

Interactive comment on “Receptor modelling of both particle composition and size distribution from a background site in London, UK – the two step approach” by David C. S. Beddows and Roy M. Harrison

Anonymous Referee #3

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The manuscript “RECEPTOR MODELLING OF BOTH PARTICLE COMPOSITION AND SIZE DISTRIBUTION FROM A BACKGROUND SITE IN LONDON, UK – THE TWO STEP APPROACH” presents a new approach (based on 2 steps) for source apportionment studies using positive matrix factorization (PMF). This method aims to properly handle dataset(s) composed of variables with multiple units (mass and number concentration, in this case). The authors claim that this new method overcomes the problem of inputting variables with heterogeneous measurement units. The authors also claim that the method is able to better detect hidden factors/sources.

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The manuscript has several elements of originality (to my knowledge, no similar methods have been already published) and directly hits a very controversial and up-to-date topic in the atmospheric sciences. Nowadays, source apportionment by PMF is amply used in both routine monitoring and research studies. Although most of them use “one-kind” variables (mostly PM chemical speciation data), an increasingly high number of studies (just a few have been cited in the manuscript, but the list should be improved) use variables with multiple units. Since the large number of available air quality measurement techniques, the merging of dataset(s) with different units is a suitable (and proven) way to better resolve the PMF source profiles and to detect unresolved sources. Essentially, additional variables may help in better detecting the edges. Under this view, a recent paper (Emami and Hopke, *Chemometr. Intell. Lab. 162* (2017) 198–202, which findings are unfortunately not considered in this manuscript), showed the effect of adding variables with different units to decrease the rotational ambiguity of PMF solutions.

Thus, the topic is suitable for the journal ACP. However, the manuscript needs revisions before to be accepted for publication.

Major points.

Essentially, the rationale behind the whole manuscript is based on the statement reported in lines 59-62: “However, while combining, for example, particle chemical composition and size distribution data in a single PMF analysis may assist source resolution, it does not allow quantitative attribution of either particle mass or particle number to the source factors.”. Later, the authors also presented a case study where they mixed variables with different units without giving quantitative results. Even if one can agree with this statement, the authors have not exhaustively explained it. Since this is a methodological manuscript, I strongly encourage the authors to better support these statements.

Another major weakness of this manuscript is the lack of sufficient details on the PMF

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analyses. This point can be easily solved by the authors, who have an extended experience with PMF analysis. This manuscript presents a new approach, so particular care should be given to details so that anyone can easily reproduce what the authors did (and test with their own data). However, details of the PMF are generally missing or they are reported in the companion paper (Beddows et al., 2015). For example, the authors should describe the method(s) used to compute the uncertainties in the 1st step (including PN0.6-10, see next point). Also, the authors should report how the raw data have been handled (if any correction was done) and the number of variables and cases inputted into the models. For example, they should report the outliers detection and how they managed the missing values (SMPS sampled every 15 min, what is the minimum number of 15 min records to have a valid 1-hour NSD value?). In addition, since the Q values are used (lines 199-203 and 215-216), they should be reported as well. Furthermore, it is unknown if the authors dealt with the rotational ambiguity of the models. Basic information on the PMF set-up is important to report. This information will allow the reader to completely understand what the authors did and (possibly) to reply the methods. It would be useful to have a quick overview of such details in the main text with the deepest description in the supplementary information.

Another unclear point is related to the “proxy-data” used to assess the PN0.6-10 variable. This is an artificial variable: it was not directly measured, but it was computed on the basis of two (three?) main assumptions: (i) particles are assumed to be spherical, and (ii) particles have fixed density. But it is not completely clear if the density is assumed constant over the time or over the whole (16-604 nm) size spectra (or both, as it should be). The authors used a density of 2 g/cm³ over all the study period, but they report a 1.8-2.5 g/cm³ range for an urban background aerosol. Consequently, the PN0.6-10 variable will be affected by a large uncertainty that cannot be well assessed. I suggest to add more details and provide an estimate of the uncertainty of this new variable.

This latter point raises another question. Why the authors did not plan to also use an

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APS to complete the size range to 10 μm ? One can argue that the sampling campaign was not planned to have an APS included (or the merging of SMPS and APS was unreliable). However, my opinion is that this point should be at least mentioned in the text, so colleagues who want to pursue the same approach are advised on the possible use of wide range particle size spectra.

The authors used R to “optimize” X to have $Q/Q_{\text{theory}} \sim 1$. More details should be reported. In particular, what does “ ~ 1 ” mean? It can be every number, but having it from 0.5 to 1.5 or from 0.99 to 1.01 makes a big difference. Please explain.

Minor comments.

Line 103. Missing bracket “)”

Subsection 2.1: Please add more details on the SMPS set-up. For example, sheath and sample flows, the status of the CPC and electrostatic classifier (serviced, calibrated?), the type of neutralizer (X-ray, 85Kr?), software/algorithm used for the data inversion (or version of the AIM software), use of multiple charge and/or diffusion loss corrections, etc. These details need to be added as supplementary information.

Line 138: $\frac{1}{4}$ hour -> 15 min

There are two equations numbered as (3), see pages 11 and 12. This should be fixed, as most of the discussion on the method refers to these equations.

Figure 3 can be easily moved to the supplementary material file.

Figure 4. NET & coarse should be NET & crustal.

Figure 6. Once printed, the labels and axes of the single plots will be likely unreadable. Please increase the font size and (if possible) please uniform the font style and size among the figures. Also, it is advisable to use a color scale that is also easily readable when the paper is printed with a black and white printer.

Figure 6 shows polarplots and polarannuli. These “openair” analyses are commonly

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reported in air quality studies and are very helpful to better interpret the data. However, a quick overview of the information provided by these two plots should be briefly reported into the materials and methods section.

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