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 #1

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The paper by Beddows et al. shows the results of source apportionment on PM mass, number size distribution and a combined database of both measurements carried out in London over a two year period. They have used PMF to find the main sources of atmospheric aerosols. Overall, they have extracted 6 factors contributing to PM mass, 4 explaining number-size distributions, and 5 for the combined set of metrics. I recommend the publication of this article. It contains valuable results on source apportionment of both mass, number and combined mass + number size distribution datasets. But before publication, the authors need to consider additional discussion on different issues, as well as to reduce the length of specific points, mostly minor comments.

1) In page 10131, line 9. A brief description (at least the acronym) of the ClearfLo campaign should be provided, or some reference.

Added both acronym and reference.

2) In page 10132, line 4. I suggest replacing the word “sources” by “emissions”.

Changed to emissions.

3) In page 10133, lines 15-16. In the same line it appears two times the word “associated”.

Replaced repeat.

4) In page 10135, lines 17-18. Recreational wood burning? Is there any reference to justify this? Could it be just that people on weekends stay during more time at home and consequently biomass domestic heating increases?

This has been modified to cite a reference for greater woodsmoke concentrations at the weekend, but not to comment on causes.

5) In page 10136, line 17. In the same line it appears two times the word “associated”.

Replaced repeat here and at other locations not mentioned.

6) From where have you identified low wind speeds associated with the Nucleation factor? From the values given in Table 2, it is just the opposite.

For marine air, 8m/s is a moderate breeze and relatively low.

7) In page 10138, lines 25-26. The sentence “greatest density of London sources” is inappropriate. I don’t think all London sources are enhanced in the west of central London but probably the area is more affected by nearby vehicular emissions and/or by the” London plume”.

The reviewer has misunderstood the statement: Since the NK site is west of the centre of London, most of the city and its emissions lies to the east of the site.

8) In page 10139-10140, lines 26-30 and 1-2. In this sentence, the fuel oil combustion factor is roughly attributed to shipping emissions, while before (for PM10), the interpretation was mainly focused on stationary sources (maybe power plants or industries): “The only constituent to show higher concentrations in the warmer months of the year

is the Fuel Oil source. This might be attributable to emission from high chimneys with more efficient mixing to ground level during the more convective summer months". Please, clarify this item.

We are grateful to the reviewer for pointing out this discrepancy. The wind sector with which the fuel oil factor is associated contains a number of refineries as well as coastal shipping. The text has been modified to recognise that both are most probably contributing.

9) In page 10142, lines 3-25. Although I agree with what is written there, I have found the discussion about CMB (a technique not used in this article) too long. I strongly suggest summarizing this couple of sentences.
This has been shortened considerably.

10) In page 10143, lines 4-7. I partially agree with this sentence, but the authors have to consider that the outcome obtained from the combined dataset is not irrelevant. Actually, from those results they (and the potential readers) are able to identify those chemicals driving each of the NSD factors. Furthermore, in terms of source contribution concerning common sources extracted by the single and combined NSD datasets, the divergences are not negligible. In this line, there is a lack of discussion in the manuscript considering these issues. Which one of the outcomes should be considered as more realistic?

The reviewer raises an interesting point. We have consequently added the following sentences to address the issue of source contributions.

"At first sight, the combined PM_{10}-NSD analysis is attributing different percentages to the components (e.g. urban background) which overlap with the individual analyses. However, the point needs to be considered that while the analysis of the PM_{10} dataset attributes PM_{10} to source factors and similarly the NSD dataset attributes particle number, it is unclear what the combined analysis is apportioning. Consequently, the apportionment results should be viewed with caution as they relate neither to particle mass nor to number alone."

11) In page 10150. It is surprising for me that the fuel-oil factor encompasses the lowest SO2 concentrations? Can the authors give an explanation about that?

The explanation appears to be that the fuel oil factor was associated with by far the highest mean wind speed (Table 2). The extent to which the SO_2 is lower is surprising, but all pollutants show very low concentrations associated with this factor.

12) In page 10150, Table 2. From the values reported there, I would be more convinced if the "diffuse urban" becomes "urban background, or urban plume". The word diffuse drive me to think is somewhat diluted, but conversely the factor encompasses the highest pollutant concentrations.

We have re-named it "urban background".

13) In page 10154, Figure 2. The total number of particles is around 5500 cm-3. In Reche et al. (2011), http://www.atmos-chem-phys.net/11/6207/2011/acp-11-6207- 2011.pdf, the annual average reported for the same site was 12100 cm-3. Can the authors comment about these differences? Is this a product of abatement strategies? Is this a consequence of changing the instrument (CPC3022 to 3775)? Are other reasons behind (meteo, etc: : :)?

No, Reche et al are referring to CPC measurements in 2009 N_{7-1000}. Our annual average for 2011/12 is also very similar at 12,776 +/- 4,210 cm-3 using the same instrument. The value of 5500 cm-3 in the paper is the average for the total SMPS count which is N_{16-604}. This is measuring a fraction of the particles measured by the CPC.

14) In page 10160, Figure 8. It is surprising the prompt increase in background concentrations of the "nucleation factor" from one year to another. From nearly 0 particles

in 2011, with peaks during events, to a regular background around 500 particles in 2012 and a more noisy plot. Can the authors comment on it? Was the SMPS system modified in any way?

The SMPS system has not been modified. As the factor is a composite parameter derived from PMF, it is very hard to diagnose why the background should have changed. Given the nature of regional nucleation processes, a background of zero would be expected, and any background is likely to be an artefact of the data analysis process and a lack of totally clean separation of the factors. Why this should apparently change between years is most unclear.

15) From this study one can conclude that most of the pollution measured in London is local in origin (from about 20 Km around). Roughly, at least 60% of the PM10 mass is from local sources (traffic+diffuse urban+mineral-road dust+fuel-oil). Obviously, part of the secondary factor is locally produced. To what point these results are in line with previous ones? In many cases I have heard and I have read that a significant proportion of the London PM comes from mainland Europe. But this study just demonstrates the opposite. I never trusted totally that history about such a dominant role of transboundary PM pollution except for specific cases. With these results the authors have the opportunity to highlight from where the problem is coming.

We partially agree with the reviewer over this, but not entirely. We do not see a justification for the statement that most of the pollution measured in London is from about 20 km around. Taking PM_{10}, the polar plots tell us that the urban background and traffic factors are highest at low wind speeds which emphasises the local contribution. The polar plots, however, do not give a quantitative differentiation between emissions on different distance scales, just an indication of where the highest concentrations might arise from. Hence, stating that at least 60% of the PM_{10} mass is from local sources is not an inference that should be drawn from the data in this paper.

The statement that a significant proportion of the London PM comes from mainland Europe derives from the fact that numerous studies using local wind direction and air mass back trajectory data have found that the highest concentrations of particulate matter at sites in southern England are typically associated with air masses arriving from mainland Europe. It should not, however, be concluded from this observation that the largest contribution to the annual mean concentration arises from mainland Europe as winds in this sector are significantly less frequent than those from westerly sectors. Consequently, attributing PM_{10} concentrations to sources in London, the rest of the UK, and mainland Europe requires an entirely different kind of study from the one that we have conducted.

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

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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 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 PM_{10} 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 period 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 ¼ 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 ™ 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 PM_{10} 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 PM_{10} 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.

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 NO$_x$ are:

Correlation coefficients with BC:

Secondary	**0.64**
Urban Background	**0.78**
Traffic	**0.53**
Nucleation	**-0.01**

Similarly for the PM factors and NO$_x$:

[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?

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 PM_{10} 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 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 in China's largest port. Atmospheric Environment 64, 277-286.

These have been added.