

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.

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1. 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.

REPLY: Although BBOA varies across different datasets, the differences among differ-

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ent BBOAs are much less than those among different OA factors, which made BBOA identified by factorization in many studies. In this study, the purpose of comparing the BBOA anchor profiles from the unconstrained PMF results with the previous ones was just to confirm their basic BBOA characteristics, providing a new way to obtain a reasonable anchor profile for the ME-2 method, without the need to rely on a priori spectra. In the revised text, we have added the analysis that using the BBOA (and other POAs) spectra generated by the unconstrained PMF run of the same local dataset was indeed better than using a priori spectra from other studies, as in the reply to the next question.

2. 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.

REPLY: Both of the PMF and ME-2 methods assume that the source profiles are invariant with time during the whole campaigns, and the invariant source profiles identified by PMF or ME-2 are the relatively best selection to the final results in terms of statistics. In order to prove the improvement of using the anchor profiles generated by the unconstrained PMF run with the same local datasets, which do not depend on other studies, we also run the ME-2 analysis using the anchor profiles in the literature, with the results shown in Table 1-2. For Qingdao, the correlations between POAs and their tracers and the Q/Qexp values using the three BBOA profiles in the literature are poorer than using the BBOA obtained in this study (Table 1). For Dongguan, the results from ME-2 using the HOA profiles in the literature are also poorer than using the HOA profiles obtained

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in this study (Table 2). Therefore, it can be seen that the method to get an anchor profile in this study is easier (it does not depend on the literature) and more valid. We have added the above analysis in section 3 in the revised manuscript.

3. 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.

REPLY: We have adjusted the relevant text structures accordingly.

4. 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.

REPLY: The process of PAH quantification is now added in Section 2.4. The input matrix for PMF/ME-2 in this study does not include PAH fitting ions. We generally use the matrix with m/z of less than 100 (or 150) as the PMF/ME-2 input data, but the PAHs ions mostly have m/z of above 150. Therefore, the PAH data do not influence the factorization.

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