

Interactive comment on “Improved source apportionment of organic aerosols in complex urban air pollution using the multilinear engine (ME-2)” by Qiao Zhu et al.

Anonymous Referee #1

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This paper details the use of a combination of unconstrained PMF runs and ME-2 to provide better factorisation of AMS/ACSM data without the need to rely on a priori spectra as much. The basics are to perform an unconstrained PMF run, select a subset of the factors that would appear to be stable and then use these as target factors for ME-2 such that the solver can be run with fewer degrees of freedom and thus give a less ambiguous output. While I could envisage that the success of this technique may vary dataset to dataset, it may be that the technique could be further refined and best practice established in due course, so I would consider this worthy as a technical paper.

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The manuscript is generally suitable for AMT and very well written and I can see the hypothetical benefit to the technique. However, I find it a little curious to see that one of the factors identified for this treatment is BBOA, which is known to vary within individual datasets (e.g. Young et al. (2015) Atmos. Chem. Phys., 15: 2429-2443, 10.5194/acp-15-2429-2015). It's also curious that they should select the optimum based on comparisons of the mass spectrum with previous studies. By doing this, I would see that what they are doing is little different to simply using the a priori reference spectrum in ME-2, thus defeating the whole purpose of the technique. The authors should comment on this.

As a more general point, I would request a few changes to be made to give the paper a better theoretical footing and also to be of more use to potential readers, which are necessary for this to be a good technical paper. It may be that this technique is more suitable for some datasets than others, so it is important to do more than simply show it working in one instance. I would recommend that the authors do the following:

1) Present a more robust theoretical case for the improvement in apportionment that could result from this method. While I would not ask the authors to submit a full mathematical proof, I would surmise that the factors that this would work best for this treatment would be the ones whose profiles are invariant (i.e. conform to the PMF data model) and produce a time series that is distinct from the other components. These should be explicitly stated and the implications of using factors that do not conform to these assumptions discussed. For instance, I would expect that if a factor has a profile that varies with time, one would expect that this would be under-represented in the unconstrained PMF solution (with some of its variability being represented by other factors) and therefore under-represented in the ME-2 solution.

2) A step-by-step recommended procedure should be unambiguously presented, for the benefit of those attempting to recreate the method. While this is kind-of done in the conclusions, it is very vague in places.

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As a final technical query, can the authors confirm that the PAH data used to validate the result were not allowed to influence the factorisation originally? It would defeat the object of the exercise if they were.

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