

We thank the referees for their comments, which helped improving the quality of our manuscript. A point by point response (in blue) to the reviewers' comments (in black, italics) will follow. Changes in the text are indicated in in black.

Anonymous Referee #4

General comments:

In this paper the concentrations of the six types of organic aerosol (OA) components (HOA, COA, BBOA, WOOA, SOOA, and SC-OA) over Switzerland are reported based on the off-line analysis of the water-soluble aerosol components in aerosol samples using an aerosol mass spectrometer (AMS). The characteristics of the retrieved OA components, e.g., the relative abundances and seasonality, are presented. Further, the uncertainty of the concentrations of the retrieved OA is discussed. The source identification of OA components based on long-term samplings at multiple locations is important, and the application of the aerosol mass spectrometry for the chemical analysis of aerosol samples collected on filters made it possible in this study. The contributions of the major sources of OA to the atmospheric concentrations in the studied area have been characterized well in view of location and seasonality. Although the results presented in this paper are highly valuable, this paper needs substantial improvement in terms of the presentation quality. The explanations for the statistical analyses are not fully comprehensive, and a part of them would be flawed. Further, the point of this study is not very clear because both the methodology of the analysis itself and the results based on it are presented and discussed. To make the point clearer, it may be better to move the discussion on the uncertainty based on the results in Figures 6 and 7 to the experimental section or the supplement. Other minor issues regarding the presentation quality include inadequate explanations, undefined abbreviations/symbols, and grammatical errors. For the reasons above, substantial improvement is required for the publication of this paper in its final form. More specific comments are listed below.

Specific comments:

Page 3, 1st paragraph: It may be better to explain more about previous source apportionment studies for organic aerosols using off-line AMS measurement techniques. The group of the first and corresponding authors reported two more studies, both of which were also for European sites (Bozzetti et al., 2017a, 2017b). There are also other source apportionment studies based on statistical analysis for the mass spectra obtained using off-line AMS techniques (Sun et al., 2011; Chen et al., 2016). Emphasis should be on which characteristics of atmospheric aerosols have not been studied tentatively even by the use the off-line AMS techniques.

In the revised version of the manuscript, we have mentioned previous work that used a similar methodology to here. While Chen et al. (2016) have used factor analysis for the mass spectra obtained using off-line AMS techniques; their focus was on the identification of chromophores. Indeed, the studies by Bozzetti et al. (2017a, b) from our group use the same methodology for a similar purpose that is the determination of spatially resolved trends of OA sources. However, the sites studied and the challenges faced in the aforementioned studies are different from here. Here, as the sites are not homogeneous the main aim of the paper is providing a methodology to satisfactorily represent the OA by few factors, with a systematic and objective assessment of the results and the underlying uncertainties.

This paragraph reads now:

“... This approach allows the retroactive investigation of specific events, e.g. haze events in China (Huang et al., 2014) as well as AMS measurements of coarse mode aerosol (Bozzetti et al., 2016) and long-term source apportionment studies (Bozzetti, 2017b, 2017a). Such an approach was also used in recent studies for identifying the different types of water-soluble chromophores (Chen et al., 2016). Additionally, such filters are routinely collected and are already available over multi-year periods at many air quality monitoring stations around the world for years/decades. ...”

Page 3, 2nd paragraph: The chemical analysis using the AMS was limited to the watersoluble component of organics in PM10, although the water-insoluble organic component was also taken into consideration in the source apportionment. This point should be addressed more explicitly.

Indeed, the chemical analysis using the AMS was limited to the water soluble fraction. However, using factor-specific recoveries determined in Daellenbach et al. (2016), the contribution of the different factors to WSOC could be scaled to OC. This is described in detail in Section 2.5.

Page 3, line 13: The site-to-site differences and time series are not explained in a specific part of this paper.

This sentence was misleading. Detailed analysis of the site-to-site differences and time series will be presented in a second paper. The sentence has been adapted to:

“...In a second paper, we will investigate the site-to-site differences and general trends in the factor time series and their relationship with external parameters. ...”

Page 4, lines 1-3: How were the mass spectra of the extracts from aerosol samples corrected for field blanks? Because the sensitivity of an AMS to aerosol components depends on the particle size, the signal intensity of organics should not be proportional to the organic mass flux from the nebulizer. For this reason, the assessment of the blank level is not straightforward. More explanation to this point is necessary.

Between two samples we measured ultrapure water. The recorded signal was subtracted from the sample spectra. In a previous study, we showed that the organic blank measurements collected by ultrapure water nebulization provide a comparable blank estimate to the organic blanks determined from the nebulization of NH_4NO_3 (Bozzetti et al., 2017a). However, we also analyzed field blanks which were extracted and measured in the same way as the exposed samples.

..

We added accordingly a statement in the manuscript on P3 L30 –P4 L7:

“... The measurement blank was determined before and after every filter sample. Each sample was recorded for 480 seconds (AMS V-mode, m/z 12-447), with a collection time for each spectrum of 30 seconds. Ultrapure water was measured for 720 seconds. Once per day, ultrapure milliQ water was nebulized with a particle filter interposed between the nebulizer and the AMS, for the determination of the gas-phase contribution to the measured mass spectrum, which was then subtracted during analysis from both blanks and filter samples. The filters from Zurich were analysed twice with a time difference of approximately 5 months to assess the measurement repeatability. High resolution mass spectral analysis was performed for each m/z (mass to charge) in the range of 12- 115. The measurement blank was subtracted from the sample spectra. In a previous study, it has been shown that the measurement blank is comparable to the organic blanks obtained from the nebulization of NH_4NO_3 (Bozzetti et al. (2017a). The interference of NH_4NO_3 on the CO_2^+ signal described by Pieber et al. (2016) was corrected as follows (Eq. 1): ...”

We have previously shown that the organic signal from the nebulization of MQ water is not statistically significantly different from the organic signals from the nebulization of NH_4NO_3 (Bozzetti et al. (2017a)), which might potentially act as a carrier seed of contaminants. Therefore, we considered the MQ water to be an adequate representation of the measurement background. In addition to the measurement blanks, we have measured field blanks following the same procedure. These samples showed WSOC and OC concentrations higher than instruments detection limits. As this contamination can contribute to different extents to different factors, data have been corrected post PMF as described in Section 2.5.

In order to account for the effect of the field blanks on the source apportionment, we subtracted the blank concentrations factor after the PMF analysis. To that purpose we performed PMF runs using $\text{PMF}_{\text{block}}$ while also the field blank measurements were included in the PMF run. Thereby, we found how much the different factors contributed to the field blanks. Finally, we subtracted this effect from the factor time series. In addition, the OC blank levels used in the previous version of the manuscript were overestimated and have now been updated. Therefore, all numbers in the manuscript related to the source apportionment analysis slightly changed (yearly average factor concentrations changed by around 15%). However, the main conclusions remain the same.

The respective text was adapted for a better readability:

“... For a limited number of PMF runs ($\text{PMF}_{\text{block}}$) also the field blank analyses were included in the PMF input data. This provides the contribution of different factors to the field blanks which were used to correct the output factor time series. Uncertainties induced by the blank subtraction were propagated. ...”

Page 4, lines 9-10: The expression in the parenthesis is unclear and needs to be reworded.

The text has been adapted in the revised version of the manuscript:

“... The correction factor $\left(\frac{\text{CO}_2, \text{meas}}{\text{NO}_3, \text{meas}}\right)_{\text{NH}_4\text{NO}_3, \text{pure}}$ was determined based on measurements of aqueous NH_4NO_3 conducted regularly during the entire measurement period and varied between ~1% and ~5% (Pieber et al., 2016). ...”

Page 5, lines 9-11: The method for rescaling here and that explained in the 2nd paragraph of page 9 does not seem identical.

While OC concentrations are available for all samples, WSOC concentrations are only available for a subset of all samples (Magadino and Zurich). Therefore, for the samples from Magadino and Zurich it was possible to evaluate mass closure, i.e. whether the sum of WSOC factor concentrations after R_k correction matched the measured OC ($\text{OC}_{i, \text{res}} = \text{OC}_{i, \text{meas}} - \sum_k \text{WSOC}_{i, k} / R_k$). For all other samples this was not possible because of the unavailability of WSOC concentrations and, therefore, these samples needed to be scaled to OC.

The text has been adapted in the revised manuscript:

“... The last criterion relates to OC mass closure. A Monte Carlo approach was applied to evaluate whether a combination of water soluble factor time series and recovery parameters would achieve OC mass closure, as described in the following. For the samples from Zurich and Magadino, for which WSOC concentrations were available (in contrast to the other samples), offline AMS measurements were scaled to the water soluble organic matter (WSOM), calculated using the WSOC measurements and OM/OC ratios from the AMS HR analysis. The water-soluble contributions from an identified aerosol source in a sample i were rescaled to their total organic matter concentrations ($OA_{i,k}$), where k represents a given factor, using combinations of factor recoveries as determined by Daellenbach et al. (2016, medians of the used combinations being: R_{HOA} : 0.11, R_{COA} : 0.54, R_{BBOA} : 0.65, and R_{OOA} : 0.89 used for WOOA and SOOA). ...

Page 5, equation 3: The constraint represented by equation 3 seems erroneous because the left and the right parts of the equation are identical.

There was indeed a typo in the equation. The equation was adapted to the presentation in Canonaco et al. (2013), since the interface presented therein was used and the same type of constraints was applied.

Page 5, lines 21-22: Were the inferred fitted ions also for constraint? Does this sentence mean all the factors other than HOA and COA were inferred from published UMR profiles?

Ions that were present in our dataset but not in the reference profiles for HOA and COA were inferred and constrained. However, such ions were given an α -value of unity. For the other factors besides HOA and COA, factor elements were not constrained but fitted by ME-2. The section was adapted to:

“... $f_{k,j}'$ is the starting value used as a priori knowledge from previous studies and $f_{k,j}$ is the resulting value in the solution. In all PMF runs (unless mentioned otherwise), we used the high resolution mass spectra for HOA and COA (cooking OA) from Crippa et al. (2013b) as constraints, i.e. two rows of $f_{k,j}$ were set equal to the mass spectra of HOA and COA. Ions that were present in our datasets but not in the reference profiles for HOA and COA were inferred from published unit mass resolution (UMR) profiles (Ng et al., 2011 and Crippa et al., 2013c). For this purpose, the fraction of signal at a specific m/z in the UMR reference spectrum ($f_{UMR,m/z}$) was compared to the fraction of signal of all ions at this m/z in the HR reference spectrum ($f_{HR,m/z}$). The difference $f_{UMR,m/z} - f_{HR,m/z}$ was used as entries in $f_{k,j}'$ for such missing ions. For these ions, an α -value of unity was set. For the other factors, the factor elements were fitted by ME-2. Alternatively, such missing ions can be also treated as ordinary factor elements, to be fitted by ME-2 with all other ordinary factor elements....”

Page 5, lines 22-24: The explanation in this sentence is not clear. This sentence should be reworded.

The respective sentence has been reworded to:

“ ... For this purpose, the fraction of signal at a specific m/z in the UMR reference spectrum ($f_{UMR,m/z}$) was compared to the fraction of signal of all ions at this m/z in the HR reference spectrum ($f_{HR,m/z}$). The difference $f_{UMR,m/z} - f_{HR,m/z}$ was used as entries in $f_{k,j}'$ for such missing ions ...”

Page 8, line 1: The values of the recoveries used in this study should be presented.

Recoveries are presented in Fig. 6 and the text was adapted to:

The water-soluble contributions from an identified aerosol source in a sample i were rescaled to its total organic matter concentration ($OA_{i,k}$), where k represents a given factor, using combinations of factor recoveries as determined by Daellenbach et al. (2016, medians of the combinations being: R_{HOA} : 0.11, R_{COA} : 0.54, R_{BBOA} : 0.65, and R_{OOA} : 0.89 used for WOOA and SOOA).

Page 8, line 2: The meaning of “the contributions of different factors to the field blank samples” is not clear. What was done is not clear, either.

The text was adapted for a better readability:

“... For a limited number of PMF runs (PMF_{block}) also the field blank analyses were included in the PMF input data. This provides the contributions of different factors to the field blanks which were used to correct the output factor time series. Uncertainties induced by the blank subtraction were propagated. ...”

Page 8, line 27: Is “ $\alpha=0.5$ ” the significance level? Fifty percent is too high.

For PMF_{block} , we performed an additional sensitivity test with $\alpha=0.05$ instead of $\alpha=0.5$. The results are described in a new section in the supplementary material and the comparison mentioned in the main text.

The sentence in the main text reads:

“... A t-test is then used to verify the significance ($\alpha=0.5$) of the average correlation coefficient between factor and marker time series, r_{avg} (Eq. 7):

$$t_{avg} = \frac{r_{avg}}{\sqrt{\frac{1-r_{avg}^2}{N-2}}}$$

(7)

Here, r_{avg} is the correlation coefficient averaged over the different stations, derived from the average z value, t_{avg} is the corresponding t-value and N is the average number of samples at the different stations. Results with significance level $\alpha=0.05$ are summarized in Fig. S8.

...”

The section in the supplementary information reads:

“ ...

For PMF_{block} , a sensitivity test with significance level of 0.05 instead of 0.5 as in the base case was performed. The factor concentrations and their corresponding uncertainties (σ_a) are compared and displayed as number density functions (Fig. S8). Changes in the estimated factor concentrations are within 10% of the factor concentrations for SCOA and smaller for all other factors. The uncertainty related to COA is decreased when lowering the significance level to 0.05, while the other factors remain largely unaffected.

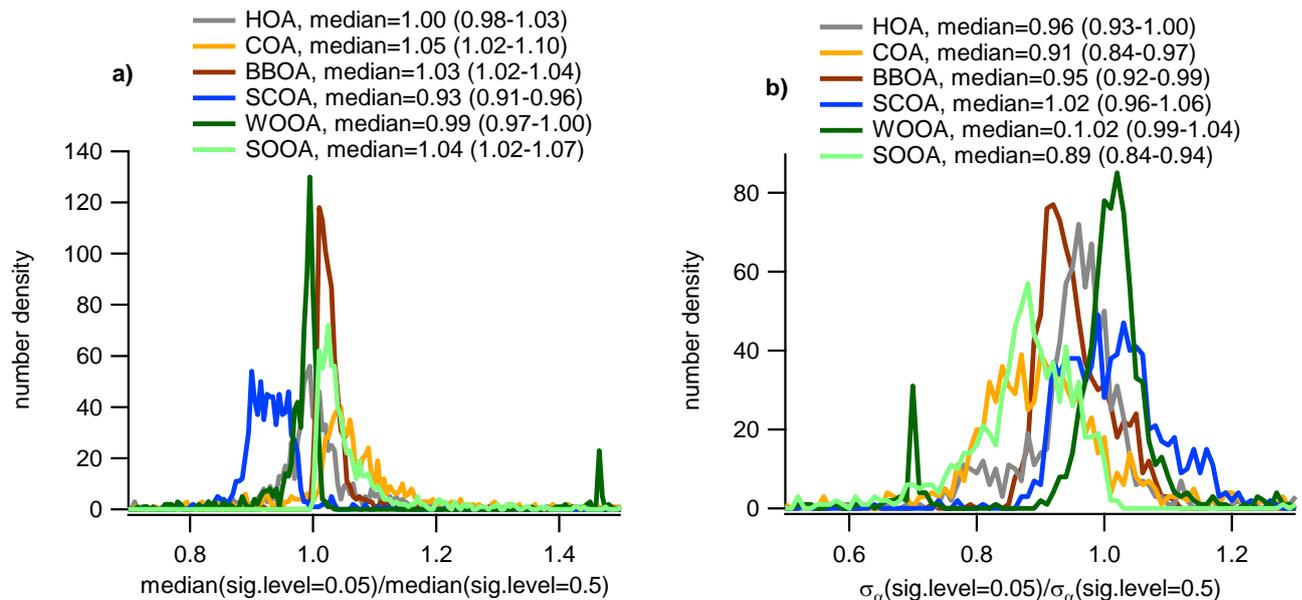


Figure S8: number density functions of source apportionment results obtained using a significance level of 0.05 normalized to results obtained using a significance level of 0.5: a) Comparison of factor concentrations b) Comparison of uncertainty estimate (σ_a).

“ ...”

Page 8, line 26-28: How the statistical analysis using the average values from different stations can be justified? The validity of this method is not obvious.

The ratios of factor concentrations to marker concentrations cannot be assumed to be the same at all sites. Therefore, correlation coefficients need to be calculated at different sites. However, to achieve an optimized system for the entire dataset the average R should be considered.

Page 9, lines 2-4: More details in the calculation should be given so that the readers can assess its validity.

We have adapted the text and added further information:

“ ...

The first two criteria (1-2) ensure an appropriate separation of HOA and COA from OOA and BBOA, respectively. Criteria 3-6 relate to the evaluation of the correlation between factor and marker time

series. This was achieved by computing the Fisher-transformed correlation coefficient z at different stations (Eq. 6):

$$z = 0.5 * \ln\left(\frac{1+r}{1-r}\right) = \arctan(r) \quad (6)$$

where r is the correlation coefficient between factor and marker at a given station. The obtained z values at the different stations are subsequently averaged and transformed back to r_{avg} before further analysis. A t-test is then used to verify the significance ($\alpha=0.5$) of the average correlation coefficient between factor and marker time series, r_{avg} (Eq. 7):

$$t_{avg} = \frac{r_{avg}}{\sqrt{\frac{1-r_{avg}^2}{N-2}}} \quad (7)$$

Here, r_{avg} is the correlation coefficient averaged over the different stations, derived from the average z value, t_{avg} is the corresponding t-value and N is the average number of samples at the different stations. Results with significance level $\alpha=0.05$ are summarized in Fig. S8.

To evaluate whether HOA correlated significantly better with NOx than COA did, the average z values obtained between HOA and NOx and between COA and NOx (Eq. 6) were compared, using a standard error on the z distribution of $1/\sqrt{N-3}$ (Zar, 1999).

...”

Page 9, lines 14-16: Is the issue really explained in the supplement?

This paragraph has been reworded and further information has been added. Now the paragraph reads:

“...The sum of $OC_{i,k}$ from all factors k (mod- OC_i) was then evaluated against the measured OC (meas- OC_i). For this, the residual OC mass (res- OC_i) for each sample was calculated (meas- $OC_i - \text{mod-}OC_i$), and the residual distributions were examined for different conditions that are specified in the Supplement. In summary, a solution was only accepted if res- OC_i were normally distributed around 0 considering all points and subsets of points: a) summer, b) winter c) Magadino, d) Zurich, e) low and high concentrations of the single factors (see Table S1). ...”

Page 10, line 2: What are the percentages of the accepted data?

We added this information to the manuscript at P10 L5:

“...Thereby Thereby, 331 PMF runs were selected for PMF_{block} (230 for PMF_{zue,isol}, 99 for PMF_{zue,repr}, and 269 for PMF_{1filter/month}). ...”

Page 11, line33 – page 12, line 1: This sentence is not clear. Does COA relate to the discussion here?

Besides the correlation between the yearly average concentrations of SC-OA and NO_x, we also present the same values for HOA vs. NO_x and COA vs. NO_x. This comparison allows the conclusion that not all anthropogenically influenced factors show a relation to NO_x.

“... SC-OA instead exhibits low background levels episodically intercepted by remarkable ten-fold enhancements, especially at urban sites affected by traffic emissions (e.g. the SC-OA contribution is significantly higher at sites with higher yearly NO_x average levels). The hypothesis of an influence of traffic activity on SC-OA is provided by the correlation of the yearly average concentrations with NO_x ($R_{s,SC-OA,NO_x} = 0.65$, $n=9$, $p<0.06$) which is, however, comparable to the correlation of HOA and COA (e.g., $R_{s,HOA,NO_x} = 0.68$, $n=9$, $p<0.05$, $R_{s,COA,NO_x} = 0.68$, $n=9$, $p<0.05$).. ...”

Page 13, line 1: The “uncertainties” here should be relative uncertainties. This should be addressed explicitly.

In Figure 7, we present the uncertainties relative to the factor concentrations. We corrected the mistake in the manuscript:

“... We note that relative uncertainties related with SOOA increase with decreasing concentrations (Fig. 7). A small error in modelling sources with high contributions (BBOA, WOOA) in winter can result in a large error of SOOA with its small contribution during winter. Furthermore, some other sources like primary biological OA (PBOA, see Sec. 4.2.2) might also mix into SOOA. ...”

Page 13, lines 2-3: The meaning of “contribution from other more significant wintertime sources” is not clear. Further, justification of the explanation in this sentence should be provided.

This sentence has been adapted and reads now:

“... A small error in modelling sources with high contributions (BBOA, WOOA) in winter can result in a large error of SOOA with its small contribution during winter. ...”

Page 13, lines 3-4: It is not clear why the mixing of some winter-time SOA into SOOA results in a larger uncertainty.

SOOA concentrations are relatively small in winter compared to BBOA or WOOA concentrations. Therefore, a small error in modelling BBOA or WOOA can result in a rather big error in SOOA. The corresponding section in the manuscript was adapted:

“... We note that relative uncertainties related with SOOA increase with decreasing concentrations (Fig. 7). A small error in modelling sources with high contributions (BBOA, WOOA) in winter can result in a large error of SOOA with its small contribution during winter. Furthermore, some other sources like primary biological OA (PBOA, see Sec. 4.2.2) might also mix into SOOA. ...”

Page 14, lines 1-2: How was σ_b calculated?

12 samples are present in all PMF datasets (PMF_{block}, PMF_{zue,iso}, PMF_{1filt/month}, PMF_{zue,reprs}). For these 12 samples we determine the median concentration for the independently treated PMF datasets. σ_b is the variability of the median concentrations of these 12 samples. This information can be found in the supplementary information (section uncertainty estimation and propagation).

We adapted the main text to the following:

“... The variability of the factor time series for the single PMF sensitivity tests (PMF_{block}, PMF_{zue,isol}, PMF_{1filter/month}, PMF_{zue,eps}) is used as an uncertainty estimate (shaded area in Fig. 4). This estimate (σ_a) depends on the measurement repeatability (10 single mass spectra included for each sample) and on the selected PMF solution/ R_k combinations and, therefore, also on the α -value. However, the variability depending (1) on the choice of input points (time and site; PMF_{block}, PMF_{zue,isol}, PMF_{1filter/month}) and (2) on the instrumental reproducibility (PMF_{zue,reprs}) of the offline AMS measurements is not accounted for. The contribution of (1) and (2) to the uncertainty is assessed through the sensitivity tests by examining the variability of the median factor time-series (σ_b). σ_b is the variability of the median factor concentrations from the PMF sensitivity tests using PMF_{block}, PMF_{zue,isol}, PMF_{1filter/month}, PMF_{zue,reprs} for the 12 samples common to all 4 PMF datasets. For the 12 filters common in all PMF datasets (PMF_{block}, PMF_{zue,isol}, PMF_{1filter/month}, PMF_{zue,reprs}), we calculate a best estimate of the overall uncertainty (err_{tot}), by propagating both error terms: σ_a and σ_b”

Page 14, line 8: The meaning of “ σ_b – rotational ambiguity” is not clear.

The part “- rotational ambiguity” has been removed and more information has been added to the text:

“... It is worthwhile to note that for major factors exhibiting a similar seasonality, i.e. WOOA and BBOA, a great part of the uncertainty arises from σ_b . Thus the variability between the PMF solutions using PMF_{block}, PMF_{zue,isol}, PMF_{1filter/month}, PMF_{zue,reprs} (σ_b) and, therefore, the sensitivity of the factor concentrations on the chosen PMF dataset significantly contribute to the uncertainty. ...”

Page 14, lines 12-15: This sentence is not very organized and needs to be reworded.

We reworded the sentence, now it reads:

“... In the present analysis, PBOA could not be separated by PMF (neither unconstrained nor using the mass spectral signature from Bozzetti et al., 2016). This inability might be caused by the low water-solubility and the absence of PM2.5 filters in the dataset. ...”

Page 14, lines 24-26: What is the definition of the site-to-site variability? Was standard deviation calculated for the average values at respective sites?

The site-to-site variability is the standard deviation of average concentrations at the different sites. This information has been added to the text and the sentence has been reworded for easier readability:

“...Using this approach, we estimate that PBOA contributes $0.30 \mu\text{g}/\text{m}^3$ during the warm months (site-to-site variability computed as standard deviation of the average concentration of all sites of $0.03 \mu\text{g}/\text{m}^3$). During the same period, SOOA concentrations are $1.78 \mu\text{g}/\text{m}^3$ (site-to-site variability of $0.18 \mu\text{g}/\text{m}^3$) and OA concentrations $4.32 \mu\text{g}/\text{m}^3$ (site-to-site variability of $0.44 \mu\text{g}/\text{m}^3$). ...”

Page 15, line 2: The use of the word “however” does not seem appropriate.

The word “however” has been removed and the sentence reworded. Now it reads:

“...The ion $C_2H_5O_2^+$ (indicator for PBOA) shows higher concentrations with increasing OC_{coarse} concentrations. ...”

Page 16, line 5: Is POA here the sum of HOA, COA and BBOA? Shouldn't it be defined here instead of line 9?

We moved the definition of POA to line 9. Now the paragraph reads:

“...In general, the seasonality of the factor time series is consistent for all the 9 sites in the entire study area (Fig. 9). In summer, SOOA is the main contributor to OA, while in winter POA (HOA+COA+BBOA) becomes more important although WOOA still contributes significantly. In comparison to the sites in northern Switzerland, OA in the southern alpine valleys is dominated by BBOA in winter, while in the north WOOA also plays a role. The different factors contribute 0.47 ± 0.12 (HOA, average and site-to-site variability), 0.31 ± 0.13 (COA), 1.37 ± 1.77 (BBOA), 0.67 ± 0.31 (SC-OA), 1.11 ± 0.23 (WOOA), 1.31 ± 0.13 (SOOA) $\mu g/m^3$ for all sites during the entire year (Table 3). In northern Switzerland, POA contributes less to OA (POA/OA=0.3) than in the southern alpine valleys where POA/OA is equal to 0.6. ...”

Figure 2: The aHOA and aCOA are not defined explicitly.

The figure caption was adapted to:

“...**Figure 1: Step-by-step outline of adopted source apportionment approach (factor recoveries R_k). a_{HOA} and a_{COA} represent the a-value applied for HOA and COA, respectively. ...”**

Figure 3: The definition of fm/z should be given.

The definition of fm/z has been added to the figure caption:

“...**Figure 2: PMF factor profiles of HOA, COA, BBOA, SOOA, WOOA, SC-OA, color-coded with ion family of PMF_{block} (average). fm/z is the relative intensity at a specific mass-to-charge ratio (m/z). ...”**

Figure 9: The definition of OAexpl is not given explicitly.

The figure caption has been adapted to:

“...**Figure 3: Map of Switzerland with yearly cycles. Negative concentrations were set to 0 prior to normalization for display. The OA mass explained by the source apportionment analysis is termed OA_{expl}”**

Page 2 (supplement): The relationship among “ $Q_i/Q_{i;exp}$ ”, “ $\Delta (Q_i/Q_{i;exp})$ ”, “ $\Delta Q_i/Q_{i;exp}$ ”, “ $(Q_i/Q_{i;exp}$ contribution)”, and “ $\Delta (Q_i/Q_{i;exp}$ contribution)” is not clear.

The nomenclature has been unified. The results refer to the distributions of $Q_{i,j}$ –(median and quartiles or average) and not to Q/Q_{exp} . $Q_{i,j}$ (referred to as Q-contribution) is computed as:

$$Q_{i,j} = \left(\frac{e_{i,j}}{s_{i,j}} \right)^2$$

We corrected the text and axis labels accordingly and refer only to Q-contribution.

“... $\Delta(Q\text{-contribution})$ shows the difference in the median Q-contributions between groups of points like sites or season. The smaller the $\Delta(Q\text{-contribution})$, the smaller are the differences in the mathematical quality of the PMF solution for the different seasons/sites. ...”

Page 3 (supplement): The definitions of “r(. . .)”, “Q25(OCres)” and “Q75(OCres)” are not given.

The table caption has been modified as:

“...**Table S1: set of acceptance criteria used. r is the correlation coefficient between a factor time series and the respective marker. Q25 is the 1st quartile and Q75 the 3rd quartile. ...**”

Page 4 (supplement): The definition of “fion” is not given.

The figure caption has been updated:

“...**Figure S6: mass spectral fingerprints of BBOA (PMF_{block}) and nebulized levoglucosan. f_{ion} is the fraction of signal of a respective ion to the sum of the total signal.**”

Technical corrections:

Page 1, line 21: Should “at” be added between “10_m” and “9 stations”?

This mistake has been corrected.

Page 3, line 20: “HiVol” should be spelled out.

Spelled out as High-Volume samplers.

Page 5, lines 29-30: The subscripts of “PMF” are not written consistently in the paper.

The PMF datasets are now consistently called:

PMF_{block}

PMF_{zue,isol}

PMF_{1filter/month}

PMF_{zue,reprs}

Page 10, line 18: Should “from” be added after “profile”?

The missing word has been added as well as the number corrected:

“...COA profile elements were constrained using the COA profile from Crippa et al. (2013b) and the obtained factor profile maintains the same features (OM/OC of 1.32, IQR 1.30-1.33, Fig. 3). ...”

Page 13, line 29: Should “Fig. 5” be “Fig. 4”?

Yes it should be Fig. 4. The mistake has been corrected.

Page 14, lines 30 and 33: Should “is in summer” be “in summer is”?

The mistake has been corrected.

Page 14, line 33: “OCcoarse” should be defined in line 30..

OCcoarse is now defined on line 30.

“... Bozzetti et al. (2016) showed that coarse OC ($OC_{\text{coarse}} = OC_{\text{PM}_{10}} - OC_{\text{PM}_{2.5}}$) in summer is dominated by PBOA for samples collected at a rural site in Switzerland (Payerne). ...”

Page 19, lines 9-10: The list of authors are incorrect.

The citation has been corrected:

“Daellenbach, K. R., Bozzetti, C., Křepelová, A., Canonaco, F., Wolf, R., Zotter, P., Fermo, P., Crippa, M., Slowik, J. G., Sosedova, Y., Zhang, Y., Huang, R.-J., Poulain, L., Szidat, S., Baltensperger, U., El Haddad, I., and Prévôt, A. S. H.: Characterization and source apportionment of organic aerosol using offline aerosol mass spectrometry, *Atmos. Meas. Tech.*, 9, 23-39, doi:10.5194/amt-9-23-2016, 2016.”

Table 1: The commas after “St. Gallen” and “San Vittore” in the column “Site (station code)”, and the periods after “m” in the column “altitude” should be omitted. The initial letter of “altitude” should be capitalized.

In the column further information on the station location have been added. Altitude has been capitalized and the periods are omitted.

Site (station code)	Classification	General location	Altitude
Basel, St. Johann (bas)	Urban/background	North of Alps/Swiss plateau	308 m
Bern, Bollwerk (ber)	Urban/traffic	North of Alps/Swiss plateau	506 m
Frauenfeld, Bahnhofstr. (fra)	Suburban/backgroun d	North of Alps/Swiss plateau	403 m
Payerne (pay)	Rural/background	North of Alps/Swiss plateau	539 m
St. Gallen, Rorschacherstr. (gal)	Urban/traffic	North of Alps/Swiss plateau	457 m
Zurich, Kaserne (zue)	Urban/background	North of Alps/Swiss plateau	457 m
Vaduz, Austrasse (vad)	Urban/traffic	North of Alps/alpine valley	706 m
Magadino, Cadenazzo (mag)	Rural/background	South of Alps	254 m
San Vittore, Zentrum (vi)	Rural/traffic	South of Alps/alpine valley	330 m

Figure 4 caption: It is better to write “HOA, COA, . . .” in the order of the corresponding panels.

The figure caption has been adapted and reads now:

“...Figure 4: HOA, COA, BBOA, WOOA, SOOA, and SC-OA and respective marker concentrations as a function of time for Zurich in 2013. Depicted are the median factor time series results for the different PMF datasets (median) including the uncertainties for PMF_{block} (first and third quartile) (green: PMF_{block} , black: $PMF_{zue,isol}$, red: $PMF_{zue,repr}$, pink bullets: $PMF_{1filter/month}$). ...”

Figure 5 caption: Should “NH4” be “NH4+”?

The figure caption has been corrected:

“...Figure 5: Scatter-plots for the different extreme sensitivity tests for Zurich and for all sites for PMF_{block} median concentrations): a) HOA vs NO_x , b) BBOA vs levoglucosan, c) SOOA vs temperature, d) WOOA vs NH_4^+”

Figure 7: Should “[” after “concentration” be “]”?

In the y-axis |concentration| refers to the absolute concentration.

Page 4 (supplement): The “interquartile range PMF block” should be represented by a symbol because it is in a mathematical formula. It may be better to write “median bootstrap solutions” as the subscript of σ .

Since σ_a and σ_b are explained in detail in the text we remove this part of the mathematical expression. The equation reads now:

“ ...

$$\text{err}_{i,k,\text{tot}} = \sqrt{\sigma_a^2 + \sigma_b^2}$$

...”

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