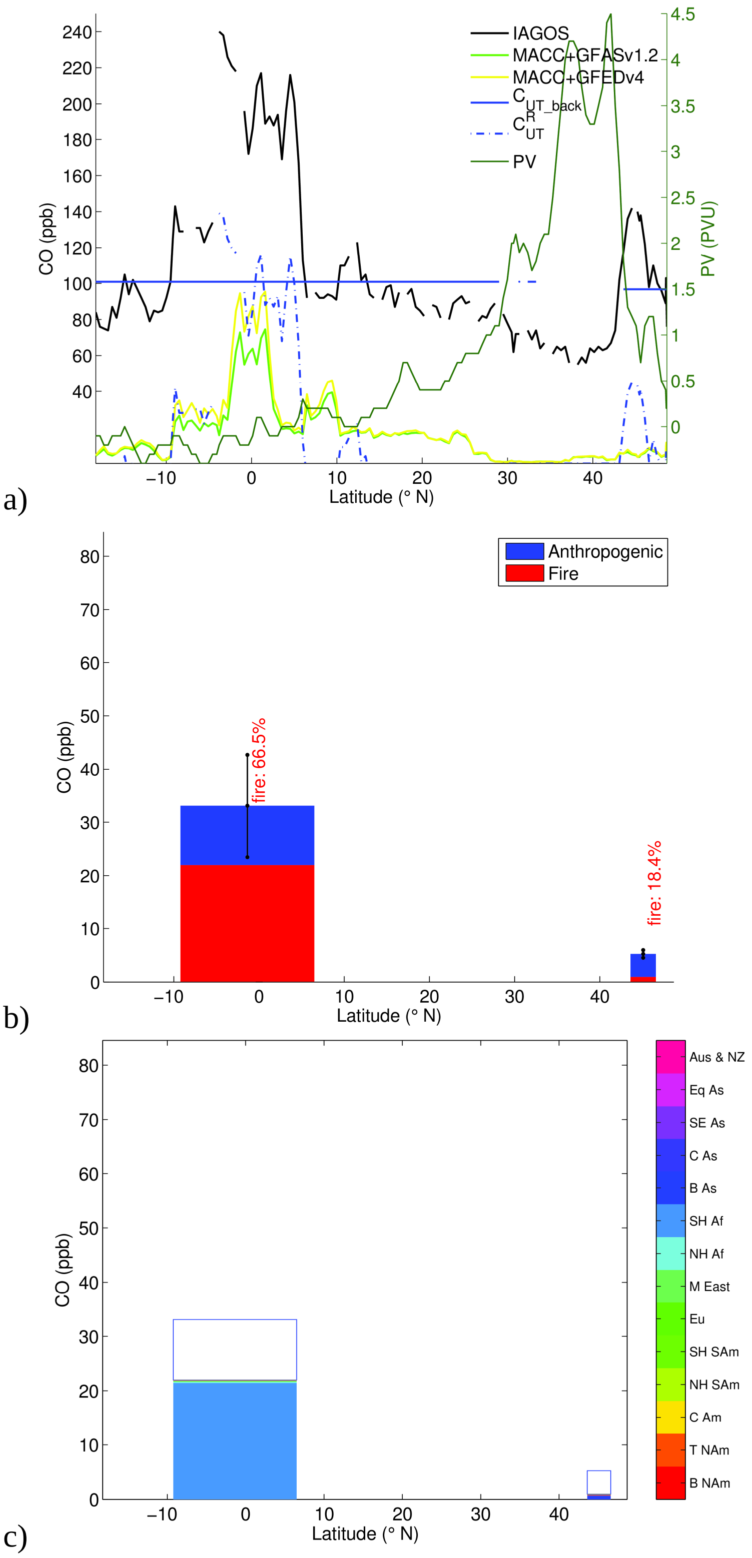
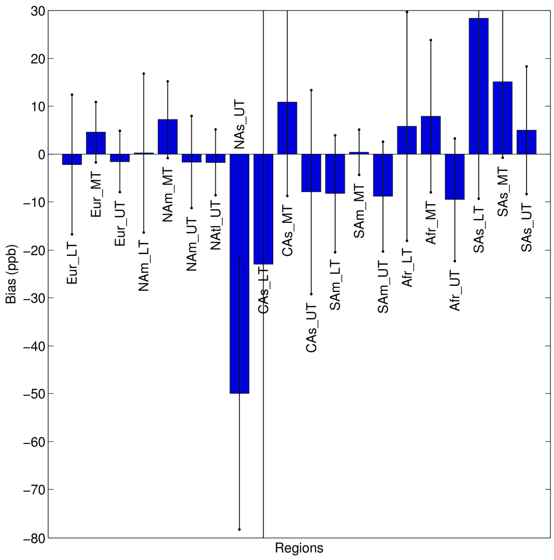
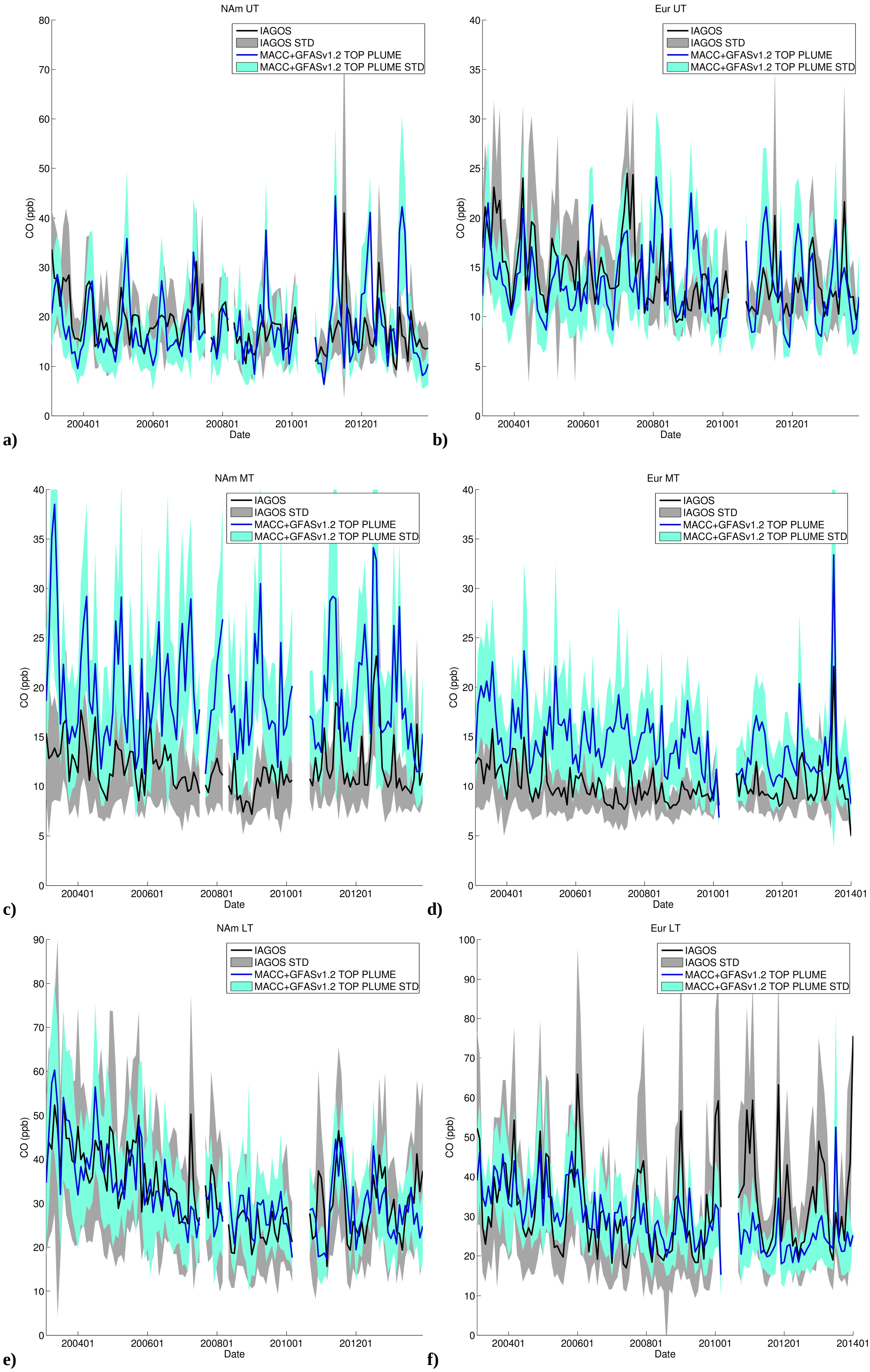


Figure 8: (a) Carbon monoxide as a function of latitude during the 30 July 2008 MOZAIC-IAGOS flight from Windhoek to Frankfurt. The black line indicates the observed CO, the blue line indicates the CO seasonal background deduced from the IAGOS data set and the dash-dotted line the residual CO mixing ratio. Light green and yellow lines indicate the simulated contributions using MACCity for anthropogenic emissions, and respectively GFAS v1.2 and GFED4 for biomass burning emissions. Dark green represents potential vorticity (pvu) from ECMWF analyses. Simulated CO is separated in (b) sources contribution (anthropogenic in blue, fires in red, standard deviation in black) and in (c) regional biomass burning origins (14 regions defined for global emission inventory, http://www.globalfiredata.org/data.html, see Fig. S1; unshaded blue square is for anthropogenic contribution), using MACCity and GFASv1.2.



a)

Figure 10: (a) Mean bias (blue) and mean standard deviation bias (black) between the modeled and observed CO anomalies ; (b) Percentiles of the modeled CO anomalies bias with respect to observations; (c) Relative contribution from anthropogenic and biomass burning sources to the modeled CO. The three graphs are for the main sampled regions (Europe, North America, North Atlantic, North Asia, Central Asia, South America, Africa, South Asia) and in three layers (LT, MT, UT), using MACCity and GFASv1.2 for the 2003-2013 period. Biomass burning vertical injection uses APT methodology.


NAm UT
Eur UT
NAm MT
Eur MT
NAm LT
Eur LT
IAGOS
IAGOS STD
MACC+GFASv1.2 TOP PLUME
MACC+GFASv1.2 TOP PLUME STD
CO (ppb)
Date
200401
200601
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a)
b)
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Figure 11: Times series (monthly means between 2003 and 2013) of the observed (black) and simulated (blue) plumes of CO enhancements for the two most documented regions (North America and Europe) in the LT (e & f), MT (c & d) and UT (a & b), using MACCity and GFASv1.2. Standard deviations are in gray (observations) and light blue (SOFT-IO). Biomass burning vertical injection uses APT methodology.

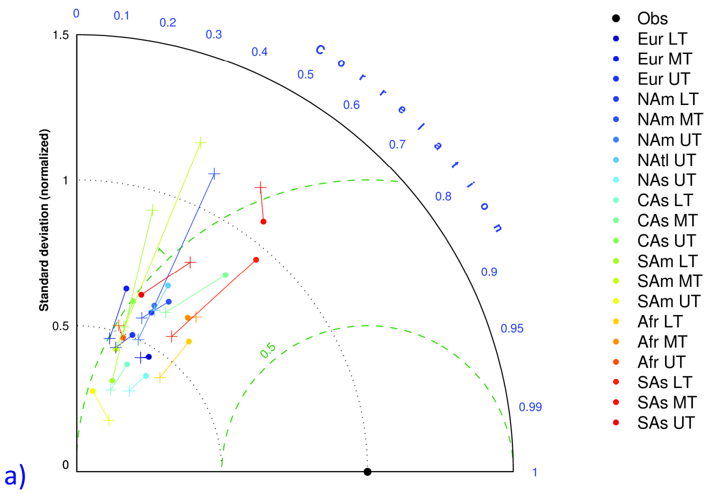
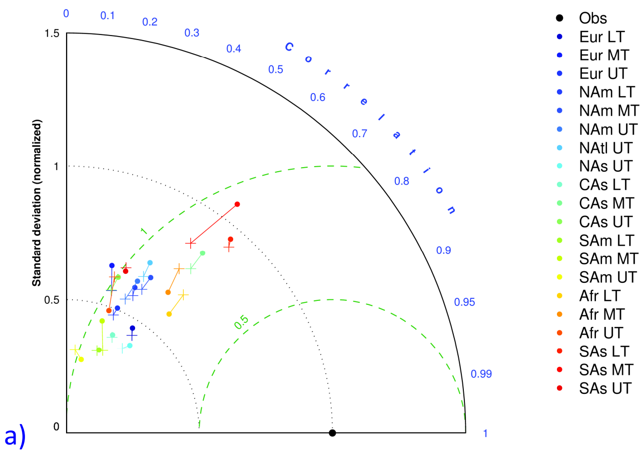
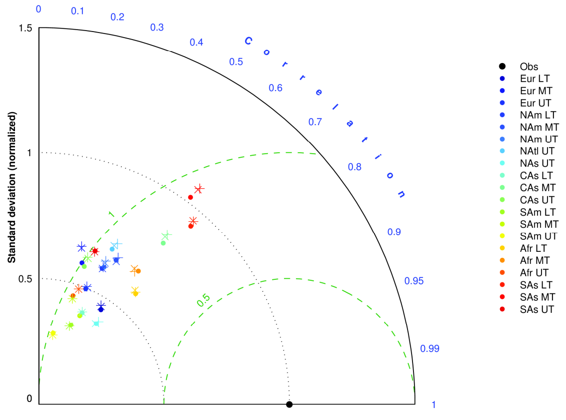


Figure 12: Comparison of the SOFT-IO anthropogenic emission influence between 2002 and 2008 (a) Taylor diagrams are obtained for the different regions and in the three vertical layers (LT, MT and UT) using MACCity (dots) and EDGARv4.2 (crosses) with GFAS (lines represent connexions between the two inventories) (b) Mean biases between the modelled (blue for MACCity + GFAS; brown for EDGARv4.2 + GFAS) and observed CO anomalies. The MIXED methodology is used for fire vertical injection





c)

Figure 13: Comparison of the SOFT-IO biomass burning emission influence between 2003 and 2013. Taylor diagrams are obtained for the different regions and in the three vertical layers (LT, MT and UT) using (a) GFASv1.2 (dots) and GFED4 (crosses) with MACCity and MIXED methodology for both GFASv1.2 and GFED4 (lines represent connexions between the two inventories); (c) GFASv1.2 and MACCity with different vertical fire injections methodologies : MIXED (dots), APT (plus) and DENTENER (crosses) (lines represent connexions between the two inventories). Mean biases between modeled and observed CO anomalies. Model is using (b) GFASv1.2 + MACCity (blue); GFED4 + MACCity (brown) and MIXED methodology for both GFASv1.2 and GFED4; (d) GFASv1.2 + MACCity and different vertical fire injections methodologies: MIXED (blue); APT (green) and DENTENER (brown)

Supplements

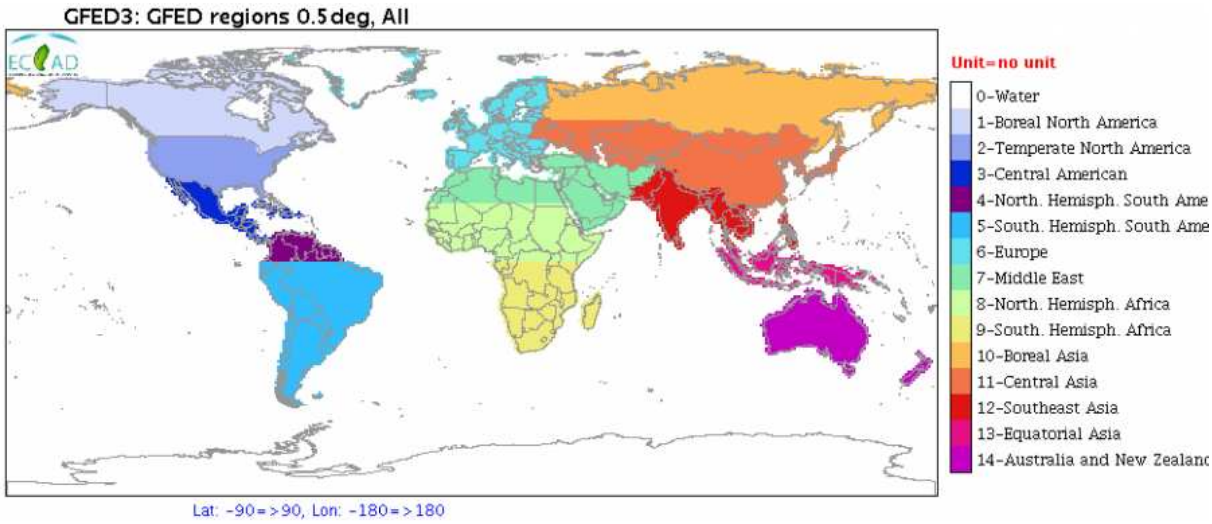
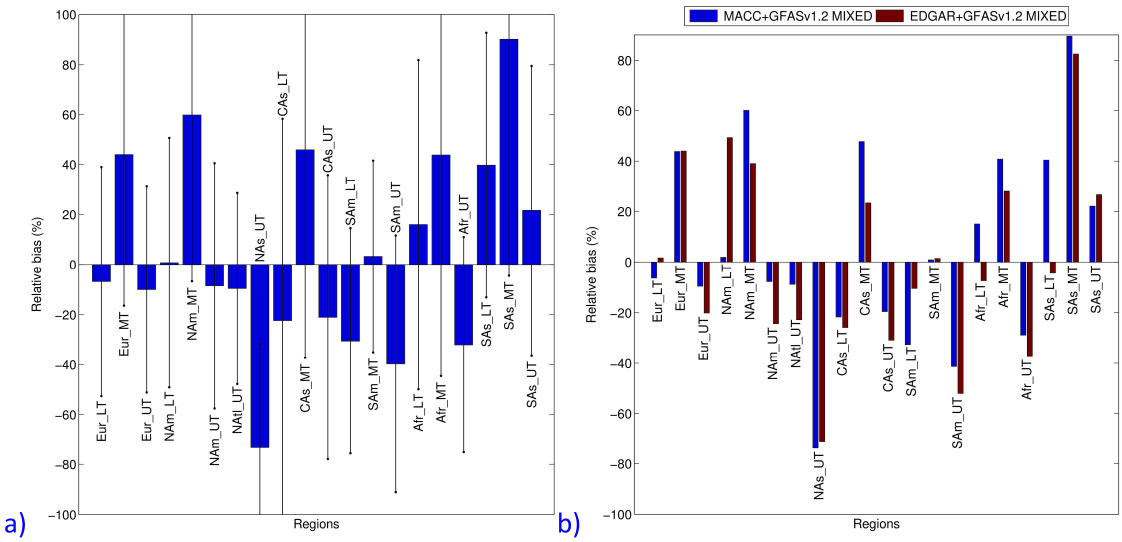


Figure S1: regions used to discriminate CO origin calculated with SOFT-IO, from http://www.globalfiredata.org/data.html



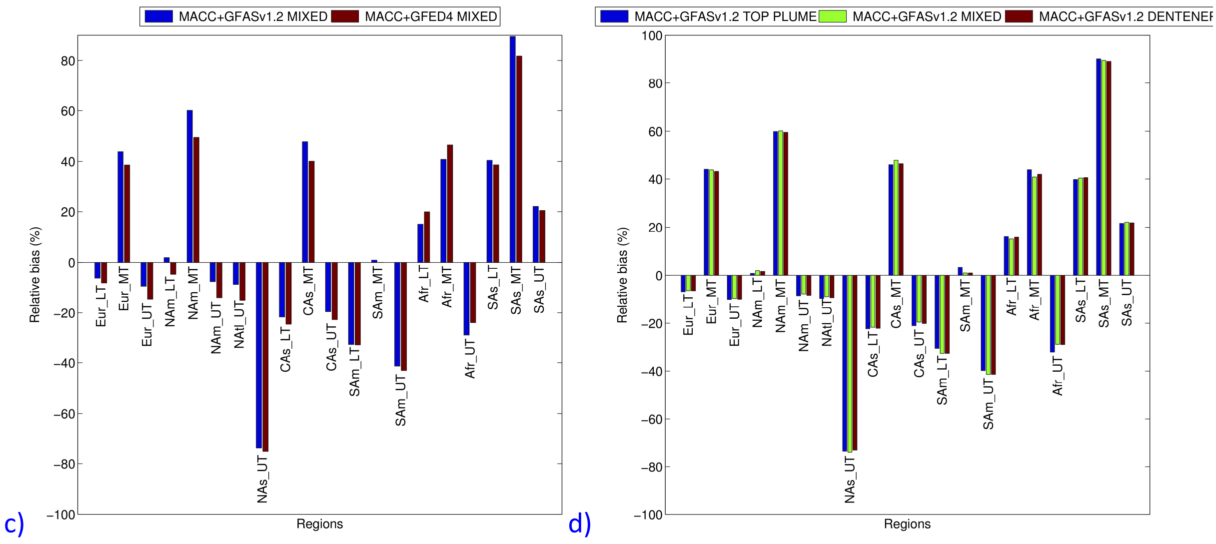


Figure S2: Same as Figs. 10a, 12a, 13b and 13d (a, b, c, d respectively) but for relative bias (%)