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Interactive comment on "Receptor modelling of both particle composition and size distribution from a background site in London, UK" *by* D. C. S. Beddows et al.
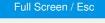
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Interactive comment on "Receptor modelling of both particle composition and size distribution from a background site in London, UK, by D.C.S. Beddows et al.

Anonymous Referee #2 Received and published: 2 June 2015 GENERAL COMMENTS

I recommend this manuscript for publication in ACP. The contents are relevant and of interest to the scientific community. The manuscript is well-organized and the findings are presented in a clear manner. There is a lot of work behind the manuscript presented

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and the conclusions are meaningful for the scientific community. Both the PM10 and the particle number sources in the London urban background have been identified and quantified. To this end two years of measurements were carried out, both for PM10 chemical composition and for particle number size distribution. The sources have been identified and quantified using Positive Matrix Factorisation (PMF). A combined PMF was also carried out. Nevertheless, some improvements should be made before the work is published in ACP. Please find some suggestions below.

1. The size range covered by NSD does not reach 10μm, hence it is obvious that the NSD source apportionment cannot identify the coarse sources, such as marine and crustal. As it is written now it seems that the only reason for not identifying these sources with the NSD data is that some sources are more important for mass and others for particle number, which is true, but if one does not measure at all in a size range, it does not matter how important is the source, it is impossible to see it. This could be pointed out already in the abstract.

We agree with the reviewer, and the sentence in the abstract has been modified as below:

"The two methods appear to be complementary, as the analysis of the PM10 chemical composition data is able to distinguish components contributing largely to particle mass whereas the number particle size distribution dataset - although limited to detecting sources of particles below the diameter upper limit of the SMPS, 604 nm - is more effective for identifying components making an appreciable contribution to particle number."

2. The investigation of back-trajectories and their relation with the variation in source contributions could be improved. Now only the days with maximum contribution of one of the factors are investigated. What happens with the second-highest day? And the third? Now, only info from Table 2 is shown for the days with the highest (>P90) contribution of each factor. The authors could classify every day of the sampling pe-

riod based on the back-trajectory and calculate the average source apportionment for each type of back-trajectory. This would confirm (or not) the explanations that are now given in the manuscript about the relation between air mass origin and source contributions. This comment applies for both the PMF results from the chemical and the NSD datasets. The comparison that is now made in page 10136 between the back trajectories for the day with highest contribution from each source depending on the database (chemistry or NSD) may change if a broader set of data is considered (as opposed to only the day with the highest contribution).

This has been dealt with by clustering all of the midday, daily back trajectories arriving at NK and calculating the contribution from each factor to the PM for those days. Descriptions have been added to the text which uses these to support the identification of the clusters. New Tables 5 and 6 express the average contribution of each source factor to the mean concentration on each trajectory (Table 5), and the contributions to the annual mean according to source factor and trajectory type (Table 6).

SPECIFIC COMMENTS 3. How often were the 24h samples collected? Daily?

Yes, this has been clarified in the first line of text in section 2.2 with the insertion of the word, "daily". Also for the NSD data it has been made clear that these were, "collected continuously every $\frac{1}{4}$ hour".

4. Page 10128, line 21. Please clarify what do you mean by "where data from the URG was not available laboratory based ion chromatography measurements on filters (Tissuquartz [TM] 2500 QAT-UP) from a Partisol 2025 were used". Do you mean lost samples? Concentrations below detection limit? Or species that can not be analysed by URG?

Points 4 and 5 have been addressed by clarifying the text.

5. Page 10128, line 23. Please clarify "Data capture over the two years ranged from 48 to 100 % as different sampling instruments varied in reliability". There are different

things to report. First: the sample collection: did you aim to collect samples daily? How many valid filter samples were collected? Second: the chemical analysis: from the collected samples, how many were analysed successfully? Explain better why do you have different % of valid samples for each species. A short explanation on the limitations of the analysis is expected, because you actually use the same URG.

Points 4 and 5 have been addressed by clarifying the text.

6. Page 10129, line 3. Please explain which sampler (and with which inlet size cut) you used to collect PM samples on quartz filters for subsequent EC and OC analysis.

The text has been modified to say that a Partisol 2025 was used with a PM10 size selective inlet.

7. Page 10129, line 6. You could say something more than only 'similar'. Add the reference for the EUSAAR_2 thermal protocol: Cavalli et al., 2010.

It is hard to quantify "similarity" in this context, which is why we reference the NPL report with the intercomparison. The Cavalli et al. reference has been added.

8. Page 10129, line 7. Please provide the size range from the SMPS and the time resolution.

This has been added to the end of section 2.2. The data was collected using 15 minute scans across 51 bins from 16.55nm – 604.3nm. This was averaged to hourly data for use in this work.

9. Page 10129, lines 11-12. Please explain which sampler did you use and with which inlet size cut to collect PM on Teflon-coated glass fibre filters

It was a Partisol with a PM10 inlet (now added).

10. Page 10130, line 15. It is not really true that the explained variation values show which constituents are the most important in each factor. What do you mean here with 'important'? It is rather that the source for which the EV is very high for one species

is the most important source for this species. And it is true that it helps identifying the sources, but maybe the authors want to re-write the sentence.

The reviewer is correct and this sentence has been reconstructed to make the point correctly.

11. Page 10130, line 18. Please re-write. The measurements are not made by the factors or sources. You can introduce the term 'contribution' here.

Word changed to 'contribution'.

12. Supplement: you wrote that you use SMPS+APS data, but you do not report any APS data in the main manuscript.

This has been replaced by 'input data'.

13. Supplement: please check thoroughly. Some typos/mistakes have been found. E.g. page 4: "within each factor as defined the in parametric form"; "For a given p factor solution there a (p+1) 'factor' is outputted

These points have been addressed and the text rewritten.

14. Supplement: there are two Figures S1.

The extra figure has been deleted.

15. Supplement: explain what is FKEY and why you need to apply it.

An explanation for FKEY has been added in the context of how we used it.

16. If CWOD is OA from wood burning, if you use both OC and CWOD for your PMF, you are using as an input something that is not independent, given that part of OC is CWOD. Why don't you use hence CWOD and then an estimation of non-wood-burning OC as a separate variable?

This is a useful suggestion, but difficult as CWOD is a surrogate variable, and several uncertain assumptions would need to be made to convert it to a mass of OC.

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17. Page 10132, line 13. Please explain better what do you mean by "consistent with exhaust emissions from road traffic resulting from factor pulling the EC and OC ratios". Consistent with previously measured EC/OC ratios closed to road traffic emissions? What do you mean by factor pulling here? If you used a constrain in the PMF for the EC/OC ratio, it should be clearly stated in the main manuscript.

We have clarified this section by explaining that we have applied factor pulling to the Traffic factor given a-priori information about OC/EC ratios for traffic. We have also added the statement that this factor has a high correlation to NOx to further substantiate our claim that this is a Traffic factor.

18. Page 10132, line 27. You say for these factors the concentrations are higher in the cooler months, but actually for January the concentrations of non-exhaust/crustal are lower than those in Sep-Nov, why?

Our statement is that the sources listed "all show signs of higher concentrations in the cooler months of the year". That was a very general statement designed to allow for exceptional months, and in the case of the non-exhaust/crustal factor, rainfall might be important and have suppressed the January concentrations.

19. Page 10133, line 8. The higher Fuel Oil contribution in summer may also be attributed to its sulfate content, since sulfate formation can be enhanced in summer due to the higher photochemical activity in summer.

This is a valuable insight and the text has been amended accordingly.

20. Page 10134, line 8. Why do the authors compare to Johnson et al. (2014)? Is there any specific similarity between Brisbane and London? There are many studies showing that shipping emissions affect ambient air concentrations of V (e.g. Pey et al., 2013; Zhao et al., 2013; Minguillón et al., 2014; Viana et al., 2014; among others).

Other references have been added.

21. Page 10135, line 16. The correlations between BC concentrations from the

aethalometer and the traffic contribution from NSD could be tested and reported to further confirm this factor.

We have run this calculation and the NSD Traffic has a correlation of 0.50 with the BC compared to a correlation of 0.82 for Urban Background and BC. Correlations of NSD factors with NOx are:

Correlation coefficients with BC:

Secondary 0.64 Urban Background 0.78 Traffic 0.53 Nucleation -0.01

Similarly for the PM factors and NOx:

[1] "Urban Background" 0.88 [2] "Marine" -0.22 [3] "Secondary" 0.33 [4] "Non.Exhaust.Traffic..and.Crustal" 0.22 [5] "Fuel.Oil" -0.102 [6] "Traffic" 0.75

And for the PM factors and BC:

[1] "Urban Background" 0.83 [2] "Marine" -0.31 [3] "Secondary" 0.26 [4] "Non.Exhaust.Traffic..and.Crustal" 0.32 [5] "Fuel.Oil" -0.04 [6] "Traffic" 0.87

We have amended the text to reflect this. 22. Page 10135, line 22. As mentioned by referee 1, the supposed wood burning increased activity during the weekend should be better justified.

A reference has been added

23. Page 10137, line 26. The study of the back-trajectories here could help. You may actually have nucleation under clean conditions for two (or more) types of air mass origin: marine air masses (your Aged Marine source) and other origin (your Nucleation source).

This is a helpful comment, and we have amended the text to reflect this possibility.

24. Page 10138. Polar plots. Where were the wind measurements carried out? At the same station as air quality measurements?

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No, these were taken from Heathrow Airport where there is less influence of buildings. A sentence has been added to clarify this.

25. Page 10138. Polar plots. How did you do the polar plots for the PM10 chemical composition data? Did you use average wind for each day? Or did you use hourly wind data and plotted the same concentration for the 24 data points for a single day? Or?

We used a vector average and this stated in the caption of Figure 10 it says "daily vector average wind velocity and direction".

26. Page 10142, line 21. The fuel oil factor was not apparent in the CMB results? What do you mean? When you mention previously which sources were included in CMB you do not mention fuel oil, hence it is obvious that it does not have any contribution, given the way CMB works. So, if it is a choice done a priori, the lack of the fuel oil source does not mean anything. Or do you mean an attempt with a fuel oil source profile was carried out with CMB and it was not well resolved?

As stated, the earlier work upon which the CMB modelling was based did not use suitable tracers for a fuel oil source and hence this was not included. The text has been amended to clarify this.

27. Page 10143. While it is true that the combination of chemical composition of PM10 and NSD data did not added much to the identification of sources, it would be worth to highlight the conclusions one can reach from the combined source apportionment, in terms of which chemical compounds are responsible to which particle size distribution ranges, as also pointed out by referee 1.

We have added, "There is a question of whether there was any advantage in combining mass composition data and number size distribution data in the source apportionment calculations. The PM10 components can be used to infer a which chemical components are most abundant for each of the number size distribution factors. For example, the nucleation mode (25 nm) is associated with nitrate and sulphate; the secondary

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mode (80 nm) is associated with OC, nitrate and sulphate, etc. This, however, needs to be viewed with caution due to the combination of data from different size ranges."

28. Page 10143, line 22. Please clarify what this 4.5% corresponds to, is it one single traffic source? The traffic contribution is present in three different sources, so the total traffic contribution is higher.

We have changed this sentences to reflect that our Traffic factor represents tailpipe emissions for both the PM10 and NSD factors. This then gives us a combined contribution of traffic to PM10 of up to 29.6% when accounting for the percentage of Traffic and Non-Exhaust and Crustal factors.

TECHNICAL CORRECTIONS - Table 1. CWOD: Please correct "using uses" Corrected.

- Page 10131, line 9: correct "campaign" Corrected from "campaign".

- Page 10137, line 12: use comma or semi-colon consistently. Corrected comma to semi-colon.

REFERENCES Cavalli et al., 2010. Toward a standardised thermal-optical protocol for measuring atmospheric organic and elemental carbon: The EUSAAR protocol. Atmospheric Measurement Techniques 3, 79-89.

Minguillón et al., 2014. Spatial variability of trace elements and sources for improved exposure assessment in Barcelona. Atmospheric Environment 89, 268-281.

Pey et al., 2013. Chemical fingerprint and impact of shipping emissions over a western Mediterranean metropolis: Primary and aged contributions. Science of the Total Environment 463-464, 497, 507.

Viana et al., 2014. Impact of maritime transport emissions on coastal air quality in Europe. Atmospheric Environment 90, 96-105.

Zhao et al., 2013. Characteristics and ship traffic source identification of air pollutants

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in China's largest port. Atmospheric Environment 64, 277-286.

These have been added.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 10123, 2015.

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