

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

## RESPONSES TO THE ANONYMOUS REFEREE #2

We 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.

The discussion of the data is somewhat qualitative and various estimates are made of injection height from the sources of the plumes and of the amount of carbon monoxide which is released to account for the concentrations detected at the interception point. The reason for the need for this is not discussed any detail or whether this is typical when modelling smoke plumes.

→ The estimation of the injection height is not so qualitative since it uses CALIOP data as it is commonly done (e.g. Labonne et al., 2007; Kahn et al., 2008; Ancellet et al., 2016). Carbon monoxide has only been used in the Flexpart model with the aim of confirming the presence of concentrations of CO greater than the background value at high altitudes.

1. In addition to carbon monoxide and black carbon the paper reports that ozone measurements were made on board the aircraft but no use is made of these measurements. This is a major omission. Many papers do comment on ozone production during long-range transport. The authors are aware of this and quote suitable references.

→ We agree and added a new paragraph entitled “4. Analysis of O<sub>3</sub> production during long-range transport” in the paper:

*The simultaneous increase of CO and O<sub>3</sub> measurements shows the production of O<sub>3</sub> inside the plume (Fig. 1 and 5). The ratio  $\Delta O_3/\Delta CO$  for the increase of the species with respect to their background values averaged over 20 minutes before and after these increases for F2 and F8, respectively, is of about 0.25 for Flight 8 and of about 0.50 for Flight 2. It has been shown that this ratio increases with the age of the plume (Jaffe and Widger, 2012). For our two flights and for boreal regions, these ratios correspond to a plume age  $\geq 5$  days (Jaffe and Widger, 2012; Parrington et al., 2013; Arnold et al., 2015). More precisely, the ratio gives an approximate plume age of 6-10 days for F8 and of 13-15 days for F2 (Jaffe and Widger, 2012), in agreement with the age of the air mass calculated with FLEXPART.*

*Jaffe, D. A., Wigder, N. L.: Ozone production from wildfires: A critical review, Atmos. Env., 51, 1-10, ISSN 1352-2310, <https://doi.org/10.1016/j.atmosenv.2011.11.063>, 2012.*

*Parrington, M., Palmer, P. I., Lewis, A. C., Lee, J. D., Rickard, A. R., Di Carlo, P., Taylor, J. W., Hopkins, J. R., Punjabi, S., Oram, D. E., Forster, G., Aruffo, E., Moller, S. J., Bauguitte, S. J.-B., Allan, J. D., Coe, H., and Leigh, R. J.: Ozone photochemistry in boreal biomass burning plumes, Atmos. Chem. Phys., 13, 7321-7341, <https://doi.org/10.5194/acp-13-7321-2013>, 2013.*

*Arnold, S. R., Emmons, L. K., Monks, S. A., Law, K. S., Ridley, D. A., Turquety, S., Tilmes, S., Thomas, J. L., Bouarar, I., Flemming, J., Huijnen, V., Mao, J., Duncan, B. N., Steenrod, S., Yoshida, Y., Langner, J., and Long, Y.: Biomass burning influence on high-latitude tropospheric ozone and reactive nitrogen in summer 2008: a multi-model analysis based on POLMIP simulations, Atmos. Chem. Phys., 15, 6047-6068, <https://doi.org/10.5194/acp-15-6047-2015>, 2015.*

2. It would be easier to understand the vertical structure of the smoke plumes if simple vertical profiles were shown rather than the complex system adopted by the authors with colour coding. The

description in the text focuses on horizontal information whereas vertical information would be just as useful since this would indicate the thickness of the layers in a more obvious form.

→ An estimation of the thickness of the biomass burning has been performed for the 6 August. It uses the measurements done during the vertical profile when descending for landing on Lampedusa (Figure 10, not shown in the manuscript). The layer is about 2.9 km thick. A sentence is added in the text, at the end of section 3.2:

*The measurements performed during this vertical profile help us determining that the thickness of the layer is 2.9 km.*

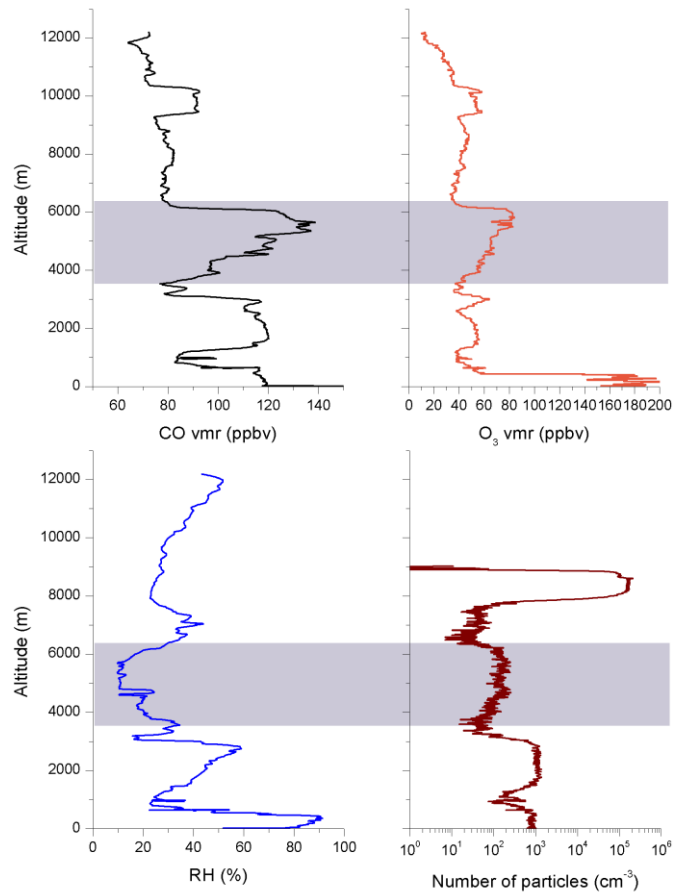


Fig. 10: Vertical profiles of CO (black) and O<sub>3</sub> (orange) vmr, RH (blue) and particles density (brown) on 6 August. The grey rectangle represents the biomass burning layer.

3. The paper focuses on the use of the trajectory model FLEXPART to identify the origin of the smoke plumes. It does however also refer to the use of the chemistry-transport model but this is only used to confirm the FLEXPART findings. It is not used to comment on any chemistry which may occur as the plume progresses around the atmosphere. Surely some comments regarding ozone production or destruction in the plumes could have been discussed.

→ To fill in this gap, we add a section in the paper dedicated to the analysis of the O<sub>3</sub> production inside the plume during its long-range transport using the MOCAGE model. See section “4.2 Analysis of O<sub>3</sub> production with MOCAGE” in the paper:

*In this section, MOCAGE simulation is used to analyse the O<sub>3</sub> production inside the biomass burning plume during long-range transport. For flight F2, the emissions are set up to an injection height of 10 km without any coefficient applied to the emissions.*

*MOCAGE simulates fairly well the O<sub>3</sub> background that is of ~40 ppbv compared to ~32 ppbv for the measurements (not shown). The simulation reproduces the variability of O<sub>3</sub> in good agreement with the measurements. For the first period of interest, between 12.0 h and 12.8 h UTC, MOCAGE simulates an increase of ~25 ppbv O<sub>3</sub> compared to ~35 ppbv for the measurements. For the second period of interest, at about 13.5 h UTC, MOCAGE simulates an increase of ~30 ppbv compared to ~50 ppbv for the measurements. Note that MOCAGE provides smoother peaks than the observations because of the finer resolution of the observations compared to the model. Considering this, MOCAGE reproduces well the measurements of flight F2 and is thus used to study the production of O<sub>3</sub> along the transport.*

*Figure 9 shows both the O<sub>3</sub> vmr and O<sub>3</sub> production on 25 July and 1 August at 5.5km altitude. The complete panels of maps from 23 July to 6 August are provided as supplementary material to follow the production (Fig. S1) and the concentrations (Fig. S2) of O<sub>3</sub> during the travel of the air mass from Siberia to the MB. It shows high O<sub>3</sub> production in the biomass burning plume up to 3 days after the emission (Fig. S1). After that, the ozone production is lowered indicating an aging of the air mass. On 25 July, the production of O<sub>3</sub> is visible above Siberia between 40°N and 70°N (Fig. 9a). Figure 9b shows this production of O<sub>3</sub> with concentrations of O<sub>3</sub> greater than 110 ppbv in the same area. Then, the air mass crosses the Pacific Ocean before arriving above Canada. On 1 August, the simulation shows the production of O<sub>3</sub> between 30°N and 90°N (Fig. 9c). The concentrations of O<sub>3</sub> in Figure 9d are more important in this area, especially around 45°N with concentrations up to ~100 ppbv and around 70°N with concentrations up to more than 120 ppbv.*

4. A minor point: The authors state in the text that on Flight 8 CO reaches 260ppb and the particle count spikes to approximately 1000 particles per ml. The majority of concentrations intercepted on Flight 8 and Flight 2 are rather similar and the higher concentration experienced on Flight 8 are only transitory. The text does not seem to convey this message.

→ We agree on this statement and added modifications in the text explaining that the increases in CO and BC measured are approximately of the same range and that the huge peak on Flight 8 is a transitory event. A sentence has been added in the text section 3.1:

*During the transect at about 9.7 km asl, an increase of CO vmr up to ~110 ppbv (from a background at ~70 ppbv) has been measured above Sardinia. A very intense and transitory increase of CO up to about 260 ppbv has been measured among this general increase of CO, correlated with a weaker increase in O<sub>3</sub> (from ~35 ppbv to ~75 ppbv) and aerosols up to about 1000 particles cm<sup>-3</sup> in the 0.21-1.1 μm diameter range, and a decrease in relative humidity (RH).*

and in section 3.2:

*The background concentrations are rather similar to the ones measured during F8, however the peak intensity of CO is lower.*

5. On Flight 2, in Figures 5 and 6, two large spikes of particles are shown around 1300 UTC, however there seems to be no increase in CO. There is no comment about this; presumably they are not associated with the fire plumes. Do they contain black carbon for instance?

→ It is not possible, from our measurements, to discriminate the type of aerosols. But, it is clear that from our simulations, we can discard a contribution from biomass burning. See point 12 of the responses to the Reviewer #1 comments.

**Other modifications in line with the editor's and referee #1 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<sub>3</sub> measurements have been added on Figures 1 and 5 in the paper.