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 Lise Bonvalot et al.

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This study presents PM_{10} aerosol data obtained in summer and winter in a valley of the French Alps. Among others, a source apportionment study has been made with the aim to distinguish sources as fossil fuel, biomass burning and biogenic emissions on the base of ¹⁴C measurements and levoglucosan. This revealed that summer samples exhibit an important relative contribution of non-fossil sources and a dominant contribution of biomass burning in winter. Interestingly, this very valuable data set and its important conclusions are similar to what was obtained in two source apportionment studies (Gelencsér et al., 2007; May et al., 2009) made on the basis of a two year round data set sampled on a weekly basis at five rural/remote sites in Europe. These detailed literature data set reflects atmospheric conditions of 2002/2003 on a European west east transect at altitudes from 40 to 3100 m asl. Given the fact that the source apportionment calculations were very similar than ins this study here, i.e. including also ¹⁴C and levoglucosan measurements to distinguish fossil, biomass burning and biogenic emissions, it might be worth that the authors have a look on this dataset and benefit by comparing their new results with these existing literature data.

References:

Gelencsér, A., B. May, D. Simpson, A. Sánchez-Ochoa, A. Kasper-Giebl, H. Puxbaum, A. Caseiro, C. Pio, and M. Legrand (2007), Source apportionment of PM2.5 organic aerosol over Europe: Primary/secondary, natural/anthropogenic, and fossil/biogenic origin, J. Geophys. Res., 112, D23S04, doi:10.1029/2006JD008094.

MAY, B., WAGENBACH, D., HAMMER, S., STEIER, P., PUXBAUM, H. and PIO, C. (2009), The anthropogenic influence on carbonaceous aerosol in the European back- ground. Tellus B, 61: 464–472. doi:10.1111/j.1600-0889.2008.00379.x

We thank Dr. Preunkert for her advice about two other papers on ${}^{14}C$ in aerosols (Gelencser et al. 2007, May et al. 2009).

These papers published in 2007 and 2009, are based on similar apportionment calculations as in Szidat et al. (2004, 2006) already cited in our paper.

Both papers cited by Dr. Preunkert are based on the same ${}^{14}C$ analyses of pooled PM_{2.5} aerosol samples for five European sites. Pooling aerosol filters reduces the number of ${}^{14}C$ analyses and allows to reach the necessary carbon amount to perform classical AMS analyses on graphite targets. Consequently, each site is only characterized by two values, one for winter and the other for summer (cf. Table 2 in Gelencsér et al. 2007, and the modified version as Table 1 in May et al. 2009).

In those papers the source apportionment is then based on the assumption of constant emission factors, e.g. $OC_{bb}/levo$ and OC_{bb}/EC_{bb} from the literature, notably based on test combustion in experimental fireplaces and oven.

By contrast to former works based on a few ¹⁴C analyses, our precise study of two close sites relies on more that one hundred of ¹⁴C analyses (duplicates of more than 50 samples), which allows to evaluate the correlation between TC, levoglucosan and ¹⁴C in many filters even characterized by low carbon contents (thanks to the low blank and detection limit reached with the gas ion source coupled to AixMICADAS).

Based on the observed linear relationship (our Fig. 7) we were able to calculate a non-fossil carbon/levoglucosan ratio independent from the literature on test combustion. As underlined in section 3.2 of our paper, the non-fossil carbon/levoglucosan ratio derived for Passy and Chamonix is compatible with the large range reported by Schmidl et al. (2008) for test combustion on various types of wood. Our value is also compatible with the central value and range assumed by Gelencser et al. (2007) and May et al. (2009) from the literature on test combustion. As noted in our paper, our measured value based on the dual radiocarbon-levoglucosan approach agrees very well with those obtained by Zotter et al. (2014) for several Swiss stations.

As far as the apportionment calculation is concerned, the novelty of our approach (section 3.2.3) is to propose to use the ratio derived from the numerous pairs of ${}^{14}C$ and levoglucosan measurements, instead of relying on an assumed and uncertain emission factor.