Interactive comment on ”Estimating contributions from biomass burning and fossil fuel combustion by means of radiocarbon analysis of carbonaceous aerosols: application to the Valley of Chamonix” by L. Bonvalot et al.

General comments

The authors present a comprehensive evaluation and validation of the novel EA-GIS-AixMICADAS facility, used to measure radiocarbon without any prior graphitization. The method is also applied to real aerosol samples from the alpine Chamonix Valley. The authors prove the accuracy and precision of the method in a satisfactory manner. Further, great benefit with this facility and method compared to other accelerator mass spectrometers (AMS) is the fact that no graphitization of aerosol samples are needed prior AMS. This makes the method more cost and time efficient. From my experience with graphitization this also means that several errors and sample losses can be avoided.

The applicability to real aerosol samples from the Chamonix Valley show satisfactory results which are in line to what one can expect in terms of source impact during different seasons. The source apportionment model to calculate TC fractions of biogenic, biomass burning and fossil fuel combustion is presented in a clear and concise way and is easily applicable by other researchers for similar studies.

The language is on a clear and high level.

The title is in my opinion to broad and general. It does not say anything about the novel radiocarbon analysis without graphitization. Further, if the authors are about the mention sources in the title as “biomass burning” and “fossil fuel combustion”, I am wondering why they don’t mention biogenic carbon? This fraction has a considerable role in the results and discussion session in the paper. Finally, the sources were not solely determined by radiocarbon, I would say that levoglucosan was equally important, so why omit levoglucosan?

I recommend this manuscript to be published in ACP.

Specfic comments

Page 1, line 15. Please explain the abbreviation AixMICADAS, if not here in the abstract then the first time the abbreviation appears in the following text.

Page 1, line 27. The unit is given in $\mu g.m^{-3}$. Why a dot “.” between µg and m^{-3}, this appears several times in the text but is not consequent. Sometimes this unit is written without a dot between.

Page 2, line 17. Can you please give a number of how large the carbonaceous fraction of PM can be, this would give important knowledge and a feeling for the numbers in this field, especially for new readers.

Page 2, line 21. It feels a bit arrogant to state that there are “obvious scientific and societal implications”, I suggest that you erase the word “obvious”.

Page 2, line 20-26. Consider re-write this paragraph, it is a bit confusing. End with "ideal test site for such measurements". Which measurements? Those you are referring to in line 20?

Page 3, line 16. The AMS AixMICADAS, state its manufacturer and model name if possible.

Page 3, line 17-18. Other studies have also shown to handle small samples 10-100 µgC with graphitization prior AMS (Genberg et al., 2010).

Page 3, line 31. What do you mean by hybrid ion source? This it can both handle graphite targets and CO_2 gas? Please clarify this.

Page 4, line 11. In what atmosphere are you heating the sample to evolve the CO_2? This should be stated. Further it is a bit confusing in which temperature the CO_2 is evolved, in 450°C or in 1050°C in the EA? Please clarify this.

Page 4, line 15. It is a bit confusing that you mention the sampled PM_{10} filters here prior to the paragraph regarding sampling of filters. Please consider putting the sampling paragraph before the AMS section.

Page 4, line 22. OxA2 is an abbreviation, please spell out the whole name of this standard.

Page 4, line 26. Why did you consider these 46 OxA2 gas samples as unknown samples when you obviously knew the $F^{14}C$ of this SRM?

Page 4, line 33. A_{SN}/A_{ON}. Please explain or omit this.

Page 5, line 11-14. I would say that the filter handling and preparation induces most contamination, do you have any reference saying that the silver boat induces large or substantial contamination?

Page 6, line 7. You should say that SRM stands for Standard Reference Material. This is not known to everyone.

Page 6, line 10. What is AGE-3 system? Reference?

Page 8, line 7. DECOMBIO, abbreviation for what?

Page 8, line 10. Please be more specific on the sampling locations. Was it on roofs of buildings? Ground level? How close to the nearest road? Surrounding landscape? Mountains, forests, pastures etc?

Page 8, line 13. Did you prebake the quartz fiber filters to avoid contamination of VOC's prior sampling? What size of filter did you use? What was the brand and model of filters and sampler?

Page 8, line 18. HPLC-PAD, abbreviation for what?

Page 8, line 22. Please state the brand and model of the TOA.

Page 8, line 23. Please state the brand and model of the TEOM-FDMS.

Page 9, line 4. Please explain the abbreviation LGGE.

Page 9, line 9. Please explain the abbreviation CEREGE.

Page 10, line 6. Please mention some meteorological conditions that may reduce photo-oxidation during winter.

Page 10, line 19. Please explain $F^{14}C_{bio}$ to the reader.

Page 11, line 3-4. "For the summer season, it is considered that all non-fossil carbon originates from organic compounds naturally released by living plants". Is this consideration true? What about organic PM from charcoal BBQs? Forest fires?

Page 13, line 15. These $F^{14}C$ values and explanation should have been presented earlier in the manuscript. At page 10 for instance.

Page 14, line 5-7. $TC_{bb}=TC_{NF}-a*[levoglucosan]$. To me, what you propose in this equation is the calculation of TC_{Bio}, i.e. $TC_{Bio}=TC_{NF}-a*[levoglucosan]$. TC_{bb} should be: $TC_{bb}=a*[levoglucosan]$, where a is the slope between TC_{NF} and levoglucosan.

With the currently proposed formula, TC_{bb} would be zero (0) during winter which seem highly unrealistic.

Page 15, line 27. Instead of using LGGE and CERGE, which I assume are labs (?), I would prefer if you state the actually used method instead, i.e. EA and TOA. This would make more sense. Either way, you need to explain the abbreviations LGGE and CERGE, which currently are adding confusion to the manuscript.

Page 24, Table 1. "X modern carbon". Please state the unit of this parameter and explain it in the caption of the table.

Page 24, Table 1. Which proportions of each SRM did you use in the mixture?

Page 24, Table 1. "Error". What type of error is this? Should be stated.

Page 24, Table 1. "Measurement after graphitization". Here should be a unit in this column.

Page 25, Table 2. "0.532 $F^{14}C$". Remove "$F^{14}C$" as it is stated in the explaining column text.

Page 26, Table 3. Please explain the "Winter $f_{NF,ref}=1.10$ $F^{14}C=1.09$ f_M" that is stated in the table. Same for summer a couple of rows further down. These should be explained in the table caption.

Page 26, Table 3. Column "± Carbon mass [$\mu g.m^{-3}$]". Please state the type of uncertainty, SD, SE, CI?

Page 27, Table 4. Please state the type of uncertainty, SD, SE, CI?

Page 28, Table 5. Please state the type of uncertainty, SD, SE, CI?

Page 29, Table 6. First row, first column. Write “Date”.

Page 29, Table 6. Please state the type of uncertainty, SD, SE, CI?

Page 30, Figure 1. Please explain PA, Oxa2, M_M and M_S in the figure caption.

Page 31, Figure 2. Please explain what you mean by “simulated” in the figure caption. From where have you derived the “Theoretical $F^{14}C$”, explain. Should further be stated in the figure caption that this graph includes measurements of SRM’s.

Page 32, Figure 3. “Blue ribbon”, looks green to me. “A large scatter is exhibit which can be caused by….” This sentence sounds erroneous.

Page 34, Figure 5. State that you are comparing EA-GIS and TOA instead of LGGE and CEREGE.

Page 35, Figure 6. Use “TC” or “Carbon Concentration” on the y-axis? Consistency.

Technical corrections

Page 5, line 16. Parenthesis error.

Page 13, line 23. Change “TableTable 5” to “Table 5”.

Page 14, line 7. Change “[levoglocosan]” to “[levoglucosan]”.

Page 25, Table 2. The font is not consistent in the table.

Page 27, Table 4. Change “masse” to “mass”.

Whole document: Please be consistent whether you use µgC or just µg. There are discrepancies throughout the whole document, in the tables and figures.

Whole document: Please be consistent whether you use [$\mu g.m^{-3}$] or [$\mu g\ m^{-3}$], same error can be found in ng (nanograms). Personally, I don’t see why you use a dot in between. There are discrepancies throughout the whole document, in the tables and figures.

References

Genberg, J., Stenstrom, K., Elfman, M., and Olsson, M.: DEVELOPMENT OF GRAPHITIZATION OF mu g-SIZED SAMPLES AT LUND UNIVERSITY, Radiocarbon, 52, 1270-1276, 2010.