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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 #3

Received and published: 6 July 2016

This manuscript presents the ME-2 factorization tool to explore the organic aerosol sources in London. The authors use the trilinear regression to compare different ME-2 solutions, and five OA sources were identified including BBOA, HOA, COA, SVOOA, and LVOOA. The authors present the ME-2 solutions and using a-values approach and tested constrained factors. There is high variability in different seasons; however, the authors did not point out the major contributions and specify the variability. The methods and results are presented very well to explore the ME-2 method for the comparisons. The triangle plot of f43: f44 and f44:f60 was applied to further examine the seasonal difference. In general, this manuscript presents lots of scientific results and data analysis, and it's publishable on Atmospheric Chemistry and Physics. Some

concerns and comments are listed.

1. The authors use ME-2 tool to analyze ACSM data to further explore the OA sources and find the best solutions, and the criteria for selecting best solution are using a-values approach, minimizing Q/Qexp, and trilinear regression analysis. However, the criteria of determining best solution are not clearly explained in each analysis (Figure S4∼Figure S7).

2. The triangle plot of f43: f44 and f44:f60 was introduced to compare the seasonal differences. The authors also pointed out that it's an oversimplification tool to address the chemical complexity of LVOOA and SVOOA component, and further work is needed to completely address the OA chemistry difference in seasonal changes. Please clarify it or add more details how this triangle plot addresses the results and conclusion. In the abstract, the authors write "the seasonal variability was explored with triangle plots of f43:f44 and f44:f60, with HOA and COA being the most suitable sources to constrain." The COA is mainly characterized by m/z 55 and m/z 57, but here the f60 is low for COA. Is it more appropriate to look at f55 and f57 for COA in seasonal differences?

Minor comments

Line 188 : Equation (4) B and C parameters were not defined.

Line 288-290: The authors write "The summer is overestimated and a strong variation of the source profiles, a situation that ME-2 is not able to capture." If there is a strong variation of the source profile, how does the ME-2 confirm the data accuracy?

Line 296: The authors write " the March-Dec dataset solution does not completely capture". What range of variability for thrilinear regression? The authors write there are seasonal variations that the dataset solution does not completely capture. How does this method completely capture it and avoid failures?

Line 320-322: " The fact that the lines are going in different directions with the seasons of year means that the factorization is identifying different aspects of the chemical

complexity." It seems that the "chemical complexity" is not clear explained why the LVOOA and SVOOA lines are different directions.

Line 330: "The fresh OA during autumn and highly oxygenated BBOA during summer" seems not convinced. It would be good if the authors could provide O/C ratios or any indicators (f44 or f43) that supports the "highly oxygenated BBOA" during summer.

Line 343-345 : Please add reference for this paragraph. The authors use the ratios of NOx/HOA, CO/HOA, and NOx/ $\triangle$CO to determine the weekdays and weekend ratios. However, there is no strong evidence show that these ratios are best indicators to analyze the impact of diesel or petrol emissions contribution. Also, when the authors conclude the heavy-duty diesel vehicle emissions are possible contributions during weekdays, but it seems not strongly supportive to conclude this.

Line 375 : " The main PM1 contributors to moderate and high PM2.5 concentrations are NO3 and LVOOA." It would be supportive if the authors could provide more information about NO3.

Line 390: In the conclusion, the author write " higher variation mainly in the SVOOA mass spectra and the BBOA; less variability was observed in LVOOA, COA and HOA." The "variability" term was used in this manuscript many times, but there is no clear range for these variations.

Table 1 : As mentioned by reviewer 1, please define clearly for the sets of target profiles.

Technical correction

Figure S1: Please label significant m/z peaks, O/C and H/C ratios in each solution. "(c)" was missing in the end of the caption of Figure S1.

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