

**“Isotope- and tracer-based measurements of fossil fuel
and biospheric carbon dioxide in Paris during winter 2010”
by M. Lopez et al.**

Answers to reviewer 2:

The authors wish to thank the anonymous referee 2 for her/his helpful and constructive comments which improved the quality of our manuscript.

General comments: This paper describes the source attribution of carbon dioxide in Paris during the MEGAPOLI campaign of January-February 2010. The authors use carbon isotopic compositions, $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$, to allocate local emissions among gas and liquid fuels and the biosphere. The fossil fuel-generated CO_2 amounts (CO_2ff) combined with CO and NO_x measurements are used to calculate emissions ratios of $\text{CO}/\text{CO}_2\text{ff}$ and $\text{NO}_x/\text{CO}_2\text{ff}$ to produce a continuous record of CO_2ff over the course of the campaign. CO_2ff depends on the meteorologic regime, being higher when the air arriving in Paris has come from the north over the industrialized continent than when it comes from the west. In the latter case, the CO_2ff is lower in abundance and is characterized by more local emissions. The tracer emissions ratios are compared to emissions inventories in order to further understand the sources of CO_2ff .

The inclusion of NO_x as a tracer for CO_2ff is new and offers an approach for identifying very local emissions, because of its very short lifetime in the atmosphere.

Although this paper is generally well-written, the Results and Discussion sections can be difficult to follow at times, especially section 3.5. A location map should be included to show the sampling sites and major wind regimes.

Answers to the general comments:

In order to improve the flow of section 3, we reorganized the results and discussion sections by moving calculation of CO_2ff to part 2.2.2 (flasks sampling strategy). We also removed less relevant information from the section 3.5 in order to further clarify this section.

We also followed the referees advice and added a map in order to show the sampling sites in Paris and Plateau of Saclay. We also included two maps showing the CO source contribution for each air masses regime.

Answers to the specific comments:

Abstract: You make no mention of the meteorological impact on observed sources, derived from your continuous record over the entire campaign.

R: We included the sentence: “The ratios $\text{CO}/\text{CO}_2\text{ff}$ and $\text{NO}_x/\text{CO}_2\text{ff}$ are functions of air mass origin”. We don't want to introduce in the abstract the different air mass regimes which are defined in the text later.

p. 2375, line 12: insert “the” after “with”.

R: Done.

p. 2376, line 2: “attended” should be “intended”.

R: Done.

line 5: delete “of”.

R: Done.

p. 2377, line 19: Clarify what you mean by “in a vented location”.

R: We deleted “in a vented location” in order to not induce confusion.

p. 2378, line 12: “suburban” (no hyphen)

R: Done.

p. 2380, line 23: Change “Semi-urban” to “suburban”.

R: Done.

p. 2381, line 23: How were the flask samples dried?

R: We added the following sentence:

At LHVP, samples were dried using a cartridge filled with magnesium perchlorate. At Jussieu, the samples were dried by passing through a glass water trap maintained in an ethanol bath at -60°C as described by Neubert et al., 2004.

Lines 25-27: Explain the sampling technique – were the samples collected into evacuated flasks or by flushing, and if flushing, what time period was integrated?

R: We reorganised this part to explain in more details how the flasks were filled and dried.

At LHVP, we flushed the flasks with dry ambient air for 10 minutes before pressurization. At Jussieu, flasks are automatically flushed for 2 hours before pressurization as we used an automatic flask sampler.

What do you mean by “flasks were regularly filled for two complete days and one night”?

R: We automatically sampled flasks every two hours during two complete and different days and one night. We replaced the sentence with: “At Jussieu, we filled 35 flasks of 2.5L volume every two hours for two complete days (09-10 February and 14-15 February) and during one night (11-12 February).”

p. 2382, lines 6-9: The first sentence should mention that you are talking about the $\Delta^{14}\text{C}$ measurements.

R: We added this mention.

line 16: Delete “with” before “in situ”.

R: Done.

p. 2383, line 19: Does “one-hour resolution” mean that you averaged the continuous data into one-hour bins?

R: The continuous data measurements have been averaged to obtain a one hour temporal resolution. We clarified this point in the manuscript.

p. 2385, lines 22-29: Include a description of the methods for the non-methane hydrocarbon analyses in the Methods sections. This discussion seems to be just tacked on and not well integrated into the arguments.

R: We think that it is not relevant to add a description of the method for the non-methane hydrocarbon analyses because the NMHC have been monitored by another team/group. We extracted the data from the MEGAPOLI database. For complete details about the measurement technique, we kindly propose to see Gros et al., 2011.

p. 2387, lines 7, 12: Source/reference for the Mace Head data? And did you average these data over the entire time period to derive a constant background?

R: We added the reference Bousquet et al., 2006 for the data selection (already cited in the same section). The background CO_2 mole fraction used is derived from the monthly maritime

background for February 2010. The CO₂ measurements are performed by our team at LSCE.

p. 2389, line 27: The $\delta^{13}\text{C}$ for liquid fuel from Widory and Javoy (2003) may be light. When was ethanol added to gasoline?

R: We used the Widory and Javoy (2003) reference as these are the only direct measurements of $\delta^{13}\text{C}$ from liquid fuel in Paris. Before 2009, ethanol was used with a 5% maximum contribution in gasoline. In 2009, a new gasoline type was commercialised with a 10% ethanol rate. Both type of gasoline are still used and the French government estimated that in 2010, the total use of biofuel is 7%. We replaced the 10% contribution by 7% in the text.

Since Widory and Javoy (2003) the use of biofuel increases from 1% to 7%. In France, 70% of cars are diesels and 30% of cars are gasoline. The biofuel added to diesel is mostly produced from rape and sunflower while the biofuel added to gasoline is partly produced from sugar-beet, wheat, corn and potatoes. Among all listed plants, only corn has a C₄ plant with a $\delta^{13}\text{C}$ signature of around -14‰ but the contribution is less than 7.5% in biofuel and 0.5% in total fuel. Therefore, we can neglect the influence of the isotopic signature from corn.

p. 2390, lines 13-22: Compare results with other cities – e.g., Dallas (Clarke-Thorne and Yapp, 2003); Salt Lake City (Pataki et al., 2003, 2006); Los Angeles (Newman et al., 2008).

R:

We did not compare our results with the study of Clarke-Thorne and Yapp, 2003 because it did not separate use of gasoline and natural gas contributions from their $^{13}\text{CO}_2$ results.

We included the comparison with Pataki et al., 2006: “Pataki et al. (2006) conducted continuous ^{13}C measurements over Salt Lake City (USA) during the early winter 2004-2005. Using the same approach as presented in this section, they found a natural gas contribution varying from 30-40 % during morning rush hour to 60-70 % at pre-dawn.”

We also included the comparison with Newman et al., (2008) as follow: “Newman et al., (2008) analysed $^{13}\text{CO}_2$ grab in flasks for the years 2002-2003 at Pasadena (Los-Angeles basin, California-USA). They found a Keeling intercept - $\delta^{13}\text{C}$ of -29.9 ± 0.2 ‰ which is less depleted than our value (-36.1 ± 2.7 ‰). This shows a stronger annual influence of liquid fuel use in the total CO₂ emission at Pasadena, compared with the winter in Paris. Based on a mass balance approach, the study of Newman et al., (2008) found that 62% of state's energy from fossil fuel is from petroleum products and 37% from natural gas consumption.”

p. 2391, line 4: The “slightly lower” value is well within the reported errors.

R: We deleted this sentence as it is not significant.

p. 2392, lines 9-18: Compare with results from other cities: e.g., Salt Lake City (Pataki et al., 2006); Heidelberg (Vogel et al., 2010); Los Angeles (Newman et al., 2012)

R:

-Newman et al., 2012, the comparison is not relevant because they published results of a spring/summer campaign in Pasadena. They found a FFCO₂ between 12 and 21 ppm overnight to midday which is less than our values.

-We already compared our results with the study of Pataki et al., 2006 (according to your previous comment). However, we cannot compare our results with this study in the p. 2392 because they do not estimate the FFCO₂.

-Vogel et al.2010: they grab 89 samples during pollution events in winter at Heidelberg between 2002-2009 show a FFCO₂ between 10 and 18 ppm. This comparison is also not relevant to our study because Heidelberg is a small city compared with Paris.

p. 2393, line 19: It is not clear what “in the sources spatial descriptions for Paris” means?

Please clarify.

R: We replaced “description” by “distribution”.

line 24: Replace “On a the French scale” with either “For France as a whole” or “On the countrywide scale”.

R: We replaced by “For France as a whole”.

lines 26-29: All of the CITEPA numbers are for years well past 1970 and 1993. Were the regulations phased in during 2005-2010, such that they could account for the factor of 2 decrease during this time?

R: The replacement of old vehicles plays a key role in this decrease as far as the increase of vehicles efficiency. In France, the average age of a car is 8 years.

p. 2394: In general difficult to follow

R: We reduced this part from details which are not important for this paper in order to make it more readable.

line 9: “four inventories” – but 7 are given in the table

R: This part has been deleted for readable reasons (see before).

line 12: Insert reference to Table 5 after “total ratio”.

R: This part has been deleted for readable reasons (see before).

line 13: “more important CO2 source” – clarify and explicitly relate to ratios.

R: The road transport sector is the major emission source of anthropogenic CO2 (77% of anthropogenic emission) while this sector contributes for about 30% of total anthropogenic CO2 emission for the other inventories.

This part has been deleted for readable reasons (see before).

p. 2395, line 12: inset “was” after “NOx”.

R: Done.

line 13: replace “show” with “reflect”.

R: Done.

Table 2: Should a particular time of day be used here – perhaps midday, during the time of greatest mixing and highest planetary boundary layer? Add a footnote stating that “Gif” is Plateau of Saclay, since this is the name you use in the Discussion. Give standard deviations or standard errors.

R: We averaged the whole time series. We added in the legend the sentence “from January 15 to February 20”.

We replaced “Gif” by “Plateau of Saclay”.

We added the standard deviations in Table 2.

Fig. 3: Plotting the standard deviations so boldly suggests that the patterns for continental and oceanic regimes are not significantly distinct. Plot standard errors (listing n in the caption) of the hourly averages instead, perhaps with shading to indicate variability by standard deviation.

R: We changed the colours for the error bars and make the curves more visible.

Fig. 4: line 4 of caption: Replace “blue dotted lines” with “blue horizontal dashed line”.

R: Done.

