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Atmospheric Measurement Techniques Discussions

Interactive comment on "ACTRIS ACSM intercomparison – Part 2: Intercomparison of ME-2 organic source apportionment results from 15 individual, co-located aerosol mass spectrometers" *by* R. Fröhlich et al.

Anonymous Referee #3

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The manuscript provided a comprehensive inter-comparison of 15 co-located aerosol mass spectrometers, 1 HR-ToF-AMS, 1 ToF-ACSM, and 13 ACSMs (ACTRIS ACSM network). It is found that while the mass spectra and time series of various species are comparable to each other, there is a fairly large variation in the f44 values across all instruments. PMF/ME-2 analysis is performed on all datasets and the results are discussed. Using these results, it is suggested that while the range of f44 values in the organics data lead to variable f44 values in the factor profiles, the mass contribution of each factor does not seem to be affected to a large extent.

This is undoubtedly an important piece of work and would be of great interest to the scientific community, particularly the ACSM users. I think this work is extensive and the analyses are thorough. However, there are two main major comments regarding the manuscript that should be addressed prior the publication of the manuscript in AMT.

Firstly, more information needs to be provided in the SI to justify the choice of PMF/ME-2 solutions. There is no mention of varying FPEAK values or seed values, thus it is not clear whether the authors have evaluated how these parameters might affect the solutions (and if FPEAK values other than zero are used). Diagnostics plots of Q/Qexp, residuals, effects of FEAPKs/seeds (if applicable), etc should be provided. It is important such information be included in the SI of the manuscript so that readers can evaluate whether the selected PMF solutions can be justified. In my opinion, an extensive section in the SI needs to be devoted to such kind of information. There are also multiple incidents where the anchor profile for ME-2 input is chosen by performing PMF analysis up a large number of factors to "extract" out that profile. The authors need to provide justification on how such a decision is made. For instance, in the PMF analysis of the HR-ToF-AMS data, it is stated that when the analysis is extended to eight factors, the HOA and COA then become unmixed and are used as anchor profiles. Please document and explain clearly what criteria are applied in making such choices. More specific comments can be found below.

Secondly, given such a rare and invaluable opportunity for inter-comparison of multiple co-located ACSMs, I think one of the very important contributions of the manuscript would be to provide recommendation for ACSMs users to evaluate how the variable f44 values in their data would affect source apportionment results. This is extensively discussed in the manuscript of course, but only from the point of view of comparing the performance of the individual ACSMs (among themselves, and with respect to HR-ToF-AMS). The results shown in Fig. 7 are promising in that the mass fractions appear to be fairly comparable across all instruments (except for the COA factor). However, all these results are the outcomes of multiple constraints that have been imposed during the

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analysis and availability of prior information about the aerosol composition at the site (e.g., absence/presence of COA, various source profiles, etc). It is not clear how other individual ACSM datasets should be analyzed in the absence of all these additional information, which is likely the case for most future studies. With such a large inter-comparison effort, could the authors provide some suggestions for scenarios where only one ACSM is available? For instance, by performing PMF analysis on each of the ACSM datasets alone, it seems that the authors can only resolve HOA, BBOA, and OOA factors. The authors specially look for the COA factor in the ACSM dataset (by constraining the source profile) based on the results from HR-ToF-AMS data. In the absence of HR-ToF-AMS, would the unconstrained ACSM PMF solution (i.e., HOA, OOA, BBOA) be "good enough", though, justified from Fig. 6, such unconstrained solutions result in a fairly large variation in the mass fractions? Or, would the authors suggest that one should always constrain the HOA factor in the analysis of ACSM data? The authors seem to suggest that the HOA anchor profile does not have a strong influence on source apportionment results. With this, if no prior studies at the same site have been conducted, should a "typical HOA profile" be used as the anchor profile for future ACSM PMF/ME-2analysis? These are just some suggestions/thoughts, but I would encourage the authors to think deeply how they could best use these valuable datasets to provide more concrete recommendations on analyzing individual ACSM datasets with variable f44 values, in the absence of additional mass spectrometers (i.e., where there is no HR-ToF-AMS or other ACSMs to compare the "answers" to). Given that this manuscript is submitted to AMT, I think that it is important and critical that more of such recommendations be included in the manuscript.

Specific comments

1. Page 1570, lines 23-28. Does this drift affect both the "filtered/background" and "ambient" data? How is the correction determined? How do the authors assure that the drift in the "filtered" data can be applied to the "ambient" data (since the background data need to be subtracted from the ambient data during analysis to get the difference

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spectra)? Please clarify.

2. Page 1571, line 7. How is eqt 6 used to downweight the weak signals? According to Ulbrich et al, the weak and bad variables are downweighted by fixed values (for instance, the weak variables are downweighted by a factor of 2), which does not involve eqt 6. Please clarify.

3. Page 1572, line 2. What is "corrected" ToF-ACSM time series? (what are they corrected for?)

4. Page 1573. Do the authors have comments/speculations on why the ToF-ACSM has the highest f44?

5. Page 1575. Section 3.3, HR-ToF-AMS source apportionment. a. It is important that more information to be provided in the SI regarding the details of the PMF analysis. Are FPEAK / seed values varied? How do the residuals for different factor solutions look like? Please include all the relevant diagnostics plots in the SI, including (not limited to) Q/Qexp, residuals, mass fractions, FPEAKs, etc. I think the authors likely have checked all these, but it is important to show them in the SI so readers can evaluate the solutions themselves and ensure that the authors' choice of the solution (i.e., 4-facotr solution) can be justified.

b. Page 1575, line 16. Is the unusually high f44 in HOA only, COA only, or both? Why is a high f44 regarded as a sign of mixing of HOA and COA?

c. Page 1575, line 16-17. The authors wrote "an extension of the analysis up to eight factors leads to an unmixing of the two factors". The HOA and COA factor profiles are fairly similar (the most outstanding difference is that COA has high f55), how do the authors decide that with eight factors, the HOA and COA are not mixed anymore? What are the criteria for "unmixing"? Please be very specific about this and explain this in details.

d. Hoes do the BBOA and OOA MS and time series obtained from the unconstrained

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PMF compared to that from the PMF/ME-2 analysis? Please discuss this. Specifically, how different are the f44 values in the BBOA and OOA factors (if any) with the unconstrained vs PMF/ME-2 analysis?

e. Page 1577, line 4. Has the flat OOA diurnal trend been observed in previous field studies conducted in the area? The correlation of OOA and sulfate (nitrate) does not appear to be particularly strong. Why? Are the diurnal trend of sulfate and nitrate also flat?

f. Page 1577, line 18. The authors noted that the high f44 in BBOA could be indicative of aging. Later in the paragraph the authors mentioned winter wood combustion. Do the authors think that this is the main source of BBOA? If so, I would think this could be local and might not be subjected to too much aging?

6. More details need to be provided regarding the choice of the COA and HOA anchor spectra for ME-2 analysis.

a. Page 1578, line 20. Why is a "verified anchor spectrum from a previous study at the nearby measurement site" used for COA? Why not use the COA spectrum obtained from the HR-ToF-AMS PMF analysis of this study? Shouldn't this be more relevant than the spectrum obtained from the previous study?

b. Page 1578, line 24. What do the authors mean by "extracted from a previous PMF solution with a higher number of factors. . ..". I suppose just like section 3.3, the authors run PMF analysis (completely unconstrained? Or with COA constrained? Please clarify) up to a "higher" number of factors, and then at some point decide that the HOA factor is "extractable"? How do the authors decide on when (which solution) the HOA factor is "good enough" to be extracted and used as input MS for ME-2 analysis? Please provide more details and clearly explain how this is done.

c. Page 1579, line 5. The authors wrote "the influence of an alternative anchor proved to be only marginal". The authors should clearly specify what influence they are re-

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ferring to. If it's the mass fraction of factors, I do not agree that the influence is "only marginal", given that they can be up to ∼30% for some factors.

7. Page 1586, line 1. ACSM #7 also seems to show a large deviation?

Technical comments 1. Page 1574, line 28. "Is is noted. . .." should be "It is noted. . .".

Interactive comment on Atmos. Meas. Tech. Discuss., 8, 1559, 2015.

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