

We want to thank Anonymous Referee #3 for their feedback and comments, which we have addressed point by point below.

Referee 3: Furthermore, the manuscript itself is clear, well written and consequently, easy to read (although the description of the procedure becomes a little long).

We thank the reviewer for the positive feedback. We think it necessary to provide a detailed description of our procedure for several reasons. A major goal of the manuscript was to provide sufficient information for replication of the method. Our OC and EC separation and trapping procedure involves multiple steps in which both timing and precision are important and therefore, we intentionally chose to be very specific when describing the procedure. Furthermore, our OC/EC analyzer is available by the Sunset Labs by special request only and no detailed manual exists for this specific model yet, which is why we included a brief description of the instrument. As OC/EC specific ^{14}C analysis is becoming more widely used, we also wanted to emphasize on the challenges and possible biases associated with the radiocarbon analysis of such small samples.

Referee 3: I only notice a weak point in the ms: in my opinion, the conclusions are extremely brief and do not contain all the essential information shown in the paper.

In the revised manuscript, we will include a more detailed conclusion section, which in addition to the original text will include a brief summary of the main results.

The revised manuscript will include the following paragraph in the conclusion section, prior to the original text:

“We used an OC/EC analyzer in a new laboratory setting to quantify the precision and accuracy of OC and EC ^{14}C measurements with the Swiss_4S TOA protocol. A set of OC and EC ^{14}C reference materials, blanks and intercomparison samples, with sizes as small as 4 μgC , were analyzed to evaluate the performance of our analytical set-up, quantify the uncertainties, and compare the consistency of our results for a shared set of ambient air samples from another laboratory. We were able to: (1) successfully separate OC and EC fractions with the Swiss_4S protocol, (2) isolate and trap the different fractions with our newly-developed set-up with high yields and low carbon blanks, and (3) measure precisely the ^{14}C content of the separate fractions by quantifying and correcting for contamination at each set of the analysis. Our results were in good agreement with ^{14}C measurements from the Laboratory of Radiochemistry and Environmental Chemistry at the University of Bern, Switzerland.”

Original:

This is the first study incorporating ^{14}C standard materials to track and quantify background carbon introduced during aerosol OC and EC ^{14}C analysis. In general, the OC and EC contents of an aerosol sample are on the order of a few micrograms (the size of typical EC sample) to tens of micrograms (the size of typical OC sample). Therefore, not accounting for the extraneous carbon introduced during the analysis can significantly

bias the results. This is particularly important for the ^{14}C measurements of the typically smaller EC fraction. For ambient aerosol studies, the correction of field blanks will also be very important. The most recent intercomparison of ^{14}C analysis of carbonaceous aerosols concluded that it is not possible to agree on common procedures of OC and EC isolation among the participating labs, and that an overarching laboratory and method intercomparison quantifying both the concentration and ^{14}C content of OC and EC is still needed (Szidat et al. 2013). Our study represents a first step towards the development of a common protocol for OC and EC ^{14}C measurements.

Referee 3: I don't understand the change of the format in the names and the order of the Tables (Table 1, Table A1, Table A2, Table 2)

The order of the tables was determined based on the journal requirements. In the final manuscript Table 1 and 2 will be in the text and will appear based on the order in which they are discussed. The appendix tables will be at the end only as auxiliary information – they are only given so the reader has access to the data for all standards, blanks and intercomparison samples used in the analysis in this manuscript.

Appendix A1 shows the radiocarbon measurements in (FM and $\Delta^{14}\text{C}$) of all OC and EC standards and blanks and has both uncorrected and corrected data for extraneous carbon.

Appendix A2 shows the radiocarbon measurements (in FM and $\Delta^{14}\text{C}$) of all intercomparison $\text{PM}_{2.5}$ samples. The uncorrected data was not relevant in the intercomparison and was therefore not shown. One column was added with information about the samples – collection location and fraction analyzed, but for simplicity, we will remove it in the revised manuscript and only distinguish between OC and EC samples. In that way the appendix tables will have comparable formatting.

Referee 3: In Table A1, are the uncertainties in the column \pm ? Why are some of them negative?

The \pm columns represent the uncertainty associated with the radiocarbon measurement reported to the left of the \pm column. For clarity, in the revised manuscript the uncertainties will be reported in the same column as the measurement.

Referee 3 brings a good point about why some of the reported uncertainties in A1 are negative. The negative values are associated with the corrected OC and EC blanks - radiocarbon free materials (adipic acid and coal, respectively). Radiocarbon free “blank” materials are used to calculate the modern fraction of the background carbon introduced during sample processing and measurement. The “modern carbon” background is an average value calculated from the measurements of the ^{14}C content of various blank materials of various sizes. Since the true fraction modern value of the radiocarbon blanks (for either OC or EC or any carbonaceous materials) is zero, once the average modern carbon background correction is applied (section 2.4 in the manuscript), some individual

blanks can yield negative values. The uncertainty determination after background correction depends on the FM value of the blank (see Santos et. al. 2007 for detailed description of ultra small samples uncertainty calculation) and therefore results can yield negative uncertainties.

Traditionally, in AMS analysis, blanks are only used to infer the amount and fraction modern signature of the extraneous carbon so samples can be corrected for it. When reporting AMS data, the blanks are therefore reported without background correction. Following conventional AMS data reporting, in the revised manuscript we will simplify our data for the OC and EC blanks and not report corrected ^{14}C data for adipic acid and coal. We will therefore simplify A1 by separating it into two tables – one showing data for the standards (both corrected and uncorrected), and one showing data for the blanks without background correction.

Referee 3: In Figure 3: Are the text in X-axis size or sample size? The right part of the figure (from 0.1 to 1) is unnecessary. You can adapt and resize the four panels to take up all the available space.

In the revised manuscript, the X-axis will be re-labeled to “Sample Size”.

The size of the plots however, was intended to show that the OC and EC samples, discussed in this manuscript, are much smaller than a regular AMS sample (~1 mgC) and therefore fall on the left side of the plots. Therefore, we think it is important to keep the size scale of Figure 3.

Reference:

Santos, G. M., Southon, J. R., Griffin, S., Beaupre, S. R., and Druffel, E. R. M.: Ultra small-mass AMS ^{14}C sample preparation and analyses at KCCAMS/UCI facility, Nucl. Instrum. Meth. B, 259, 293–302, 2007