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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 #1

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Review of Reyes-Villegas et al., "Organic aerosol source apportionment in London 2013 with ME-2: exploring the solution space with annual and seasonal analysis

General comments

This paper presents a thorough investigation of the possible factors contributing to OA in London using both traditional PMF and multilinear engine (supervised algorithm). This analysis is useful in that many groups are using PMF and beginning to use ME-2, but so far little validation has been done on the merits or pitfalls of ME-2. Futher, despite the subtlety of the findings and methods, the authors do a reasonable job of guiding the reader through the manuscript. One significant shortcoming of this work is the use of AMS-dervied target profiles to deconvolute ACSM data. While spectra are

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similar, important differences have been reported. This is described in detail below. The major concern regarding the use AMS target profiles is that future work will rely on these findings to justify the use of seasonal target profiles (for example). Currently, a number of sections are difficult to read and a few key details are missing. These are relatively minor changes and I recommend this paper for publication following these changes as well as a discussion of the caveats involved in using AMS target profiles with ACSM data.

Specific comments

Abstract line 19: the phrase "seasonal data sets showed a higher seasonal variability" is confusing. After reading the rest of the paper, I think I understand what the authors are saying, but this should be rephrased for clarity. I believe the point is that some sources are seasonal, and therefore the seasons should be evaluated independently rather than running PMF or ME-2 on the full data set.

Line 156: Use of target profiles from AMS and not ACSM may hinder this work, given that Ng et al. 2011 reported a markedly different collection efficiency for HOA and OOA by the ACSM, leading to underestimation of HOA by the ACSM. Further, Ng et al. 2011 reported that when HOA containing particles are externally mixed from OOA particles, this differential collection efficiency could be significant. Do the authors have any insight as to whether this could be important for the London work? At the very least, this shortcoming should be presented clearly at the outset of the work. If ACSM target profiles are available, those would be preferable to test the performance of ME-2.

Line 216: The authors write, "When analysing different solutions from the same dataset (Fig. 2.b), it is possible to observe that the use of different a-values does not imply a high variation, ranging between 1.88-2.2, suggesting that all the solutions are mathematically acceptable." I do not understand the point of this sentence, and I think the main issue is the phrase "does not imply a variation." Please clarify.

Line 220: The authors write "however, [the] PMF solution has a high Chi square and

negative slope for COA (Fig. 2.a), suggesting that this solution is not environmentally acceptable, thus it is necessary to analyse all the different parameters in fig. 2 in order to select the solution that best identifies the OA sources." It is useful to use the trilinear regression to determine if the PMF factors are environmentally realistic. However, the authors should provide more detail here if they intend to use this justification to eliminate the PMF solution. Specifically,for which "y" is the slope negative? BC, CO, NOx or all three? Please indicate here. And, was this true for all the PMF solution rotations under 5 factors? Or just the one with the lowest Q/Qexp? What about seeds? Was this negative slope true for those as well?

Line 292 (first entire paragraph in 4.1.2): I could not parse this paragraph. I think the term "variation" is misused and causes confusion. I understand the value of using the trilinear regression, but this description is nearly incomprehensible. Please revise.

Line 322: This finding, that SVOOA is not always in the same position in the triangle relative to LVOOA seems BIG. Perhaps it could be emphasized more, or earlier? In the abstract?

Table 1: This would be far more useful if the acronyms were avoided or defined.

Figure 2 (a): The color scale is useful to compare residuals, though I question the use of different colors to indicate positive and negative residuals. It would be easier to read the graph if either the best solutions were darkest (since the dark pink looks close to the dark green) OR if the same color was use for both positive and negative. It would also be useful to separate or demarcate the break between "a" and "c" as well as "c" and "w." This would make it clearer that we are comparing those larger groups instead of individual factors.

Technical corrections

Abstract: Please spell out PSI.

Abstract and throughout: Is "interphase" a British form of "interface"? (Line 72)

Line 30: "comprising" should be "composing"

Line 216: insert a space before the sentence beginning "When"

Line 220: should read "the PMF solution" instead of "PMF solution"

Line 298: "All this analysis carried out in the section 4" needs editing for grammar. Perhaps "The analysis presented in Section 4"

Line 380: "found secondary aerosols to dominate high concentrations over the year" should be "found secondary aerosols to be the predominant source of PM over the year"

Line 392: "infer" is misused here, instead the authors could use "support"

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