

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 #1

Received and published: 23 March 2015

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

C357

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?

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.

There is a major caveat that must be applied to this work in that the methods used

C358

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.

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?

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.

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

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

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.

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

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.

C359

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

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.

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).

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

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

P1579, L16: The inclusion of the R2 between OOA and SO4 in the basis for selecting solutions is questionable. While correlations between these have been 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.

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).

P1587, L11: It should also be noted that this does not account for any variation in

C360

CE of the different sources, something that other analysis has possibly hinted at (e.g. doi:10.5194/acp-15-2139-2015).

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