We thank all four referees for their comments and suggestions. We would like to provide the following responses, where referee comments are in normal font and our responses are italicised.

Anonymous Referee #1

1) General comment: The fact that two SFOA factors are identifiable, arguably due to different combustion efficiencies is very important, in this reviewer's opinion since emissions from solid fuel combustion sources are too frequently lumped together in a single category. I would argue that this should be stressed in the paper where appropriate, since they represent two different sources (i.e., solid fuel with different combustion efficiencies).
    a. *In Section 4.2, particularly paragraph 2 on page 20859 and the end of the second paragraph on page 20860, we clearly discuss that it is not possible to distinguish between two different/discrete sources but rather we discuss the possibility that the two SFOA factors represent a continuum of end members with source profiles that vary.*
2) Abstract, page 20847, lines 13-16: From the remainder of the paper, it is evident that SFOA1 is largely attributed to southern sources and SFOA2 is largely attributed to easterly or westerly air masses, but I find it somewhat confusing with the directional clarifier in the abstract. I would recommend removing the text "in the south" and "in the east and west" such that this sentences reads "The split between the two factors is likely governed…where SFOA1 best represents more efficient combustion and SFOA2 represents less efficient combustion". This should also be addressed in the Conclusion section.
    a. *This has been addressed in the revised manuscript as suggested.*
3) As a follow-up comment, the authors, at the top of page 20858, state that the meteorological data "do not provide representative insight in to spatial differences in the SFOA concentrations". Further, the SFOA2 factor also appears to extend to the south. It is not clear why a discussion of the directional measurements is included when the authors appear to discredit their robustness.
    a. *The suitability of the meteorological data from North Kensington are questioned due to the influence of the surrounding large buildings thus is used to justify the use of meteorological data from Heathrow airport as opposed to the local wind data. The analysis using meteorological data show that at low wind speeds there is an area source. However, this analysis does not allow differentiation of the source(s) of the two SFOA factors.*
4) Section 4.3, pages 20860-20861: It is unclear to this reviewer why "Role of fuel type" and "Role of burn conditions" are subsections under "Role of atmospheric processing". To me, it would make sense to re-organize these sections (in their current order) as Sections 4.2.1, 4.2.2, and 4.2.3 since they all relate back to Section 4.2, Investigating the behavior of SFOA.
    a. *This has been amended in the revised manuscript.*
5) Page 20864, lines 1-5: I find these two sentences to be confusing, since in the first sentence, the authors -state that they could not identify SFOA from the data, yet SFOA are still significant with respect to total OA. If the SFOA do not exist in the data set, how certain can you be that the factor is significant? Perhaps I am simply misinterpreting the authors' meaning, in which case I would request that these sentences be re-written for clarification.
    a. *From the year round data it was possible to derive the SFOA factor as two winter periods were covered by the measurements when solid fuel burning activities are likely to be prominent thus enabling the PMF algorithm to detect SFOA as a factor throughout the year. However, SFOA is a small contributor during the summer thus when this period is looked at in isolation in both the cToF-AMS and HR-ToF-AMS datasets, SFOA is not identified. The term "significant" has been removed from this sentence in the revised manuscript to become: "However, as SFOA is still found to*

contribute to the total organic mass during this period it could be that this factor represents a different activity to domestic space heating during the winter."

Anonymous Referee #2

Comments:

1) In section 3.1, the use of CO and NOx as gas-phase tracers for comparison to the PMF solution is discussed. The authors state that these two gases come from more than one source, which is why they were compared to summed PMF solutions. Did the authors consider source apportionment on the gases? If not, explain why separating the gas-phase signals from different sources was not carried out.
    a. *This manuscript focuses on particulates, using additional data such as gas-phase tracers to aid the interpretation of the aerosol data. Including both gas-phase and particle-phase data in PMF analysis is a substantial amount of work (e.g. Slowik et al., 2009 –doi:10.5194/acp-10-1969-2010; Crippa et al., 2013 – doi: 10.5194/acp-13-8411-2013) and is outside the scope of this paper.*

2) In section 4, the 'role of fuel type' and 'role of burn conditions' are under the heading of 'role of atmospheric processing'. It would be better to have the sub sections as their own sections.
    a. *This has been addressed in the manuscript as detailed in 4a from Referee #1.*

Minor revisions:

1) Page 20854, Line 17-18: When discussing OOA1 and 2, also include the SVOOA and LVOOA terms since they are often used in other AMS publications.
    a. *In the manuscript, an additional sentence will be added in line 18 to say "In the literature, OOA1 and OOA2 are often referred to as low volatility oxygenated organic aerosol (LV-OOA) and semi-volatile oxygenated organic aerosol (SV-OOA) respectively."*
2) Page 20860, Line 22: There is a typo. The reviewer thinks the author indented to say, "…well as f44 compared to SFOA2.", instead of SFOA1.
    a. *This has been amended accordingly in the manuscript.*
3) Figure 1 caption: The reviewer realizes the PMF solution is in a complimentary paper; however, it would be good to include the fraction of mass that is remaining in the residuals. If it is zero, please state it in the figure caption, or alternatively, guide the reader to the complimentary paper.
    a. *The caption for Figure 1 has been edited to include the relevant information regarding the residual: "*Figure 1. **(a)** Time series of the 5-factor-PMF solution from the HR-ToF-AMS PMF. **(b)** Average fractional contribution to the total submicron organic aerosol mass, where the residual accounts for 1.24% of the mass (also see Figure S27 in Section 5.1.2 in the supplementary material to Young et al. (2014), the complimentary paper). **(c)** Median diurnal profiles for each of the 5 factors and the diurnal profile of the difference between the two SFOA factors (SFOA2-SFOA1, inset)."
4) Figure 2 caption: The author discusses south and east-west patterns in the paper and in figure 3. For clarify, explain the grid-lines in the wind-direction sub plot in the caption of figure 2.
    a. *The caption for Figure 2 will be changed to "Time series of SFOA1 and SFOA2 concentration and meteorological data from Heathrow. The horizontal grid lines in the wind direction sub plot from top to bottom represent westerly, southerly, and easterly wind directions, respectively."*

5) Figure 4: For quick reference, include the O:C in the SFOA 1 and SFOA 2 sub plots.
    a. *The O:C ratios have been included in the subplots of figure 4 as suggested.*

R. M. Healy (Referee)

1) Title: perhaps "Investigating a two-component model....."
    a. *The authors agree with this comment and will change the title accordingly.*
2) Page 20855: Line 9. This event could be removed as it may be leading to overrepresentation of HOA at this site. This would lead to a less representative apportionment of residential urban background conditions.
    a. *The authors believe that this comment is regarding line 9 on page 20856. Having looked back through the records of the experiment, there is no record of a single vehicle being on site for the 2-hour period that this event occurs over so the event cannot be justifiably removed from the data. However, a vehicle could still be the cause of the large concentration but we cannot reliably say that it was. This sentence therefore has been reworded in the revised manuscript to be "As HOA is related to traffic emissions, this event could be the result of a vehicle parked close to the site with its engine running."*
3) Page 20859, line 15: Pearson's r values for comparison of the two SFOA factors with a BBOA reference mass spectrum are discussed. The SFOA factors could presumably also be compared with more recently published reference spectra for SFOA and coal combustion that the authors refer to in the manuscript. Was this investigated?
    a. *There are not any more recently published reference spectra on the database. However, some spectra from the literature have been obtained and compared. For example both SFOA1 and SFOA2 were compared with the coal combustion factor identified in Cork, Ireland (Dall'Osto et al., 2013) with Pearson's r of 0.60 and 0.65, respectively. In addition, SFOA1 and SFOA2 were compared with the SFOA factor identified in London, UK during the REPARTEE project (Allan et al., 2010), with Pearson's r values of 0.88 and 0.92, respectively. However, due to differences in the operation of the instruments, location of the measurements as well as differences in meteorology, fuel types and conditions and more (e.g. Crippa et al., 2013 – doi: 10.5194/acp-13-961-2013) , comparisons of MS should only be used qualitatively or used with caution. These results have therefore not been included in the manuscript.*
4) Page 20859, Line 24-29: Rephrase these sentences replacing the "missing" descriptions, the section is difficult to follow.
    a. *These lines could be amended in the manuscript to: "For example, the peaks above the line are primarily composed of oxidised hydrocarbons at m/z 43, 44, 57, and 60 and the peaks below the line are primarily composed of reduced hydrocarbons. Prominent peaks in the SFOA1 MS such as m/z 41 and 55 are not present above the line in the difference MS and peaks at m/z 43 and 57 in the SFOA2 MS are not present below the line in the difference MS.". However, reviewer #4 also makes a similar comment so this paragraph has been amended to address both these comments. Please see response to reviewer 4's comment below.*
5) Page 20860, Line 20-22, one of these should be SFOA2.
    a. *The authors agree and has been addressed as part of minor comment 2 from referee #2.*
6) Page 20862, line 11: "different phases of combustion occurring under similar conditions"? Please clarify.
    a. *The authors mean that the same burner type could be used, for example, but the combustion phase is different such as flaming or smouldering.*

        i. *The following sentence has been added into the manuscript, line 11 on page 20862: This could mean that the same burner type is being used, for example, but the combustion phase is different such as flaming or smouldering burns.*

7) Page 20862, line 27: "low signals at m/z 60 and 73" But these are higher for SFOA1 than for SFOA2?
    a. *The meaning of line 27 was that for the SFOA1 MS the signals of m/z 60 and 73 are relatively low compared to that of m/z 44 rather than compared to the signals in the MS of SFOA2. As discussed on page 20860, line 11, the type and phase of combustion can also affect the signal at m/z 44 and is being shown to possibly be the case here.*
        i. *The sentence has been edited in the revised manuscript to "This is in keeping with the mass spectral profile of SFOA1 exhibiting a large signal at m/z 44, indicative of fast combustion which results in the conversion of organic matter to $CO_2(g)$."*
        ii. *The following has been added to line 3 on page 20863: "Similar to the findings of Weimer et al. (2008), levoglucosan signals in the MS of SFOA1 i.e. m/zs 60 and 73, are enhanced compared to those in the MS of SFOA2 suggesting SFOA1 represents higher temperature burns as it is expected that higher thermal breakdown occurs at higher temperatures in flaming combustion (Weimer et al., 2008)." Please also see the author's response to Reviewer #4, comment 5.*
8) Section 4.3.2: At the end of this section, reinforce that potentially different burn efficiency for SFOA1 and SFOA2 is the point here.
    a. *The following has been added to line 3, page 20863 as suggested: "In summary, differences in burn efficiency are potentially governing the split between the two SFOA factors."*
9) Can the authors provide an explanation as to why one residential area to the south would be characterised by generally different burn conditions than areas to the east and west? It could be expected that large residential areas in any part of London would contain the same cross-section of less efficient and more efficient burners. This is a central point to the manuscript and it would be nice to expand on it. Is there a greater install base for more efficient burners in different residential areas? Are these data available?
    a. *Inventory data is either not reliable or available to enable these questions to be answered. Data on the emissions of solid fuel organic aerosols is poor and involves a substantial amount of speculation. An additional comment about the poor quality of inventory data in this area has been added to the manuscript at the end of Section 4.2.3: "Inventory data for the emissions from solid fuel combustion is of poor quality and other data such as regarding burner installations for example, are not available thus cause for differences in burn efficiency can only be speculated on."*
10) Page 20863, line 14: "mass resolution"
    a. *Line 14 will be changed to the following: "Only one SFOA factor was derived from PMF analysis of the cToF-AMS data, due to the lower mass resolution of this version of the instrument compared to the HR-ToF-AMS where different ions at the same nominal m/z can be distinguished."*
11) Page 20863, line 18: perhaps replace SOA with OOA for consistency
    a. *The line has been amended in the manuscript to read "If significantly aged, some SFOA may be apportioned to SOA (OOA) by PMF due to the chemical similarity."*
12) Page 20865, line 22: "Changes in economy" is quite broad. Perhaps a statement on a potential increase in oil or gas prices relative to solid fuel prices would be more appropriate
    a. *This line has been amended as suggested to: "Changes in the economy, including increases in fossil fuel prices, may lead to an increase in the contribution of SFOA to*

the total NR-PM_1 aerosol burden, which could have significant influences in forming future air quality policies and mitigation strategies due to the associations between fine combustion aerosols and adverse health, air quality, and climate effects.

13) Fig. 3 caption: Remove clause after (Carslaw, 2013). "openeair" typo?
    a. *The clause has been removed in the manuscript as suggested and the typo has been addressed – should be openair.*
14) Fig. 5 Typo in "Cubison" on graph
    a. The typo will be addressed in the figure in the manuscript. The year of the publication is also incorrect. The figure has been edited in the revised manuscript.

Anonymous Referee #4

Major comments:

1. The authors use the term "SFOA" to describe solid-fuel OA, emphasizing that other fuels such as coal contribute significantly (31% in the case of coal) to PM1 in the UK. However, both of the SFOA mass spectra presented contain significant signals at *m/z* 60. This ion is a tracer for anhydrosugars such as levoglucosan, which are not expected to be abundant in coal-combustion smoke (Oros and Simoneit, 2000; Wang et al., 2014). So, is (a) the OA contribution of coal combustion being apportioned by PMF into the other factors, (b) is coal-combustion OA negligible, or (c) is the coal used in London generating a surprisingly high amount of anhydrosugars (or some other confounding molecule)? If (a) is true, the factors presented here are not SFOA factors. If (b) is true, there is no need to call these factors SFOA rather than BBOA (biomass burning OA). If (c) is true, then more data would be needed to demonstrate it.
    a. *The authors use the term "SFOA" to cover contributions from all possible sources to this mass spectral signature, which could include biomass burning and coal combustion. We have a quantitative marker for the integrated sum of emissions from solid fuel combustion where the anhydrous sugar marker is greater from wood but the source(s) of the other markers in the mass spectrum are not known and are not necessarily from wood. Even though the SFOA factors contain levoglucosan, this does not mean that coal combustion generates levoglucosan. The mass spectra are therefore convolved together and the organic aerosols temporally co-vary. When the concentration of SFOA is compared to that of a wood smoke factor derived from Chemical Mass Balance (CMB) methods (see Yin et al., 2014) as well as wood smoke plus coal, the correlation is improved for the latter with an increased* r^2 *when summed, which indicates that the SFOA factors derived here contain a coal contribution thus implying that the umbrella term of solid fuel OA is more appropriate for this study than BBOA. This is discussed in the manuscript on page 20861, lines 15-21. The contributions from other fuels are to PM1 emissions from domestic fuel burning as mentioned in the manuscript, rather than contributions to PM1 as mentioned in the reviewer's comment.*

2. The fact that the SFOA/BBOA generated by an entire city may not be adequately represented by a single mass spectrum, or even by two mass spectra, is undoubtedly a major challenge for an atmospheric study such as this one. The authors make a number of clear statements to this effect, but there are still some points at which the discussion might be reworded to reflect this fact.

   First, since SFOA/BBOA is not a single chemical component nor a single source, statements such as "the mass spectral profile of SFOA is highly variable, the findings from this study may have implications for improving future source apportionment and factor analyses" (Abstract, line 18 p 20847) would be much more accurate if they were reworded along the lines of "the SFOA profiles reported in this study may provide a basis for the identification of SFOA in other environments". This also applies to the conclusion (p 20865, lines 4-11).

a. *As mentioned in the previous response, SFOA is a continuum of different sources and range of behaviours. The authors did not intentionally refer to SFOA as distinct profiles and do not agree with the reviewer's suggested improvement sentence so the relevant sentences will not be changed in the manuscript.*

Second, the discussion on page 20859 lines 19-29 and page 20860 lines 5-7 does not make sense to me. The language of page 20859 lines 19-29 suggests that mass-spectral differences between the two PMF factors were not expected (probably unintentionally). The discussion of page 20860 lines 5-7 suggests that the two factors represent the endpoints of a continuum, by analogy to OOA (presumably referring to the continuum of volatilities or oxygenation observed for OOA). But this analogy breaks down because SFOA/BBOA composition may change with burn conditions, fuel type, age, and dilution. Over which dimension should SFOA1 and SFOA2 represent endpoints? And what evidence is there that they are endpoints, rather than simply two of the most-variable contributors to London OA during this particular period?

a. *The comment regarding the language in lines 19-29 on page 20859 is in addition to comment 4 from R. Healy. The authors have therefore removed most of this paragraph from the main manuscript and include in the caption of figure 4. This section will become "The difference in the mass spectra (MS) of the two factors highlights the variation within SFOA, which likely led to the derivation of a split factor by the PMF algorithm. The main chemical differences between the two factors are shown in the difference MS in Fig. 4, where the chemical groups appear to be affected differently by what is driving the split between the factors. Therefore, to better understand these differences the roles of atmospheric processing, fuel type, and burn conditions, including burn phase, in varying the MS of SFOA and their influences in governing the split into two factors are investigated in the following sections."*
b. *Regarding the comment about possibly unintentionally not expecting mass-spectral differences between the two PMF factors, the authors would like to mention that from the literature, there was no precedence to expect two factors as previous studies have only derived one SFOA factor (e.g. Allan et al., 2010).*
c. *The two SFOA factors described using the OOA analogy are not discrete end members but rather extreme mass spectral profiles, which are expected to vary in between. The authors use the OOA analogy as it is the one that the AMS community understands (see Morgan et al., 2010 and Ng et al., 2010).*

Third, PMF factors are occasionally discussed as though they are real entities, existing independently of the analysis itself. On page 20854, line 14, the statement "it is not always possible to separate OOA in to its two subtypes if ambient temperature and photochemistry are not sufficiently variable such as in the winter" is strange, because if ambient temperature is constant and photochemistry is negligible, then there is no meaning to the concept of two OOA subtypes anyway.

d. *The authors agree with the reviewer's comment that OOA should not be discussed as a real entity in the atmosphere but in the example sentence given by the reviewer OOA is not treated as a real entity. This sentence was to mean that is the variables don't vary then the two factors cannot be unpicked. The authors have removed* *'photo' from the term 'photochemistry' in the sentence given as the environment is not constant but the chemistry is definitely there throughout the year.*

3. The reasoning behind the correlations of SFOA with NOx and CO appears to be circular. First, it is noted that multiple NOx and CO sources exist, including solid fuel burning and vehicle engines. Then, it is noted that the sum of SFOA1 and SFOA2 correlates better with NOx and CO than the separate factors. Why is this noteworthy? One would also expect that summing all NOx/CO sources would improve the correlation even more (neglecting differences in atmospheric lifetime).

   The multiple linear regression fit also reads as though it follows circular reasoning. First, the fit is performed so that the regression coefficients explain as much of the variability in NOx or CO as possible. Then, the fitted function is correlated with NOx or CO. Since the goal of the fit was to explain more of the NOx or CO variability in the first place, then the fitted function will necessarily be better correlated with those parameters.

   Also, it is not clear exactly how the fit was performed. Here, the fit is described as a "multi-linear regression fit", citing Allan et al. (2010), who described the same analysis as a "bilinear fit". The written function indicates that multiple linear regression was performed with HOA and SFOA as predictors, but does not explicitly show how CO or NOx were used. I presume that either CO or NOx were used as the response variables? If this is the case, please use more standard regression language (multiple linear regression, predictor variables, response variable). Finally, the term "arbitrary" must be removed when describing the fit coefficients A and B; these coefficients are not arbitrary if their values were determined by least-squares. Perhaps it was the choice of a linear regression function that was arbitrary.

   In my opinion, this entire section would be made clearer by simply stating the correlations between SFOA1 and SFOA2 with the gas tracers, and moving on to the final paragraph (Section 3.1), which states that both solutions could be valid (as expected, for such a PMF analysis) and states that the 5-factor solution was chosen based on physical meaning. The only value added by the regression seems to be that the assumed sources of SFOA1 and SFOA2 were allowed to have different OA/CO emission ratios.

   - *The authors feel that the reasons as to why this test was being run were not conveyed in the manuscript. The primary reason was to compare the Pearson's r values as these cannot be obtained from a multi-linear regression fit. The NOx or CO concentrations were estimated using the fit values from the multi-linear regression and subsequently correlated with the measured NOx or CO and fit to obtain 'r'. The goal of maximising 'r' is also what this test was used for in Allan et al. (2010) with the production of grid-scale emission factors being a secondary product of this. An additional explanation has been included in the revised manuscript.*
   - *The terms suggested by the reviewer have been used to describe the variables in the revised manuscript, where HOA and SFOA are the independent (predictor) variables and CO and NOx are the dependent (response) variables.*
   - *The term "arbitrary" has been removed from the manuscript as suggested.*
   - *The paragraph beginning on line 7 on page 20855 now reads: "where [HOA] and [SFOA] are the predictor variables and are concentrations of the HOA and SFOA PMF factors. A, B, and C are fitting parameters optimised to minimise the squared difference between f (HOA, SFOA) and NO_x or CO, where NO_x and CO are the response variables. This multi-linear regression fit was performed on the HOA and SFOA factors from the 4-factor solution and the HOA and combined SFOA factors*

from the 5-factor solution. Using the multi-linear regression fit, the CO and NOx concentrations were estimated and subsequently compared to the measured trace gas concentrations. A Pearson's r value could then be obtained and compared to the r values from the linear regression."

- *An additional multi-linear regression has been performed whereby SFOA1 and SFOA2 are separate independent variables with the following Pearson's r values obtained:*
    - *0.79 for CO and 0.74 for NO_x*
    - *The Pearson's r for CO is slightly greater when SFOA1 and SFOA2 are not summed and is also greater than that for when the fit was performed on the 4-factor solution. The Pearson's r for NO_x is the same as that for the 4-factor solution but is slightly greater than that for when the two SFOA factors are combined from the 5-factor solution.*
    - *The following text has been added to line 16 on page 20855 and the corresponding values have been added to Table 1: "An additional multi-linear regression fit was performed with SFOA1 and SFOA2 as separate predictor variables, similar to the method used by Liu et al. (2014). The Pearson's r values are shown in Table 1. The correlations for both CO and NO_x are improved when the two factors are included as separate variables."*

4. "SFOA1 and SFOA2 differ by degree of oxygenation and therefore age" on line 10, page 20860 is not a sound argument as different fuels are known to produce BBOA of different degrees of oxygenation (e.g. Schneider et al., 2006, and the introduction of Section 4.3.2). Also, the estimated O:C ratios should include a citation of the method used, and should note the limitation that the AMS fragmentation table may not be valid for these POA samples.
    a. *The authors agree with the reviewer about the sentence on line 10 of page 20860 and have subsequently edited the sentence to be "SFOA1 and SFOA2 differ by degree of oxygenation, which could hypothetically be a function of age". This is a hypothesis regarding the split between the two SFOA factors, which the authors then go on to rule out.*
    b. *The elemental analysis software is detailed in Section 4.1 of the complimentary paper (Young et al., 2014 - doi:10.5194/acpd-14-18739-2014). The relevant papers describing the algorithms used in the analysis software have been cited in the revised manuscript in line 9 on page 20860: "Here, the O:C ratio is based on the algorithms described in Aiken et al. (2007, 2008) and is 0.41 for SFOA1, whereas for SFOA2 it is 0.15 and 0.53 for OOA."*

5. p 20863, line 1: Why should the higher temperatures of flaming combustion produce more-functionalized OA? Why should lower temperatures give more alkanes? A citation would help here.
    a. *Higher temperatures would increase oxygenation whereby flaming burns would increase fragmentation and thus more functionalization, which has been observed in experiments investigating combustion chemistry (e.g. Weimer et al., 2008). However, the results obtained in this study are not directly comparable to those of Weimer et al. (2008), evident in the very different mass spectra that were obtained, as different methods were used to obtain the mass spectra in the two studies.*
        i. *In the revised manuscript, the following has been added to line 3, page 20852: "Furthermore, different methods are used in these studies, resulting in very different mass spectra being obtained. This could be due to different fuel types, burner, or changes to the profile that occur between the source and the receptor, thus mass spectra are not necessarily directly comparable."*

ii. *The following has been added to line 3 on page 20863 in the revised manuscript: "Similar to the findings of Weimer et al. (2008), levoglucosan signals in the MS of SFOA1 i.e. m/zs 60 and 73, are enhanced compared to those in the MS of SFOA2 suggesting SFOA1 represents higher temperature burns as it is expected that higher thermal breakdown occurs at higher temperatures in flaming combustion (Weimer et al., 2008)."*

iii. *Finally, the term "alkanes" has been replaced with "aliphatic chains" in the revised manuscript on line 2, page 20863.*

6. p 20864, lines 1-7: If the sources of SFOA were drastically different in the sumer, then the diurnal trends should be considerably different to those of the winter. Including more information on the summer SFOA might significantly strengthen the arguments and interpretation of the winter data. (Would enhanced summer photochemistry be a plausible reason why HR-ToF-AMS SFOA was not identifiable?)
   a. *The diurnal trend of SFOA in the summer would have been looked at but because SFOA in the summer was identified from the long-term cToF-AMS dataset it is not possible to look at the diurnal trends as the data are daily averaged concentrations. Regarding the comment about HR-ToF-AMS SFOA not being identifiable, please see the authors' response to Reviewer #1, comment 5.*

Minor comments:

- In general, ions are discussed in terms of their integer m/z (m/z 44, 60, 73, etc) rather than their actual identity, the identification of which is presumably the reason that the HR-ToF-AMS data are presented in a separate paper to the UMR AMS data. The actual ion being referred to should be named at least once in these cases.
  - *This has been addressed in the revised manuscript. For example, line 16 on page 20859 has been changed to "Although both factors comprise several similar peaks, such as at m/z 43 ($C_3H_7^+$, $C_2H_3O^+$), 55 ($C_4H_7^+$, $C_3H_3O^+$), 57 ($C_4H_9^+$, $C_3H_5O^+$), 60 ($C_2H_4O_2^+$), and 73 ($C_3H_5O_2^+$), SFOA1 also has a greater signal at m/z 44. Furthermore, SFOA1 also comprises more oxygenated compounds than SFOA2."*

- The authors frequently switch between the past and present tense. For example, the paragraph on line 15, page 20856 is in the present tense while the remainder of that section is usually in the past. Line 6 on that page begins "the ... concentration was" and continues "this event is the result of...".
  - *This has been addressed in the revised manuscript.*

- The abstract (line 15-16 of page 20847) argues that the SFOA1/SFOA2 split is due to differences in wind direction, then also argues that the split may be due to differences in burn phase. How can burn phase be related to wind direction? Is the implication that poorer stove technologies are used in a poorer region of London, so that less flaming combustion occurs? I find it hard to make sense of this combination of statements.
  - *The wind directions have been removed from the abstract in response to a comment from Reviewer #1.*

- The abstract (line 21-23 of page 20847) claims that 20% of the OA mass is more than 18%. I find this hard to believe given the uncertainties inherent in PMF.
  - *This statement has been amended in the manuscript to: "During the winter, SFOA was found to contribute 38% to the total submicron organic aerosol (OA) mass, with*

    similar contributions from both SFOA factors (20% from SFOA1 and 18% from SFOA2).”

- Page 20849 paragraph 2, please define all categories and not just solid fuel and biofuel. Also, it is stated that distinguishing sources in this manner is important without explaining why.
    - *Fossil fuel has been defined in the revised manuscript.*

- page 20849, paragraph 3, please spell out UK.
    - *This has been addressed in the manuscript as requested.*

- p 20850, paragraph 3: the wording implies that POAs are only important to climate and health when associated with BC.
    - *The final sentence in this paragraph has been edited in the revised manuscript to address this comment: “Furthermore, as the contribution of solid fuel and other primary organic aerosols to the total aerosol loading is increasing, there are significant implications on human health (Bølling et al., 2009), air quality, and climate.”*

- p 20850, last paragraph: This would be clearer if you spelt out that the citations are from the same field study discussed here.
    - *This has been addressed in the revised manuscript as suggested.*

- p 20852, first paragraph: I think the true time resolution is the opposite: 30 minutes, obtained over 5 minutes; and not 5 minutes obtained only once per 30 minutes.
    - *This sentence has been changed in the revised manuscript to: “In this study, 4 minute averaged data were obtained by the HR-ToF-AMS every 30 minutes, as sampling occurred in an alternating sequence with other black carbon and aerosol volatility measurements using a thermodenuder (Huffman et al., 2008).”*

- p 20853, 2nd paragraph: This is not "potassium ion" but rather water-soluble potassium.
    - *This has been addressed in the revised manuscript.*

- p 20853, the first paragraph of Section 3.1 reads as though the PMF analysis was highly subjective, as it only includes partial statements of the analysis. In contrast, the actual cited description makes it clear that the analysis was objective and thorough. The discussion also may fit better in a Methods section.
    - *As part of PMF analysis one must go through a process to determine the most suitable number of factors to derive which explain the dataset (Ulbrich et al., 2009). As this process involves a number of steps the details of the PMF analysis, most of which have been covered in a separate publication as noted by the reviewer and in the manuscript, we show the additional steps taken to determine the appropriate number of factors as both the 4- and 5-factor solution sets were deemed valid. The factors derived from PMF analysis within the AMS community are conventionally reported and discussed in the results section (e.g. Mohr et al., 2010; Ng et al., 2010; Crippa et al., 2013) so this discussion will remain in this section in the manuscript. An additional sentence has been included in the start of Section 3.1. of the revised manuscript: “As part of PMF analysis one must go through a process to determine the most suitable number of factors (Ulbrich et al., 2009). Here, we show the steps we took to determine the number of factors where four main components were identified from PMF analysis on the organic fraction of the HR-ToF-AMS data from*

  the winter IOP: hydrocarbon-like OA (HOA), cooking OA (COA), solid fuel OA (SFOA), and oxygenated OA (OOA), where an additional SFOA factor was identified from the 5-factor solution set."

- p 20854, line 7: please cite Ulbrich et al. (2009) regarding factor splitting.
    - *This has been included in the manuscript as requested.*

- p 20854, line 9: what is "divergence" in this context? Splitting?
    - *Divergence in this context is the general term used to describe the behaviour in computational solvers whereby numerical instability causes the residual to increase with iterations, resulting in a solution not being found.*

- p 20854, line 22: this argument seems incomplete. Was it meant that all 3 (NOx, CO and HOA) are emitted from vehicle engines?
    - *This sentence has been edited in the revised manuscript to clarify the argument: "It has been shown that* NO_x *and CO are strongly related to HOA (Zhang et al., 2005) as they are all emitted from fuel combustion in vehicle engines."*

- Section 3.2, p 20856: Why are averages and standard deviations presented, if the data are not scattered about their mean? The interquartile range would be more meaningful than the standard deviations.
    - *The standard deviations are presented as it is more conventional to report standard deviations to demonstrate variability. Standard deviations are presented in this paper to give an overview of the variability in the dataset and it is convention to report these values (e.g. Lanz et al., 2010 - doi:10.5194/acp-10-10453-2010; Dall'Osto et al., 2013 - doi:10.5194/acp-13-4997-2013; Zhang et al., 2014 - doi:10.5194/acp-14-2887-2014).*

- p 20856, line 19: Does "commercial cooking activities" mean restaurants, or something more?
    - *Commercial cooking activities does mean restaurants.*

- p 20857, line 18: "Imported" is a strange word for pollution.
    - *This term has been replaced with "advected" in the revised manuscript.*

- p 20857, line 21: black carbon is not always produced by combustion. Also, please add "In their study" to the following sentence to be clearer.
    - *This has been addressed in the manuscript as requested.*

- p 20858, line 6: it would be more meaningful to state that the composition of these OA are not constant, rather than the mass spectra.
    - *This sentence has been amended in the manuscript to "The composition of biomass burning and solid fuel OA, and therefore the mass spectra, are not constant (DeCarlo et al., 2010)."*

- Section 3.2: The diurnal trend plot would be improved by information on the variability of the data, such as shading the interquartile ranges. At the moment, there appear to be three peaks in the OOA which are not discussed.
    - *Figure 1c is already a busy plot so adding the suggested shading would make it difficult to interpret the data and also detract from the point of the plot. The peaks in*

  - *the OOA diurnal trend are not discussed as this manuscript focuses on the SFOA factors.*

- Section 4.2 discusses the variability of AMS mass spectra from different combustion sources. It seems to me that the point is about the actual variability in composition of the material produced. The section could benefit from some non-AMS citations and discussion.
  - *The following sentence with a non-AMS citation, has been included in the revised manuscript on line 9 of page 20858, at the start of Section 4.2: "This is consistent with changes in composition measured by various techniques (e.g. Schauer et al., 1996)."*

- At p 20858, line 24, and p 20859, line 14-15: the SFOA factors are discussed in terms of other studies' BBOA factors. That the SFOA factors are mass-spectrally similar to BBOA raises the problem of definition described above.
  - *There are currently no reference mass spectra for SFOA from the database used (http://cires.colorado.edu/jimenez-group/HRAMSsd/ and http://cires.colorado.edu/jimenez-group/AMSsd/) As biomass burning likely contributes to the composition of the SFOA factor identified in this study, the reference spectrum of BBOA from these databases are the only available comparison. The strong correlations found are likely due to the comparisons been made in UMR space where the reference MS was from the Q-AMS and the profiles in the study are from the HR-AMS, where more detailed chemical composition information can be gleaned from the HR-AMS data. Similarly in response to R. Healy's comment (see above), SFOA1 and SFOA2 were compared with the SFOA factor identified in London, UK during the REPARTEE project (Allan et al., 2010), with Pearson's r values of 0.88 and 0.92, respectively. These strong correlations are encouraging, particularly as the measurement project both took place in London, although in slightly different locations and years. The authors believe this strengthens the case for using the term SFOA rather than BBOA.*

- p 20859, line 19-29: the statements "there are differences in the structure of the peaks above and below the line" (of the normalized difference spectra), and "this suggests that the chemical groups are affected differently by what is driving the split between the factors" seem to be circular, because without differences in the temporal trends of these ions, PMF would not have been able to report two SFOA/BBOA factors.
  - *This has been addressed in the revised manuscript as part of major comment number 2 and in response to R. Healy's review.*

- p 20861, line 7: please specify "wood, peat and coal" with "combustion", if applicable.
  - *This has been addressed in the revised manuscript as suggested.*

- p 20861, line 17: please clarify if the stronger correlation is referring to Yin et al's data.
  - *This has been addressed in the manuscript as requested.*

- p 20862, line 28: typo: the conversion of organic matter to CO_2(g), not CO_2^+.
  - *The manuscript has been amended accordingly.*

- p 20863, line 15: "integer" m/z, not nominal.
  - *The authors feel 'integer' is not suitable because it might imply that the signals at non-integer m/z are not considered. By using the term 'nominal' (as is standard*

  - *practice in the HR-AMS community), we indicate that all signals close to the m/z are included. The manuscript will therefore not be altered here.*

- p 20863, line 24: please state that REPARTEE took place in London.
  - *This has been addressed in the manuscript as suggested.*

- p 20864, line 18: how can SFOA1 contain more organic matter than SFOA2? Both are organic components.
  - *This line has been amended in the manuscript to "SFOA1 was found to contain more oxygenated organic matter than SFOA, however, correlations with ancillary data suggested that the split in SFOA was not likely to be primarily driven by differences in fuel type or degree of processing."*

- Please capitalize the letters making up acronyms when introducing them.
  - *The authors will address this comment in the manuscript where appropriate as convention is followed for some such as the introduction of PMF factors.*

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Oros, D. and B. Simoneit. Identification and emission rates of molecular tracers in coal smoke particulate matter. Fuel, 79, 515-536, 2000.

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