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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 1

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 supplement. There, additions are shown in **blue**, deletions in **red**.

This paper outlines a comparison of ME-2 analysis as applied to multiple ACSM instru-

ments as part of the ACTRIS intercomparison exercise at SIRTA. This has the potential to be a hugely significant technical paper; while the exact techniques of ME-2 analysis of ACSM data are still the subject of ongoing development, this paper conducts the first thorough intercomparison of several instruments and quantifies the instrument-to instrument variabilities. This has implications on not just how ACSM (and, by extension, AMS) data are compared, but it also quantifies exactly how much confidence can be attached to the factorisation associated with instrument-to-instrument variabilities and general random variations in the datasets. As such, this is very well suited to AMT and I recommend it be published subject to the comments outlined below. Probably the single biggest shortfall of this paper is that I find the application of ME- 2 to be inadequately documented as regards the choices made when analysing the individual instruments' datasets. That is not to say that it is 'wrong' (or even 'right' for that matter), but for this paper to fulfil its stated purpose, it must be demonstrated that the analysis was robustly performed in as consistent and as objective a manner as possible. At the moment, some of the criteria are not very well explained.

Major comments: I do not consider sufficient detail to be given concerning the HR-AMS analysis in section 3.3 and this is a problem because the accuracy of all subsequent conclusions depends on this. While I understand conceptually what the authors are doing, they need to make sure that everything is properly documented so that they can demonstrate that what they are doing does not introduce subjectivity. To this end, they should present (perhaps as supplementary material) more diagnostics for the 8-factor solution, (e.g. seed dependence) and the systematically tabulated results of the correlations with the external tracers for the 4-factor unconstrained, the 8-factor unconstrained and the 4- factor constrained solutions. Also, was a constrained 5-factor run attempted?

We agree with the referee that not sufficient detail about the procedures how the best ME-2 solution was determined were given in the Supplementary so far. A reason was that the focus of this work was set more onto performing the same source apportion-

ment for each instrument and examine if the model yields the same results instead of presenting a source apportionment for the Paris region which was already done before by Crippa et al. and others. But of course nevertheless great efforts were made to find the best possible solution. This is crucial for the possibility to perform a good intercomparison.

The seed diagnostics and correlations to external data mentioned by the referee were used to this end in addition to a thorough investigation of the model residuals and a large number of factors and different constraints (e.g. anchor profiles).

We added Chapters 3 and 4 (17 pages, 10 figures) to the Supplementary containing a detailed discussion of these parameters and justification why the constrained 4 factor solution was selected.

Further from this, the authors describe that they applied different target profiles and a values for the different ACSM instruments, but because this is documented in a verbose descriptive manner in section 3.4 (P1579 L27 onwards I find particularly confusing), it is not completely clear to me how the same decision-making process was robustly applied across all of the instruments to obtain the 'best' solutions. I would strongly recommend that the authors document this process in a more procedural form, outlining it in a more step-by-step manner that leaves no room for ambiguity. It would be very useful if this procedure could be outlined in the form of a flowchart.

The text P1579 L27 was updated and clarified. In addition a flowchart of the procedure was added to the supplement (Fig. S8).

There is a major caveat that must be applied to this work in that the methods used to obtain the optimal solutions were based on comparisons with other instruments. While this has probably meant that the most reliable solutions were indeed obtained, the overall outcome would probably have been much less favourable if these external comparisons were not available. Furthermore, if these external data were not accurate (for whatever reason), this would have compromised all of the solutions. While all of

these are implicitly or explicitly acknowledged in the text, I feel that this is an important enough point that the fact that external data was used to select the ME-2 solutions should be mentioned in the text of the abstract.

We agree that correlating the PMF results with external data to find the best solutions depends on the reliability of these external measurements. However, this caveat applies to almost all PMF source apportionments presented in the literature, since PMF results always have to be justified and checked against reality. One very common way is to check the results against external data. Results that don't agree are typically discarded. In this manuscript we basically do the same. Specifically, for this study the maximisation of the correlation to external data assures that for each instrument an equivalent solution is selected. Even if this was not the "best solution" if the external data was flawed we would make sure to not compare apples and oranges. A hypothetical absence of external data would render this comparison much more difficult but does not mean that a good PMF analysis in such a case would not be possible. We do not want to imply that.

We changed the L19 in the abstract from "Individual application and optimisation of the ME-2 boundary 20 conditions (profile constraints) are discussed together with. . ." to "ME-2 boundary conditions (profile constraints) were optimised individually by means of correlation to external data in order to achieve equivalent / comparable solutions for all ACSM instruments and the results are discussed together with. . ."

Minor/technical comments:

P1564, L11: The authors list the period that the data is from. Is this the entire measurement period? If not, why was this period selected?

It is the period all instruments were running and measuring in parallel, which is important for the comparability of solutions. There still were some short breaks within this period for some instruments but they were deemed negligible since the breaks were fixed after short time by the on-site staff. In total the study was conducted over a pe-

riod of three weeks (from the 10th Nov to 2nd Dec) but the first week was reserved for setup, calibration, and inlet preparation.

We added the parenthesis "(the full period of parallel measurements of all instruments)" to P1564 L 11.

P1564, L23: I find the list of additional instruments a bit unwieldy. The authors should restrict the list to the ones pertinent to this study and refer the reader to the other paper for a complete list.

We reduced the list and refer to the companion paper.

P1565, L11: Why is it ~30 min? Is it not exactly 30 min? If not, why not?

The time resolution of the ACSM depends on the settings of the quadrupole scan speed. Here (and in many other ACSM campaigns as well) the settings are selected to be as close to 30 min as possible but in reality are not exactly 30 min.

P1565, L15: The vaporiser should be specified as being made of porous tungsten and be in an inverted cone shape.

Thank you for the comment, we clarified that in the text.

P1565, L27: Again, the authors should be a bit more specific when they quote approximate values for the flows. This could be taken to mean that they don't know the actual flows, or that they varied within the measurement campaign or between instruments. They should specify what they mean by using the '~' symbol.

The inlet flows of the ACSM can differ slightly between instruments (depending on exact size of the critical orifice or the turbomolecular pumps) and also change during a campaign. The small changes during a campaign however are taken into account by the standard airbeam correction (we added a sentence about that to the manuscript) and the differences between instruments are taken into account by the IE calibration. We removed the symbols, because we think that on the order of precision shown here

(one digit after the comma) we are having the same flows in all instruments.

P1566, L2: The chopper slit is not used for the backgrounds; the beam is fully blocked for the background.

Revised.

P1570, L23: I don't understand the problem described with the TOF-ACSM, or the reasons given for why it was occurring. The authors should describe this in more detail (probably in the supplement), showing graphs for what they describe.

A few more lines explaining the applied correction and reasoning were added to the text and a graph was added to the supplement (Fig. S1).

P1571, L25: Was the median data calculated on a point-by-point basis, or was this the organic time series of the median instrument?

Revised. It was calculated on a point by point basis.

P1571, L13: I do not consider an influence of vaporiser temperature to be 'ruled out' by the study of reference standards because it has never been conclusively proven that reference standards behave exactly the same way as ambient organic matter. I think it would be appropriate to tone down this conclusion to state "we do not believe that vaporiser temperature is responsible. . ." or something.

We changed the statement from "ruled out" to "deemed unlikely" and deleted the word reference since we tested several real ambient aerosol mixtures that were collected on filters and re-extracted by the cited nebulisation technique.

P1576, L6: The majority of the signal in HOA is thought to come not from the fossil fuels themselves but the lubricating oils used in diesel engines (e.g. Canagaratna et al., Aerosol Sci. Technol., 38, 555-573, 2004).

Revised. We thank the referee for the clarification.

P1577, L12: The signal originates not just from levoglucosan, but also other anhydrous sugars (e.g. mannosan and galactosan) that are co-emitted.

Revised. We thank the referee for the clarification.

P1577, L17: Variations in the BBOA mass spectrum can be ascribed to ageing, but can also occur at source; see doi:10.5194/acp-15-2429-2015

We included the suggested citation to the main text.

P1579, L16: The inclusion of the R2 between OOA and SO4 in the basis for selecting solutions is questionable. While correlations between these have noted to have been high in a number of datasets (owing to them often being from regional anthropogenic sources) there are also examples where this has not been the case (e.g. doi:10.5194/acp-15-2429-2015). While I have nothing against documenting the correlations, for the authors to include it in the criteria for selecting solutions, they must present a robust case (e.g. based on previous measurements at this site) for why a good correlation is expected here.

Crippa et al (2013): doi:10.5194/acp-13-961-2013 showed covariance of OOA and SO4 at the same site for winter 2010 (R^2=0.63). A comment and a reference to that paper was added to the main text.

P1584, L16: Hypothetically, could the a-value be relaxed for m/z=44 specifically, rather than being applied to the whole mass spectrum? This could be a recommendation for a future development of the analysis software (this is kind of said later on).

Yes, basically the ME-2 solver allows that. It is just a matter of how to correctly prepare the input data. As the referee mentions we suggest one way in the recommendations towards the end of the manuscript.

P1587, L11: It should also be noted that this does not account for any variation in CE of the different sources, something that other analysis has possibly hinted at (e.g. doi:10.5194/acp-15-2139-2015).

We included the suggested citation and note to the main text.

Please also note the supplement to this comment:
http://www.atmos-meas-tech-discuss.net/8/C1043/2015/amtd-8-C1043-2015-supplement.zip

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