Author’s response:

We thank Referee #2 for the careful revision and comments which helped in improving the overall quality of the manuscript.
A point-by-point answer (in regular typeset) to the referees’ remarks (in the *italic typeset*) follows, while changes to the manuscript are indicated in blue font.

Anonymous Referee #2

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The manuscript presents results from an analysis of atmospheric filter samples collected during 2013 and 2014 in Switzerland using offline HR-ToF-AMS and carbon 14 measurements. The results give increased insights into the sources and types of aerosols observed. Especially interesting is the focus on the type/source of the precursor for the factors instead of the more commonly used degree of oxidation or volatility. The methods and the descriptions of the data analysis are very thorough and a good deal of work is done in calculating and communicating the uncertainties. This manuscript presents results that follow expected trends in the formation and processing of atmospheric aerosols and thus serves as a good demonstration of the feasibility of combining these two analyses. I recommend addressing two minor issues.

1) *A mention of blanks is made with respect to the radiocarbon analysis, but there is no discussion of how blanks were handled for the AMS analysis. Were blanks extracted and prepared in the same manner as AMS samples? How did the authors account for the fact that dilute solutions may not show aerosol signal in the AMS when atomized, despite there being some level of organic material in the solution?*

Indeed, in the offline AMS analysis the field blanks were extracted and prepared in the same way as with the samples. In several studies in the past (Bozzetti et al., 2016, 2017a, Daellenbach et al., 2017) field blanks were measured and compared to the nebulized ultrapure water. The resulting signal of the field blank, as in our case, was not statistically different from that of nebulized Milli-Q water.
To ensure that particles generated from dilute solutions are not smaller than the AMS lens transmission and could be measured, we have nebulized NH_4NO_3 and $(NH_4)_2SO_4$ solutions (1ppm), providing additional material in the blank. For a number of *m/z* (45%), the resulting signals are statistically significantly higher than nebulized Milli-Q water (by up to a factor of two), but remain negligible compared to ambient filter signals (on average by a factor of 120). As some of this signal can arise from additional operations during solution preparation (e.g. impurities in the salts or different materials (glassware) used for the salt solution preparation compared to the sample preparation) and as the associated signals are negligible (<1% of the signals), we do not correct the filter measurements for the blanks obtained using nebulized NH_4NO_3 and $(NH_4)_2SO_4$ solutions.

2) *The authors could increase readability of the manuscript by providing the names corresponding to acronyms in the text the first time the acronyms are used. This includes the factors as well as all components in equations. Also, the letter labels (a, b, c, and d) are missing on Figure 3. It would also be beneficial to have names for the factors in all of the corresponding figure captions.*

Corrected as suggested in:

Page 4 Lines 35 and 36: $\max ATN_{S_3}$ is the maximum attenuation in step three, while $initial\ ATN_{S_2}$ and $initial\ ATN_{S_1}$ are the initial attenuations in step two and one, respectively.

Page 6 Line 11: water soluble organic matter ($WSOM_i$)

Page 7 Line 7: and $\left(\frac{OM}{OC}\right)_{bulk}$ is estimated from the input data matrix for the PMF.

Page 7 Line 11: Where $(\frac{OM}{OC})_k$ is calculated from each factor profile.

Page 9 Line 36: Hydrocarbon like OA (HOA)

Page 9 Line 46: Biomass burning OA (BBOA)

Page 10 Line 12: Sulphur containing OA (SCOA)

Page 10 Line 23: Primary biological OA (PBOA)

Page 10 Line 40; note here as well the changed nomenclature: anthropogenic OOA (AOOA).

Page 10 Line 47: Summer oxygenated OA (SOOA)

Page 11 Line 12: Named after its seasonal behavior (Daellenbach et al. 2017), the third oxygenated factor, winter oxygenated OA (WOOA)

Page 12 Line 22: The fossil fractions of SOOC ($SOOC_f$) and WOOC ($WOOC_f$)

Page 12 Line 27: From the non-fossil sources, apart from non-fossil SCOC ($SCOC_{nf}$) and non-fossil OOC (OOC_{nf})
Page 12 Lines 30, 31: SOOC was 79% non-fossil which supported the AMS/PMF results: the significance of non-fossil SOOC ($SOOC_{nf}$)
Page 12 Line 40: Non-fossil WOOC ($WOOC_{nf}$)

Figure 3 was also corrected and names of factors in all corresponding figure captions were added.

Main manuscript, Figure 4 caption: Probability density functions of factor recoveries: hydrocarbon like OA (HOA) in grey, biomass burning OA (BBOA) in dark brown, sulphur containing OA (SCOA) in blue, primary biological OA (PBOA) in green, anthropogenic oxygenated OA (AOOA) in purple, summer oxygenated OA (SOOA) in yellow and winter oxygenated OA (WOOA) in light brown.
Main manuscript, Figure 6 caption: Factor (in red for PM_{10} and blue for $PM_{2.5}$) and external marker (in grey markers) time-series for the two size fractions: HOC and NOx, BBOC and levoglucosan, SCOC, PBOC and cellulose, AOOC and OC_f, SOOC and temperature and WOOC and NH_4^+.
Main manuscript, Figure 8 caption: Probability density functions of the fitting coefficients of the relative fossil contributions: SCOC in blue, AOOC in purple, SOOC in yellow and WOOC in light brown.
Main manuscript, Figure 9 caption: Relative contributions to the fossil OC per factor (PM_{10}) (a) and to the non-fossil OC per factor (PM_{10}) (b): BBOC in dark brown, $SCOC_f$ and $SCOC_{nf}$ in blue, PBOC in green, $AOOC_f$ and $AOOC_{nf}$ in purple, $SOOC_f$ and $SOOC_{nf}$ in yellow and $WOOC_f$ and WOO_{nf} in light brown. Note that the total non-fossil concentrations (dark green markers) are on average 6 times higher compared to the fossil ones (dark grey markers).
Main manuscript, Figure 10 caption: Yearly cycles of fossil PM_{10} (a), non-fossil PM_{10} (b), fossil $PM_{2.5}$ (c), and non-fossil $PM_{2.5}$ (d) OC factors: BBOC in dark brown, $SCOC_f$ and $SCOC_{nf}$ in blue, PBOC in green, $AOOC_f$ and $AOOC_{nf}$ in purple, $SOOC_f$ and $SOOC_{nf}$ in yellow and $WOOC_f$ and WOO_{nf} in light brown. Note that the covered time periods in (a/b) and (c/d) are different.

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