Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-465-RC2, 2016

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Organic Aerosol source apportionment in London 2013 with ME-2: exploring the solution space with annual and seasonal analysis" *by* Ernesto Reyes-Villegas et al

Anonymous Referee #2

Received and published: 1 July 2016

The authors present a source apportionment analysis for 10 months of ACSM measurements in North Kensington, London. ME-2 is used to explore solutions and sensitivity of results to algorithm parameters (seed, number of factors), segregation of data set (whole dataset or by season), and target profiles known a priori. The authors use a regression approach to help determine the final solutions and explore the variability in factor compositions obtained. The authors find that HOA and COA are robust while the other OA to be more variable from the perspective of mass fragment ratios, and that seasonal decomposition results in higher variability (also considering mass fragment ratios). Source apportionment is a difficult problem with a large number of possible solutions, and the authors make a worthwhile contribution in understanding

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how to explore this space and the variability in solutions generated from ME-2 using ACSM measurements. The use of trilinear regression is a useful addition for this data set. The results are clearly presented and the topic is suitable for Atmospheric Chemistry and Physics. The manuscript is recommended for publication after the following comments are addressed.

1. Much of the variability in solutions are expressed in terms of fragment ratios. What is the range in OA mass estimated for the set of remaining plausible solutions?

2. The conclusions regarding the superiority of the outlined approach are perhaps stated too strongly by the authors. For instance, in the abstract and conclusions: trilinear regression is said to "objectively determine" the solution, but it is perhaps not truly not objective in that the authors are imposing their prior assumption regarding the nature of cooking emissions and their relation to combustion tracers. And even then, the protocol does not uniquely determine the "best" solution. On this point, it would be useful to plot confidence intervals on the regression coefficients in Figure 2 (and Figure S6) as there are a number of solutions which fulfill this criterion.

Similarly, the statement about ME-2 being "robust" or "using appropriate mass spectra" being important may be revisited. As pointed out by Reviewer 1, there are some concerns about using AMS profiles to constrain ACSM. More generally, introducing source profiles can help reduce the range of solutions compared to PMF. However, it is not made clear in this manuscript that the solutions obtained by this approach are necessarily better than a subset of solutions that can be obtained by PMF (which are derived solely from the ACSM data). Some claims might be made regarding the appropriateness of AMS profiles to the extent that the physical expectations set forth by the authors are met using them (i.e., there are solutions for which COA is not correlated with combustion tracers), but there are still many questions remaining to make too strong a conclusion.

Minor comments:

In Figure 4, it would be helpful to plot points or contours for the range observed in ambient samples for comparison against ME-2 profiles.

Section 4.4: Some discussion of the NO3 and LV-OOA percentages in the text would be useful.

The authors write the essential equations for PMF but not equations for how factor constraints are introduced using the "a-factor" by ME-2, while the rest of the manuscript is dedicated to presenting ME-2 solutions.

Additional minor comments were pointed out by Reviewer 1.

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