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Interactive comment on "Sources and contributions of wood smoke during winter in London: assessing local and regional influences" *by* L. R. Crilley et al.

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RESPONSE TO REVIEWERS

REVIEWER #1 This paper deals with the study of carbonaceous aerosol in the great London area, by parallel sampling and measurement at 2 central London urban background sites, one on the top of a high tower, and at two rural neighbouring locations. Twenty four hours integrated aerosol samples were measured for carbon (OC, EC,

14C), levoglucosan and K+; BC was also measured semi continuously with multi-wavelength aethalometers. Using measured data and an array of source apportionment methodologies the carbonaceous component of the aerosols were quantitatively attributed to traffic emissions, biomass burning or gas to particle transformation. The paper contains a large collection of experimental information, which is thoroughly treated, compared and discussed, in order to justify and demonstrate the results and interpretations provided.

The paper is quite extended and long, and a bit tiresome to read and follow. Most of the sections in the paper are justified but I wonder if some of the sections could not be shortened to make the manuscript more linear and easy to read and interpret. I agree with most of the arguing and conclusions but some of the arguments are somehow repeated across sections. Also some of the discussion involves direct mentioning of figures and tables in an Annex section which in my opinion should be avoided (if figures or tables are part of the discussion they should be included in the main manuscript). As an example of sections that could be shortened without much loss of information is the discussion about the BT tower measurements. RESPONSE: We have carefully reviewed the paper and have tightened the text at a number of places throughout the paper. The main reductions were in Section 4, the long term measurements at BT tower. We have removed the section on the sources of gaseous species at BT and have shortened the section on sources of BC at BT, so that it now reads: "The two likely major sources of BC (traffic emissions and biomass burning) in urban areas have distinct diurnal cycles, and hence the mean diurnal variation of BC for the London sites for the long-term measurement period is shown in Figure 10. At each of the sites, the normalised BC diurnal cycles (Figure 12) demonstrated similar cycles to that of NOx and CO (Figure S9, Supporting Information), thus implicating a similar source, likely traffic emissions based on the high fraction of fossil EC found in Section 3.3.1. One feature of Figure 10 and Figure S9 is a one hour lag in the morning concentration peak at the BT tower compared to the ground-level site, possibly due to the time for the locally emitted pollutants to mix to the top of the tower (Despiau and Croci, 2007). The main

difference in the diurnal cycles shown in Figure 10 was in the absolute concentrations of BC at each site, which varied depending on the distance from the main local source, traffic emissions. BC concentrations at BT tower were, on average a factor of 2-3 times lower than at NK (Figure 10). EC has been observed to follow similar trends in previous measurements at BT tower and NK (Harrison et al., 2012b). Concentrations of BC at NK and BT tower were correlated (r2 of 0.6), indicating that the urban background as represented by the NK site was the main source of pollutants at the elevated BT site.

CPF analysis was applied to the observed BC concentrations at NK and the BT tower for the long-term measurements, to examine whether local or regional sources were the more significant contributors, with the results shown in Figure 11 and was similar to the plots for NOx and CO (Figure S10, Supporting Information). In Figure 11, similar trends were observed at NK and the BT tower, with the highest BC concentrations being associated with low wind speeds, which indicate that the major contributions were from local sources, likely vehicle emissions based on the observed diurnal cycles (Figure 12). Overall, it appears that the main influences on BC concentrations at BT tower are local ground-level sources (traffic emissions), with some influence from regional sources, investigated further in subsequent sections."

We have moved Figure S8 from the supplement to the main manuscript as it is referred to a number of times in the discussion. Comment 1: I agree with most of the discussion and interpretation of data and data treatment results which are well explained and well demonstrated. Only a few points were less clear for me. I have some doubts about the correctness of the approximation to OC SOA being only originated from biogenic VOCs gas to particle conversion. Although that may be reasonable in non-urban areas, at urban central areas contribution from fossil fuel burned VOCs may not be negligible, principally taking into account that Primary OC/ECff ratios used for primary ff source evaluations were on the lower range of bibliographic data, more characteristic of tunnel conditions, not taking into account the relatively rapid condensation processes (with VOCff ??) that follow these fresh emissions which bring urban OC/EC ratios to 0.7-0.8

values. In my opinion this approximation should be discussed a bit further, now that in some points of the paper it was used for explaining discrepancies between results (for example paragraph 5 in page 27484). Figure 4 shows that OC SOA average concentration estimated at NK is almost the double of average values calculated for the rural sites. If OC SOA is only from biogenic origin which is the explanation for this fact? Higher biogenic VOC precursor concentrations in the urban centre? More effective gas to particle transformation processes in the urban environment? Or more effective gas to particle conversion of fossil fuel VOCs? RESPONSE: The primary OC/EC ratio for vehicle emissions used for Figure 4 was 0.58, which is notably similar to recent ambient measurements in London of vehicle emissions (Pant et al., 2014), see also page 27478, line 19). Therefore we feel that we have used an OC/EC ratio that is reasonable estimate for vehicle emissions under ambient conditions for this study. Regarding the higher average concentration of OCSOA at NK compared to the rural sites, biogenic VOC precursors were not the likely sole source of this OC fraction as stated at page 27473 line 9-11: "The remaining OC is from biogenic, cooking (both primary and secondary) or other secondary sources that can include secondary organic aerosols formed from fossil fuel combustion and this fraction is referred to as OCSOA." Thus other sources such as cooking were also contributing and increased cooking emissions may have contributed to the observed higher OCSOA concentration as NK is an urban site. Furthermore, recent work in the UK found that SOA arising from fossil fuel emissions was a small fraction of the total particulate OC (Heal et al., 2011), and was thus likely a negligible fraction of the OCSOA (See page 27474, line 5-15), though may have also contributed to the observed higher OCSOA fraction concentration at NK compared to the rural sites. Comment 2: Another small point that is less clear for me- last sentence of paragraph 5, page 27482- I did not understand the explanation for the high correlation observed between BCtr and levoglucosan in NK. Do you mean that the dominance of traffic emissions degrade the capability of the evaluation methodology to separate between BCtr and BCwb? Another possible explanation could be that peak concentrations of pollutants (which mostly influence correlation values) are mostly de-

pendent of low dispersion periods of inversion. RESPONSE: We were implying that the methodology to separate BCtr and BCwb may have been influenced by the dominance of traffic emissions, as was reported by Harrison et al. (2013). We do not think that that meteorology was a major factor (e.g. boundary layer height), as then it would be expected that a high correlation between levoglucosan and BCtr would also have been observed at the rural sites, but this was not the case. Furthermore, levoglucosan concentrations were not found to be related to wind speed in this work (See Fig S4 and Page 27478, lines 1-5), which suggests that peak concentrations of levoglucosan were more related to changes in emissions rather than reduced dispersion. To clarify we have thus amended the text at line 7, page 27482 to read: "The high correlation observed between BCtr and levoglucosan at NK is surprising and may be due to dominance of traffic emissions in London affecting the ability of applied BC source apportionment methodology to separate sources." References: Harrison, R.M., Beddows, D.C.S., Jones, A.M., Calvo, A., Alves, C. and Pio, C.: An evaluation of some issues regarding the use of aethalometers to measure woodsmoke concentrations, Atmos. Environ., 80, 540-548, 2013.

Heal, M. R., Naysmith, P., Cook, G. T., Xu, S., Duran, T. R. and Harrison, R. M.: Application of 14C analyses to source apportionment of carbonaceous PM2.5 in the UK, Atmos. Environ., 45, 2341-2348, 2011.

Pant, P., Yin, J. and Harrison, R. M.: Sensitivity of a Chemical Mass Balance model to different molecular marker traffic source profiles, Atmos. Environ., 82, 238-249, 2014.

REVIEWER #2 General comments The paper assesses the influence of wood burning on the aerosol load in the London area as well as the contribution of other sources such as traffic. The analysis is based on an extensive dataset combined several independent approaches to the topic. The data are presented in great detail and the discussion of the data is very thorough, however, the manuscript would benefit from tightening some sections and making it overall more concise.

Specific comments – The abstract is fairly long and already contains a wealth of details. Think about shortening it.

RESPONSE: The abstract has been shortened to remove some of the details and now reads:

"Determining the contribution of wood smoke to air pollution in large cities such as London is becoming increasingly important due to the changing nature of domestic heating in urban areas. During winter, biomass burning emissions have been identified as a major cause of exceedences of European air quality limits. The aim of this work was to quantify the contribution of biomass burning in London to concentrations of PM2.5 and determine whether local emissions or regional contributions were the main source of biomass smoke. To achieve this, a number of biomass burning chemical tracers were analysed at a site within central London and two sites in surrounding rural areas. Concentrations of levoglucosan, elemental carbon (EC), organic carbon (OC) and K+ were generally well correlated across the three sites. At all the sites, biomass burning was found to be a source of OC and EC, with the largest contribution of EC from traffic emissions, while for OC the dominant fraction likely included contributions from secondary organic aerosols, primary biogenic and cooking sources. Source apportionment of the EC and OC was found to give reasonable estimation of the total carbon from non-fossil and fossil fuel sources based upon comparison with estimates derived from 14C analysis. Aethalometer derived black carbon data were also apportioned into the contributions from biomass burning and traffic, and showed similar trends to that observed for EC. Mean wood smoke mass at the sites was estimated to range from 0.78-1.0 μg m-3 during the campaign in January-February 2012. Measurements on a 160 metre tower in London suggested a similar ratio of brown to black carbon (reflecting wood burning and traffic respectively) in regional and London air. Peaks in the levoglucosan and K+ concentrations were observed to coincide with low ambient temperature, consistent with domestic heating as a major contributing local source in London. Overall, the source of biomass smoke in London was concluded to be a background regional

source overlaid by contributions from local domestic burning emissions. This could have implications when considering future emission control strategies during winter and may be the focus of future work in order to better determine the contributing local sources."

– The text uses a lot of acronyms and abbreviations, which makes some paragraphs hard to read. I suggest to carefully check if all of them are needed, even though they may be commonly used in the literature. Some of them are not used throughout the full text but only once or twice in one paragraph, those could be omitted.

RESPONSE: We have carefully considered all of the acronyms used and have removed the following as they are only used within one paragraph. AMS for aerosol mass spectrometers in the introduction MAC for mass absorption cross-section in Section 2.4.2

– Harmonize the spelling of 2/7 wavelength, 7-wavelength, two/seven wavelength, Aethalometer/aethalometer.

RESPONSE: We have harmonised the spelling to 2 and 7 wavelength Aethalometer throughout.

– The presentation of the measurement sites and the descriptions which measurement took place when and where are very difficult to follow. In the abstract three sites are mentioned, but section 2.1. lists four sites, whereas figure 1 shows six locations without further explanation in the caption, e.g. which ones are lidar sites and that the lidar had been moved. Not being familiar with the sites I found it difficult to follow the discussion, especially since they are rather different regarding the measurement setup.

RESPONSE: To improve presentation of the measurements sites we have altered the text for Section 2.1 to read to provide an overview of the measurement sites.

"The measurements for this paper were conducted as a part of the NERC-funded ClearfLo project (Clean Air for London, www.clearflo.ac.uk), to investigate boundary

layer pollution in London. An overview of the project can be found in Bohnenstengel et al. (2014). Four sites were selected for chemical sampling in this particular study; an urban background and elevated site within London and at two sites in surrounding rural areas, with details summarised in the following sections. Site locations are shown in Figure 1. Furthermore, additional measurements with a scanning Doppler lidar were also conducted for this study at sites within central London as indicated in Figure 1. Details on the lidar measurements performed can be found in Section 2.2.3."

The caption for Figure 1 has also been altered to highlight which measurements were conducted and when.

"Figure 1: Map of the study area with the sampling sites (indicated with circles) and lidar locations (designated with triangles) shown. Note for the start of the winter IOP the lidar operated from the Westminster City Council (WCC) building and then moved to Imperial College London (ICL). During the summer IOP the lidar operated from the North Kensington (NK) site"

Full description of the instrumentation deployed for chemical sampling at each site is given in Section 2.2, and the text in Section 2.1.1 has been changed to clarify this: "The North Kensington (NK) site is located within a school grounds in a residential area (51°31'15" N, 0°12'49" W) and is classified as urban background. Central London lies 7 km to the east and a major road is located approximately 500 m to the south of the site. A more complete description of the site and air pollution climate is available in Bigi and Harrison (2010); the site is considered representative of the background air quality within much of London. This site is part of the Automatic Urban and Rural Network (AURN), with further details on the instrumentation and sampling procedures available in Section 2.2."

Introduction – p 27463 L20 and L 21: replace "2" with "two" – p 27464 L6: a reference to Wagener et al 2011 and/or 2012 should be included here for the measurements in Berlin – p 27464 L9: matching closing bracket is missing – p 27464 L13: . is missing

RESPONSE: The above errors have been fixed and reference to Wagener et al. (2012) added where recommended. Version IMethod – p 27464 L2: why "METHOD" is capitalized? – p 27471 L23: wrong numbering

RESPONSE: The above two errors have been fixed.

– p 27472 L10: What was the time resolution of the meteorological data? 1hour averages?

RESPONSE: The time resolution of the meteorological data from the Met office is 1 hour intervals, and to clarify this we have added the following text in Section 2.2.3, page 27468

“The meteorological data obtained from the Met Office had a 1 hour time resolution”

– p 27473 L4 ff: what do you mean by "the ratio of levoglucosan to OC for biomass burning"? an emission factor? same for "OC/EC ratio for biomass burning / for vehicle emissions" – which values were used?

RESPONSE: The ratios used were ratios from previous measurements. Thus we have changed the text to read in order to clarify this:

“In this method, the primary OC concentration from biomass burning (OCbb) is estimated based upon the concentration of levoglucosan and the ratio of levoglucosan to OC measured for biomass burning emissions from the literature. The EC concentration from biomass burning (ECbb) is calculated from the OCbb using previously observed OC/EC ratio for biomass burning emissions and the remainder of EC is apportioned to EC from fossil fuel emissions (ECff). Primary OC from fossil fuel emissions (OCff) is estimated using previously observed OC/EC ratios for vehicle emissions.”

– would there be a significant influence of ethanol in the fuel on the fossil/non-fossil in partitioning?

RESPONSE: In the UK, ethanol blended fuels were not widely available during the

sampling period, and therefore was not likely to have a significant influence on fossil/non-fossil partitioning.

– p 27474 L20: change "suffers" to "suffer" Results – p 27477 L1–2: term "references therein" is doubled

RESPONSE: The above two errors have been fixed. – p 27477 L15/16: The wording here implies "regional" to refer to mainland Europe, however, later in text regional seems to refer to the closer surroundings of London. Please clarify.

RESPONSE: In this section of the text we are suggesting that the regional sources were from mainland Europe. We are not sure to where in text the reviewer is referring to when we suggest regional sources were surrounding areas of London. While the term ‘regional sources’ was used as a general term to indicate non-local sources in some instances where appropriate, as we used the term regional sources to refer to sources that were not local and from further afield. Overall the results pointed to the long-range transport of emissions from mainland Europe as the source, as described at page 27484, line 23:

“This therefore suggests that the major source was regional biomass burning at all of the sites. Furthermore, the observed high correlations between levoglucosan and OC at all the sites (r2 of 0.71–0.88) along with the majority of the OC being found to be secondary in origin in Sect. 3.3.1, suggests a common regional source, possibly including a contribution from mainland Europe (Charron et al., 2013)”

And at page 27486, line 26:

“A regional influence was also evident at Detling, with the highest contributions to the BC concentrations associated with high wind speeds from the east, in agreement with that observed at Harwell and probably influenced by advection from mainland Europe”

– p 27480: I suggest to include a graphical represention of the values in table 4 with the supplementary material

RESPONSE: We have included a figure in the supplement: Figure S6: Daily average percentage of non-fossil total carbon fraction (TCnf) and the total carbon fossil fraction (TCff) as calculated using different source ratios. For details on the source ratios used, refer to the case ID given in Table 4.

– p 27480 L24: What do you mean by "diurnal trends"? An evolution of the diurnal cycle with time? My reading is that you talk about the average diurnal cycles, using of the word "trend" in that context is misleading as it is commonly used for interpretation of time series (applies to other parts of the text as well, in particular the conclusions section)

RESPONSE: When referring to diurnal trends we were referring to average diurnal cycle. Thus to avoid confusion we have swapped the term diurnal trend for diurnal cycle, in this section of the text and elsewhere in the manuscript. For example, the text on page 27480, line 21-26 now reads;

"Diurnal cycles of the BC concentrations at Harwell, NK and Detling during the winter campaign measured by 2W AE are shown in Figure 5, with the cycles for the 7W AE given in Figure S6, Supporting Information. Similar cycles and concentrations were observed by the 2W AE and 7W AE at NK while at Detling the diurnal cycles were slightly different, which may have been due the instruments not being co-located. The observed diurnal cycles indicate that traffic was the likely main source of BC at NK and Detling, as the classic bimodal cycle coinciding with peak traffic times was observed at both sites." Discussion Paper – p 27481 L24: I do not agree that there is noticeable morning peak at HAR, the feature is rather weak.

RESPONSE: We agree that there is no morning peak in the UVPM diurnal cycle at Harwell, with a noticeable morning peak in the UVPM diurnal cycle only observed at North Kensington, as we mentioned page 27481 line 19:

"In addition to the evening concentration peak observed as expected in the UVPM cycle at NK for both instruments, there was also a peak at around 07.00, suggesting a traffic

influence in the observed UVPM concentrations at NK."

– p 27481: could you comment on the difference in the cycles calculated from the 2wave and the 7wave aethalometer? what is the cause of the higher values for column A and the wider confidence interval?

RESPONSE: We do not agree that there is a significant difference in UVPM concentration as determined by the 2 and 7 wavelength Aethalometer, with the similar peak and minimum concentrations observed at NK and Detling for the two instruments. The wider confidence interval may be due to the different sampling intervals used for the two instruments.

– p 27481 L21: "07.00" should read "7:00" – p 27482 L18: missing "than" – p 27483 L17 and L20: comma missing following "However"

RESPONSE: The above three errors have been fixed.

– p 27484 L2–3: wording unclear

RESPONSE: The text has been changed to:

"From Table 5, the closest match to for ECbb to the BCwb percentage contributions was found using the mean levoglucosan/OC and OC/ECveh and minimum OC/ECbb (Table 4, F). However, this combination of source ratios did not match 14C source apportionment results as well as other combinations (See Table 4)."

– p 27484 L6: missing "a"

RESPONSE: Fixed.

– p 27487 L4–22: This paragraphs refers in detail to figures from the supplement. In my opinion the supplement should only contain material which is not discussed in detail in the text but should be a collection of additional graphs or tables with the main text being self-consistent.

RESPONSE: Figure S8 has been moved from the supplement to the main text.

Long-term Measurements – p 27488: Although it was mentioned before, it would be worthwhile to remind the reader of the location altitude and the tower height in the first paragraph.

RESPONSE: We have added the sampling height at the BT tower at page 27487, line 26, as shown below:

“One method that has been used previously is to sample from an elevated site such as the BT tower (160 m a.g.l.) (Harrison et al., 2012b).”

– p 27488 L13–18: Is there an explanation for the evening peak being lower relative to the mean at BT while the morning peak is at almost the same level for both sites?

RESPONSE: We are not sure but may be due to the higher boundary layer height, particularly during the warmer months during the evening, resulting in increased dispersion.

Figures In several graphs axes are labelled "concentration" only. Understanding what is plotted would be facilitated by specifying the compound in the axis labels. Same e. g. with "normalised level" in figures S9 and S10. Labels and annotation in many figures use a too small font size.

RESPONSE: For Figures 5, 6, 12, S7 we have added the species to the axis label and increased the font size where required.

– Figure 1: differentiate in the figure caption and by symbol which sites hosted the lidar and a note about the move of the lidar

RESPONSE: The lidar sampling locations have now been differentiated from the sampling sites in the map, with text added on the changing locations in the caption:

“Figure 1: Map of the study area with the sampling sites (indicated with circles) and lidar locations (designated with triangles) shown. Note for the start of the winter IOP

the lidar operated from the Westminster City Council (WCC) building and then moved to Imperial College London (ICL). During the summer IOP the lidar operated from the North Kensington (NK) site”

– Figure 6: brackets in figure caption do not match

RESPONSE: We have fixed the caption to read:

“Figure 6: Mean diurnal cycles of UVPM during the winter campaign as determined by 2W AE (Column A) and 7W AE (Column B) for the sites where the data was available. Shaded areas indicate the 95% confidence intervals. In the key HAR and DET represents Harwell and Detling, respectively.”

– Figure 14: enhance figure size

RESPONSE: We have increased the size of these plots to fill out a page.

– Figures S1, S2, S3: label "m sôĂĂĂ1" did not come out properly

RESPONSE: A formatting issue we have tried to fix this but just in case have added to the caption that the wind speed is in m s-1.

– Figures S1, S12, S13: annotation is cut off

RESPONSE: Again a formatting issue not present in the figures we submitted that we have addressed and will check in final version.

– Fig S18 – S22: "NOx concentration (ppb)" – Quantities given in ppb are not concentrations but mixing ratios.

RESPONSE: The captions have been changed to “NOx mixing ratio (ppb)”.

References Bigi, A. and Harrison, R. M.: Analysis of the air pollution climate at a central urban background site, Atmospheric Environment, 44, 2004-2012, http://dx.doi.org/10.1016/j.atmosenv.2010.02.028, 2010.

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