Response to Reviewer #3

We kindly thank the referee for taking our manuscript into consideration and we value the comments raised to improve the manuscript. A point-to-point response to the issues raised is enclosed below.

Visser and co-authors describe the Multilinear Engine implementation of the Positive Matrix Factorization model used on datasets of trace metals collected in three size ranges (PM10-2.5, PM2.5-1.0 and PM1.0-0.3) at three sites in London during the ClearfLo campaign in winter 2012. The implementation of the model was conducted on datasets comprising all three sites but segregated by size. This approach is very useful for the separation of sources with high temporal covariance but significant spatial variability. The main contribution of this study is the use of "anchor profiles", which were retrieved by analyzing data subsets in which a particular source was evident. The author's used those anchor profiles in ME-2 for rotational control of the solution. This approach although it introduces some subjectivity in the analysis is in my opinion a very nice and useful approach. Rotational ambiguity is the main source of uncertainty in this kind of analysis, so a technique that helps to control the rotation using profiles/information that derive from the datasets and not some external source can be very useful if properly implemented. The ME-2 analysis on the datasets resulted in a total of nine source profiles, three for each size fraction, which were namely brake wear, sea salt, resuspended dust, secondary sulphate, fuel combustion and industrial emissions. The attribution of the factor profiles to sources is well justified in all cases. The final results include information about the relative contribution and the spatial variability of the sources as well. Overall I find this study to be very well written and scientifically sound. For these reasons, I recommend it for publication with a few minor suggestions.

Comment #1:

I suggest adding a paragraph comparing the results of the unconstrained run of the model with that of the constrained run. It will help the reader to understand the benefits of using an anchor profile, especially for the profiles that were not well defined on the initial run. In addition to that it will help the authors justify why they selected those specific sources to apply the constraints.

Response:

We have added the following discussion to the end of the synthesis section (Section 3.2):

"The analysis herein clearly shows the advantages of rotationally controlled analyses relative to an unconstrained PMF solution. Figures S2-S5 show the best solutions retrieved from unconstrained analyses for the separate size fractions (4-, 4-, and 5-factor solutions for $PM_{10-2.5}$, $PM_{2.5-1.0}$, and $PM_{1.0-0.3}$, respectively). The unconstrained $PM_{10-2.5}$ solution (Figs. S2 and S5) yields high residuals of Ni, Cr, and Mo and does not resolve a brake wear factor. The unconstrained $PM_{2.5-1.0}$ solution (Figs. S3 and S5) likewise does not yield brake wear and additionally fails to resolve aged (reacted) sea salt from regionally transported sulphate and solid fuel, despite strong evidence for this processing in the raw time series. Finally, the unconstrained $PM_{1.0-0.3}$ solution mixes secondary sulphur and solid fuel sources. It also fails to explain major events contained in the Cl-rich factor, apportioning significant Na to these events, leading to high Na residuals. Higher order solutions do not resolve these problems, instead leading to uninterpretable splitting of the dust factor, factors consisting only of single elements, and unstable solutions that are highly dependent on algorithm initialization (seed)."

Comment #2:

Page 12, lines 359-364: Have the authors considered the possibility to check the Si/Ca ratio to investigate possible influence from construction work?

Response:

The reviewer refers to the scaled residuals (θ_{ij}/σ_{ij}) ratios that exceed ±3 for Na, Si and Ca (coarse), Na, Al, Si and Ca (intermediate) and Al and Si (fine) and/or that are skewed at the sites relative to each other. This spread in the scaled residuals may indicate different dust profiles across sites. Although we were not able to separate different dust profiles, it is indeed possible that the resuspension dust profile in the city is influenced by other dust-generating activities, such as construction work.

This point has been clarified by changing lines 62-64, p. 12 into:

"This is potentially caused by varying dust compositions or emission processes. Resuspension in the city is dominated by road dust influenced by anthropogenic activities and by other dust-generating activities, such as construction works, in contrast to influences from natural soils at DE."

Comment #3:

Page 16, lines 397-399: Maybe not all aged sea salt is resuspended. At least a part of it might be fresh sea salt reacting with HNO3 in the atmosphere. Thus it would be expected that the availability of HNO3 would affect this source at least partially. Because HNO3 is expected to have higher concentration in polluted areas, this source might not be site-independent.

Response:

We agree with the reviewer and noted in Section 3.1.3 (paragraph 2) of the original manuscript that the aged sea salt likely originated from both the reaction of HNO_3 with fresh sea salt (based on trajectory modelling) as well as from resuspended sea salt (based on diurnal patterns and concentration gradients between higher/lower-traffic sites). This paragraph has been reorganized for clarity as follows:

"The data suggests that a fraction of the aged sea salt is directly transported from the sea, while part comes from resuspended sea salt particles after deposition on roads. Direct transport is indicated by the diurnal variations (Figs. 7 and 8), which have no obvious pattern – peaks occur at different hours of the day throughout the entire time series, whereas resuspension would likely peak during the day with vehicle use. Additional support is provided by NAME dispersion modelling and wind direction analyses, which indicate that high concentration episodes in the aged sea salt factor coincide with air masses from the sea. The sea salt concentrations also increase with increasing wind speed, consistent with other Na observations in the UK (Supplement Fig. S12; Twigg et al., 2015). However, the $PM_{10-2.5}$ concentrations of the aged sea salt factor are enhanced by a factor of 1.3 and 2.2 at the kerbside (MR) site relative to the urban background (NK) and rural (DE) sites, respectively. This suggests that aged sea salt concentrations are also significantly modulated by human activity in the form of resuspension."

References

Twigg, M. M., Di Marco, C. F., Leeson, S., van Dijk, N., Jones, M. R., Leith, I. D., Morrison, E., Coyle, M., Proost, R., Peeters, A. N. M., Lemon, E., Frelink, T., Braban, C. F., Nemitz, E., and Cape, J. N.: Water soluble aerosols and gases at a UK background site. Part 1: Aerosols, Atmos. Chem. Phys. Discuss., under review, 2015.