Manuscript title: Intercontinental transport of biomass burning pollutants over the Mediteranean Basin during the summer 2014 ChArMEx-GLAM airborne campaign by Brocchi et al.

RESPONSES TO THE ANONYMOUS REFEREE #1

We first thank the reviewer for his thoughtful comments that were helpful in improving the manuscript. Changes have been made in response to his specific comments listed below (in black). Our responses appear in red, changes in the revised manuscript in italic.

R. Zbinden has been added to the co-authors due to her participation in the campaign and her collaboration in the data analyses.

1. The methodology of the study is centred on the use of the Lagrangian particle model FLEXPART backward in time, and the related potential emission sensitivity (PES) tool. I found it difficult to understand the details of this calculation, from the description given in section 2.4. Although I understand more details are given probably in other papers, at least the minimal information to understand the results of this paper needs some clarification. The method is based on the release of particles from the point of interest (peak of concentrations measured from the aircraft, in this case) and moving back in time. The result is illustrated as a map showing the PES quantity, apparently measured in seconds (s), which intuitively suggest the most "visited" places by the particles. It is unclear, however, how the information from the emission inventory is used: is PES calculated as the time spent in any grid point having a non-zero emissions? The author states that the PES quantity is 3-D (from the surface up to 10 km here) but the map is 2-D (lat-lon): is the quantity shown the vertical integral of this PES? If it is a time quantity, perhaps it is the average? The authors are asked to add more details on these calculations, in order to make fully understandable their results.

→ It is true that more details are given in the papers cited. However, more explanations are added to the manuscript. One has to distinguish two available products with FLEXPART. The first product is a residence time of the particles in the total atmospheric column while the second one is the PES in a footprint layer (with the altitude of the layer chosen according to the study done: a few hundred meters for anthropogenic emissions and a few km for biomass burning for instance). It is commonly accepted to look at the residence time of the particles in the total column to visualize the most “visited” places (having it in seconds makes it more readable) while the PES (in $s.m^3.kg^{-1}$) coupled with the emission inventories ($kg.m^{-2}.s^{-1}$) gives access to the contribution of the emissions in a specific layer (Stohl et al., 2003). It appears that, in our case, which deals with pyroconvective biomass burning, the footprint layer is more or less equivalent to the total column.

The PES quantity is 3-D but the map presented in the manuscript shows the vertical integrated values (from 0 to 10 km). For each day of simulations, the FLEXPART calculations are daily averaged. Sentences are modified or added in the manuscript giving more details, as follows:

> *This backward mode gives access to two products. The first one is a residence time of the particles in the total atmospheric column while the second one, defined as the potential emission sensitivity (PES), informs on the location where the sampled air mass has been impacted by the surface emissions.*
>
> *The thickness of the PES layer is chosen consistently with the altitude (vertically integrated values from 0 to 10 km).*

Outputs are averaged every 24 hours with a horizontal resolution of 0.5°×0.5° globally.

2. The description of models and data used does not always report the version number. Where the information is missing, please add the version number of the model and the version number, identification code and url from where data (emission inventories, satellite data, etc.) are taken.

→ The requested information has been added. The version R2.15.0 for MOCAGE is specified. The inventories of emissions used in MOCAGE can be found at eccad.sedoo.fr. GFAS data v1.2 can be found at http://apps.ecmwf.int/datasets/data/cams-gfas/. Measurements from AIRS instrument can be found at https://giovanni.gsfc.nasa.gov/giovanni/.

3. The meteorological fields used to run FLEXPART are chosen at 0.5° x 0.5° resolution. In section 2.3 it is however mentioned that the same dataset (ERA-Interim) is used at 0.125° x 0.125°. Please add a note why a degraded resolution is used for the FLEXPART simulation.

→ The time of calculation would be too long with a resolution of 0.125° at global scale (considering the capacity of our computers, of standard quality). However, in section 2.3, studies have been performed at local scale and only wind direction has been visualized. This is the reason why the highest horizontal resolution available with ECMWF has been used. We modified the text as follows:

Wind fields from ERA-INTERIM are 1) extracted at 6-hourly intervals (0000, 0600, 1200, 1800 UTC) and 3-hour forecasts (0300, 0900, 1500, 2100 UTC) with a horizontal resolution of 0.125°×0.125°, high enough to specify the wind direction at the local scale, and 2) selected at the dates of the presence of the aircraft in the vicinity.

Data are extracted at 6-hourly intervals (0000, 0600, 1200, 1800 UTC) and 3-hour forecasts (0300, 0900, 1500, 2100 UTC) with a resolution of 0.5°×0.5° in latitude and longitude, a compromise at the global scale between computing cost and the trajectory accuracy.

4. On line 109, "ro-vibrational" is probably "roto-vibrational".

→ It is not a typo. The term "Ro-vibrational" is also currently used and admitted in the spectroscopy community.

5. At lines 142-143, the authors claim "no significant difference" between aircraft and ground-based CO concentrations. The term "significant" should be accompanied by a statistical measure such as the p-values, derived by a standard statistical test (e.g. t-test or other non parametric tests). I suggest to include this information, or rephrase avoiding the used of the term "significant". For example, it can be just said that the difference is within the measurement uncertainty.

→ As suggested by the reviewer, we do not use "significant" anymore, and follow the recommendation to use "*the difference is within measurement uncertainty*" in the following rewritten sentence:

the difference (-3.5 to +5.1 ppb) observed is within the total estimated uncertainties reported for both instruments (4.7 to 7.9 ppb).

6. On line 165, the resolution of GEOS-Chem OH field is said to be 3° x 5°, but it is probably 4°x 5°: please check.

→ The technical note for FLEXPART (v8.2, http://flexpart.eu/downloads/26) explains that: «A monthly averaged 3°×5° resolution [OH] field averaged to 7 atmospheric levels is used». But it is true that the resolution given in Bey et al. (2001) is 4°×5°. As we do not know whether it is a typo error or a new interpolation of the [OH] field, we have decided to change the reference in the manuscript and give the one of the technical note, downloadable at http://flexpart.eu/downloads/26.

7. On line 250, "The map of CO contribution to biomass burning ...", "to" is probably "from".

→ Yes, we corrected accordingly.

8. On line 293 and Figure 3a, the authors illustrate a sensitivity test on fire emission intensity from Canada: is the factor of 2 used here within the expected uncertainty of the related fire emission inventory?

→ Validation of fire emission inventories is really difficult. We got in touch with Johannes Kaiser, lead author of the GFAS paper (Kaiser et al., 2012), who indicated that the CAMS validation report (Eskes, H. J., et al.: Validation report of the CAMS near-real-time global atmospheric composition service: December 2016 – February 2017, Copernicus Atmosphere Monitoring Service (CAMS) Report, CAMS84_2015SC2_D84.1.1.7_2017DJF_v1.pdf, 2017) gives errors of 30-50% generally for individual fire events. If we apply this range of errors in our case, we are therefore within the expected uncertainty of GFAS inventory. The following sentence has been added:

> *As errors depend on individual fire events because the fire space-borne observations depend on the instrument sampling, as e.g. cloud-free observations (J. Kaiser, pers. com.), the underestimation detected in our study cannot be considered as a general statement that has to be applied in every case study using GFAS CO emissions, but only in ours.*

9. Also on the "factor of 2" sensitivity test: in Figure 3a the simulated peak of CO mixing ratio is certainly closer to observations, however also the background values outside the peak are increased, and they are higher than the observations. The factor of 2 multiplicative factor seems to be thus unjustified. The model probably does not capture the intensity of the peak, because of low resolution or numerical diffusion. I this suggest to smooth the statements regarding the possible underestimation by a factor of 2 of the fire emission inventory.

→In line with point 8., we smoothed our statements by mentioning that errors are dependent on each fire and thus, this multiplicative factor is not a factor that has to be applied globally to GFAS emissions. It only corresponds to our results on a particular case study.

10. Figure 1: the caption reports "Time series of aerosol concentrations ...", I would better call them "aerosol total number concentrations".

→ The caption has been rephrased accordingly. It has also been modified in the caption of Figure 5.

11. Figure 2: there seems to be significant fire activity also in southern Russia (north of the Black Sea), which may potential contribute to the air masses captured by the aircraft instruments. I would not expect a significant contribution on the episode of August 10, but perhaps it may play a role on that of August 6, since the contribution from Siberia is found to be larger than that from North America in Figure 6. I suggest to briefly discuss it or revise the calculation for the August 6 episode.

→ New simulations were performed in order to estimate the contribution of the fire activity in Southern Russia (north of the Black Sea) both on the 10 and 6 August (Figure 10 below). The CO

contribution is calculated in an area defined from 25°E to 40°E and 40°N to 55°N. In both cases, it appears that the contribution of CO from biomass burning located in this area is close to zero and, thus has no impact on our results. A sentence has been added in the manuscript but the Black Sea contribution has not been shown in the Figure presented in the revised manuscript:

The biomass burning contribution in Southern Russia (North of the Black Sea) has also been studied but leads to a contribution close to zero (not shown).

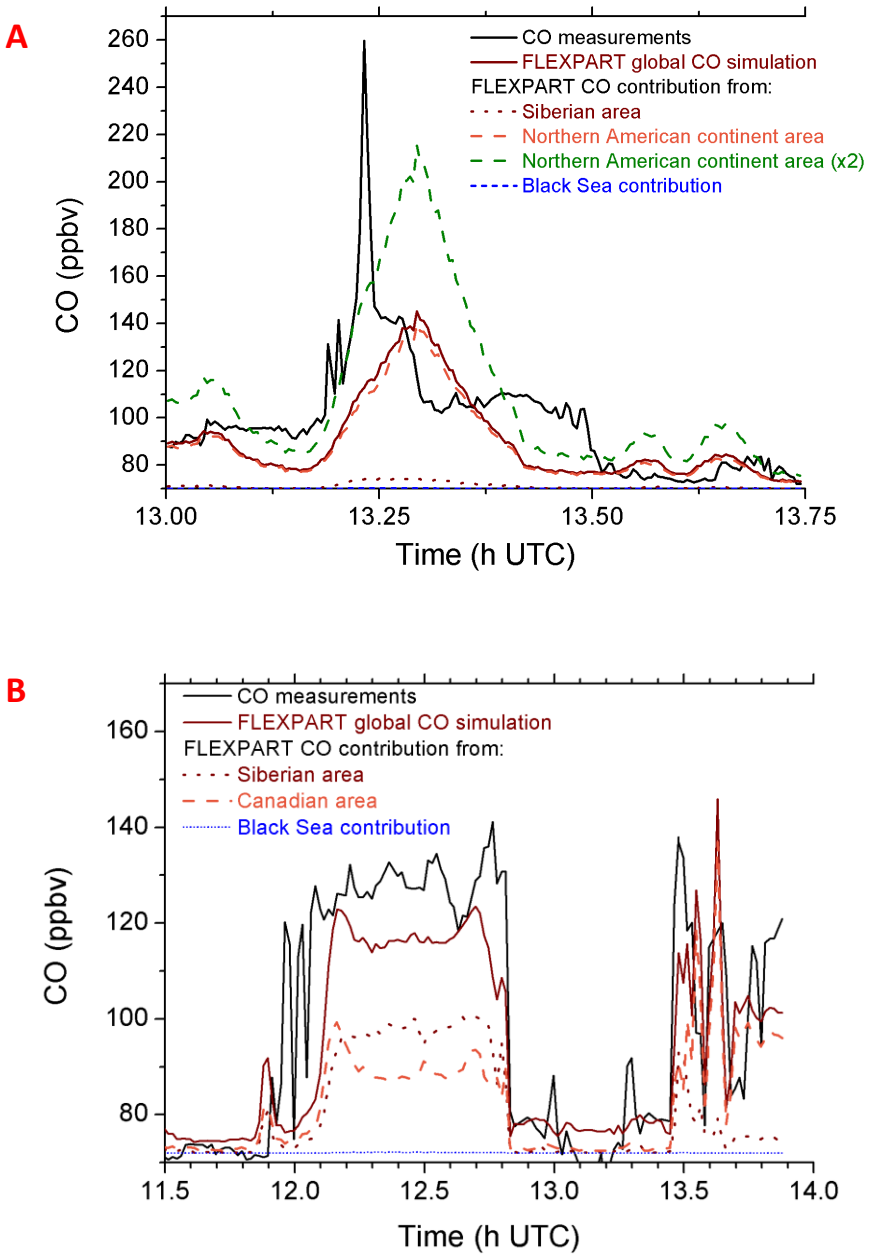

Figure 10. (**A**) Same as Figure 3a in the manuscript with the contribution of CO from biomass burning calculated in the Southern Russia area, named Black Sea contribution in the figure. (**B**) Same as Figure 6a in the manuscript with the contribution of CO from biomass burning calculated in the Southern Russia area, named Black Sea contribution in the figure.

12. Figure 6: there are two peaks around time 13.0 and 13.5 in both CO and BC. Those of BC are larger than the signal discussed in the paper (between times 12-13). These peaks are apparently completely unrelated to forest fires, because are not minimally reproduced by FLEXPART. I suggest to

add a note on these peaks in the text, perhaps leaving them for future study or suggesting some speculative hypothesis on their origin (anthropogenic?).

→ A coupling between the PES and EDGAR inventory (for anthropogenic emissions) has been performed in order to evaluate the CO contribution from anthropogenic emissions to those two peaks (Figure 11 below). The Figure shows that anthropogenic emissions contribute to about 4 ppbv (compared to an excess of CO of about 15 ppbv) for the first and second peaks. Thus, it cannot only explain the peaks measured but a background of anthropogenic emissions can contribute to it. A hypothesis for the aerosols peaks could be the presence of dust particles although the size presented in the paper (0.21-1.1 µm) is a bit small for this category. We add a note in the text as follows:

> *Two peaks of aerosols are also measured around 13.1h and 13.4h UTC (Fig. 5). FLEXPART simulation shows that these peaks are not related to biomass burning as it is not reproduced by the model (Fig. 6c). These two spikes of aerosols could be considered as being dust particles although the associated size (0.21-1.1 µm) is rather small for this type of particles.*

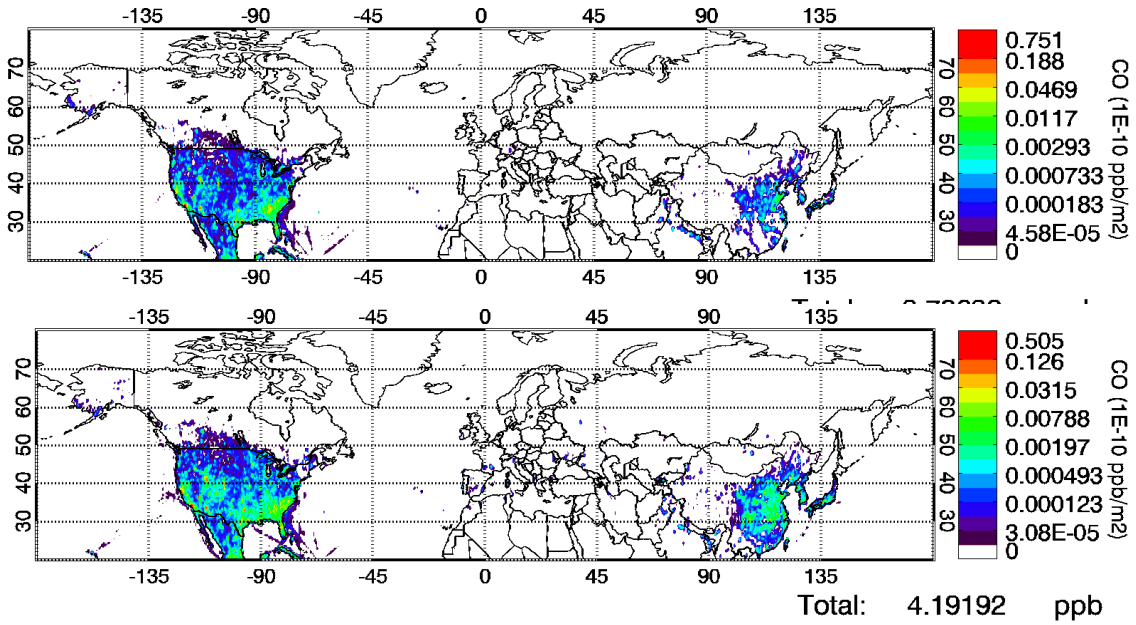

Figure 11. Global CO anthropogenic contribution in a 0-200 m layer on 6 August for the first peak (upper panel) and second peak (lower panel).

Other modifications in line with the editor's and referee #2 advices:

- Figure 3c and Figure 6c have been modified. The legend that was first 'BC measurements' becomes 'Aerosol measurements' as we cannot discriminate the type of aerosols with the PCASP instrument.
- The O_3 measurements have been added on Figures 1 and 5 in the paper.