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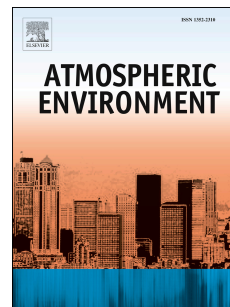
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5 **Sensitivity of a Chemical Mass Balance Model**
6 **for PM_{2.5} to Source Profiles for Differing**
7 **Styles of Cooking**
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21 **ABSTRACT**

22 Use of a Chemical Mass Balance model is one of the two most commonly used approaches to
23 estimating atmospheric concentrations of cooking aerosol. Such models require the input of
24 chemical profiles for each of the main sources contributing to particulate matter mass and there is
25 appreciable evidence from the literature that not only the mass emission but also the chemical
26 composition of particulate matter varies according to the food being prepared and the style of
27 cooking. In this study, aerosol has been sampled in the laboratory from four different styles of
28 cooking, i.e. Indian, Chinese, Western and African cooking. The chemical profiles of molecular
29 markers have been quantified and are used individually within a Chemical Mass Balance model
30 applied to air samples collected in a multi-ethnic area of Birmingham, UK. The model results give
31 a source contribution estimate for cooking aerosol which is consistent with other comparable UK
32 studies, but also shows a very low sensitivity of the model to the cooking aerosol profile utilised. A
33 survey of local restaurants suggested a wide range of cooking styles taking place which may explain
34 why no one profile gives an appreciably better fit in the CMB model.

35
36 **Keywords:** Chemical Mass Balance model; cooking aerosol; source apportionment; molecular
37 markers

38

39 INTRODUCTION

40 More studies are published almost daily on the source apportionment of airborne particulate matter
41 (PM), usually expressed as mass concentration within a particle size range, typically PM_{2.5} or PM₁₀.
42 Such studies are an essential pre-requisite to the development of cost-effective mitigation options
43 for PM. While it has long been known that cooking processes are a source of airborne PM, the
44 capability to estimate concentrations by aerosol mass spectrometry (AMS) has led to cooking
45 aerosol featuring as a contributor to many source apportionment estimates (e.g. Mohr et al., 2009;
46 2012; Allan et al., 2010). Identification of the cooking organic aerosol (COA) factor in most AMS
47 studies is dependent upon recognition of a temporal signature (Lanz et al., 2007), often with
48 characteristic mass spectral features also being recognisable (Mohr et al., 2009). However, some
49 recent studies have cast doubt upon the attribution of the COA factor to food cooking as the sole
50 source, and have concluded that other sources may also contribute to this factor (Hayes et al., 2013;
51 Dall'Osto et al., 2015).

52
53 Chemical Mass Balance (CMB) models provide an alternative means of estimation of cooking
54 aerosol (Schauer et al., 1996; Robinson et al., 2006), but the number of studies is relatively few,
55 and some attribute relatively small contributions to cooking aerosol (e.g. Yin et al., 2010). The
56 study by Yin et al. (2015) was unique in making a direct comparison of cooking aerosol derived
57 from a CMB model with an estimate derived from simultaneous measurements by AMS, with
58 application of Positive Matrix Factorization (PMF) to identify source-related factors and quantify
59 their contributions. A comparison of estimates of 24-hour average concentrations derived from the
60 two techniques revealed a close correlation ($r^2 = 0.80$), but a considerably higher estimate from the
61 AMS data (y) than from CMB (x), with a regression equation of $y = 2.24x - 0.33 \mu\text{g m}^{-3}$. Various
62 possible explanations for the discrepancy were advanced by Yin et al. (2015), the most plausible
63 appearing to be that cooking organic aerosol is collected with close to 100% efficiency by the AMS,
64 rather than the normally assumed efficiency of 50%, accounting for a possible over-estimation of

65 the AMS method by a factor of two. Such high collection efficiencies for particles from cooking
66 are reported by Pandis (2016). However, the CMB model approach has weaknesses, and is liable to
67 give incorrect source contribution estimates if the adopted source profile input to the model does
68 not well match that in the actual emissions from the cooking source.

69

70 It is clear from the literature on the speciation of cooking emissions, reviewed recently by Abdullahi
71 et al. (2013), that some emission profiles may differ substantially between different methods (e.g.
72 boiling versus frying) and styles (e.g. Chinese versus Indian) of cooking. In this study, we have
73 measured source profiles from the cooking of a number of dishes characteristic of different cooking
74 styles and have used them in a CMB model to test sensitivity to the input profile. There are many
75 cooking styles used around the world, and to sample them all would not be feasible. Consequently,
76 we have selected four types of national cuisine which are very common in the United Kingdom, as
77 well as in other parts of the world.

78

79 **EXPERIMENTAL**

80 **Sampling from Cooking Experiments**

81 Cooking on a gas or electric hotplate took place in a trailer located on the University of
82 Birmingham campus. At a vertical distance of approximately 61 cm above the cooking fume source
83 the aerosol entered a 70 cm diameter steel extraction hood through which air was drawn at 495-500
84 $\text{m}^3 \text{h}^{-1}$. From the hood, the sampled air passed along a steel ducting of 20 cm diameter from which
85 it was sub-sampled isokinetically through a stainless steel probe at 30 L min^{-1} with aerosol particle
86 collection on a 47 mm quartz (Whatman GF/A) filter. Samples were used for characterisation of
87 source profiles, with no estimation of emission factors. For full details, please see the
88 Supplementary Information.

89

90 The cooking styles and food options selected are described in Table 1. Six samples were taken for
 91 each cooking style, with individual sample collection lasting from the start to end of the cooking
 92 cycle.

93
 94 **Table 1:** Cooking styles and food options selected.
 95

Cooking style	Dish	Method
Chinese	Chicken kun pao with rice	Stir fry
Western	Chicken, eggs and chips	Deep fry
Indian	Chicken tikka masala with rice	Stew
African	Chicken in tomato stew with rice and plantain	Deep frying, stew

96 97 **Sampling in the Atmosphere**

98 Air sampling took place in a measurement station operated by Birmingham City Council on
 99 Stratford Road, Birmingham, UK. The location, together with that of local restaurants appears in
 100 Figure 1. A list of local restaurants, together with their predominant cuisine and distance from the
 101 sampler appears in Table S2. Air sampling took place using a Digital high volume sampler
 102 operated at 500 L min^{-1} with a $\text{PM}_{2.5}$ inlet. Quartz fibre filters (15 cm) were pre-baked at 500°C for
 103 5 hours to reduce the blank, and exposed for 24-hours in the air sampler. Samples were collected
 104 between 10-18 December 2014 and 9-18 January 2015. For further details, please see the
 105 Supplementary Information.



106
 107
 108 **Figure 1:** Map of Stratford Road showing restaurants and sampling site.
 109
 110

111 Chemical Analysis of Samples

112 After sampling, circles of 1.5 cm diameter were taken by punch from the Digitel quartz filters and
113 analysed for elemental (EC) and organic (OC) carbon with a Sunset Laboratory thermal-optical
114 analyser using transmission for pyrolysis correction and the EUSAAR 2 temperature protocol
115 (Cavalli et al., 2010). Subsequently, prior to extraction, the filters were spiked with isotopically
116 labelled standards including octacosane-d58, hexatriacontane-d74, dibenz(a,h)anthracene-d14, aaa-
117 20R-cholesterol-d4, heptadecanoic acid-d33, cholesterol-2,2,3,4,4,6-d6 and levoglucosan-U13C6.
118 The filters were extracted with dichloromethane (twice) followed by methanol (twice) according to
119 the method of Yin et al. (2010), which is based upon Sheesley et al. (2004). Organic acids were
120 derivatised according to the method of Podlech (1998) and Aldai et al. (2005), and sterols
121 derivatised by the method of Yue and Fraser (2004), all as described previously by Yin et al.
122 (2010). One field blank was collected and analysed for each six field samples.

123
124 After drying, preconcentration and blowing down with nitrogen, the extracts were split in three
125 aliquots for either a) direct analysis, b) derivatisation with 2M trimethylsilyldiazomethane (TMS-
126 DM) in diethyl ether (for analysis of acids) or c) derivatisation with N,O-bis (trimethylsilyl)
127 trifluoroacetamide/trimethylchlorosilane (BSTFA-TMCS) (for sterols). Analysis was by GC-MS
128 using an Agilent Technologies 6890N gas chromatograph and 5973N MSD fitted with a HP-5MS
129 column (30 m; 0.25 mm dia; 0.25 μ m thickness). Calibration was with natural standards, using the
130 internal deuterated standards to correct for recovery.

131

132 Chemical Mass Balance (CMB) Modelling

133 Chemical Mass Balance modelling used the USEPA version 8.2 CMB model, in a manner
134 following that outlined by Pant et al. (2014), to estimate contributions to PM_{2.5}-OC. The source
135 profiles adopted were these described in Yin et al. (2010) and Yin et al. (2015). The UK-derived
136 road traffic profile derived by Pant et al. (2014) from a twin-site study was employed. The various

137 cooking source profiles derived in this study were each used individually in model runs and the
138 CMB model outputs were used to assess the quality of fit to the atmospheric composition
139 measurements. In all cases, the profiles derived from cooking with gas were used, as this is more
140 widely used in local restaurants/take-aways, and is also available and widely used in local domestic
141 properties.

142

143 **RESULTS AND DISCUSSION**

144 **Measurement of Source Profiles**

145 The mass concentrations of particles collected on the filter using the gas hotplate ranged from $81 \pm$
146 12 (s.d.) $\mu\text{g m}^{-3}$ for African cooking to $368 \pm 83 \mu\text{g m}^{-3}$ for Chinese cooking. The range for the
147 electric hotplate was similar, from $99 \pm 19 \mu\text{g m}^{-3}$ for Indian cooking to $470 \pm 263 \mu\text{g m}^{-3}$ for
148 Chinese cooking. The mean concentrations of the organic compounds analysed appear in Table S3
149 for the gas hotplate and Table S4 for the electric hotplate. Each cooking experiment was replicated
150 four times and an example of the range of concentrations of alkanes and PAH appears in Table S5.
151 It may be seen that relative standard deviations of individual compounds were typically in the range
152 of 10-20% for alkanes and 30-50% for PAH. Table S6 shows source profiles for gas cooking
153 expressed as $\mu\text{g} (\mu\text{g OC})^{-1}$. Total concentrations of the groups of compounds appear in Table 2
154 which shows that Chinese cooking exceeds the other cooking styles for all groups of compounds, in
155 some cases by a substantial margin. African cooking emits the least, in all but one compound class.
156 Correlations between cooking styles were tested within the alkane, PAH and acid classes, with the
157 results expressed as Spearman's rho from rank correlation appearing in Table S7. Correlations are
158 typically low (< 0.20) to modest (0.5-0.7) with a few higher. In particular, the acids group tend to
159 correlate strongly between cooking styles (Table S7(c)) with many correlations > 0.80 . The
160 coefficient of divergence, defined as in Liu and Harrison (2011) describes the similarity between
161 datasets, with values occupying a range from 0-1, with zero indicating total similarity and one a
162 high degree of difference. The values obtained appear in Table S8.

163 **Table 2:** Total concentrations of compounds (alkane, PAH, sterol, glyceride and acids) at cooking
 164 source ($\mu\text{g}/\text{m}^3$).
 165

($\mu\text{g}/\text{m}^3$)	INDIAN	WESTERN	AFRICAN	CHINESE
Total n-alkanes	12.41	11.66	4.67	12.99
Total PAH	5.35	9.31	2.92	12.74
Total acid	6.65	9.87	6.83	21.61
Total sterols	1.18	0.94	0.37	1.34
Total monoglyceride	3.38	10.33	1.48	11.52

166

167 Normalisation by the concentration of organic carbon (OC) gives a better test of similarity of
 168 profiles, and regression plots between cooking styles appear in Figure S5. There is sufficient co-
 169 linearity between the profiles that each was used in a separate run of the model, rather than
 170 attempting to include multiple profiles in one run of the model. It may be seen from Table S2 that
 171 the restaurants in the locality serve a variety of cuisine, with Indian restaurants being the most
 172 common. The population of the area is also culturally diverse, with a substantial community with
 173 ethnic origins in the Indian sub-continent.

174

175 Concentrations of organic carbon were apportioned in the model, with four primary sources
 176 showing a good fit: woodsmoke, dirt/soil, traffic and cooking aerosol. The criterion used for model
 177 fitting were the χ -squared and r^2 values, the ratio of the source contribution and standard error (t_{stat}),
 178 and the ratio of calculated to measured concentration. The contributions of the four sources
 179 according to the cooking style used in the model appear in Figure 2, and show little sensitivity to
 180 the input source profile for cooking. There is a large unaccounted mass of OC, labelled in the
 181 figure as “other”, which we believe is comprised mainly of secondary organic carbon, which is
 182 known to make a substantial contribution to OC at UK sites (Harrison and Yin, 2008; Yin et al.,
 183 2010; Pio et al., 2011).

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Figure 2: Average source contribution estimates for OC according to style of cooking source profile.

192 Average values of χ -squared and r^2 for the model fits appear in Table 3, and show no significant
193 difference for the compositional profiles tested. Examination of results for individual days showed
194 differences not only between the day-to-day apportionment to sources, but also the source
195 contribution estimates obtained when using different source profiles for cooking (see Figure S6).
196 However, variations in the model fit as revealed by χ -squared and r^2 values within a day according
197 to source profile were fairly minor (Table 3). The day with greatest variation showed a range of r^2
198 for the different cooking styles of only 0.02, whereas the variation between days (of 0.67 to 0.94)
199 was far greater. Similarly there was more day-to-day variation in χ -squared than in the within-day
200 values for cooking styles.

201

Table 3: Quality of fit parameters for CMB model according to style of cooking source profile.

202
203

Cooking profile	West	Indian	Chinese	African
R^2	0.80±0.08 (n=14)	0.80±0.08 (n=14)	0.78±0.07 (n=10)	0.80±0.08 (n=13)
χ -squared	0.09±0.02 (n=14)	0.08±0.02 (n=14)	0.09±0.01 (n=10)	0.09±0.02 (n=13)

204 The measured concentration for $PM_{2.5}$ on the days of sampling averaged 6.9 ± 1.6 (s.d.) $\mu\text{g m}^{-3}$.
205 This was a period of unusually clean air for the time of year. The annual mean for the nearest
206 AURN (national network) station of Acocks Green for $PM_{2.5}$ was $12 \mu\text{g m}^{-3}$ in 2014 and $9 \mu\text{g m}^{-3}$ in
207 2015. The mean concentration of organic carbon apportioned to cooking aerosol was $0.12 \mu\text{g m}^{-3}$
208 (using the Indian and African cooking source profiles) and $0.13 \mu\text{g m}^{-3}$ (from the Western and
209 Chinese profiles). This converts to $0.21\text{-}0.23 \mu\text{g m}^{-3}$ organic matter, equivalent to the mass of
210 cooking aerosol particles, contributing $3.0\text{-}3.3\%$ of $PM_{2.5}$ mass. This figure compares with a mean
211 mass concentration of OC of $0.39 \mu\text{g m}^{-3}$, equivalent to $0.69 \mu\text{g m}^{-3}$ of cooking aerosol, comprising
212 4.4% of $PM_{2.5}$ measured at North Kensington, London by Yin et al. (2015) using a CMB model.
213 The Stratford Road, Birmingham samples showed an average contribution from road traffic of 0.37
214 $\mu\text{g m}^{-3}$ to OC concentrations, equivalent to $0.64 \mu\text{g m}^{-3}$ (9.3%) of $PM_{2.5}$. This compares with 0.73
215 $\mu\text{g m}^{-3}$ of OC, equivalent to $1.26 \mu\text{g m}^{-3}$ (8.0%) of $PM_{2.5}$ at London, North Kensington. These
216 results thus appear very consistent when allowing for the relatively clean air period which was
217 sampled at Stratford Road, Birmingham.

218
219 Ots et al. (2016) have used AMS measurements of cooking aerosol to estimate a source strength,
220 from which concentrations across the UK have been modelled. Their model predicts a mean
221 concentration of COA in 2012 of $0.5 \mu\text{g m}^{-3}$ for the model grid cell showing highest concentration.
222 The annual mean $PM_{2.5}$ at Birmingham, Acocks Green in 2012 was $11 \mu\text{g m}^{-3}$. If the cooking
223 aerosol estimated for Stratford Road by CMB is scaled by $11/6.9$ to make it equivalent to mean
224 annual conditions for 2012, the concentration is $0.35 \mu\text{g m}^{-3}$ (taking the mean from all cooking
225 styles). Given the results of comparison of AMS and CMB by Yin et al. (2015) and the possible
226 over-estimation of COA by AMS by a factor of up to two, discussed in detail by Ots et al. (2016),
227 the scaled concentration of $0.35 \mu\text{g m}^{-3}$ compares well with the model estimate of $0.5 \mu\text{g m}^{-3}$.

228

229

230 **CONCLUSIONS**

231 Although the main purpose of this research was not to estimate the magnitude of cooking aerosol
232 concentrations, the comparison with earlier measurements from London (Yin et al., 2015) and with
233 the model results of Ots et al. (2016) show a strong consistency. This suggests that in recent years
234 in major UK cities, cooking aerosol represents about 3-4% of measured $PM_{2.5}$. The comparison
235 with the numerical model results of Ots et al. (2016) is again suggestive of an over-estimation of
236 COA by the AMS-PMF technique relative to the CMB model results, although in this case the ratio
237 is less than the two suggested by Ots et al. (2016) as a maximum.

238

239 The main objective of the research was to compare the estimates of cooking aerosol from the CMB
240 model using source profiles typical of our different cooking styles: Indian, Chinese, Western and
241 African. Despite some differences in the profiles, the CMB model results from each profile are
242 very similar. This may be because in a multi-ethnic cosmopolitan city such as Birmingham no one
243 cooking style is dominant, or because there is sufficient colinearity in the profiles that each leads to
244 a similar estimate, whatever the predominant source of the cooking. The evidence from a survey of
245 local restaurants is that they cater for a very wide range of cuisine, which seems likely to be a
246 dominant factor in this case.

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341 **TABLE LEGENDS**

342

343 **Table 1:** Cooking styles and food options selected.

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345 **Table 2:** Total concentrations of compounds (alkane, PAH, sterol, glyceride and acids) at
346 cooking source ($\mu\text{g}/\text{m}^3$).

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348 **Table 3:** Quality of fit parameters for CMB model according to style of cooking source profile.

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352 **FIGURE LEGENDS**

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354 **Figure 1:** Map of Stratford Road showing restaurants and sampling site.

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356 **Figure 2:** Average source contribution estimates for OC according to style of cooking source
357 profile.

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360

SENSITIVITY OF A CHEMICAL MASS BALANCE MODEL FOR PM_{2.5} TO SOURCE PROFILES FOR DIFFERING STYLES OF COOKING**AUTHORS: K.L. Abdullahi, J.M. Delgado-Saborit and Roy M. Harrison****HIGHLIGHTS**

- Chemical profiles of four cooking styles have been measured
- Profiles show some marked differences
- Each profile has been used in a CMB model
- No single profile shows clearly a better fit in the model
- Estimated cooking aerosol mass is consistent with other methods