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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Advanced source apportionment of size-resolved trace elements at multiple sites in London during winter" *by* S. Visser et al.

Anonymous Referee #1

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Visser and co-authors describe a positive matrix factorization analysis of trace metal SR-XRF data (for rotating drum impactor samples) collected in three size ranges at three sites in London during the ClearfLo campaign in winter 2012. For each size range, data from all three sites were combined into a single dataset prior to analysis by ME2-PMF. This is a nice approach because sources that have high spatial gradients can be identified even if they co-vary temporally when the sites are compared to each other. The Multilinear Engine approach allowed for the introduction of representative "anchor profiles" associated with physically meaningful sources in the analysis. Some of the final factors resolved were constrained to have relative intensities for marker met-

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als within a user defined range of the anchor profiles used. The approach used here involved iterations of ME-2-PMF analysis of subsets of the data with high S/N ratios to identify periods where factors are well resolved, and subsequently applying these factor profiles as a basis set when analysing the entire dataset. The methodology is reasonably objective, but with user input in some cases where physically meaningful factors are extracted only at unsatisfactorily high values of p, for example. The final PMF results are used to infer conclusions about the spatial variability of trace metal sources across the three sites, the size dependence of the different sources, and the relative mass contributions of the different sources to total metal mass concentration at all three sites in all three size fractions. The authors identify and apportion sources associated with brake wear, resuspended dust, sea salt, secondary sulphate, solid fuel combustion and industrial emissions. The size dependence, temporality and spatial distribution of the mass contributions of the different factors supports their assignments. For example brake-wear and resuspended road dust exhibit the highest mass concentrations at the roadside site and lowest at the rural site, while secondary sulphate transported from continental Europe exhibits similar mass concentrations and temporality across all sites. Correlations with relevant tracers for traffic and solid fuel sources are also investigated. Overall, I find this effort to be very well written and scientifically rigorous with extensive sensitivity analyses. It thus represents a useful template for future source apportionment analyses of trace metals. I have only minor suggestions below:

General comments:

The description of the modified ME-PMF approach (Section 2.3) is quite dense, and I had to read through it three times to fully grasp the steps. Rewriting parts of this section will almost certainly help. It may be worthwhile to change the naming conventions of the steps to more immediately tangible titles for readers. The meaning of “ME-2 all” is self-evident but “PROF-nonres” and “SENS” are not necessarily useful when examining Fig. 1, for example. A brief discussion of which factors that could not be resolved or identified using the unconstrained PMF analysis could also be added to the discussion

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or conclusions section to demonstrate the value of the ME2-PMF approach used here.

One aspect I'm still not clear on is whether anchor profiles are used for all factors prior to the final ME2 analysis of the total dataset. For example, in Table 1, criteria for only a handful of the factors are listed for each size range. Were there no constraints for the other 3-4 factors in each size range? This should be explained in the text or caption. How was it decided which factors should or should not be constrained in each size range?

I suggest moving the map from the Supplement to the main manuscript as the site locations are helpful for interpreting Figs 3, 4, 11 and 14. Also changing the map to an image will help to demonstrate how "rural" the DE site is.

Minor comments:

Page 14737, line 13: should be "and PM2 data"

Page 14373, line 24: state the limit value

Page 14741, eq 3: But how does one decide how many factors have the "a" constraint applied?

Page 14742, line 14: "e.g."? Were other offset sample numbers investigated?

Page 14744, line 2-3 "Consistent with existing measurements"? Do you mean previously reported source profiles?

Page 14753, line 23-28: Rewrite this part for clarity

Conclusions: It would be helpful to mention briefly which sources could not be identified using the unconstrained PMF either here or earlier in the discussion

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 14733, 2015.