

## CARBONACEOUS PARTICLES EMITTED FROM COOKING ACTIVITIES IN PORTUGAL

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### ABSTRACT

PM<sub>2.5</sub> samples were collected from the exhaust stacks on the roofs of a university canteen, a charcoal-grilled chicken restaurant and a wood-oven roasted piglet restaurant. The use of thick exhaust hood filters and also gas and electricity as energy sources for cooking in the kitchen of the university canteen contributed to the emission of much lower PM<sub>2.5</sub> levels (24-127 µg m<sup>-3</sup>) compared to the restaurants. The charcoal-grilled chicken restaurant emitted particles at concentrations from 26 to 127 mg m<sup>-3</sup>. In the exhaust of the wood-oven roasted piglet restaurant, PM<sub>2.5</sub> ranged from 192 mg m<sup>-3</sup>, when vine cuttings were used to fire up the ovens, to 203 µg m<sup>-3</sup>, at the final stage of roasting. Organic carbon (OC) represented, on average, 49% of the PM<sub>2.5</sub> mass in samples from the canteen, while elemental carbon (EC) accounted for 1-6%. The OC and EC contents in aerosols from the charcoal-grilled chicken restaurant ranged from 73 to almost 95% and from 0.4 to 1%, respectively. Very high organic contents in the smoke exhausted from the piglet restaurant were also determined. However, the EC mass percentages in PM<sub>2.5</sub> from this restaurant are higher than those found in samples from the other establishments.

**Keywords:** Portugal, Cooking Emissions, PM<sub>2.5</sub>, Organic Carbon, Elemental Carbon

### 1. Introduction

Atmospheric particles are of major scientific interest due to their demonstrated role in climate change and their effect on human health (Calvo *et al.*, 2013; Silva *et al.*, 2013). Current European Directive on Air Quality 2008/50/CE targets the mass concentration of airborne particulate matter under 2.5 µm (PM<sub>2.5</sub>). The particle threshold values are frequently exceeded in many urban areas (e.g. Eeftens *et al.*, 2012; Maraziotis *et al.*, 2008; Triantafyllou and Biskos, 2012). Source apportionment studies performed worldwide indicated that, along with vehicle and biomass burning emissions, cooking activities may represent an important source of PM<sub>2.5</sub>, which have been ignored until now (Alves *et al.*, 2012; McDonald *et al.*, 2003; Mohr *et al.*, 2012; Robinson *et al.*, 2006). In addition, it has long been known that cooking can create high concentrations of aerosol indoors (Abdullahi *et al.*, 2013). It has been demonstrated recently that the health effects of cooking emissions are relevant to people across the globe (Kim *et al.*, 2011). Some emission profiles have been obtained for American (McDonald *et al.*, 2003; Mohr *et al.*, 2009; Schauer *et al.*, 1999a,b), and Chinese cooking styles (Chen *et al.*, 2007; He *et al.*, 2004, 2010; Li *et al.*, 2003; Zhao *et al.*, 2007). However, emissions depend strongly on the cooking method and food ingredients, and nothing is known about particle emissions from the Mediterranean cuisine.

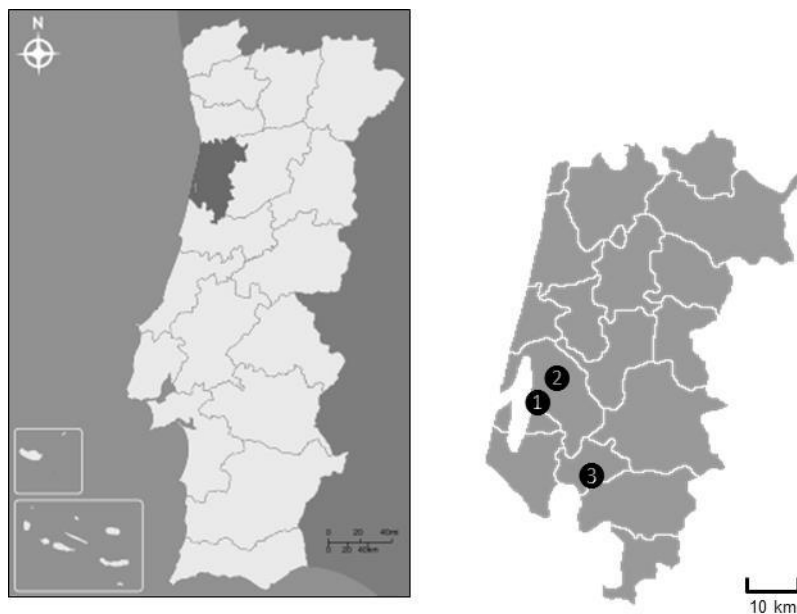
The main goal of this study consisted in obtaining particle concentrations and the respective carbonaceous

content in the exhaust fumes from representative restaurants serving traditional Portuguese meals. Quantitative knowledge of individual source emissions to the atmosphere is crucial to devise cost-effective abatement strategies.

## 2. Methodologies

### 2.1. Sampling

In 2012, low-volume PM<sub>2.5</sub> samples were collected from the exhaust stacks on the roofs of a university canteen, a charcoal-grilled chicken restaurant and a wood-oven roasted piglet restaurant (Figure 1), during the preparation of lunches and/or dinners. The roof heights were 9, 5 and 6 m, respectively. The University of Aveiro owns 4 canteens where 5000-6000 meals are daily prepared. The selected canteen serves about half of the total meals. The cooking equipment is electrically or gas powered. The island canopy exhaust hoods possess 10 cm thick filters.



**Figure 1.** Location of the 3 cooking establishments in the Aveiro district:

- ① university canteen (Aveiro municipality), ② charcoal-grilled chicken restaurant (Aveiro municipality), and ③ wood-oven roasted piglet restaurant (Oliveira do Bairro municipality)

Throughout Portugal there are numerous restaurants located in towns, cities, and also by roadside on national highways offering the typical fare of barbecued chicken. The selected restaurant provides table service and takeaway. The wall-mounted canopy hood does not contain any filtration system. A fan is installed at the termination point of the round chimney.

Despite being made across the country, the Bairrada region, in central Portugal, is specialised in serving up spit-roasted, crispy skinned and pepper-spiced piglets to thousands of visitors. In this region, around 15 tons of sucking pigs are eaten a day. Piglets are roasted in brick ovens. As in most restaurants of this kind, the selected establishment did not have any mechanical exhaust system. Ventilation for hot flue gases or smoke is provided by a simple masonry, parallelepipedic in shape, chimney stack.

The PM<sub>2.5</sub> samples were collected onto pre-baked (6 h at 500 °C) 47 mm diameter quartz filters at a flow of 2.3 m<sup>3</sup> h<sup>-1</sup> using 4 Echo TCR Tecora samplers, following the EN 14907 norm. The regression slopes for the results obtained from parallel measurements involving the various samplers over one week, before the sampling campaigns, differed by less than 5%. The correlation coefficients were in the range 0.975-0.992. Two pairs of parallel samples were collected directly from the exit of each exhaust during variable periods, depending on the smoke loads. Two filters were collected in parallel with the aim of obtaining enough

amount of sample to allow, in a near future, the speciation of organic compounds by chromatographic techniques. However, due to malfunctioning of one of the samplers, which had to be replaced by another, the first three samples of the piglet restaurant do not have the corresponding pair. In the case of the university canteen, the sampling campaign lasted for several weeks and collection times ranged from about 2 to 12 hours. The longest periods are associated with sampling of fumes from the preparation of boiled dishes, for which it was necessary to accumulate the emissions of two or three lunches and/or dinners onto the same filters. Because of the very dense smoke emitted through the stacks, sampling times for the other two establishments ranged from 5 to 75 minutes. Samples were taken at ambient temperature, in the case of the university canteen. For the two restaurants, the smoke temperatures were between 40 and 50 °C at the vent of exhausts. All sampled volumes were corrected to standard conditions. Taking into account that the sampling heads were deployed at the vent of exhausts, the organic carbon contents represent lower limits of the true atmospheric levels since, as the emissions are cooled and diluted, gas-to-particle partitioning is likely to occur.

## 2.2. Analytical techniques

Before weighing, the filters were conditioned in a desiccator at least for 24 h in a temperature- and humidity-controlled room. Previous to and after sampling, gravimetric determination was performed with a microbalance Mettler Toledo AG245 (readability 0.1 mg/0.01 mg). Filter weights were obtained from the average of several measurements, with weight variations less than 5%.

After gravimetric determination of the PM<sub>2.5</sub> mass, samples were analysed by a thermo-optical transmission system in order to obtain the carbonaceous content (OC and EC). The thermo-optical system comprises a quartz oven with two distinct heating zones. It also includes a helium-neon laser (632.8 nm) and a detector connected to a transducer. Associated with the laser, there is a chopper whose function is to eliminate interferences that may occur due to the existence of other light sources. To detect the amount of carbon in various stages of heating, the system encompasses a calibrated infrared non-dispersive CO<sub>2</sub> analyser. The temperature programme is imposed by a controller. To control the flow of gases (N<sub>2</sub> and O<sub>2</sub>) passing through the oven, a mass flow meter is used. A computer terminal makes the data acquisition every second, recording various parameters, such as temperature, flow rate, and CO<sub>2</sub> concentrations. For each filter, two 9 mm diameter punches were used in each analytical run. For samples of the chicken restaurant, due to the very high loads and saturation problems, 5 mm diameter punches were used. Thus, in this last case, the results may be more imprecise. At least, three replicate analytical runs were applied to each filter. To start the analysis, a purge to remove all traces of CO<sub>2</sub> is carried out until zero is read by the analyser. The data acquisition can be started as soon as the purge is completed. The analysis is done in two stages. In the first phase, the sample is subjected to an anoxic environment with the presence of only nitrogen. Controlled heating in these anoxic conditions is performed to separate OC into two fractions of increasing volatility. The first fraction corresponds to the volatilisation at T<200 °C of lower molecular weight organics. The second fraction is related to the decomposition and oxidation of higher molecular weight species at temperatures ranging from 200 to 600 °C. The last fraction of OC is identified by transmittance and corresponds to pyrolysed organic carbon (PC) produced in the previous heating steps. The second stage starts by opening a valve that introduces air into the oven. Oxygen will join the existing stream of nitrogen and will transform the atmosphere of the first heating zone in an oxidising atmosphere. This factor, coupled with a further increase in temperature, promotes the oxidation and volatilisation of EC. The last carbon fraction contains initial EC plus OC that has pyrolysed during heating under an inert atmosphere. The interference between PC and EC can be controlled by continuous evaluation of the blackening of the filter using a laser beam and a photodetector measuring the filter light transmittance. The split between the PC and EC is assigned when the initial (baseline) value of the filter transmittance is reached. All carbon removed before the split is considered organic, and that removed after the split is considered elemental. The walls of the second heating zone are coated with a catalyst (CuO), which guarantees the conversion of any carbonaceous material to CO<sub>2</sub>.

### 3. Results and discussion

The use of either thick exhaust hood filters, which were changed at the beginning of the first day of sampling, or gas and electricity as energy sources for cooking in the kitchen of the university canteen, contributed to the emission of much lower PM<sub>2.5</sub> levels compared to the restaurants (Table 1). Irregular wind flows in some few days on the roof of the canteen may have contributed to an uneven distribution of particles between parallel filters, resulting in higher dispersion around the means. The PM<sub>2.5</sub> samples from the canteen exhausts presented concentrations between 24 and 127  $\mu\text{g m}^{-3}$ . The highest concentrations were obtained in samples from the fry and stew stations, while the lowest corresponded to the boiled food station. The charcoal-grilled chicken restaurant emitted particles at concentrations from 26 to 127  $\text{mg m}^{-3}$ , averaging 71  $\text{mg m}^{-3}$ . In the exhaust of wood-oven roasted piglet restaurant, PM<sub>2.5</sub> ranged from 52-78  $\text{mg m}^{-3}$ , when vine cuttings and pine slats were used to fire up the brick ovens in which piglets were roasted, to 203  $\mu\text{g m}^{-3}$ , at the final stage of roasting. This confirms the results reported in a recent review paper in which it was demonstrated that the elevated concentrations derive largely from the combustion of the fuel used for cooking rather than from the cooking itself (Abdullahi *et al.*, 2013). In accordance with other works (Buonanno *et al.*, 2009), in the present study it was also observed that, in general, foods containing a higher percentage of fat generate higher emission rates than those with less fat percentage. See and Balasubramanian (2008) analysed various cooking methods (steaming, boiling, stir-frying, pan-frying and deep-frying). The authors observed that the highest concentration of PM<sub>2.5</sub> measured at 20 cm from the cooker was produced during deep frying (190  $\text{mg m}^{-3}$ ) and the lowest level was registered during steaming (72  $\mu\text{g m}^{-3}$ ). It has been also demonstrated that cooking with oil contributes to the generation of more particles than cooking with water (Buonanno *et al.*, 2009; He *et al.*, 2004; See and Balasubramanian, 2008). In a comparison of emissions from Chinese, Indian and Malay food stalls (See *et al.*, 2006), it was reported that the highest mass concentrations of PM<sub>2.5</sub> were measured in the Malay stall (245  $\mu\text{g m}^{-3}$ ), while the lowest were observed in the Indian stall (187  $\mu\text{g m}^{-3}$ ). It should be noted, however, that these studies were carried out in the kitchens of residential houses or in domestic kitchens set-up inside a laboratory, and do not refer to stack emissions.

OC represented, on average, 49% of the PM<sub>2.5</sub> mass in samples from the canteen, whilst EC accounted for 1-6%. The OC and EC particle mass fractions from the charcoal-grilled chicken restaurant ranged from 73 to around 95% and from 0.4 to 1%, respectively. Very high organic contents in the smoke exhausted from the piglet restaurant were also determined. However, the EC mass percentages in PM<sub>2.5</sub> from the latter place are higher than those found in samples from the other eateries.

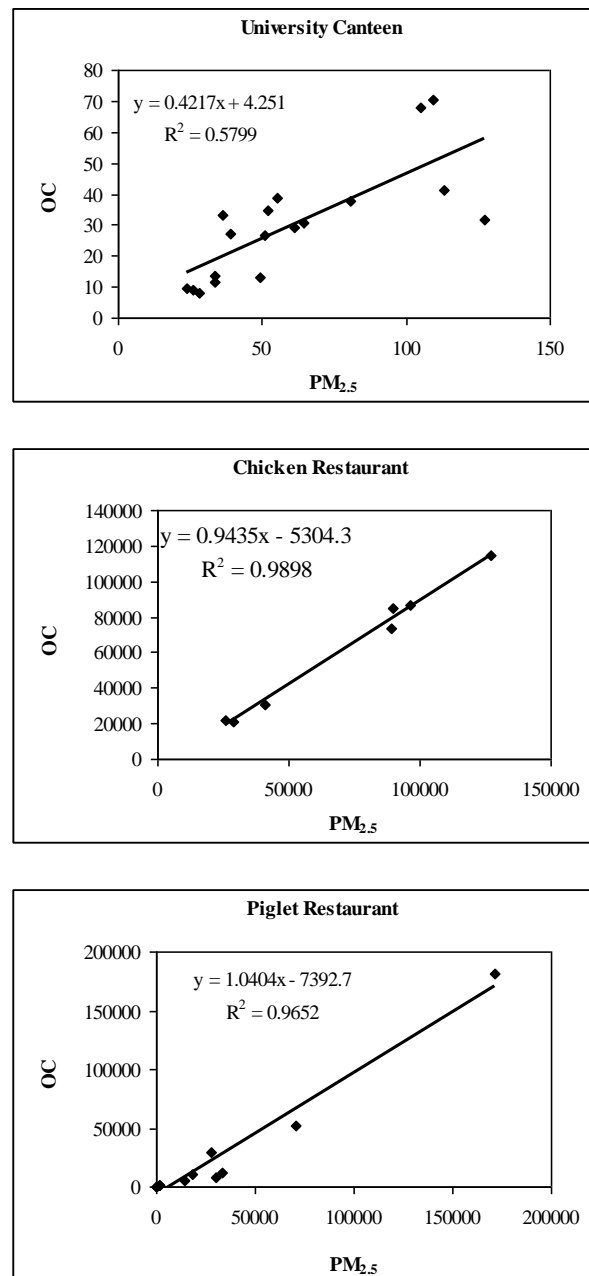
The *r*-square of the correlations between PM<sub>2.5</sub> and OC indicates that this carbonaceous component can explain 58, 99 and 97% of the variation of the particle concentrations in the exhaust of the university canteen, chicken restaurant and piglet restaurant, respectively (Figure 2). Previous studies have also indicated that OC carbon is the major constituent found in cooking fumes (Chow *et al.*, 2004; Kleeman *et al.*, 1999, 2006; Schauer *et al.*, 1999a,b; See and Balasubramanian, 2008). Comparison of different culinary methods (Abdullahi *et al.*, 2013) showed that those using oil release higher concentrations of EC and OC than those using boiling (i.e. boiling and steaming).

In a recent study in a rural area in the Indo-Gangetic-Plains, it was observed that black carbon (BC) levels, both indoors and outdoors, have anomalously large twice-daily peak concentrations reaching 60  $\mu\text{g m}^{-3}$  for indoor and 30  $\mu\text{g m}^{-3}$  for outdoor, during the early morning, 05:00-08:00, and early evening, 17:00-19:00, coinciding with the morning and evening cooking hours (Rehman *et al.*, 2011). The peak indoor BC concentrations reached as high as 1000  $\mu\text{g m}^{-3}$ . The large diurnal peaks, due to cooking activities, seen in this research lead to the conclusion that satellite based aerosol studies that rely on once-daily daytime measurements may severely underestimate the BC loading of the atmosphere. The concentration of OC was a factor of 5 larger than BC and furthermore optical data show that absorbing brown carbon was a major component of the OC. It should be stressed that the term EC or BC is defined operationally in the literature and refers to the same absorbing component (i.e. dark-coloured "soot") in carbonaceous aerosols.

**Table 1.** Concentrations of particles, organic carbon and elemental carbon in the stack exhausts from cooking operations

Description	PM <sub>2.5</sub>	OC	EC
UNIVERSITY CANTEEN ( <i>n</i> = 20; units: $\mu\text{g m}^{-3}$ )			
Boiled chicken with vegetables	41±11	14±0.33	1.8±0.27
Fried fish; seafood rice	97±23	39±2.5	1.4±0.32
Veal stroganoff stew	63±2.3	30±0.79	1.5±0.34
Boiled pork meat with vegetables	27±1.5	8.5±0.57	1.0±0.19
Roasted ribs	127±2.0	33±0.58	6.0±0.01
Alentejo pork meat with clams; stewed pork chops with sausages	45±8.1	27±0.28	1.8±0.43
Grilled pork	107±3.1	69±1.8	1.2±0.58
Beef with rice; pork with boiled potatoes	29±6.7	10.4±1.3	1.7±0.66
Grilled and boiled fish	44±11	34±1.1	1.3±0.02
Chicken stew; turkey stew with rice	55±4.0	39±1.2	2.1±0.01
CHARCOAL-GRILLED CHICKEN RESTAURANT ( <i>n</i> = 7; units: $\text{mg m}^{-3}$ )			
Smoke-roasted chicken	71±39	62±37	0.37±0.12
WOOD-OVEN ROASTED PIGLET RESTAURANT ( <i>n</i> = 11)			
Over preheating with remains of pruning vines and pine slats	70 $\text{mg m}^{-3}$	52 $\text{mg m}^{-3}$	7.5 $\text{mg m}^{-3}$
Burning of pruning vines and pine slats (flaming)	31 $\text{mg m}^{-3}$	28 $\text{mg m}^{-3}$	1.24 $\text{mg m}^{-3}$
Pine slats burning like in a fireplace (door open) after refueling; flaming phase and beginning of smouldering phase	192 $\text{mg m}^{-3}$	178 $\text{mg m}^{-3}$	9.24 $\text{mg m}^{-3}$
Smouldering and glowing embers	16.0±2.58 $\text{mg m}^{-3}$	8.13±3.07 $\text{mg m}^{-3}$	5.02±0.739 $\text{mg m}^{-3}$
Tray with rice soaked in chicken's blood; glowing embers at the bottom; door open occasionally to spread ashes on the embers	31.4±2.24 $\text{mg m}^{-3}$	10.3±2.29 $\text{mg m}^{-3}$	5.02±0.739 $\text{mg m}^{-3}$
Three piglets in the oven; glowing embers at the bottom; door open	1393±142 $\mu\text{g m}^{-3}$	809±106 $\mu\text{g m}^{-3}$	107±21 $\mu\text{g m}^{-3}$
Final phase of roasting; oven door closed	207±6.1 $\mu\text{g m}^{-3}$	158±29 $\mu\text{g m}^{-3}$	< 5 $\mu\text{g m}^{-3}$

Destructive analytical techniques (such as thermal-optical method) that use the thermal resistance, oxidative resistant and chemical inert nature of highly polymerised graphitic-like fraction of soot termed it as EC. Non-destructive analytical techniques (such as optical methods) that use the light absorbing characteristic of soot termed it as BC. The definition of BC includes EC as well as all components in the soot that absorb light such as some OC fractions (known as brown carbon) that show strong absorption in near-UV region of solar spectrum (Andreae and Gelencser, 2006).



**Figure 2.** Relationships between the particle and organic carbon concentrations in the exhaust emissions. Values are given in  $\mu\text{g m}^{-3}$ .

The OC/EC ratio can be helpful in differentiating between aerosol sources. Typically mean OC/EC ratios for fossil fuel dominated aerosol in urban cities (range from about 1 to 3), while in regions impacted by biomass burning emissions these ratios are substantially higher (Novakov *et al.*, 2000). OC and EC measurements performed in a busy roadway tunnel in central Lisbon showed an OC/EC ratio in the range of

0.3-0.4, which reflected the composition of fresh vehicular exhaust emissions (Pio *et al.*, 2011). Schauer *et al.* (2001) reported OC/EC values in the range 16.8-40.0 for wood combustion. Schmidl *et al.* (2008) found OC/EC ratios between 2.7-3.3 for hard- and 2.6-5.7 for softwood during the combustion of mid-European Alpine species in a domestic tiled stove. Values ranging from 3.99 to 52.6 were obtained by Gonçalves *et al.* (2011) in PM<sub>2.5</sub> emissions from fireplace and woodstove combustion of typical Portuguese wood species. Roden *et al.* (2006, 2009) reported OC/EC ratios between 0.5 and 13 (mean value of 2.3) during field measurements from a traditional Honduran wood-fired cookstove. The authors argued that the large amount of EC emitted during cook stove combustion may not be able to distinguish between fossil and biofuel combustion aerosols when cooking with biofuel is dominant.

High OC/EC ratios (19.3±15.7) were obtained in our study for the cooking emissions of the university canteen. Much higher ratios, between 76.9 and 247 (average = 155), were found in the very greasy fumes of the charcoal-grilled chicken restaurant. Since chicken skin is very high in fat, cooking it on an underfired charbroiler yields high amounts of OC, which superimpose the EC emitted from the combustion of charcoal. OC/EC values in the range 2.5-10.5 have been reported for residential coal burning (Chen *et al.*, 2006). Emissions from the piglet restaurant produced an average OC/EC of 16.3 once the thin vine twigs were lit, and small pieces of pine wood were added, achieving a very intense flaming phase. The ratio decreased to values of 1-2 after heating the oven up to high temperatures and glowing coals covered the bottom. During the roasting process values around 8 were achieved. A large fork is used to place the pig's carcass in the oven, while hot water is applied to it to cause the skin to tighten. The piglet is roasted at high temperature until meat is tender and skin is crispy. Ratios from 32.9 to 81.6, i.e. between those of the piglet and chicken restaurants, have been described in the literature for kitchen emissions (He *et al.*, 2004).

#### 4. Conclusions

Commercial cooking is a surprisingly large source of particle emissions that could pose risks to human health and the environment. While that mouth-watering smell may whet our appetite, emissions likely involving hazardous pollutants in the smoke from the cooking processes are ejected into the air that we breathe. Mechanical and environmental engineering solutions aiming at mitigating emissions recommend the combined application of air pollution control equipments (e.g. grease filter, electrostatic precipitator, hydrovent, etc.) together with a suitably located exhaust outlet. The outlets should be located at a place where ventilation is good and sufficient separation distance from the neighbourhood is maintained. The emission from the exhaust system should be adequately dispersed and should not be restricted or deflected by any plates, caps or other obstacles.

The emissions of cooking aerosol are highly diverse, depending upon factors such as the raw-food composition, cooking temperature, cooking style and energy sources. Although it is possible to clearly distinguish the OC/EC ratios of vehicle exhausts from those of cooking emissions, the separation between the latter and biomass burning is tricky, because overlapping of values may occur.

While further knowledge the chemical composition of cooking emissions would be beneficial, quantitative source apportionment will remain imprecise and potentially inaccurate in situations with mixed cooking source types contributing to atmospheric concentrations. This provides a considerable challenge to receptor models, such as Chemical Mass Balance (CMB), where it is necessary to input a source profile.

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