

Interactive comment on “Long-term chemical analysis and organic aerosol source apportionment at 9 sites in Central Europe: Source identification and uncertainty assessment” by Kaspar R. Daellenbach et al.

Anonymous Referee #4

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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

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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.

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

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be addressed more explicitly.

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

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.

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

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

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

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?

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

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

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.

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

Page 8, line 26-28: How the statistical analysis using the average values from different

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stations can be justified? The validity of this method is not obvious.

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

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

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

Page 11, line 33 – page 12, line 1: This sentence is not clear. Does COA relate to the discussion here?

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

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.

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

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

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

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

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?

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

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

Figure 2: The a_{HOA} and a_{COA} are not defined explicitly.

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Figure 3: The definition of $f_{m/z}$ should be given.

Figure 9: The definition of OA_{expl} is not given explicitly.

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.

Page 3 (supplement): The definitions of " $r(\dots)$ ", " $Q_{25}(OC_{res})$ " and " $Q_{75}(OC_{res})$ " are not given.

Page 4 (supplement): The definition of " f_{ion} " is not given.

Technical corrections:

Page 1, line 21: Should "at" be added between "10 μm " and "9 stations"?

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

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

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

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

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

Page 14, line 33: " OC_{coarse} " should be defined in line 30..

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

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.

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

Figure 5 caption: Should " NH_4 " be " NH_4^+ "?

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Figure 7: Should "[" after "concentration" be "]"?

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 .

References:

Bozzetti, C., Sosedova, Y., Xiao, M., Daellenbach, K. R., Ulevicius, V., Dudoitis, V., Mordas, G., Byčenkienė, S., Plauškaitė, K., Vlachou, A., Golly, B., Chazeau, B., Besombes, J.-L., Baltensperger, U., Jaffrezo, J.-L., Slowik, J. G., El Haddad, I., and Prévôt, A. S. H.: Argon offline-AMS source apportionment of organic aerosol over yearly cycles for an urban, rural, and marine site in northern Europe, *Atmos. Chem. Phys.*, 17, 117–141, doi:10.5194/acp-17-117-2017, 2017a.

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