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Interactive comment on "Receptor modelling of fine particles in Southern England using CMB including comparison with AMS-PMF factors" *by* J. Yin et al.

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AUTHOR COMMENTS

The authors are most grateful to the reviewers for their thorough and thoughtful commentaries on this paper. We are pleased to respond as follows:

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Anonymous Referee #1

(a) If the uncertainties can be estimated for each method, they should be included on Figure 5 and discussed in section 3.3. RESPONSE: The uncertainties are difficult to express quantitatively. However, Table 4 expresses them for the CMB model results as a standard deviation which accounts for uncertainties in the analytical data, the source profiles and the extent of colinearity among the different source profiles. It is very difficult to quantify the uncertainties inherent in the analysis of the AMS data. These are discussed at some length in Section 4, and new text has been added to amplify the point and to draw attention to the uncertainties in the CMB results.

(b) At the end of section 3.2.2, the authors state that 55% of SOA is created from biogenic influence. Please specifically state that CMB method, which determined this fraction, can address the origin and composition, but not necessarily formation mechanism (unless the author can propose a mechanism). RESPONSE: A sentence has been added to make this point.

(c) Section 3.3.1, line 20, "It is interesting to note that both CMB-WS and CMB. . . are correlated more closely . . .at low levels. . ." Can the author provide an explanation as to why the two methods relate better at lower mass concentrations that at higher concentrations? Is it a result of measurement technique? Or something different? RESPONSE: It is unlikely to be the measurement technique, and is more probably a reflection of a different composition when concentrations are higher. A sentence has been added to state this.

(d) In the conclusions, the authors make a general statement, "Work needs to be performed. . ." Specifically state what is needed to better constrain these issues. RESPONSE: Some suggestions have been added.

(e) Technical Comments: Organization comments: The authors include a brief comparison of the methodology of CMB vs PMF in the conclusions section; however, it would be best to include that also in the introduction. The receptor model differences

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are discussed briefly in paragraph three of the introduction. Since the comparison of the results between the receptor models is the major result in the paper, it merits a more thorough discussion, for example outlining the similarities and differences, and the benefits and/or limitations to each method. RESPONSE: A further paragraph has been added to the Introduction.

(f) Since the AMS-PMF method discussion and results are being published in Young et al 2014, it would be better to put the CMB model section 2.2.5 before the AMS data analysis section 2.2.3 section. RESPONSE: The order has been reversed, as suggested.

(g) Regarding the AMS 2.2.3 section, since details are presented in this paper regarding the AMS-PMF solution, the author must cite the Young et al. 2014 paper at the beginning of the section. RESPONSE: The Young et al. (2014) paper is already cited in the first paragraph of Section 2.2.3 (now 2.2.4).

(h) Why is the 2.2.4 section on its own? Perhaps these two paragraphs should be moved to section four, where the PMF analysis uncertainties are discussed. If not, reference this section 2.2.4 in section 4 discussion. RESPONSE: A cross-reference has been introduced into Section 4 (Discussion).

(i) Other comments: Figure 3 has a text box that says, 'NK (b)' in it. What does the (b) refer to? Figure 5 – Make the text readable in this figure. Also, the lines surrounding the graphs, the axis, and the grid-line spacing are inconsistent – make it uniform.# RESPONSE: The text box has been removed from Figure 3, and Figure 5 has been improved as recommended.

Anonymous Referee #2

(j) Section 3.3 Comparison between CMB and AMS-PMF results. Generally This comparison is only possible for the urban NK site as measurements at HAR were made in the absence of a collocated AMS. As comparison of CMB results from NK and HAR are

made earlier in the manuscript (section 3.2), the inclusion of data from HAR in section 3.3 (e.g., p. 24543, l. 3-4, p. 24544, l. 9-10, p. 24545, l. 5-6, p. 24545, l. 19-23, etc.) is unnecessary and distracts from the CMB/AMS-PMF comparisons. I would recommend reorganizing to the extent necessary in order to move the HAR numbers to section 3.2 (if they are not reported there already) and limit the discussion in section 3.3 to only the NK site. RESPONSE: We disagree with the reviewer. We think it useful to discuss fully the CMB data from both sites before the sections in which the intercomparison with AMS is described. Different readers will wish to focus on different parts of the paper, and the current format permits that. In fact, the discussion of the Harwell results is overall quite brief. The sections have been re-numbered and re-titled to make this more accessible to the reader.

(k) In order to compare results of CMB (reported as OC) with PMF (reported as OM), CMB concentrations were first converted to OM using appropriate OM/OC factors (ref. p. 24542, l. 3-5). Are these the same conversion factors that were used to calculate the source contributions to PM2.5 as listed in Table 4 as "OC/PM2.5 or OC/OM CF"? If so, it would be helpful to the reader to state as much in the text here as this conversion is a critically important first step in comparing these two data sets. RESPONSE: The reviewer raises a good point, which has now been clarified in the text, at the end of (the now numbered) Section 4.

(l) It would also be worthwhile for the authors to note in the text whether the AMS-PMF derived component concentrations have been adjusted to account for the organic mass difference between PM1 and PM2.5, if such difference existed. Authors casually mention this difference on p. 24548, l. 14-18 and also that AMS volume concentrations were compared with DMPS (presumably differential mobility particle sizer?, not defined) measurements in order to validate the use of a time- and composition-dependent AMS collection efficiency (ref. p. 24530, l. 25-26) but do not provide any additional information with regard to correcting for any potential organic mass difference. RESPONSE: It is made very clear in the discussion of the intercomparison that

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no such adjustment was made. No data were available from which to make the adjustment.

(m) Section 3.3.1 Woodsmoke Among the largest differences between CMB and AMS-PMF results is for the contribution of woodsmoke with AMS-PMF reporting nearly double that reported by CMB with the CMB value presumably calculated with an OM/OC value of 1.20 as reported in Table 4. As this source presents the largest difference between CMB and AMS-PMF and considering the critical reliance on the CMB number to the appropriate conversion factor, I would recommend including a sensitivity analysis for the CMB value varying the OM/OC ratio similar to that included by the authors for source contribution calculations (ref. section 3.2.2). For example, Turpin and Lim (2001) report an OM/OC_1.9 value for woodsmoke. Using this OM/OC value would increase the CMB average value to ∼1.35 μg m-3, much more in line with the AMS-PMF determined value. Consequentially, this would also decrease the CMB-determined secondary OM value as this fraction was determined as the residual between classified source concentrations and measured PM2.5 OC. RESPONSE: Another good point. However, the OM/OC factor used was in fact 1.6, but still a little below that suggested by the reviewer. This point is now expanded in the text and values derived from the AMS data are also included.

(n) Section 3.3.2 Food cooking particles Similar to woodsmoke, the AMS-PMF-determined contribution of food cooking to PM1 (∼20% of PM1) was higher than the CMB-determined value (9% of PM2.5 OC; 4% of PM2.5). The authors provide additional literature values for the contribution of food cooking obtained from CMB (overall average ∼8.5%, range ∼7-12% of PM2.5 OC) and AMS-PMF (overall average ∼22%, range 10.4-30% of PM1 OM) from different locations to conclude that there is a "systematic difference" between CMB- and AMSPMF- derived food cooking contributions. It is worthwhile to note that these literature values were reported for widely different geographical locations and none of the referenced studies used both CMB and AMS-PMF as was done in the current study. While additional studies may reveal a sys-

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tematic difference along with its underlying causes, the data available at present is better described as a trend of either AMS-PMF overestimating or CMB underestimating food cooking contributions to OM. RESPONSE: The words "systematic difference" have been replaced with the softer term "trend" in line with the reviewer recommendation.

(o) Section 4 Overview of CMB comparison with AMS-PMF results CMB and AMS-PMF analyses reported similar results for select sources such as traffic while substantial deviation was observed for others including wood burning, food cooking, and secondary organics. The authors broadly attribute these differences to either (in whole or in part): 1) differing effective particle mass ranges used in CMB and PMF analyses (i.e., CMB apportioned PM2.5 while PMF apportioned PM1); 2) an ambiguous collection efficiency (CE) on the part of the AMS; 3) rotational ambiguities inherent in the PMF analysis which may cause mass to be inadequately apportioned among the various factors resolved by PMF, or; 4) inaccuracies on the part of CMB. The authors do not, however, take any steps to narrow among those possibilities the most likely source(s) of the reported discrepancies to the extent that is at all possible. For example, in discussing the large difference between CMB- and AMS-PMF determined secondary organics, (ref. p. 24548, l. 17-18), the authors speculate that the larger CMB-derived value could be due to condensation of secondary organics onto supermicron particles. Could this not be investigated by looking at AMS and DMPS volume concentrations throughout the sampling period with the aim of identifying whether such differences exist in the supermicron region? While a null result would not negate this possibility outright, it would at least decrease the likelihood that differences in the effective mass range sampled in each case contributes significantly to the observed differences. RESPONSE: The suggestion made by the reviewer is not feasible as neither the AMS nor the DMPS measure into the supermicron region.

(p) Similarly, the authors suggest that an ambiguous AMS CE may lead to an overestimation of food cooking in the AMS-PMF analysis assuming both that the particles are

externally mixed and that the food cooking particles are more liquid in nature thereby leading to a higher CE for this particular mass fraction. Investigating the AMS and DMPS time series (along with their diurnal profiles) may provide additional information regarding those time periods for which CE significantly deviated from the time and composition-dependent CE employed in the analysis. Those periods could then be superimposed on top of the AMS-PMF time series of, for example, the food cooking factor to examine the correlation between the two. Similar to the above example, while this possibility could not be completely discounted in the event of a null result, it would provide additional direction for others interested in this comparison to advance in the future. RESPONSE: We do not believe that this additional work would lead to the new insights suggested due to the extreme complexity of the aerosol, which is at no time dominated by an individual source such as food cooking.

Tables and Figures (q) Figure 1. Recommend using the same scale on the y-axis to represent difference in total mass measured at each location. Also make clear in caption that results do not include secondary biogenic components at NK to make a clear contrast with Figure 2. RESPONSE: The caption has been modified as suggested. We do not regard re-drawing the figure to be useful as the y-axis is clearly labelled.

(r) Figure 5. Recommend modifying this figure to make the axes titles as well as figure legends easier to read. RESPONSE: This has been done.

(s) Technical corrections: p. 24534, l. 1: change "measurement" to "measurements" p. 24538, l. 10: a reference should be added to support use of OM/OC ratio=1.8 for secondary biogenic sources p. 24539, l. 28: change "comparable magnitude" to "comparable in magnitude" p 24542, l. 11-12: based on the numbers provided (1.63 and 0.85) AMS-PMF woodsmoke concentration is 2.0 times the CMB woodsmoke only if you round to the tenths place in which case the numbers should be change accordingly otherwise the ratio should be corrected p. 24544, l. 19: "South-Eastern" should be changed to "Southeastern" RESPONSE: All points have been corrected.

Anonymous Referee #3

(t) 1) Section 2.2.2: Why were the secondary biogenic markers not measured for the Harwell filters? The text says "(NK only)" without any explanation. It seems like this would be an important part of the comparison between NK and HAR. RESPONSE: This was a resource limitation. The funding was insufficient to carry out these analyses for both sites.

(u) 2) Section 2.2 Methodologies: This section would flow better if you put the CMB analysis sections after the marker measurements and moved the AMS analysis to the end. RESPONSE: Agreed. This has been done.

(v) 3) Section 2.2.3 AMS data analysis: The third paragraph about W-mode data and ME-2 seems unnecessary in this paper. Presumably these choices are discussed in detail in Young et al. (2014) and don't need to be rehashed here. RESPONSE: We feel that this brief section is valuable in explaining which AMS data were used and why.

(w) 4) Section 3.1.1, first paragraph. Lists of numbers are difficult to read in text. Since the numbers are given in Table 2, maybe the text could just discuss the comparison. RESPONSE: Agreed. The numbers have been removed and the text rearranged.

(x) 5) Section 2.3.1. Are there markers for secondary anthropogenic aerosol that could be used in this analysis in the same way that markers for secondary biogenic are applied? It would be really interesting if you could get closure on the organic mass loading. Also in this section, is there a reason to prefer Hysplit back trajectories (many of which originate at high elevations) to the NAME back trajectories that were calculated for the ClearfLo campaign? RESPONSE: Molecular markers have been used in other studies for the oxidation products of some anthropogenic VOC. To capture the diversity of precursor compounds it is necessary to analyse several such markers, and resource was insufficient to do so.

(y) 6) Page 24539, lines 5 to 9, "The derivations. . ..final choice." This sentence is long

and confusing. Can you revise it into shorter sentences that make your points clear? RESPONSE: The sentence has been split into two and modified extensively.

(z) 7) Section 3.3 Comparison between CMB and AMS-PMF estimates. "Comparison has been made. . .Fig. 5.": Reference to Table 5 should be to Table 4. I think Table 4 should have separate columns for PM2.5/OC and OM/OC since these are different conversions for some sources, e.g., traffic. The discussion of OC to OM conversion should be moved to this section from its current location in the third paragraph of Section 4. I found Section 3.3 to be very confusing until you explained what you meant by OM and PM2.5 in Section 4. RESPONSE: This re-ordering has been carried out, with some amendment of text.

(aa) 8) page 24542, lines 16 to 20: Is 0.78 a "stronger correlation" than 0.75? Looking at Figure 5, the data looks very similar. RESPONSE: The word "slightly" has been inserted.

(bb) 9) page 24545, line 10: insert "such as" after "variables," Do you have a reference for this sentence? RESPONSE: Amended, as suggested.

(cc) 10) Section 4, third paragraph, converting OC to OM: One can get OC directly from HR-AMS measurements so why not compare the CMB OC directly to AMS OC? This would avoid some of the uncertainty in the OC to OM conversions. RESPONSE: While OC can be derived based on AMS data using the method of Aiken et al. (2007), it must be borne in mind that this is an estimate, not a direct measurement and a currently in-press paper by Canagaratna et al. (2014) provides a refinement to this technique. Furthermore, it is necessary to convert CMB OC to OM anyway, such that it can be compared with the PM2.5 mass budget. Nevertheless, we can report that the OM:OC values estimated using the Aiken et al. (2007) method are 1.69 and 1.33 respectively. By the Canagaratna et al. (2014) method, they are 1.91 and 1.40 respectively. These data have been added to the text.

(dd) 11) page 24548, line 5: "other constituents" is really "refractory constituents".

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RESPONSE: The comment was meant to say that the AMS-reported factor does not include mineral components that would be included with a mass-based profile such as soil. However, given that the AMS does not detect such a factor anyway, this sentence is largely irrelevant and has been removed.

(ee) 12) page 24548, lines17-18, "It is conceivable that. . .": This seems like an unlikely explanation since the difference in organic mass loading between PM1 and PM2.5 is usually only 10 to 20%. This doesn't explain a factor of 3 difference between CMB and AMS. RESPONSE: It could be a contributing factor; the wording has been amended to reflect this.

(ff) 13) page 24548 line 19: I would not say that the AMS CE is ambiguous. It is relatively constant over many field campaigns in many locations, although it can be different for particles with extreme compositions (e.g., very acidic). RESPONSE: While there have been many studies reporting on the consistency of the AMS CE in the ambient atmosphere, these largely report on a mixed organic/inorganic particulate dominated by the accumulation mode, which is not necessarily relevant to an externally mixed population of primary organic particles. There are currently very few data reported on the CE of primary particulates from sources such as biomass burning and cooking and furthermore, because the AMS measures in bulk, the quantitative mixing state of these organics with the accumulation mode is ambiguous, hence our statement that this is currently ambiguous.

(gg) 14) page 24548, lines 25 to 30: I'm not sure what the point is of this discussion about some constituents of cooking aerosol being liquid at room temperature. The cooking aerosol agrees fairly well between PMF and CMB. The biggest disagreement between PMF and CMB for the primary particles is for biomass burning, but the liquid particle argument doesn't apply there. RESPONSE: While the correlation for cooking is good, the slope is not unity, implying there is a multiplicative factor causing disagreement between the two techniques. A variation in CE caused by liquid particles (amongst the other possible causes) would be a plausible explanation for this.

(hh) 15) page 24549, line 26-27: Loss of levoglucosan from biomass burning particles during atmospheric transport is well-documented. Could this account for some of the discrepancy in SFOA between PMF and CMB? RESPONSE: The reviewer makes a good point which has been added to the text.

(ii) 16) References: There are a lot of typos in the references – please proofread them carefully. RESPONSE: This has been done.

(jj) 17) Table 6: The first source is called biomass burning here, but SFOA or WS in other tables and figures. Maybe call it SFOA/WS for consistency. RESPONSE: Amended to woodsmoke.

(kk) 18) Figure 3 and 4: The colors for Seasalt and Other OM are different between these two figures. Please use the same colors! RESPONSE: Amended as requested.

(ll) 19) Figure 5: This figure is very hard to read. Please make the axis labels and legend much larger. It would help to number the panels and describe them in the caption. Why are the slopes in Figure 5 very different than the ratio of the numbers in Table 6 (e.g., slope of SFOA vs WS is 2.8, but the ratio from Table 6 is 1.9)? Also, the intercept of SFOA vs WS has a significant offset on the WS axis. Does this suggest an interference in the CMB extraction of the WS source? RESPONSE: The figure has been improved. The presence of intercepts can contribute to the differences between ratios and gradients. The reason for them is currently a matter for speculation.

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