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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.

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Author reply to: Anonymous Referee 2

Referee comments are written in **green** Author replys are written in **black**

Changes in the text of the article and the supplement are shown in the attached pdf documents (see supplementary to this comment). There, additions are shown in **blue**, deletions in **red**.

The manuscript describes the analysis of a data set with respect to source apportionment taken by 13 of the new Aerodyne ACSM monitors, which are supposed to monitor particle composition on ACTRICE sites. In addition an ACSM-TOF and a HR-TOF were simultaneously operated. The HR-TOF was used as guide to extent the statistical analysis, performed by PMF and ME-2. This is a very important study for methodology and quality assurance of aerosol monitoring. The manuscript focus on the results achieved with PMF and variations, ME-2, and their intercomparability, when applied to a data set from different instruments. Unfortunately the companion experimental paper was not available although the authors refer several times to it. Since the manuscript focus on methods of evaluation, this can be tolerated, but it would have been helpful to check for some boundary conditions. The manuscript is published in AMTD, so, as mentioned before, the focus is not the result itself, i.e. the source apportionment, but the coherence of PMF and ME-2 analysis on a dataset monitored the same place at the same time with the set of instruments. The manuscript is well written and interesting to read. Figures and Tables are ok and sufficient. The Figures are detailed and for that detail somewhat too small (if printed). Up to here, I have only minor points which will follow below. What is presented in the manuscript can be published in AMT after minor revisions.

However, before publishing the authors may want to consider the following major revision: In my opinion the manuscript has a conceptual weakness. Instead of doing a careful analysis with ACSM data alone, push it to edge, and then compare with the HR-AMS data, the authors chose the opposite way and used the results of analysis of the HRAMS data as a guideline to optimize performance of the analysis of the ACSM data. (The authors clearly state that procedure and this not my point.

We understand the point raised by the referee and the suggested procedure was pondered at the beginning of the project. But the strength of the dataset at hand is not to evaluate the limits of PMF on ACSM datasets, for that purpose several different datasets from different locations, ideally with an existing reference source apportion-

ment (SA) to compare would be better suited. The large ACSM database created by the European ACTRIS project provides the opportunity for another study investigating that. For a detailed analysis of the limits of ACSM SA it would notably make sense to investigate different PMF inputs (e.g. different time resolution of the same dataset, different m/z ranges of the same dataset, different time intervals).

In our opinion the real advantage of the parallel operation of the ACSMs in Paris is to investigate how comparable are SA on data from different ACSMs if the SA was done exactly the same way, i.e. find the uncertainties of a specific SA depending on the ACSM instrument which was used to record the data. To do so, it is important to find a way to make sure that the presented solutions can be compared (e.g. if we had found 3 factors in one instrument and 4 in the other we would know that one of them is wrong and the solutions are not comparable). A solution-target - which is thought to best represent the OA sources during the study period - has thus to be agreed on. With a much larger temporal and spectral resolution than ACSM, the HR-AMS provides the best chances to separate all present OA sources and thus to furnish the best agreed solution. Moreover, to be achieved properly, SA studies need significant variability (in time and contributions). Since the study period was only two weeks long, it was already evident from the beginning that ACSM datasets may be at the lower size-limit allowing reasonable SA. Fortunately the ambient conditions in Paris during these two weeks created enough source variability allowing for a satisfying SA analysis, using both HR-AMS and ACSM systems.

Although, even in the analysis of HR-AMS data already a lot of detailed work and know-how had to put in to clearly separate the four factors. And I asked myself in how far is this objective or already depending on the specific skills (or 'opinions' what to expect) by the operator.)

We agree with the notion of the referee that skill and experience of the SA operator are very important factors for the determination of the validity of SA results. To make the made choices more comprehensible, we added an extensive section of PMF diagnos-

tics to the Supplementary Information (page 20 onwards).

Another important study which can profit from the dataset of the intercomparison is trying to assess the influence of the operator's skills and experience on the SA results. But a thorough investigation of that is considered to be out of the bounds and goals of this manuscript.

I think the a posteriori analysis is not the real test for the ACSM source apportionment and the described results (even considering the careful discussion) do not really help the case.

The case was not to define the limits of ACSM SA, but to investigate uncertainties.

My question is, what can we expect if we have a single instrument at an ACTRICE site? Then Figure S6 is the truth and the question is what does this 3 factor result mean when compared to the more detailed HR-AMS results. Do I have a clear HOA and BBOA factor and the OOA factor is a mixture of OOA and COA?

The lower time and spectral resolution of the ACSM may just not provide sufficient source resolution to resolve all OA factors with unconstrained PMF. This is discussed in the text (e.g. P1580). A longer measurement period would potentially have increased the source resolution but the investigation of this is a task for an additional study as mentioned above.

Since the HR-AMS unconstrained PMF analysis showed the presence of COA-like aerosols, it was quite logical to investigate how much ACSM constrained/unconstrained PMF analysis is able to evidence this type of aerosols, and to evaluate the related uncertainties.

A short additional comment to Fig S6: In many cases when a source cannot be separated, a careful analysis of all factors may reveal points the operator may identify as source mixing and motivate him to dig deeper e.g. by investigating the solution space with additional constraints to the ME-2 algorithm. Fig. S6 shows the unconstrained

3 factor PMF results before applying constraints. In most ACSMs an investigation of e.g. the residuals at m/z 55 or diurnal cycles and profiles of the 3,4 and 5 factor PMF shows signs for COA presence. I.e. reasons for including a COA anchor can also be found with the ACSM data. Just the possibility to isolate the COA without using the constraints option of ME-2 like it was possible for the HR AMS was not given.

If I want to refine the ACSM analysis, is there a general procedure (that is independent of the experience/'opinion' of the operator)? For example, can I consider always a certain set of anchor profiles and apply ME-2?

Crippa et al. (2014) doi:10.5194/acp-14-6159-2014 investigated AMS datasets from 25 sites using ME-2. From this analysis she compiled a set of recommendations / a standard procedure. Generally this can also be applied to ACSM data. But from this intercomparison it is not possible to give similar standard procedures, since we have only one dataset, however from 15 instruments, to test them on. A future comprehensive SA study, based for instance on the ACTRIS ACSM datasets, may be able to answer that question.

Which anchor profiles for which site?

In P1587 L22 we recommend that profiles extracted from the dataset at hand should be preferred to external profiles. Else we cannot make any statement which profiles are recommended for different sites, because this study only works with data from one site. What we could show is that if the a value is optimised, the resulting contributions between the extracted HOA anchor and a COA anchor from the database are similar in most cases (Fig 7).

These are important questions, just like all the others above, which, as the referee already noted, cannot be fully answered from this study alone. But it should be noted here that even if a non-real constrained factor yielded non-zero contributions the operator still would have to provide additional evidence that this factor is real (correlations with external data, stability of the solutions, explainable features in the diurnal cycles

or total time series etc.).

What would happen to your comparison if you allow only 3 factors in PMF of HR-AMS ? I.e. skip e.g. the weak expressed COA factor?

See replies to last comment and above.

Do I need some HR-AMS analysis at each site? For how long - a few weeks, a full seasonal cycle?

No, we show that a careful ACSM analysis can also resolve the same sources as the AMS. Ideally then a period longer than two weeks is available making the separation easier and more clear. However this all depends strongly on the site and the variability of sources present. Since the AMS typically records a data point every few minutes while the ACSM only every 30 min and the spectrum is much more separated and reaches to higher m/z, the AMS may need a measurement period that is only about a tenth or less to produce similar source resolution as the ACSM. Also see replies above.

Of course these questions cannot be answered overall within this manuscript, but I urgently suggest to more discuss the consequences of Fig S6 in relation to the HR-AMS result and at least sketch generalized ways how to treat ACSM data in a self-consistent way - without input by HR-AMS results. The presented manuscript can be either shortened to make room for this addition, but can also stay as it is and the proposed aspect can be added on top of it.

As detailed in the reply to the first comment this study cannot provide recommendations for a generalised way. Such recommendations could rather be drawn from the analysis of many different data sets and in any case should be treated with caution since every PMF will still need a very careful individual analysis.

A new paragraph was added after P1578 L18 commenting on the three factor PMF solution of Fig. S6.

Minor points: p1564, l 4: "Overviews of wintertime. . .". This seems to be misplaced, it

belongs to Site description.

Revised.

p1570, l19: move definition of RIT from p1584, l22to here.

Revised.

p1572, l14: This sentence sounds strange.

Revised.

p1574, l27: move reference behind “setting”.

Revised.

p1575, l26: the word “classified as POA” seems to be strange, I would suggest “attributed to POA sources”. Similar for “classified as SOA”.

Revised using the suggested wording.

p1578, l13: In the beginning the simple PMF is noted as unconstrained PMF, which is ok. From here on the notation “pure” PMF is used. I suggest a consistent use of “unconstrained” PMF, for the simple case.

Revised.

p1578, l22: . . .subscript “Paris”. . ., not subscript.

Revised.

p1579, l12 l24: I suggest to move those two paragraphs to the Method section, it feels misplaced here in such detail.

Revised.

p1583, l10: high f44 in HOA of #13, is strange. But also #1 and #5 have significant contributions of f44 in the HOA profile, but do not have high f44 in general? By the way,

the organization of the Figures S13 and S14 is unfortunate, as the same instruments are not appearing at the spot part in the figure. Move the explanations from the center of the figure to the beginning of each factor.

We thank for that very useful suggestion and have rearranged the plots accordingly.

p1585, l15: Crippa et al.(2014) derived the HOA(Paris) profile from HR-AMS ? Why can it not applied in the HR analysis ?

This argument is correct. We did not show it, however, because the lists of fitted ions used within the Crippa et al. spectrum and within our HR analysis did not overlap completely.

p1586, l14: How can we be sure that the COA factor is real? If you would skip the COA factor in HR-AMS analysis would that unify the ACSM and HR-results? Compare to major concerns.

As mentioned above, also from the ACSM data there is indication that a COA-like factor should be introduced, it just cannot be resolved by unconstrained PMF. A skip of this fourth factor would lead to larger uncertainties and less seed stability. For more justification on the source attribution for this factor, please also check the newly added Sect. 3 of the Supplementary Information.

Please also note the supplement to this comment:
http://www.atmos-meas-tech-discuss.net/8/C1051/2015/amtd-8-C1051-2015-supplement.zip

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