

Author's response:

We thank Referee #1 for the careful revision and comments which helped improving the overall quality of the manuscript. A point-by-point answer (in regular typeset) to the referees' remarks (in the *italic typeset*) follows. Changes to the manuscript are indicated in blue font.

Anonymous Referee #1

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The manuscript presents the application of two methodologies representing fundamentally different principles and time resolutions. In a sense, the two distinct methods are complementing each other as one gives information on bulk carbon (a significant part of which is non-soluble) whereas off-line AMS technique represents the water-soluble organic and inorganic fractions. It is a real challenge to combine the results of such distinct methodologies to get valuable insight into major factors determining PM source apportionment at that particular location, but it is done correctly and in a scientifically correct way in the manuscript. The methods including statistical processing of the results are up-to-date and well-founded, uncertainties are handled properly and the conclusions drawn are self-consistent and in a sense rather trivial and correspond to what can be dictated by common sense. There are, however, two minor issues that leave some degree of discomfort in the referee upon reviewing the manuscript.

- 1) *The first is that in the Introduction it is explicitly implied by the strongly biased selection of references (Page 2, Line 35) that the whole story of using miniaturised radiocarbon measurements for source apportionment of carbonaceous aerosols has started around 2010 only and been carried out exclusively by groups affiliated to the authors of this manuscript. The fact is that such studies have started around 2000 (see e.g. Lemire et al. JGR 2002), and were also carried out in Europe already at that time (in fact by the group of the authors themselves Szidat et al., 2004) and even within a large scale European project (e.g. Gelencser et al., 2007 JGR). The major conclusions of the latter study were very much in tune with the main findings of this manuscript. Apart from the radiocarbon-based source apportionment studies there have been other studies based on other principles such as specific tracers, OC/EC ratios, inverse modelling and the like, which also pinpoint to the growing contribution of biomass burning to PM aerosols even in highly urbanized areas in Europe. It would be fair to quote some of them in the manuscript, which would also strengthen the conclusions of the manuscript.*

According to the suggestions of anonymous referee#1 we changed the text in the introduction (Page 2, 4th paragraph) as follows:

The radiocarbon (¹⁴C) analysis of particulate matter has proven to be a powerful technique providing an unequivocal distinction between non-fossil (e.g. biomass burning and biogenic emissions) and fossil (e.g. traffic exhaust emissions and coal burning) sources (Lemire et al., 2002, Szidat et al., 2004, 2009). The measurement of the ¹⁴C content of total carbon (TC), which comprises the elemental carbon (EC) originating from combustion sources and the organic carbon (OC), had been the subject of many studies (Schichtel et al., 2008, Glasius et al., 2011, Genberg et al., 2011, Zotter et al., 2014b, Zhang et al., 2012, 2016, Bonvalot et al., 2016). Results have shown that in European sites especially in Alpine valleys, the non-fossil sources play an important role during winter due to biomass burning and in summer due to biogenic sources (Gelencsér et al., 2007, Zotter et al., 2014b). Moreover, at regional background sites close to urbanised areas in Europe (Dusek et al., 2017) as well as in megacities like Los Angeles and Beijing fossil OA may also exhibit significant contributions to the total OA (Zotter et al, 2014a, Zhang et al., 2017). However, the determination of the ¹⁴C content in EC and OC separately is challenging and therefore not often attempted for extended datasets.

In Page 8 Line 22 we added two more citations in the text: Genberg et al. (2011) who reported yearly cycles and used in addition levoglucosan measurements and a chemical transport model and Gilardoni et al. (2011) who as well reported yearly cycles and used back trajectories analysis in addition to the radiocarbon and marker analysis.

So far radiocarbon results have been reported mostly for relatively short periods of time (Bonvallot et al., 2016), mainly describing high concentration events and only few studies report measurements on a yearly basis (Genberg et al., 2011, Gilardoni et al., 2011, Zotter et al., 2014b, Zhang et al., 2016; Zhang et al., 2017; Dusek et al., 2017). Here, for a subset of 33 PM₁₀ filters from the year 2014, we present yearly contributions of OC_{nf}, OC_f, EC_{nf} and EC_f.

To compare our results of the residential wood burning with other studies that not only used ^{14}C analysis but other methods as well, we used the following citations in Page 8 Line 36: Jaffrezo et al., 2005 and Favez et al., 2010 and added Puxbaum et al., 2007 and Sandradewi et al., 2008 (for the aethalometer model).

OC_{nf} was the dominant part of TC throughout the year with contributions of up to 80% in winter and 71% in summer (Fig. 2b) and average concentrations of $8.5\pm 4.2 \mu\text{g m}^{-3}$ and $2.4\pm 0.6 \mu\text{g m}^{-3}$ in winter and summer, respectively (Fig. 3b). Such high contributions in winter strongly indicate that biomass burning (BB) from residential heating is the main source of carbonaceous aerosols in this region, similar to previous reports (Jaffrezo et al., 2005, Puxbaum et al., 2007, Sandradewi et al., 2008, Favez et al., 2010, Zotter et al., 2014b). The coefficient of determination R^2 between OC_{nf} and levoglucosan, a characteristic marker for BB, was 0.92 (Fig. S7a) and the slope ($\text{OC}_{\text{nf}}/\text{levoglucosan} = 4.8\pm 0.3$) lies within the reported range by Zotter et al. (2014b) for Magadino (which was 6.9 ± 2.6).

- 2) *The second is that since this study is confined to a single location with specific orography and local meteorology and covers a sufficiently long period of time, it is more than tempting that the major findings of the study be tested against the results of inverse modelling using (local) emission inventories. I understand that such an approach is outside the scope of the present manuscript, but maybe a follow-up paper would make use of the very same data and would yield extremely valuable information for such exercises.*

We agree with the reviewer that comparing our results to a modelling study is valuable. However, modelling meteorological parameters over a mountainous region is challenging due to spatial resolution limitations for example, a potential alternation of the type of land within one grid. Moreover, in some cases during winter the planetary boundary layer height ends below the measurement stations and therefore a mismatch between measurements and model often occurs in such regions (Ciarelli et al., 2016, Freney et al., 2011). For these reasons, such comparisons are rarely conducted for Alpine regions and would need the development of highly resolved models for specifically this region.

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