

Interactive comment on “Better constraints on sources of carbonaceous aerosols using a combined ¹⁴C – macro tracer analysis in a European rural background site” by S. Gilardoni et al.

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Answer to Referee #3

R: This manuscript presents a comprehensive and all-year-round assessment of PM_{2.5} aerosol sources at a rural background site. It combines methodologies that are not new to the field and their use has been successfully demonstrated in recent years for similar purposes. Therefore the reviewer does not feel that the first word of the title ‘Better constraints on: : :’ can be justified. In this sense, the title of any scientific paper that

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is published in high-standard journals should start with this word in order to indicate that it indeed adds something new to already existing knowledge. Nevertheless it is a useful manuscript the statements of which corroborate previous European results of similar nature and lead to very similar conclusions and recommendations.

A: The words “better constraints” want to highlight the lowering of source strength uncertainty compared to the uncertainties that characterized emission factors and reference fractions used to solve the source apportionment problem.

R: Page 2506 Line 10 suggest ‘particulate matter’

A: “Particle matter” was corrected as “particulate matter”.

R: Page 2506 Line 22 In fact, no tracer method is capable of ‘accurately’ and ‘completely’ characterize any single source. All source apportionment studies are just estimates of potential source contribution relying on a number of simplifying assumptions.

A: Sentence is modified as follow: “However, these tracer methods have not completely characterized organic aerosol and especially Secondary Organic Aerosol (SOA).”

R: Page 2507 Line 13 ‘More recently’ – above the authors quote papers from 2009 and 2010. What could be ‘more recent’ than those? In fact, the quoted simplified approaches are no longer the ‘state-of-the-art’ methodologies in source apportionment, despite their recent publications. Either this should be put into a more historical perspective or simply omitted.

A: Sentence is rephrased as: “Using a different approach, source apportionment studies have been integrated with ¹⁴C measurements to distinguish fossil from non-fossil carbon”

R: Page 2507 Line 20 rephrase ‘: : :is spread throughout the atmosphere: : :’

A: Sentence is rephrased as: “The ¹⁴C produced is quickly oxidized to carbon dioxide which is taken up by plants through photosynthesis in the troposphere”.

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R: Page 2507 Line 22 rephrase ‘.atmospheric values of contemporary 14C: : :’

A: Sentence is rephrased as: “Thus, 14C is incorporated into all land-living organisms.”

R: Page 2508 Line 9 suggest ‘: : associated with: : :’

A: Sentence is corrected.

R: Page 2508 Line 20 In addition to what? The reference 14C/12C values were already used as variables already in the two quoted papers.

A: “In addition” introduces the sentence that explains how, differently from the quoted papers, the present study defines the reference fraction of non-fossil carbon as a function of the reference fraction of biomass burning carbon.

R: Page 2508 Line 25 Why the ‘representativeness of results’ is guaranteed if the method is applied on a larger dataset?

A: The statistical representativeness of any analysis increases when the number of samples used to investigate the system is increased. Sentence is rephrased as: “This enhances the representativeness of results and the significance of seasonal differences.”

R: Page 2510 Line 7 May the selection criteria (high loadings of TC) distort the conclusions of the study strongly in favour of anthropogenic sources? May higher biogenic contributions be expected at days with low TC?

A: The authors thank the referees to underline the need to better clarify the representativeness of data subset. The following sentence is added at page 2520 line 16: “TC concentrations of the subset of samples ranges from 3 to 53 ug m⁻³, while the range corresponding to the entire dataset is 0.4 – 53 ug m⁻³. The subset TC concentrations correspond to the range 15th percentile – 100th percentile, indicating that clean days were included in the source apportionment analysis, as well as polluted days.”

R: Page 2514 Line 20 ‘discrimination’

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A: Text is corrected.

R: Page 2518 Line 5 Usually with atmospheric dilution the temperature is also changing (decreasing, especially when convection take space). So it is not at all evident that the primary components are ‘overestimated’: semi-volatile compounds may also condense on primary particles without being oxidized in the gas-phase.

A: The authors agree that cooling of emissions increases the OC to EC ratio, because of condensation of semi-volatile species. On the contrary, the ratio is lowered by dilution, due to gas-particle partitioning of primary organic aerosol (Robinson et al. 2007). Emission factors are usually measured at dilution that reduces the temperature to ambient level, while the aerosol concentration is order of magnitude higher than atmospheric level (Shrivastava et al. 2006). Measurement of emission factors under these conditions leads to overestimation of the OC to EC emission ratios.

R: Page 2519 Line 13 change ‘reduces’

A: Text is corrected.

R: Page 2522 Line 6-8 Why do small concentrations of levoglucosan in fall indicate that wood burning in the dominant source in winter? It should be trivial anyway, but not from the logic of this sentence.

A: Sentence is rephrased as: “In the study area the possible sources of biomass burning carbon are wildfires, agricultural waste burning, and wood burning for residential heating. The absence of this source during summer, when dryer weather conditions could facilitate wildfires in the surrounding regions, and the small concentrations during fall, when agricultural waste burning takes place, indicate that the main source of biomass burning carbon observed in Ispra was wood burning for residential heating.”

R: Page 2524 Line 9-10 Why did the authors use the same OM/OC ratios for primary and secondary OC for all categories? I would have expected larger values for SOCs: e.g. 2.1 for SOCb

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A: The OM is recalculated using the OM to OC ratio equal to 1.8 and 1.4 for SOA and POA, respectively.

R: Page 2525 Line23 It is reasonable to combine primary and secondary BB and FF sources. From the point of control, it does not make much difference but introduce large uncertainties which make the results less 'credible' for decision-makers.

A: To combine primary and secondary organic carbon makes more difficult the evaluation of pollution reduction strategies. On the other hand, the combination of primary and secondary OC results in a reduction of numerical uncertainties.

References

Shrivastava, M., Lipski, E., Stainer, C. O., and Robinson, A.L.: Modeling semivolatile organic aerosol mass emissions from combustion systems, *Environmental Science and Technology*, 40, 2671-2677, 2006.

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