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Abdullahi, K. L.; Delgado-Saborit, J. M.; Harrison, Roy M.

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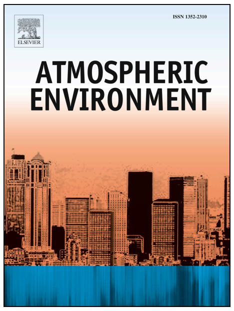

Sensitivity of a Chemical Mass Balance model for $PM_{2.5}$ to source profiles for differing styles of cooking

K.L. Abdullahi, J.M. Delgado-Saborit, Roy M. Harrison

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Sensitivity of a Chemical Mass Balance Model for $PM_{2.5}$ to Source Profiles for Differing Styles of Cooking

K.L. Abdullahi, J.M. Delgado-Saborit and
Roy M. Harrison[*][†]

Division of Environmental Health and Risk Management
School of Geography, Earth and Environmental Sciences
University of Birmingham
Edgbaston, Birmingham B15 2TT
United Kingdom

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[*] To whom correspondence should be addressed.
Tele: +44 121 414 3494; Fax: +44 121 414 3709; Email: r.m.harrison@bham.ac.uk

[†] Also at: Department of Environmental Sciences / Center of Excellence in Environmental Studies, King Abdulaziz University, PO Box 80203, Jeddah, 21589, Saudi Arabia

ABSTRACT

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

Keywords: Chemical Mass Balance model; cooking aerosol; source apportionment; molecular markers

INTRODUCTION

More studies are published almost daily on the source apportionment of airborne particulate matter (PM), usually expressed as mass concentration within a particle size range, typically $PM_{2.5}$ or PM_{10}. Such studies are an essential pre-requisite to the development of cost-effective mitigation options for PM. While it has long been known that cooking processes are a source of airborne PM, the capability to estimate concentrations by aerosol mass spectrometry (AMS) has led to cooking aerosol featuring as a contributor to many source apportionment estimates (e.g. Mohr et al., 2009; 2012; Allan et al., 2010). Identification of the cooking organic aerosol (COA) factor in most AMS studies is dependent upon recognition of a temporal signature (Lanz et al., 2007), often with characteristic mass spectral features also being recognisable (Mohr et al., 2009). However, some recent studies have cast doubt upon the attribution of the COA factor to food cooking as the sole source, and have concluded that other sources may also contribute to this factor (Hayes et al., 2013; Dall'Osto et al., 2015).

Chemical Mass Balance (CMB) models provide an alternative means of estimation of cooking aerosol (Schauer et al., 1996; Robinson et al., 2006), but the number of studies is relatively few, and some attribute relatively small contributions to cooking aerosol (e.g. Yin et al., 2010). The study by Yin et al. (2015) was unique in making a direct comparison of cooking aerosol derived from a CMB model with an estimate derived from simultaneous measurements by AMS, with application of Positive Matrix Factorization (PMF) to identify source-related factors and quantify their contributions. A comparison of estimates of 24-hour average concentrations derived from the two techniques revealed a close correlation ($r^2 = 0.80$), but a considerably higher estimate from the AMS data (y) than from CMB (x), with a regression equation of $y = 2.24x – 0.33\ \mu g\ m^{-3}$. Various possible explanations for the discrepancy were advanced by Yin et al. (2015), the most plausible appearing to be that cooking organic aerosol is collected with close to 100% efficiency by the AMS, rather than the normally assumed efficiency of 50%, accounting for a possible over-estimation of the AMS method by a factor of two. Such high collection efficiencies for particles from cooking are reported by Pandis (2016). However, the CMB model approach has weaknesses, and is liable to give incorrect source contribution estimates if the adopted source profile input to the model does not well match that in the actual emissions from the cooking source.

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

EXPERIMENTAL

Sampling from Cooking Experiments

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

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

Table 1: Cooking styles and food options selected.

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

Sampling in the Atmosphere

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

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

Chemical Analysis of Samples

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

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

Chemical Mass Balance (CMB) Modelling

Chemical Mass Balance modelling used the USEPA version 8.2 CMB model, in a manner following that outlined by Pant et al. (2014), to estimate contributions to $PM_{2.5}$-OC. The source profiles adopted were these described in Yin et al. (2010) and Yin et al. (2015). The UK-derived road traffic profile derived by Pant et al. (2014) from a twin-site study was employed. The various cooking source profiles derived in this study were each used individually in model runs and the CMB model outputs were used to assess the quality of fit to the atmospheric composition measurements. In all cases, the profiles derived from cooking with gas were used, as this is more widely used in local restaurants/take-aways, and is also available and widely used in local domestic properties.

RESULTS AND DISCUSSION

Measurement of Source Profiles

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

Table 2: Total concentrations of compounds (alkane, PAH, sterol, glyceride and acids) at cooking source (µg/m^3).

(µg/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

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

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

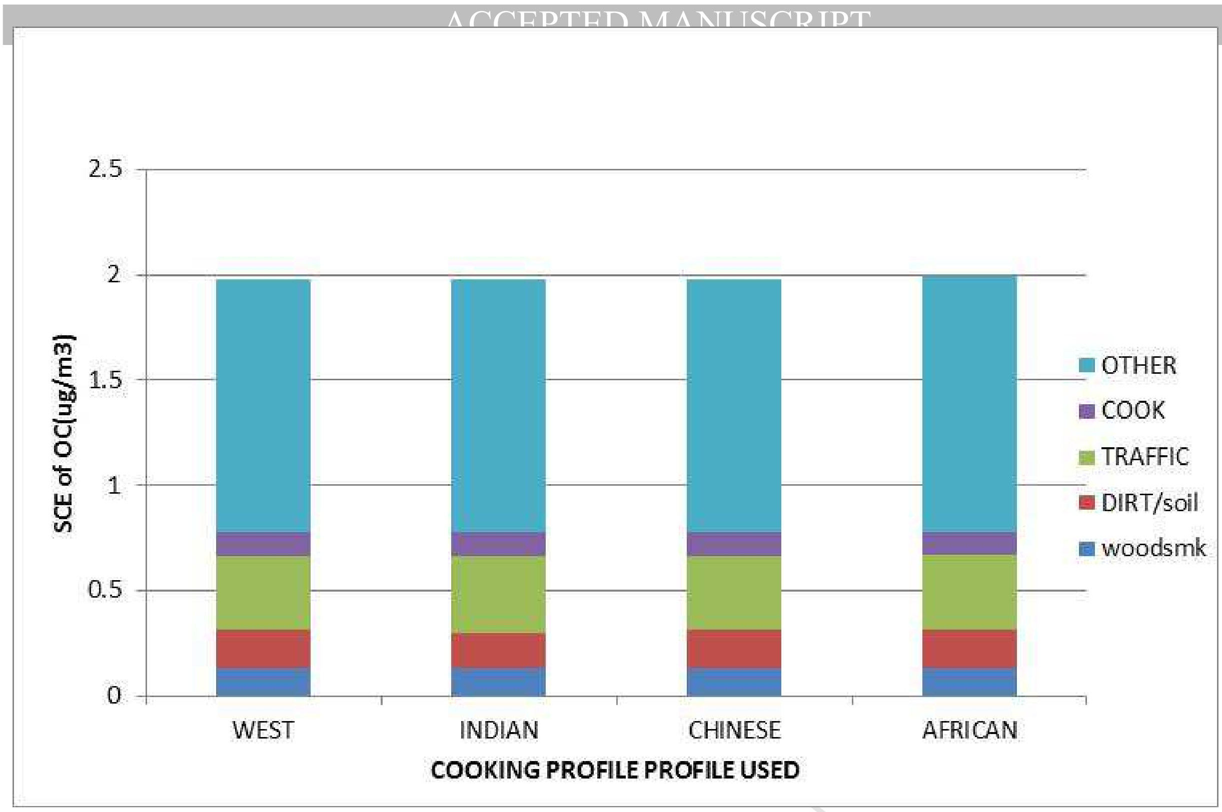

Figure 2: Average source contribution estimates for OC according to style of cooking source profile.

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

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

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)

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

Ots et al. (2016) have used AMS measurements of cooking aerosol to estimate a source strength, from which concentrations across the UK have been modelled. Their model predicts a mean concentration of COA in 2012 of 0.5 $\mu g\ m^{-3}$ for the model grid cell showing highest concentration. The annual mean $PM_{2.5}$ at Birmingham, Acocks Green in 2012 was 11 $\mu g\ m^{-3}$. If the cooking aerosol estimated for Stratford Road by CMB is scaled by 11/6.9 to make it equivalent to mean annual conditions for 2012, the concentration is 0.35 $\mu g\ m^{-3}$ (taking the mean from all cooking styles). Given the results of comparison of AMS and CMB by Yin et al. (2105) and the possible over-estimation of COA by AMS by a factor of up to two, discussed in detail by Ots et al. (2016), the scaled concentration of 0.35 $\mu g\ m^{-3}$ compares well with the model estimate of 0.5 $\mu g\ m^{-3}$.

CONCLUSIONS

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

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

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TABLE LEGENDS

Table 1: Cooking styles and food options selected.

Table 2: Total concentrations of compounds (alkane, PAH, sterol, glyceride and acids) at cooking source (µg/m^3).

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

FIGURE LEGENDS

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

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

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