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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

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General Comments:

In Receptor modelling of fine particles in Southern England using CMB including comparison with AMS-PMF factors, Yin et al. present results of chemical mass balance (CMB) source apportionment of fine (PM2.5) particle mass sampled during winter months (January and February, 2012) at two locations in Southern England: a background urban location at North Kensington (NK) and a rural location at Harwell (HAR). CMB identified similar sources of fine particle mass and its organic fraction from both locations albeit with differing contributions at each sampling site due to their different geographical location. Overall, primary sources including traffic, woodsmoke, food

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cooking, coal combustion, vegetative detritus, natural gas, and dust/soil account for just over half of the organic carbon (OC) at each location while the inclusion of a secondary biogenic source in the CMB model for measurements at NK accounted for another ~25%. CMB results from each location are compared with similar CMB studies results from European locations and are in reasonable agreement. Finally, results of CMB from the NK site are compared with results of positive matrix factorization (PMF) of high resolution mass spectra obtained from a collocated high resolution time-of-flight aerosol mass spectrometer (AMS). Despite the widely different approach of CMB and PMF in apportioning mass to various PM2.5 sources, there was broad agreement between each in the identification of several sources (e.g., traffic) while, for others, the two approaches present significant differences (e.g., biomass burning and secondary OC), the potential causes of which are discussed by the authors.

Overall, the manuscript is well written, reasoned, and referenced. In this reviewer's opinion, however, the numerous comparisons of this study's CMB results with those from other European locations unnecessarily adds length to the manuscript without adding substantively to the discussion regarding the work at issue leading the paper to read more like a review in a few sections. I would recommend that the authors examine whether this material could be included in a more succinct format, such as a table included in the main text (e.g., Table 3 in the main text) or perhaps, more appropriately, in the Supporting Information section. One example of this can be found on p. 24544, lines 11-26 when discussing previously reported literature values for food cooking obtained by CMB and PMF of AMS data. The inclusion of these and similar literature values in a tabular form would have a greater impact and would substantially add to the readability of the paper as a whole.

Notwithstanding this aspect the manuscript is technically sound and of interest to the atmospheric science community at large, and worthy of publication in Atmospheric Chemistry and Physics after addressing the specific comments listed below.

Specific comments:

ACPD
14, C8649–C8654, 2014

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.

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.

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.

Section 3.3.1 Woodsmoke

Among the largest differences between CMB and AMS-PMF results is for the contri-

ACPD

14, C8649–C8654, 2014

bution 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$\sim$1.9 value for woodsmoke. Using this OM/OC value would increase the CMB average value to $\sim$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.

Section 3.3.2 Food cooking particles

Similar to woodsmoke, the AMS-PMF-determined contribution of food cooking to PM1 ($\sim$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 $\sim$8.5%, range $\sim$7-12% of PM2.5 OC) and AMS-PMF (overall average $\sim$22%, range 10.4-30% of PM1 OM) from different locations to conclude that there is a "systematic difference" between CMB- and AMS-PMF-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 systematic 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.

Section 4 Overview of CMB comparison with AMS-PMF results

CMB and AMS-PMF analyses reported similar results for select sources such as traffic

ACPD
14, C8649–C8654, 2014

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 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.

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

ACPD
14, C8649–C8654, 2014

provide additional direction for others interested in this comparison to advance in the future.

Tables and Figures

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.

Figure 5. Recommend modifying this figure to make the axes titles as well as figure legends easier to read.

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”

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 24523, 2014.