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Fingerprinting and emission rates of particulate organic compounds from typical restaurants in Portugal

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Abstract

The aim of this study was a detailed chemical characterisation of the particles released during the preparation of popular Portuguese dishes. $PM_{2.5}$ 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 speciation of organic compounds was carried out by gas chromatography-mass spectrometry. The canteen was responsible for the lowest emissions of $PM_{2.5}$, while emissions from the roasted piglet restaurant were the highest. Naphthalene was quantified as the most abundant aromatic compound in particle emissions from the canteen, while phenanthrene, fluoranthene, pyrene and chrysene were the dominant polycyclic aromatic hydrocarbons in samples from the other establishments. Benzo[a]pyrene equivalent concentrations obtained for the charcoal-grilled chicken and piglet restaurant indicate a dangerous carcinogenic potential to human health. Cholesterol was the prevalent sterol. Its highest values were obtained in particles from the charcoal-grilled chicken restaurant (621±233 µg g^{-1} $PM_{2.5}$). Oleic and palmitoleic were the unsaturated fatty acids identified at highest concentrations (from trace levels to 34.4 and to 6.89 mg g^{-1} $PM_{2.5}$, respectively). Resin acids, such as dehydroabietic and abietic, were detected in all samples from the wood-oven roasted piglet restaurant. Nicotinamide was the amide detected at highest amount in emissions from the university canteen during the preparation of stews (7.67 mg g^{-1} $PM_{2.5}$). Levoglucosan and its isomers were identified in all samples from the roasted piglet restaurant, but only the first monosaccharide anhydride was present in emissions from the university canteen and the charcoal-grilled chicken restaurant. Additionally, emission rates were estimated for the most representative compounds, taking into account the specific activity of each restaurant.

Keywords: Cooking emissions, $PM_{2.5}$, organic compounds, PAHs, emission rates

Introduction

Atmospheric particles remain a growing field of scientific interest due to their established role in climate change and their effect on human health. Besides traffic and biomass burning for heating purposes, cooking emissions have been found to represent a great contributor to atmospheric pollution levels, and especially to particulate matter (PM) (Crippa et al., 2013; Guo, 2012; McDonald et al., 2003; Robinson et al., 2006; Wang et al., 2009; Zhao and Zhao, 2018), although in most areas, to date, this source has been overlooked. Cooking emissions were estimated to account for up to 40% of the total organic mass of PM in several cities, e.g. Paris (Crippa et al., 2013), London and Manchester (Allan et al., 2010), Barcelona (Mohr et al., 2012), Zurich (Canonaco et al., 2013; Lanz et al., 2007), Toronto (Slowik et al., 2010), Beijing (Huang et al., 2010), New York (Sun et al., 2011), Pittsburgh (Robinson et al., 2006), Pasadena (Zotter et al., 2014) and Fresno (Ge et al., 2012).

It has been demonstrated that the health effects of cooking emissions are relevant to people across the world (Kim et al., 2011). Pollutants from food mainly result from heating and cooking operations through which organic substances in the food are volatilised. The amount and composition of pollutants emitted from those sources, and consequently the health effects, would greatly depend on the cooking stuff, cooking styles and even on cooking fuel (Kabir and Kim, 2011). Cooking activities emit other constituents, namely volatile organic compounds (Alves et al., 2015), polycyclic aromatic hydrocarbons (PAHs) (Abdel-Shafy and Mansour, 2016; Abdullahi et al., 2013; Chen et al., 2019; Munyeza et al., 2020), fatty acids and carbonyls (He et al., 2004a; Peng et al., 2017; Schauer et al., 2002; Zhao et al., 2007a). Schauer et al. (2002) demonstrated that, using different seed oils in the preparation of food, compositional differences in the emissions from distinct cooking operations are originated. Composition of cooking emissions can be largely affected by the food ingredients. He et al. (2004a) detected β-sitosterol, stigmasterol, levoglucosan and azelaic acid in emissions from Chinese cooking, but these compounds were absent in samples from American meat cooking. See et al. (2006) reported higher emissions of PAHs from Malay food stall compared to those from both Chinese and Indian food stalls. The differences in PAH emissions were associated with the deep-frying method used and the quantity of food cooked. PAHs have been a main concern due to their reported carcinogenic and mutagenic characteristics (Abdel-Shafy and Mansour, 2016; Li et al., 2003). Epidemiological investigations have revealed that exposure to PAHs contribute significantly to the development of human cancers (Rengarajan et al., 2015; and references therein). PAHs, when emitted from sources, can be dispersed over considerable distances before deposition, which has raised global concern (Kim et al., 2013; Ravindra et al., 2008). Chen et al. (2012) measured PAHs from cooking emissions in the exhaust stacks of Chinese, Western and barbecue restaurants, and evaluated the incremental lifetime cancer risk for people directly exposed in the receptor area during a substantial period of time. Extremely high incremental lifetime cancer risks were obtained.

Emissions depend greatly on the cooking method and food ingredients, but almost nothing is known about the composition of fumes from the Latin cuisine or other styles. Some chemical emission profiles were obtained for American (McDonald et al., 2003; Roe et al., 2005; Schauer et al., 2002) and Chinese (He et al., 2004a; Zhao and Zhao, 2018) cooking styles. Cooking with rapeseed and soybean oil at high temperatures in woks is typical in the Chinese cuisine. One of the main differences between the USA and Mediterranean diets lies in the fact that Americans eat more red meat, while consumption of fish, poultry, vegetables and olive oil is prominent in Latin countries (Abdullahi et al., 2013).

Receptor models, which attribute observed concentrations to sources through statistical and/or meteorological interpretation of data, are based on mass conservation (or balance) of chemical species (Hopke et al., 2003). In the USA, the Chemical Mass Balance (CMB) model is routinely used. It requires *a priori* knowledge of major sources and their emission characteristics in the study area (Amato et al., 2009; Cesari et al., 2016; Karanasiou et al., 2009). CMB has been rarely applied in Europe and Latin America due to poor knowledge of the composition of particles from specific sources. Thus, due to the lack of a detailed chemical characterisation of emissions from cooking in Latin countries, the application of CMB to environmental databases is unfeasible and, thus, it is very difficult to estimate the contribution of this source to the atmospheric levels. Yin et al. (2010) applied emission profiles obtained for the USA to measurements carried in the UK, but no mass concentration could be assigned to particles from cooking. In 2015, the same authors applied source profiles obtained from Western-style fast food cooking to receptor modelling of fine particles in southern England, stressing the need of obtaining regional source profiles (Yin et al., 2015). In Europe, receptor modelling using Aerosol Mass Spectrometer (AMS) data has been applied to determine cooking contributions to ambient organic aerosols (Crippa et al., 2013; Mohr et al., 2012). However, difficulties have been reported when using AMS datasets to separate cooking from other source emissions (Crippa et al., 2014; Mohr et al., 2009; Xu et al., 2015). Therefore, in order to minimise the uncertainties resulting from AMS source apportionment linked to cooking emissions, the development of new source emissions profiles for cooking activities to be used in the CMB method is essential (Dall'Osto et al., 2015; Reyes-Villegas et al., 2018; Xu et al., 2021; Yin et al., 2015). It must be taken into account that the emission profiles obtained with continuous instruments, such as AMS, cannot be used as input to source apportionment models when they are applied to long-term environmental databases based on the collection of PM on filters. Moreover, a large collection of research papers has been focused on characterising emissions from different cooking styles, especially Chinese cuisine, and their effects on indoor air quality, as shown in several review articles on the topic (Abdullahi et al., 2013; Kim et al., 2011; Torkmahalleh et al., 2017; Zhao and Zhao, 2018; Zhao et al., 2019). However, these databases can also not be directly compared to those obtained in the outdoor air or at the exit of the chimneys.

The innovative character of the present work lies in the development of so far unavailable Portuguese source emission profiles for cooking that can be used in source assignment in the outdoor air. Such information is important as it appends to the emergent speciation profiles for major sources in Europe (SPECIEUROPE). Since its release, new source profiles have been added to the SPECIEUROPE database. However, of the ca. 200 profiles that currently comprise SPECIEUROPE distributed across different source categories, none of them concerns cooking emissions. The aim of this study was to characterise the emissions from different cooking styles, to roughly calculate the emission rates and to estimate the carcinogenic potency associated with particle-bound PAHs. As far as we know, nothing is known about the organic speciation of particle emissions from different methods of preparing dishes that are common to several Latin countries. This work complements previously reported information for volatile organic compounds (Alves et al., 2015) and carbonaceous particles (Alves et al., 2014) emitted by the same cooking activities.

Methodology

Sampling

In May and June 2012, samples were collected on the roofs of a university canteen, a charcoal-grilled chicken restaurant and a wood-oven roasted piglet restaurant, all located in the region of Aveiro, Portugal. The University of Aveiro has four canteens that are responsible for preparing 5000-6000 daily meals. Sampling was conducted near the vents of the main canteen during the preparation of lunches and dinners. Barbecue chicken is a central recipe to many restaurants in Portugal. It is served all over the country but is also very popular in states with Portuguese communities, such as Canada, Australia, the United States, Venezuela, Brazil and South Africa. According to National Statistics, the annual internal consumption of chicken meat is around 300 kton, which illustrates the high demand for this kind of food. Portugal is the largest per capita consumer of poultry in Europe. On average, the annual per capita consumption is 37.5 kg compared with 22.5 kg in other EU countries. The restaurant chosen for this study provide table service and take away. The circular-shaped chimney is equipped with an electric draft inducing fan but does not have any filtration unit or other treatment system of flue gases. The third restaurant covered in this study is specialised in another typical Portuguese dish: spit-roasted, crispy skinned and pepper-spiced piglet. The suckling pig is found on the menu all over the country, but the Bairrada wine region in central Portugal is best known for this speciality, which was nominated one of the seven wonders of the Portuguese cuisine. Traditional ovens are fuelled initially with wood and then with branches pruned from local vines, which impart a subtle but definite flavour to the meat. According to calculations of the sector, two to three thousand piglets (about 15 tons) per day are sold in the restaurants of the Bairrada region, generating more than 87 million euros per year. During sampling in the chimney of the selected restaurant, the collection of PM filters was segregated according to the various phases of preparation of the dish. Thus, sequential filters were sampled to distinguish between the oven heat-up phase and the roasting phase. The chimney has a parallelepiped configuration and does not have any forced gas extraction or filtration system.

Particulate matter lower than 2.5 µm ($PM_{2.5}$) was collected on 47 mm diameter quartz fibre filters previously baked at 500 ºC for 6 hours using Echo TCR Tecora samplers with a standard European inlet head. For the 3 food establishments, two pairs of parallel samples were collected directly from the exit of exhausts during variable periods, depending on the smoke loads. In the case of the university canteen, the sampling campaign lasted for several weeks with sampling times varying from around 2 to 12 hours. The longest samplings were carried out during the preparation of boiled dishes, for which it was required to accumulate the emissions of two or three lunches and/or dinners onto the same filters. Due to the heavy emission loads, sampling times for the other two establishments oscillated between 5 and 75 minutes. Two to five replicate samples were taken for the emissions from the preparation of each type of menu.

Analytical techniques

The gravimetric determination was performed using a microbalance (Metter Toledo AG245 with readability 0.1 mg/0.01 mg). Filter weights were obtained from the average of at least six readings, with weight variations between them of less than 5%. Before weighing, the filters were conditioned for 24 h, in a room with controlled temperature and humidity (50% and 20 ºC). In addition, some control filters (blank filters) are kept in the weighing room, which are randomly weighed on different days to check that conditions have not changed significantly.

Punches of replicate filters were joined into a pooled sample. Each one was extracted by refluxing 300 mL of dichloromethane for 24 hours and then extracted 3 times with methanol (75 mL for 10 min, each extraction) in an ultrasonic bath. After filtration, all the 4 extracts were combined, vacuum concentrated and dried under a gentle nitrogen stream. The total organic extracts were separated into 5 different organic fractions by flash chromatography in a silica gel (230 – 400 mesh, 60 Å Merck Grade 9385) column using various solvents of increasing polarity (Alves et al., 2011). After separation of components, the resulting 5 organic fractions were vacuum concentrated and evaporated by a gentle ultra-pure nitrogen stream. Before chromatographic injection, fractions that included compounds with hydroxylic and carboxylic groups were derivatised in sealed vials with *N,O*-bis(trimethylsilyl)trifluoroacetamide (BSTFA) + trimethylchlorosilane (TMCS) (99:1, Supelco). All the organic extracts were analysed by gas chromatography-mass spectrometry (GC-MS). The GC-MS calibration was based on a total of 200 standards in different concentration levels with relative response factors determined individually for most compounds. Compound identification was performed by comparison with authentic standards, using the Wiley and NIST spectral libraries and analysis of fragmentation patterns. For those compounds for which no authentic standards were available, relative response factors were obtained as an average of the relative response factors from the overall homologous series or from compounds of similar chemical structure and retention time. Standards and samples were both co-injected with two internal standards: tetracosane-D_{50}, 1-chlorohexadecane or 1-chlorododecane. For the extracts containing PAHs, the EPA 8270 semi-volatile internal standard mix, with six deuterated compounds, was used: 1,4-dichlorobenzene-D_4, naphthalene-D_8, acenaphthene-D_{10}, phenanthrene-D_{10}, chrysene-D_{12} and perylene-D_{12}.

Emission rates

The emission rates of organic compounds during operation hours were calculated as follows:

$$\text{Emission rate (mg h}^{-1}\text{)} = \text{concentration (mg m}^{-3}\text{)} \times L \times W \times v \times 3600 \text{ (s h}^{-1}\text{)} \quad (1)$$

where L = exhaust duct height (m), W = exhaust duct width (m) and v = exhaust velocity (m s^{-1}) (Alves et al., 2015; Chen et al., 2007). In the case of circular structures, the expression $L \times W$ is replaced by $(1/4) \times \pi \times d^2$, where d represents the diameter (m). The cross-sectional area of the exhaust of the university canteen, chicken restaurant, and piglet restaurant, was 0.23, 0.79 and 0.72 m^2, respectively. The exhaust velocities for the three facilities were 14, 8.9 and 4.7 m s^{-1}, respectively. These velocities were measured using a Pitot tube.

Estimation of PAH carcinogenic potential

For the calculation of the total B[a]P equivalent (B[a]P_{eq}) concentration for the identified PAHs, the methodology based on the procedures of the Office of Environmental Health Hazard Assessment (OEHHA) of the California Environmental Protection Agency (CalEPA) was followed. The calculation of B[a]P_{eq} concentrations requires the concentration of individual PAHs in each sample and the respective toxicity equivalent factor (TEF) (Alves et al., 2017).

Results and Discussion

Particulate matter

$PM_{2.5}$ concentrations between 27 and 127 µg m^{-3} were obtained in samples from the university canteen (Table 1), whose levels were much lower compared with those of the chicken restaurant (71±39 mg m^{-3}) and wood oven roasted piglet restaurant (from 0.207 to 236 mg m^{-3}). The lower emission levels of the canteen are due to two main reasons: i) use of effective exhaust hood filters, and ii) use of cleaner energy sources, such as gas and electricity. Higher concentrations were recorded for samples from the fry and stew stations compared to the boiled food station. In accordance with previous observations, exhaust emissions from frying operations using electricity as source of energy generated lower $PM_{2.5}$ concentrations compared to the same process using natural gas as fuel (Buonanno et al., 2009).

Concentrations in the exhaust outlet of the charcoal-grilled chicken restaurant were, on average, 71±39 mg m^{-3}. The highest concentrations were recorded at the beginning of the process, when the chickens were still all raw.

The initial heating stage of the brick oven in which piglets were roasted showed a high variation in particulate emissions. However, the remaining stages of preparation of the roasted piglet revealed significantly lower variations. The highest $PM_{2.5}$ levels were obtained during the heating-up phase of the oven, when vine cuttings and pine slats were added to light the fire, reaching a value of 236 mg m^{-3} (Heating Phase - HP). At the final stages of roasting, concentrations decreased to 0.207 mg m^{-3} (Roasting Phase - RP). Abdullahi et al. (2013) argued, in a review paper, that high concentrations derive largely from the combustion of the fuel used for cooking rather than from the cooking itself. It has been shown that particle mass concentrations are higher when cooking with gas equipment compared to electric ranges (Zhao and Zhao, 2018; and references therein). It has been proposed that foods with a great amount of fat have higher particle emissions and therefore more organic carbon (Buonanno et al., 2009; Ramdahl, 1983). Different types of oils also contribute to significant variations in PM emissions. After testing three different types of oils (olive oil, peanut oil and sunflower oil), Buonanno et al. (2009) concluded that cooking with sunflower oil and olive oil produced the lowest and the highest particle emission levels, respectively. See et al. (2006) sampled emissions in the Singapura University for Chinese, Malaya and Indian cuisine. The highest $PM_{2.5}$ emissions were observed for the Malaya (245.3 µg m^{-3}), followed by the Chinese (201.8 µg m^{-3}) and Indian (186.9 µg m^{-3}) cuisines. Discrepancies in particle concentrations were linked to the different cooking styles.

Particulate organic speciation

Aliphatic compounds

Aliphatics, one of the dominant organic classes, were emitted at concentrations of 1.86 and 697 µg m^{-3} by the stacks of the canteen and charcoal-grilled chicken restaurant, respectively, representing $PM_{2.5}$ mass fractions of 52.9 and 8.22 mg g^{-1}. The level of this group of compounds was 55.0 µg m^{-3} when vine cuttings were used to heat-up the oven of the piglet restaurant, whilst at the final stage of the roasting process, the mean concentration was 13.6 µg m^{-3}. These concentrations corresponded to mass fractions of 0.742 and 3.53 mg g^{-1} $PM_{2.5}$, respectively. The *n*-alkane homologous series in the samples from the three places ranged from decane (C_{10}) to octatriacontane (C_{38}). He et al. (2004a) reported a similar *n*-alkane distribution, with homologous ranging from C_{14} to C_{35}, for two types of Chinese cuisine. For Hunan style, the mass concentration of *n*-alkanes varied from 11.5 to 81.7 ng m^{-3}, whilst for Cantonese style emissions ranged from 2.55 to 218 ng m^{-3}. However, these compounds represented only 0.32 to 0.43% of the total quantified organic mass emitted. Little difference has been found by Wang et al. (2015) between the mass fractions of *n*-alkanes in $PM_{2.5}$ from four cooking styles: home cooking, Shandong cuisine, Hunan cuisine and barbecue. Furthermore, the highest concentrations occurred at C_{16}-C_{20}, which represented 61.8-69.0% of the total *n*-alkanes. In the present study, the *n*-alkane homologous series maximised at hentriacontane (68.5 mg g^{-1} $PM_{2.5}$) in samples collected from the university canteen during the preparation of pork with boiled potatoes. The most abundant compounds were in the interval from C_{23} to C_{34}. In emissions from the charcoal-grilled chicken restaurant, *n*-alkane homologous series ranged from undetectable levels to 8.73 mg g^{-1} $PM_{2.5}$ (nonacosane-C_{29}), maximising at C_{29}-C_{35}. Individual mass fractions in samples from the roasted piglet restaurant ranged between untraceable values and 0.792 mg g^{-1} $PM_{2.5}$ (hentriacontane-C_{31} during the roasting phase), peaking for the homologous from C_{22} to C_{32} (Supplementary material, Tables S1 and S2). He et al. (2004a) obtained *n*-alkane distributions maximising at C_{25}, C_{29} and C_{31} in samples emitted from Western-style fast food cooking. As observed in the present work, it has been reported that emissions of *n*-alkanes are severely dependent on the cooking style. Hildemann et al. (1991) documented that the *n*-alkane emission rates increased from frying to charbroiling of meat. Moreover, extra lean meat released less constituents than regular meat. Rogge et al. (1991) found that charbroiling produced three times the mass of *n*-alkanes than frying of meat, while charbroiling regular meat released four times the mass compared to extra lean meat. Thus, it seems that, in addition to the preparation mode, the fat content of meat plays a key role in the emission of these compounds. The homologous series of *n*-alkanes emitted from Chinese cooking also differs from the distribution from meat cooking (He et al., 2004a; Rogge et al., 1991; Schauer et al., 1999), but resembles the patterns from frying vegetables in seed oils (Schauer et al., 2002, 1999).

The estimated hourly emissions for the most representative *n*-alkanes are depicted in Figure 2. In the university canteen, hentriacontane (C_{31}), dotriacontane (C_{32}) and triacontane (C_{30}) during the preparation of grilled pork were the *n*-alkanes with the highest hourly emissions with a similar individual value around 20.0 mg h^{-1}. For the charcoal-grilled chicken and piglet restaurants the highest emissions were, respectively, 16.2±16.8 g h^{-1} (nonacosane-C_{29}) and 290 mg h^{-1} (tetracosane-C_{24}) for the beginning of the heating phase.

The homologous series of *n*-alkenes identified in samples ranged between dodecene (C_{12}) and tritiacontene (C_{33}). He et al. (2004b) also identified *n*-alkenes in fine particles emitted from two Chinese restaurants (C_9 to C_{17}). In emissions from the university canteen, individual concentrations up to 21.4 ng m^{-3} were determined, which represented a mass fraction of 168 µg g^{-1} $PM_{2.5}$. The highest mass fractions were obtained for octacosene (C_{28}), followed by heptacosene (C_{27}), during the preparation of a meat stew. *n*-Alkene individual concentrations in emissions from the charcoal-grilled chicken restaurant varied from undetectable levels to 94.3 µg m^{-3} (nonacosene-C_{29}). The most representative homologues were C_{29}, C_{35} and C_{20}. Samples from the roasted piglet restaurant presented individual mass fractions from untraceable values to 39.0 (heneicosene-C_{21}) and to 102 µg g^{-1} $PM_{2.5}$ (tricosene-C_{23}) for the heating and food preparation stages, respectively. The estimated emissions of alkenes identified for the piglet and charcoal-grilled chicken restaurants were 0.653-67.2 mg h^{-1} and 4.61±0.417 g h^{-1}, respectively.

Hopanes and steranes were only detected in emissions from the university canteen. The triterpenoid hydrocarbons were quantified by the key ion *m/z* 191, with individual concentration varying between 0.026 and 13.1 ng m^{-3}, which correspond to $PM_{2.5}$ mass fractions from 0.243 to 146 µg g^{-1}. The series of tricyclic terpanes ranged from C_{23} to C_{30}. The hopanes with the highest concentrations were 17α(H),21β(H)-30-norhopane, 17α(H)-diahopane, 18α(H)-22,29,30-trisnorneohopane and C30 tricyclic terpane (22R). The *m/z* 217 mass fragmentograms enabled the identification of the series of 20R and 20S C_{27}-C_{28}-C_{29} homologous sterane series. The mass concentrations obtained in samples from the university canteen ranged between untraceable values to 8.01 ng m^{-3}. The highest mass fraction was obtained for 13β(H), 17α(H), 20S-cholestane during the preparation of boiled pork meat with vegetables (Table 2). These groups of compounds are normally present in mineral oil products (motor oils, hydraulic oils, lubricating oils, Vaseline) and coals (Alves, 2008; Wang et al., 1999). The widespread contamination of foodstuff by mineral oils, such as hazelnuts, almonds, cocoa beans, rice, coffee, milk, salami, cheese, candies, vegetable oils, mussels, clams, fish and olive pomace has been reported (Moret et al., 2003; Populin et al., 2004). Migration into food from polystyrene containers, waxed paper and paperboard, can coatings, and other packaging materials has been described by several authors (e.g. Castle et al., 1991, 1993a, 1993b, 1994). Mineral paraffins used as diluents in printing inks, which are applied to cardboard boxes, are also prone to be transferred to packed foods even if they are wrapped in plastic or other protection material (Droz and Grob, 1997). The detection of triterpenoid hydrocarbons only in the emissions from the canteen restaurant may be related to contamination of food by packing materials, since this establishment makes a greater variety of dishes and uses a wider range of food packaging and containers. Another hypothesis for the detection of these two groups of compounds are connected to the fact that sampling times in the university canteen were much longer than those of the other restaurants and, consequently, the influence of ambient air was much higher. The possible contribution of the lube oil from the forced-air ventilation system cannot be ruled out either. The use of gas-fired kitchen appliances may also have contributed to the emission of this type of compounds. The presence of these compounds in emissions from cooking activities requires further studies.

Polycyclic aromatic compounds

PAHs are formed by incomplete combustion processes (McDonald et al., 2003). Humans are exposed to PAHs during the cooking processes, either by inhalation of hazardous fumes or by ingestion of overcooked foodstuffs, especially those processed at high temperatures (grilling, roasting, frying) (Kim et al., 2013, 2011). PAHs and alkyl PAHs were detected in samples from the three eateries. Naphthalene was one of the compounds emitted at higher rates by the stacks of the university canteen (up to 21.1 ng m^{-3}). Particulate mass fractions up to 767 µg g^{-1} PM$_{2.5}$ were obtained for this low molecular weight aromatic compound (Table 3). Due to its high volatility, this compound is predominantly found in the gaseous phase. The highest concentration of naphthalene in the particulate phase suggests that real emissions of this compound should be significantly higher than those obtained in this study. Li et al. (2003) reported PAH values systematically higher in the gaseous phase than in the particulate phase. In a study conducted at the University of California, naphthalene was also found to be the main compound in emissions from food preparation (McDonald et al., 2003). See et al. (2006) detected higher concentrations of the more volatile PAHs in a kitchen when low temperature cooking was employed at the Indian food stall, while higher-molecular weight compounds dominated in PM$_{2.5}$ samples collected during frying operations, stir- and deep-frying at the Chinese and Malay stalls. The most abundant PAHs in emissions from the charcoal-grilled chicken restaurant were phenanthrene, fluoranthene, pyrene and chrysene (Table 3). In the study conducted by McDonald et al. (2003), phenanthrene, acenaphthylene and pyrene were the most common PAHs in emissions from charbroiling and grilling of chicken and beef, while Rogge et al. (1991) reported chrysene/triphenylene, followed by fluoranthene, pyrene and benzo[a]anthracene, as the most representative aromatic compounds in emissions from charbroilers and meat cooking operations with the use of natural gas. The most significant

PAHs in $PM_{2.5}$ from the charcoal-grilled chicken restaurant were also detected in emissions of the various stages of preparation in the roasted piglet restaurant. Nevertheless, for this latter site, the highest values were obtained during the biomass combustion phase (oven heating) (Table 3). Emissions of naphthalene during the biomass combustion stage were very low, whilst very high values, ranging between 1.66 and 6.71 µg m^{-3}, were observed for the roasting phase. These concentrations corresponded to mass fractions from 0.214 to 8.02 mg g^{-1} $PM_{2.5}$. The PAHs present in larger amounts in fine particles emitted from Chinese cuisine were pyrene and fluoranthene with concentrations from 11.0 to 18.7 ng m^{-3} and from 8.8 to 10.4 ng m^{-3}, respectively, depending on the cooking style (Hunan or Cantonese) (He et al., 2004a). PAH concentrations were found to be higher in $PM_{2.5}$ emitted during the preparation of Cantonese dishes (Bowman and Johnston, 2005; He et al., 2004a; Zhao et al., 2007a). The analysis of particles emitted during the preparation of Malaysian cuisine dishes in Singapore revealed a higher concentration of PAHs (609 ng m^{-3}, representing 2.5 mg g^{-1} of $PM_{2.5}$) compared with the other cuisine types (See et al., 2006). See et al. (2006) reported PAH concentrations of 141 and 37.9 ng m^{-3} in emissions from Chinese and Indian cuisine, which accounted for 0.7 and 0.2 mg g^{-1} of $PM_{2.5}$, respectively. In the present study, total PAH represented 0.09-1.04, 1.08 and 5.77-49.3 mg g^{-1} of the $PM_{2.5}$, for the university canteen, charcoal-grilled chicken and roasted piglet restaurants, respectively.

Diagnostic ratios are used as a tool for assessing the possible contribution of different sources (Alves, 2008). However, the PAH ratios obtained for particulate matter emitted by the different food establishments overlap those of biomass burning and vehicle exhausts, making questionable their use as source assignment tool (Supplementary material, Table S3).

For the university canteen, the highest estimated hourly emissions were obtained for naphthalene (256 mg h^{-1}), phenanthrene (17.2 mg h^{-1}) and fluoranthene (12.2 mg h^{-1}) for the preparation of grilled and stewed pork, respectively. Emissions of 131±91.6 and 94.1±65.5 mg h^{-1} were estimated for phenanthrene and fluoranthene from the charcoal-grilled chicken restaurant. Benzo[a]pyrene accounted for about 2.57-6.88% and 0.646-2.16% of the estimated total hourly emissions of PAHs for the heating and roasting phases, respectively.

The B[a]P_{eq} concentrations for the university canteen and the charcoal-grilled chicken restaurant were 0.011-0.301 and 834±523 ng m^{-3}, respectively. See et al. (2006) obtained values in the same range in samples collected in Indian stalls. Chen et al. (2012) obtained B[a]P_{eq} concentrations of 1.82±2.24 µg m^{-3} in samples collected from the stacks of Chinese restaurants. For the roasted piglet restaurant, the values of B[a]P_{eq} obtained during the heating and roasting phases were, respectively, 17.6-1090 and 0.073-7.43 mg m^{-3}, which accounted for up to 4.63 (HP) and 0.35 (RP) mg g^{-1} of $PM_{2.5}$, respectively. During the heating phase, the major input to total B[a]P_{eq} came from benzo[a]pyrene (around 70%), while benzo[a]pyrene, together with fluoranthene, were the compounds with greatest contributions during the roasting phase (around 50 and 20%, respectively).

Alkylated PAHs were identified in samples from the three restaurants. These compounds are characterised according to the number of parent-PAH rings and the carbon number of alkyl substituents. This group of compounds is more abundant and persistent in the environment than the parent PAHs (Lian et al., 2009). In the university canteen, the concentrations of alkylated homologues varied from untraceable values to 19.9 ng m^{-3} (C_1-fluorene and C_2-phenanthrene, respectively), representing particulate mass fractions up to 362 µg g^{-1} $PM_{2.5}$. The highest mass fractions were obtained when stewed and roasted meat dishes were cooked. Concentration of 43.1±30.3 µg m^{-3} were emitted from the charcoal-grilled chicken restaurant, where the most abundant alkylated PAHs were C_1-phenanthrene and C_1-fluoranthene/pyrene. Likewise, these constituents, along with C_3-dibenzothiophene, were also the most abundant in emissions of the various stages of preparation in the roasted piglet restaurant (Table 3). During the biomass combustion stage, the concentrations of C_1-phenanthrene and C_1-fluoranthene/pyrene were, respectively, around 60 and 20-fold higher in relation to the roasting stage. Alkyl-(C_1 to C_3) fluorene were only detected in the heating phase. It was estimated that the emissions of C_1-phananthrene and C_1-fluoranthene/pyrene for the charcoal-grilled chicken and roasted piglet restaurants accounted for about 26.5 and 14.1%, and 0.086-5.23 and 5.02-27.9% of the total PAH emissions, respectively. Alkyl-(C_2 and C_3) dibenzothiophenes (sulphur-containing heterocyclic compounds) were detected in particles from the university canteen and the roasted piglet restaurants (Table 3). The highest mass fraction was obtained during the preparation of chicken and turkey stew. In the roasted piglet restaurant, C_2-dibenzothiophene was only detected in low concentration in the final stage of roasting (0.609-10.9 ng m^{-3}). However, C_3-dibenzothiophene was detected in both stages of the preparation of roasted piglet with substantially higher concentrations and mass fractions: 171-1560 (HP) and 0.714-10.7 (RP) µg m^{-3} (0.726-6.62 and 0.003-0.012 mg g^{-1} $PM_{2.5}$, respectively), for the heating and roasting phases, respectively. In the charcoal-grilled chicken restaurant only C_3-dibenzothiophene was detected. For this site, the concentration was of the same order as that found in the roasting phase of piglets. The C_3-dibenzothiophene emissions for the heating phase of the roasted piglet were estimated to range between 2.08 and 19.0 g h^{-1}. A PAH total emission of 2.18 g h^{-1} was estimated for the charcoal-grilled chicken restaurant. Very different total PAH emission values were obtained for the two sequential phases in the ovens of the piglet restaurant: 5.48-141 g h^{-1} (HP) and 0.052-2.20 g h^{-1} (RP).

Ketones and aldehydes

The *n*-alkenone homologous series obtained for the 3 sites ranged from C_{13} to C_{18}. 2-Tridecanone and 2-octadecanone were the dominant ketones in $PM_{2.5}$ emitted from the charcoal-grilled chicken and roasted piglet restaurants. The highest mean concentration was obtained in fumes from the chicken restaurant for 2-tridecanone (38.8±30.7 µg m^{-3}), contributing to mass fractions of 1.14±1.02 mg g^{-1} $PM_{2.5}$. He et al. (2004b) reported a value of 6.51 µg m^{-3} in samples from Chinese cooking emissions. Acetosyringone was identified in the three restaurants of the present study (Table 4). So far, this ketone has been documented as a marker compound of hardwood combustion (Bari et al., 2011; and references therein). The polycyclic aromatic ketone 9-fluorenone was identified in emissions from the three sampling places. However, the highest concentrations and mass fractions were found in the roasted piglet restaurant during the heating phase (up to 138 µg m^{-3} and 0.585 mg g^{-1} $PM_{2.5}$). This compound has been related to emissions from biomass combustion (Shen et al., 2013), but its detection in samples from cooking processes not involving biomass burning leads to assume multiple formation and emission mechanisms. The polycyclic aromatic ketones and aldehydes, 9,10-anthracenedione, benzo[a]anthracene-7,12-dione, 7H-benzo[de]anthracen-7-one, cyclopenta[def]phenanthrenone, 2-naphthalenecarboxaldehyde, 2-fluorenecarboxaldehyde, 9-anthracenecarboxaldehyde and 1-pyrenecarboxaldehyde were only detected in samples from the roasted piglet restaurant. These oxygenated PAHs can be released to the environment from incomplete combustion or as secondary products of photo-oxidation reactions of PAHs with atmospheric oxidants (Knecht et al., 2013; and references therein). The highest concentrations were obtained during the heating phase with individual values from undetectable to 201 µg m^{-3}, while in the roasting phase, concentrations were significantly lower, up to 27.5 µg m^{-3} (Figure 1). The mass fractions obtained for the heating and roasting phase varied from undetectable levels to 12.5 and to 2.39 mg g^{-1} $PM_{2.5}$, respectively. The methoxyphenol sinapinaldehyde was present in samples from the three establishments. The highest concentration was found in samples collected in the piglet restaurant (Table 4). The emissions of this compound for the heating and roasting phases were estimated to be 0.300-2.29 and 0.004-0.130 g h^{-1}, respectively. Methoxyphenols have been identified as thermal degradation products of lignin (Hawthorne et al., 1988). Their detection can be related to the vine cuttings and pine slats used as fuel to fire up the brick ovens in which piglets were roasted. In the university canteen, the highest contributor to the emissions was acetosyringone during the preparation of chicken and turkey stew (15.8 mg h^{-1}). 2-Tridecanone accounted for about 86% of the total emissions of *n*-alkanones from the chicken restaurant.

Alcohols, sterols and other compounds with OH group

Alcohols and sterols have been detected in aerosols of many urban and rural areas around the world (Alves et al., 2012; Oliveira et al., 2007). The oxidation reactions to which alcohols are subjected can lead to the corresponding sterols (Benesch et al., 2011).

Cholesterol (cholest-5-en-3β-ol) was a representative compound in emissions of the three food establishments with individual concentrations ranging between untraceable values and 75.7 µg

m^{-3}, symbolising up to 13.1 mg g^{-1} $PM_{2.5}$. In the university canteen, the most abundant concentration was found during the preparation of grilled pork (181 ng m^{-3}), while the highest $PM_{2.5}$ mass fraction was obtained for the preparation of grilled and boiled fish (2.07 mg g^{-1}). The highest concentration was obtained in emissions from the charcoal-grilled chicken restaurant (38.1±23.1 µg m^{-3}). However, the highest mass fraction was found in samples from the piglet restaurant during the roasting phase, when the three piglets were inside the oven with glowing embers at the bottom and the door open (13.1 mg g^{-1} $PM_{2.5}$) (Table 4). This compound belongs to the group of steroids and is present in all animal tissues, since it is biosynthesised by the body (McDonald et al., 2003; Rogge et al., 1991). Many authors have defined cholesterol as one of the most useful tracers for the aerosol released from meat cooking (Gysel et al., 2018; McDonald et al., 2003; Rogge et al., 1991). He et al. (2004a) reported values of 738 and 248 ng m^{-3} for Hunan and Cantonese cooking styles included in the regional Chinese cuisine. McDonald et al. (2003), in a work carried out in the US, observed higher amounts of cholesterol in emissions from charbroiling of chicken and beef than in emissions from electric grilling of the same foods. In studies conducted in Guangzhou and Shenzhen, cholesterol was also detected in emissions from Chinese cuisine, although the sterol compound with the highest mass fraction in the emitted particulate matter was β-sitosterol (1350-2610 mg g^{-1} $PM_{2.5}$). Stigmasterol was also identified as an important compound in emissions of the Chinese cuisine (He et al., 2004a; Zhao et al., 2007a). In the present study, β-sitosterol and stigmasterol were observed in samples from the three restaurants with individual concentrations ranging, respectively, from untraceable values to 5.11 µg m^{-3} and to 2.54 µg m^{-3} (Table 4). Myrtenol was detected in all the samples from the charcoal-grilled chicken restaurant (1.56±0.991 µg m^{-3}; 33.8±30.6 µg g^{-1} $PM_{2.5}$). This monoterpene compound is found in essential oils of several plants and is often used as a fragrance ingredient in a wide range of cosmetic and non-cosmetic products (such as toilet soaps, household cleaners and detergents) (Bhatia et al., 2008).

In the present study, the estimated emissions of cholesterol for the piglet restaurant were up to 12.9 (HP) and 9.27-222 (RP) mg h^{-1}, respectively. Emissions of this compound from the chicken restaurant was estimated to be 1.52±0.919 g h^{-1}.

n-Alcohols were also analysed (C_{10} to C_{30}). For most menus in the canteen, decanol was one of the most abundantly emitted compounds, while pentadecanol and octacosanol were the most common in particles from the charcoal-grilled chicken and piglet restaurants, respectively. Emissions of decanol for the university canteen and pentadecanol for the charcoal-grilled chicken restaurant accounted for 7.5-90% and 42% of the total estimated *n*-alkanol emissions, respectively. For the piglet restaurant, pentadecanol accounted for 31-75% (RP), while octacosanol represented 15-67% (HP) and 14-46% (RP) of the total n-alkanol emissions. Rogge et al. (1991) only detected and quantified three alcohols in emissions from charbroiler and meat cooking operations, with pentadecanol as one of the most abundant compounds.

Acids

The homologous series of *n*-alkanoic acids ranged from C_8 to C_{22}, with an even-to-odd carbon number preference that maximised at C_{16} (hexadecanoic acid), followed by C_{18} (octadecanoic acid) and C_{14} (tetradecanoic acid) for the three restaurants. This pattern has been reported in emissions from Chinese cooking, meat cooking and Western-style fast food (He et al., 2004a; Rogge et al., 1991; Zhao et al., 2007b, 2007a). In $PM_{2.5}$ from the university canteen, the individual concentrations for these representative compounds were up to 3.07, 1.57 and 0.379 $\mu g\ m^{-3}$, respectively. The highest mass fractions were achieved in samples collected in the exhaust from the fry and stew stations, while the lowest corresponded to the boiled food station (55.7, 28.5 and 8.95 $mg\ g^{-1}$ $PM_{2.5}$, respectively). Regardless of the menus, palmitic acid was always one of dominant fatty acids. The concentrations of these compounds obtained in the charcoal-grilled chicken restaurant were 574±302, 47.0±47.9 and 71.5±43.0 $\mu g\ m^{-3}$, respectively, accounting for 9.73±1.96, 2.19±1.23 and 1.20±0.262 $mg\ g^{-1}$ $PM_{2.5}$. In the piglet restaurant, palmitic acid was the most representative compound in samples collected during the heating and roasting phase (Table 4). He et al. (2004a) reported higher mass fractions for the same compounds in emissions of fine particles from Chinese cooking. Emissions of palmitic acid from the university canteen and charcoal-grilled chicken restaurant accounted for 47-67 and 62% of the estimated emissions of total *n*-alkanoic acids, respectively. For the piglet restaurant, the contribution of this compound to the total n-alkanoic acids emissions represented a share of about 65%.

The unsaturated fatty acids identified at highest concentrations were oleic and palmitoleic acids. These compounds are formed through the combustion of triglycerides and phospholipids from seed oil, vegetable oil, and animal fats (Abdullahi et al., 2013; Rogge et al., 1991; Schauer et al., 2002). The concentration of oleic acid quantified in the emissions from the roasted piglet restaurant during the heating phase was found to be 16 and 80 times lower than the levels measured in emissions from the charcoal-grilled chicken restaurant. However, at the beginning of the roasting phase, and in the final stage of the piglet's roasting, concentrations of oleic acid were, respectively, 5 and 46-fold lower than those obtained in the chicken restaurant. Additionally, the highest mass fractions of palmitoleic and oleic acids were obtained in samples from the piglet restaurant, especially in the final stage of the roasting process, with values ranging between 0.389-3.96 and 2.11-34.4 mg g^{-1} $PM_{2.5}$, respectively. For the charcoal-grilled chicken restaurant the obtained mass fraction of palmitoleic acid was of the same order as that found at the roasting phase of piglets, while the mass fraction of oleic acid was substantially lower (7.26±2.68 mg g^{-1} $PM_{2.5}$). In the university canteen, the highest concentration and mass fraction of oleic acid were measured in emissions from the preparation of chicken and turkey stew (1.43 µg m^{-3} and 26.1 mg g^{-1} $PM_{2.5}$, respectively). The emission of oleic acid estimated for the preparation of charcoal-grilled chicken was 16.2±7.64 g h^{-1}.

Resin acids, such as dehydroabietic and abietic, were detected in all samples from the wood-oven roasted piglet restaurant. The highest concentrations were attained in samples of the heating phase when vine cuttings and pine slats were used to fire up the brick oven (5.45-28.5 and 0.351-17.1 µg m^{-3}, respectively) and decreased in samples corresponding to the beginning of the roasting phase (0.207-1.07 and 0.003-0.694 µg m^{-3}, respectively). $PM_{2.5}$ mass fractions obtained for these compounds during the heating phase were 0.121-0.340 and 0.022-0.073 mg g^{-1}, while for the roasting stage the values were 0.034-1.00 and 0.015-0.040 mg g^{-1}, respectively.

Saccharides and polyols

Levoglucosan was identified in samples from the three restaurants. Its concentrations varied between untraceable values and 293 µg m^{-3}, representing values up to 8.86 mg g^{-1} $PM_{2.5}$. This anhydrosugar has long been pointed out as a tracer of the thermal breakdown of cellulose, which is a major component of biomass (Alves et al., 2019; Vicente et al., 2020). However, it has been speculated that the high temperature during cooking processes would also lead to the emission of levoglucosan due to thermal degradation of vegetable cellulose (He et al., 2004a) or decomposition of spices used as condiments (Hou et al., 2008). Levoglucosan was also detected in emissions from Chinese and Western-style fast food cooking (He et al., 2004a; Zhao et al., 2007b). The highest concentration (293 µg m^{-3}) of this compound was obtained in samples from the piglet restaurant during flaming conditions, when the oven was heated-up. Concentrations in samples taken during the roasting phase ranged between 0.317 (final stage) and 20.8 µg m^{-3} (initial stage). The mass fractions found in the heating and roasting phase were 1.24-3.98 and 0.664-8.86 mg g^{-1} $PM_{2.5}$, respectively. The highest particulate mass fraction obtained under roasting conditions was taken at the time that three piglets were in the oven. The levoglucosan isomers, galactosan and mannosan, were only detected in particles emitted from the piglet restaurant (0.638-0.488 and 0.106-0.786 mg g^{-1} $PM_{2.5}$, respectively). The emissions of galactosan, mannosan and levoglucosan from the piglet restaurant were estimated to be up to 1.40, 2.26 and 3.58 g h^{-1}, respectively. For the chicken restaurant, the emission of levoglucosan was estimated to be 435±214 g h^{-1}. In the university canteen, the highest hourly emission was obtained for the preparation of roasted ribs (2.54 mg h^{-1}). Other saccharidic compounds identified in the exhaust fumes from the three restaurants included the monosaccharides lyxose, arabinose, ribose, fructose, xylose and glucose and the disaccharide maltose. Medeiros et al. (2006) reported several natural sources for monosaccharides, namely, microorganisms, vascular plants and animals. Lyxose and fructose were only detected in samples from the university canteen during the preparation of roasted and grilled meat (up to 88.7 ng m^{-3}, representing 829 µg g^{-1} $PM_{2.5}$). Arabinose and glucose were the most abundant monosaccharides in the three restaurants. The highest concentrations of these compounds were obtained during the heating phase of the roasted piglet confection (up to 491 and 4.52 µg m^{-3}, respectively). The highest mass fractions of arabinose and glucose, obtained in the heating-roasting phases, were 2.08-3.55 and 0.055-0.127 mg g^{-1} $PM_{2.5}$, respectively. Among sugar alcohols (polyols), xylitol and sorbitol were detected in the present study. Polyols have been pointed out as components of bacteria, fungi, lichens, lower plants and invertebrates, acting as stress inhibitors or carbohydrate providers (Graham et al., 2003). Xylitol was present in samples from the three eateries. However, sorbitol was only quantified in particles from the piglet restaurant. In this latter restaurant, the highest concentrations of these two polyols were found in samples collected from the heating phase (5.39 and 1.25 µg m^{-3}, respectively). The mass fractions of sorbitol for the heating and roasting phases varied from untraceable values to 0.078 and to 0.096 mg g^{-1} $PM_{2.5}$, respectively.

The unsaturated monoglyceride 2-monoolein was identified in samples from the three food establishments. Monoglycerides derive from the hydrolysis of triglycerides during meat cooking (Abdullahi et al., 2013). This compound is employed in different industries, such as food, cosmetic and pharmaceutical (Miao and Lin, 2019; Nitbani and Jumina, 2020). The highest concentrations and mass fractions were measured in samples from the charcoal-grilled chicken restaurant (53.5±42.5 µg m^{-3} and 1.14±0.940 mg g^{-1} $PM_{2.5}$, respectively). In samples from the piglet restaurant, this monoglyceride was only detected during the roasting phase, with concentrations between 1.03 and 2.36 µg m^{-3}, representing $PM_{2.5}$ mass fractions from 0.075 to 4.96 mg g^{-1}. In the university canteen the highest concentration and mass fraction were obtained during the preparation of grilled pork (77.8 ng m^{-3} and 727 µg g^{-1} $PM_{2.5}$).

Nitrogen compounds

Amide compounds have been ascribed to cooking process (Abdullahi et al., 2013; and references therein). Amides were only detected in samples from the university canteen. The compounds with highest concentrations and mass fractions were nicotinamide, followed by hexadecanamide and oleamide (up to 482, 474 and 368 ng m^{-3}, respectively) (Supplementary material, Tables S1 and S2). The highest mass fractions of these compounds were detected during the preparation of stew meat (7.67, 7.53 and 5.86 mg g^{-1} $PM_{2.5}$) (Table 5). Nicotinamide is present in food of animal origin. It is a hydrolysed product of vitamin B_3 (present in the raw food) during cooking processes. The highest nicotinamide hourly emissions were estimated to be 5.59 mg h^{-1} from the preparation of meat stew. Zhao et al. (2007a) detected hexadecanamide and oleamide in fine particles emitted from Chinese cooking. Palmitonitrile was identified in emissions from the three food places. The highest emission of this compound was estimated for the charcoal-grilled chicken restaurant (657±453 mg h^{-1}). Rogge et al. (1991) identified the presence of oleamide and palmitonitrile in emissions from charbroilers and meat cooking operations. The emissions from fried hamburgers were significantly higher than the emissions from lean meat. Emissions of these compounds increase with increasing fat contents in meat. Simoneit et al. (2003) suggested that alkyl amides and alkyl nitriles result from the reaction of fatty acids with ammonia or ammonium salts during combustion. The nitrogen heterocyclic compounds thymine was also identified in samples from the three restaurants. However, adenine and alanine were only detected in samples from the university canteen during the preparation of grilled meat and fish. As far as we know, these compounds were never detected as components of the particulate material emitted during food cooking.

Conclusions

The results of this study showed much lower emissions to ambient air from the university canteen, reflecting the use of cleaner energy sources and hood filters. However, hopanes and steranes were only identified in the fumes emitted from the stacks of the university canteen, which can be related to the use gas-fired cooking equipment, contamination of food by petroleum-based packaging or emissions of lubricating oils from ventilation systems. At this university establishment, total PAH emissions during the preparation of meat stews were two times higher than those from cooking of the various boiled meat dishes. In the wood-oven roasted piglet restaurant, significant differences in emissions were observed from the various preparation stages of the roasting piglet. The use of biomass as fuel for heating the oven clearly influenced the composition and particulate mass fractions of the compounds for this type of restaurant. Polycyclic aromatic ketones and aldehydes were only identified in the fumes from the piglet restaurant. Furthermore, the total mass fractions of these compounds emitted during the heating phase of the oven were 4 times higher than those obtained when roasting the piglets, highlighting the importance of fuel in emissions from cooking activities. Besides the fuel, it was also clear that the cooking ingredients also contributed to the diversity of particulate mass fractions obtained from the tree eateries. In addition, the identification of levoglucosan and the pattern of *n*-alkanes in the exhaust fumes of the university canteen may be related to the inclusion vegetables, especially those containing cellulose, in the menu. The present study highlights the great diversity in the composition of fumes emitted by cooking activities and their dependence on the type of food ingredients and cooking style, as well as the energy sources. Given the enormous variety of cooking styles, many additional works are needed to increase the representativeness of the emission profiles of this source. Commercial cooking activities largely contribute to the emission of pollutants to the atmosphere and cannot be overlooked in emission inventories. The database obtained in this study represents an input to improved emission inventories and can contribute to support the development of air quality management policies. It is also important as it appends to the emergent speciation profiles for major sources in Europe (SPECIEUROPE), helping to make more accurate estimates of the contribution of cooking emissions to particulate matter levels at receptor sites when applying source apportionment models. It should be noted that the identification in this study of some organic compounds, such as levoglucosan and hopanes, which so far have been pointed out as markers of wood burning and vehicular exhaust, respectively, suggest that the contribution of these sources may be overestimated when running source apportionment models, such as the CMB. Thus, sensitivity tests may be required to estimate the wood burning or vehicular contributions by running the CMB model with and without including cooking markers like cholesterol.

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Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Figure 1. Particulate mass fractions of polycyclic aromatic ketones and aldehydes in samples collected from the exhaust stack of the roasted piglet restaurant (S1 and S2 samples – heating phase; S3 to S5 samples – roasting phase).

Figure 2. Hourly emission rates for the most representative organic compounds identified in the different categories of restaurants (U1 to U10 samples from the University Canteen left Y axis; [a] right Y axis representing the emission rates of the CkR sample from the charcoal-grilled chicken restaurant and S1 to S5 samples from the Piglet restaurant – S1 to S2 heating phase, S3 to S5 roasting phase).

Table 1. Type of cooked food and $PM_{2.5}$ concentrations emitted by different categories of restaurants.

		Description	$PM_{2.5}$
		University Canteen	*($\mu g\ m^{-3}$)*
U1		Boiled chicken with vegetables	41±11
U2		Fried fish; seafood rice	97±23
U3		Veal stroganoff stew	63±2.3
U4		Boiled pork meat with vegetables	27±1.5
U5		Roasted ribs	127±2.0
U6		"Alentejo" pork meat with clams; stewed pork chops with sausages	45±8.1
U7		Grilled pork	107±3.1
U8		Beef with rice; pork with boiled potatoes	29±6.7
U9		Grilled and boiled fish	44±11
U10		Chicken stew; turkey stew with rice	55±4.0
		Charcoal-Grilled Chicken Restaurant	*($mg\ m^{-3}$)*
		Smoke-roasted chicken	71±39
		Wood-Oven Roasted Piglet Restaurant	*($mg\ m^{-3}$)*
HP	S1	Oven preheating with remains of pruning vines and pine slats Burning of pruning vines and pine slats (flaming) Pine slats burning like in a fireplace (door open) after refueling; flaming phase and beginning of smouldering phase	236±208
	S2	Smouldering and glowing embers	16.0±2.58
RP	S3	Glowing embers at the bottom; door open occasionally to spread ashes on the embers	31.4±2.24
	S4	Three piglets in the oven; glowing embers at the bottom; door open	1.39±0.142
	S5	Final phase of roasting; door closed	0.207±0.006

U1-U10 Samples; HP – Heating phase; RP - Roasting phase; S1-S5 Samples

Table 2. Mass fractions of hopanes and steranes in samples collected from the exhaust stack of the university canteen (µg g^{-1} $PM_{2.5}$).

	University Canteen									
	Boiled chicken with vegetables	Fried fish; seafood rice	Veal stroganoff stew	Boiled pork with vegetables	Roasted ribs	Alentejo pork with clams; stewed pork; chops with sausages	Grilled pork	Beef with rice; pork with boiled potatoes	Grilled and boiled fish	Chicken stew; turkey stew with rice
Hopanes										
C_{23} Tricyclic terpane	--	--	33.9	--	--	--	0.243	--	0.781	11.1
C_{24} Tricyclic terpane	--	--	26.0	--	--	--	4.54	20.1	19.5	--
C_{26} Tricyclic terpane (22R)	--	--	--	--	--	--	--	--	8.98	--
C_{26} Tricyclic terpane (22S)	--	--	--	--	--	--	--	--	14.3	--
18α(H)-22,29,30-trisnorneohopane	--	--	53.2	80.1	48.0	50.5	4.19	--	--	--
C_{30} Tricyclic terpane (22R)	--	--	75.9	79.4	43.0	50.9	4.64	--	12.8	19.8
17α(H),21β(H)-30-norhopane	--	--	123	146	103	72.9	1.75	--	3.50	18.7
18α(H)-30-norneohopane	--	--	--	--	--	24.5	--	--	--	--
17α(H)-diahopane	--	--	97.2	146	69.1	103	2.56	51.4	7.66	28.0
22S 17α(H),21β(H)-22S-homohopane	--	--	38.0	71.0	25.2	--	--	--	--	--
22R 17α(H),21β(H)-22R-homohopane	--	--	34.6	53.2	24.5	--	--	--	--	--
22S 17α(H),21β(H)-22S-bishomohopane	--	--	32.3	--	--	--	--	--	--	--
Σ Hopanes	--	--	514	575	313	302	17.9	71.5	67.5	77.5
Steranes										
13β(H), 17α(H), 20S-Cholestane (diasterane) (C_{27})	--	--	66.4	89.2	54.6	61.9	5.28	--	19.7	24.6
13β(H), 17α(H), 20R-Cholestane (diasterane) (C_{27})	--	--	48.3	52.8	30.0	41.8	2.92	--	--	16.3

13α(H), 17β(H), 20R-Cholestane (diasterane) (C_{27})	--	--	--	33.5	--	--	--	--	--	--
13α(H), 17β(H), 20S-Cholestane (diasterane) (C_{27})	--	--	32.4	42.7	27.9	38.1	--	--	--	--
ααα (20S)-Cholestane (C_{27})	--	--	38.2	65.4	--	--	--	--	--	--
αββ (20R)-Cholestane (C_{27})	--	--	--	75.0	--	59.9	--	--	--	--
αββ (20S)-Cholestane (C_{27})	--	--	47.0	68.9	38.9	68.2	--	--	--	14.3
ααα (20R)-Cholestane (C_{27})	--	--	--	64.3	62.9	87.3	--	--	--	23.0
24-Methyl-5α(H), 14α(H), 17α(H) 20S-Cholestane (20S,24S) (C_{28})	--	--	25.9	--	20.5	--	--	--	--	--
24-Methyl-5α(H), 14β(H), 17β(H) 20R-Cholestane (20R,24S) (C_{28})	--	--	--	--	25.4	--	--	--	--	--
24-Methyl-5α(H), 14β(H), 17β(H) 20S-Cholestane (20S,24R) (C_{28})	--	--	25.1	42.3	21.6	31.3	--	--	--	--
24-Methyl-5α(H), 14α(H), 17α(H) 20R-Cholestane (20R,24R/S) (C_{28})	--	--	--	51.7	--	36.5	--	--	--	--
24-Ethyl-5α(H), 14α(H), 17α(H) 20S-Cholestane (20S,24R) (C_{29})	--	--	27.8	43.9	20.2	29.6	--	--	--	--
24-Ethyl-5α(H), 14β(H), 17β(H) 20R-Cholestane (20R,24R) (C_{29})	--	--	30.2	50.1	--	40.4	--	--	--	--
24-Ethyl-5α(H), 14β(H), 17β(H) 20S-Cholestane (20S,24R) (C_{29})	--	--	28.5	43.6	--	25.0	--	--	--	--
24-Ethyl-5α(H), 14α(H), 17α(H) 20R-Cholestane (20R,24R) (C_{29})	--	--	30.0	45.9	--	46.7	--	--	--	--
Σ Steranes	--	--	400	769	302	567	8.20	--	19.7	78.2

-- below detection limit or not detected

Table 3. $PM_{2.5}$ mass fractions of PAHs for the three restaurants.

	University Canteen										Charcoal-Grilled Chicken Restaurant	Wood-Oven Roasted Piglet Restaurant				
												HP		RP		
	U1	U2	U3	U4	U5	U6	U7	U8	U9	U10		S1	S2	S3	S4	S5
	µg g^{-1}										µg g^{-1}	mg g^{-1}				
Naphthalene	38.9	--	--	767	--	--	60.5	733	--	4.03	--	--	0.006	0.214	3.26	8.02
Acenaphthylene	--	--	--	--	--	--	--	--	--	--	-- -0.244	0.672	--	--	--	--
Acenaphthene	--	--	--	2.00	--	--	0.537	--	--	--	-- -0.897	0.513	--	0.013	0.055	0.180
Fluorene	--	--	--	--	--	--	--	--	--	--	0.980-7.52	0.595	--	0.002	--	--
Phenanthrene	--	--	--	2.49	2.08	14.5	13.2	--	6.54	5.88	35.1-63.1	4.78	0.016	0.104	0.064	0.084
Anthracene	--	--	--	--	1.13	1.03	1.01	1.42	1.15	1.14	3.09-10.5	1.29	0.003	0.015	0.009	0.010
Fluoranthene	3.24	2.61	4.19	6.19	7.14	22.4	6.82	--	0.850	5.65	22.0-53.2	5.35	0.432	0.862	0.480	1.54
Pyrene	2.29	0.726	1.13	--	--	2.81	0.118	--	--	3.04	20.6-40.0	5.41	0.544	0.809	0.510	1.42
p-Terphenyl	--	--	--	--	--	--	--	--	--	--	2.25-2.95	0.022	0.111	0.028	0.021	0.034
Retene	1.56	--	--	--	0.706	0.745	1.22	1.53	0.353	--	--	0.023	0.169	0.143	0.034	0.198
Benzo[a]anthracene	2.32	0.889	1.49	0.952	1.93	1.91	1.14	0.880	--	2.28	8.19-20.1	2.35	0.828	0.325	0.314	0.866
Chrysene	2.34	0.337	1.04	0.313	0.711	0.988	0.289	--	--	2.43	14.4-37.9	2.17	0.866	0.393	0.374	0.933
Benzo[b]fluoranthene	7.07	1.38	1.87	2.02	3.84	2.67	1.42	3.59	0.717	4.64	8.73-20.5	1.68	0.739	0.179	0.200	0.184
Benzo[j]fluoranthene	2.67	0.640	1.09	1.06	2.19	1.83	0.617	1.53	0.387	1.67	0.773-1.41	0.172	--	0.018	0.036	0.037
Benzo[k]fluoranthene	2.96	0.809	1.19	0.884	0.903	1.13	0.745	1.58	0.243	0.898	2.37-4.75	1.97	0.740	0.095	0.144	0.079
Benzo[e]pyrene	--	--	--	--	--	--	1.90	8.61	0.833	--	7.09-28.5	1.63	0.572	0.162	0.162	0.165
Benzo[a]pyrene	1.88	--	1.31	--	0.998	1.05	0.392	--	--	--	4.27-10.7	3.39	0.721	0.124	0.155	0.132
Cyclopenta[cd]pyrene		--	--	--	--	--	--	--	--	--	--	3.02	0.720	--	--	--
Perylene	--	--	--	--	--	--	--	--	0.132	--	--	0.350	0.092	0.012	0.020	0.015
Indeno[1,2,3-cd]pyrene	6.07	1.03	1.76	1.99	2.82	3.35	1.16	3.95	0.528	2.66	2.81-6.45	2.08	0.576	0.075	0.137	0.178

Dibenzo[a,h]anthracene	0.768	--	0.306	--	--	--	--	--	--	--	0.942-1.34	0.235	0.099	0.015	0.032	0.044
Benzo[ghi]perylene	4.71	0.735	1.89	2.13	2.38	3.54	1.18	6.71	0.395	2.78	2.42-9.5	1.71	0.357	0.075	0.104	0.170
Alkyl-PAHs																
C_1-naphthalene	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
C_2-naphthalene	--	--	--	--	--	--	--	--	--	--	-- -2.40	0.010	--	--	--	--
C_3-naphthalene	--	--	--	--	--	--	--	--	--	--	-- -3.59	0.011	--	--	--	--
C_1-phenantrene	260	23.1	25.2	40.1	93.4	--	36.3	--	111	270	60.2-541	2.33	0.017	0.040	0.004	--
C_2-phenantrene	304	53.7	51.9	80.8	127	--	41.8	--	255	363	7.31-239	0.230	0.010	0.026	0.002	--
C_3-phenantrene	--	--	--	--	--	12.4	9.60	42.1	--	117	-- -113	0.003	0.002	0.008	--	--
C_4-phenantrene	--	--	--	--	--	--	--	--	--	--	--	--	--	0.030	--	--
C_2-dibenzothiophene	--	--	--	39.7	25.2	38.6	14.3	98.2	23.1	143	--	--	--	--	--	--
C_3-dibenzothiophene	--	--	--	--	--	--	--	--	--	16.1	33.8-190	6.62	0.726	0.012	0.045	0.003
C_1-fluorene	--	--	--	--	--	--	--	--	--	7.87	-- -115	0.300	--	--	--	--
C_2-fluorene	--	--	--	--	--	--	--	--	--	21.2	-- -84.4	0.077	--	--	--	--
C_3-fluorene	--	--	--	--	--	--	--	--	--	--	--	0.044	--	--	--	--
C_1-chrysene	--	--	--	--	--	--	--	--	--	--	-- -92.2	0.156	0.155	0.006	0.005	--
C_1-fluoranthenes/C_1-pyrenes	--	--	--	--	--	--	--	--	--	60.2	-- -673	3.06	0.532	0.155	0.005	0.002
Other PAHs																
Benzothiazole	--	--	--	34.0	--	--	--	--	--	--	--	0.001	--	0.001	0.011	0.028
Dibenzothiophene	--	--	--	--	--	0.310	0.599	--	--	--	-- -0.531	0.005	--	0.001	--	0.035
Carbazole	0.975	0.051	0.257	0.361	0.361	--	0.817	--	0.743	--	3.16-16.5	0.029	0.010	0.029	--	0.023
Σ PAHs	642.4	86.0	94.6	982	273	109	196	903	401	1035	1083	52.3	9.04	3.97	6.18	14.4

-- below detection limit or not detected; U1-U10 Samples; HP Heating phase; RP Roasting phase; S1-S5 Samples

Table 4. PM$_{2.5}$ mass fractions of various groups of organic compounds for the three restaurants.

	University Canteen										Charcoal-Grilled Chicken Restaurant	Wood-Oven Roasted Piglet Restaurant				
												HP			RP	
	U1	**U2**	**U3**	**U4**	**U5**	**U6**	**U7**	**U8**	**U9**	**U10**		**S1**	**S2**	**S3**	**S4**	**S5**
	mg g^{-1}										**mg g^{-1}**	**mg g^{-1}**				
Ketones and Aldehydes																
2-Tridecanone	--	--	--	--	--	--	--	--	--	--	-- -2.37	0.051	0.019	0.414	0.318	0.115
2-Octadecanone	0.012	0.001	0.005	--	--	0.001	0.003	0.012	0.018	0.002	0.068-0.144	0.002	0.011	0.131	0.411	0.761
Acetosyringone	--	0.012	--	--	--	--	--	--	--	0.243	0.004-0.036	--	--	0.001	0.001	--
Sinapinaldehyde	0.013	0.002	0.002	--	--	0.004	0.002	0.002	0.008	0.009	0.009-0.112	0.798	1.54	0.340	0.846	1.45
Alcohols, sterols and other compounds with OH group																
Decanol	0.015	0.089	0.049	0.017	0.014	0.285	0.651	0.149	0.302	0.401	0.008-0.013	--	--	0.001	0.007	0.053
Pentadecanol	0.031	0.586	0.015	0.014	0.013	0.011	0.016	0.027	0.023	0.018	0.011-0.019	--	--	0.026	0.115	0.033
Docosanol	0.018	0.003	0.011	0.004	0.017	0.014	0.003	0.005	0.015	0.011	-- -0.008	0.005	--	--	0.038	--
Tricosanol	--	0.387	--	--	--	0.014	0.027	--	0.165	0.021	0.001-0.011	0.014	0.001	0.001	0.003	--
Pentacosanol	--	0.093	--	--	--	0.005	0.013	--	0.081	0.008	-- -0.006	0.006	0.005	0.001	0.005	--
Heptacosanol	0.002	0.011	0.001	0.001	0.002	0.005	0.003	--	0.018	0.004	-- -0.003	--	0.008	--	0.005	--
Octacosanol	0.025	0.011	0.033	0.021	0.056	0.009	0.004	0.007	0.007	0.016	-- -0.002	0.004	0.151	0.006	0.170	0.015
Triacontanol	0.010	0.007	0.016	0.016	0.024	0.006	0.003	0.005	0.001	0.009	--	0.001	0.062	--	0.027	--
Cholesterol	0.057	0.337	0.475	0.034	0.399	0.171	1.69	0.116	2.07	0.097	0.410-0.979	0.004	--	0.102	13.1	3.67
β-Sitosterol	0.008	0.590	0.294	0.007	0.024	0.278	0.210	0.039	0.298	0.367	-- -0.001	0.022	0.050	--	0.082	--
Stigmasterol	0.027	0.174	0.232	0.006	0.027	0.094	0.144	0.009	0.206	0.100	-- -0.028	0.003	0.001	0.004	0.056	--

Acids																
Tetradecanoic acid	--	--	--	3.42	2.97	5.94	2.68	8.95	4.77	5.16	0.691-1.39	0.031	0.325	0.701	2.94	6.99
Hexadecanoic acid	--	--	--	13.7	22.9	25.4	17.0	33.6	28.5	55.7	6.06-11.8	0.308	7.18	6.11	19.9	66.9
Octadecanoic acid	--	--	--	2.27	7.36	4.87	10.2	7.28	5.81	28.5	1.63-3.13	--	2.06	1.60	8.88	20.7
Palmitoleic acid (C16:1)	--	--	--	--	0.095	0.197	0.218	--	--	--	3.00-6.89	--	0.025	0.389	3.25	3.96
Oleic acid (C18:1)	--	--	--	0.060	0.951	3.79	4.44	0.069	1.16	26.1	4.03-11.5	0.088	0.256	2.11	17.8	34.4
Abietic acid	--	--	--	--	--	0.006	--	--	--	--	--	0.073	0.022	0.022	0.040	0.015
Dehydroabietic acid	0.026	--	--	--	--	--	--	--	--	--	--	0.121	0.340	0.034	0.236	1.00

-- below detection limit or not detected; U1-U10 Samples; HP Heating phase; RP Roasting phase; S1-S5 Samples

Table 5. Mass fractions of nitrogen compounds in samples collected from the exhaust stack of the university canteen (mg g^{-1} $PM_{2.5}$).

	University Canteen									
	Boiled chicken with vegetables	Fried fish; seafood rice	Veal stroganoff stew	Boiled pork with vegetables	Roasted ribs	Alentejo pork with clams; stewed pork; chops with sausages	Grilled pork	Beef with rice; pork with boiled potatoes	Grilled and boiled fish	Chicken stew; turkey stew with rice
Nitrogen compounds										
Nicotinamide	--	0.464	7.67	--	--	2.93	--	--	--	--
Oleamide	0.375	2.13	5.86	--	--	0.446	--	--	--	--
Dodecanamide	--	0.117	0.798	--	--	--	--	--	--	--
Hexadecanamide	--	0.349	7.53	--	--	0.496	1.07	--	--	--
Palmitonitrile	--	--	--	--	--	0.259	0.311	0.328	--	0.495
Oleanitrile	--	--	--	--	--	--	--	--	--	--
Thymine	--	--	--	--	--	--	0.260	--	0.440	--
N,N-Dimethyldodecylamine	6.04	0.605	2.35	--	--	1.38	--	--	--	--
L-alanine	--	--	--	--	--	--	0.516	--	0.195	--
Adenine	--	--	--	--	--	--	3.30	--	4.23	--

-- below detection limit or not detected;

Highlights

- Organic speciation of $PM_{2.5}$ in smoke from a variety of Latin cuisine was obtained
- Polycyclic aromatic ketones and aldehydes were linked to the use of biomass as fuel
- Cholesterol was the dominant sterol
- Fatty acids were one of the dominant organic class
- The diversity of organic mass fractions greatly depended on the ingredients

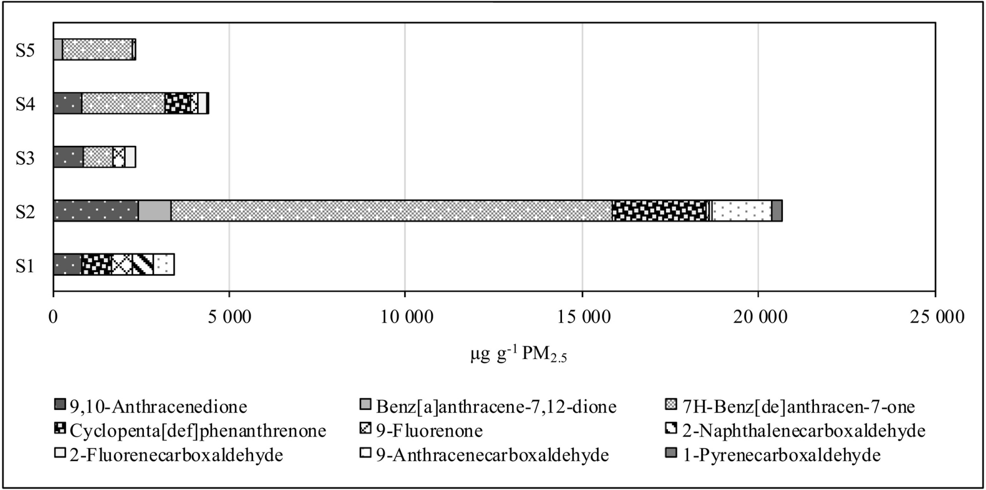
S5
S4
S3
S2
S1
0
5 000
10 000
15 000
20 000
25 000
μg g-1 PM2.5
9,10-Anthracenedione
Benz[a]anthracene-7,12-dione
7H-Benz[de]anthracen-7-one
Cyclopenta[def]phenanthrenone
9-Fluorenone
2-Naphthalenecarboxaldehyde
2-Fluorenecarboxaldehyde
9-Anthracenecarboxaldehyde
1-Pyrenecarboxaldehyde

Figure 1

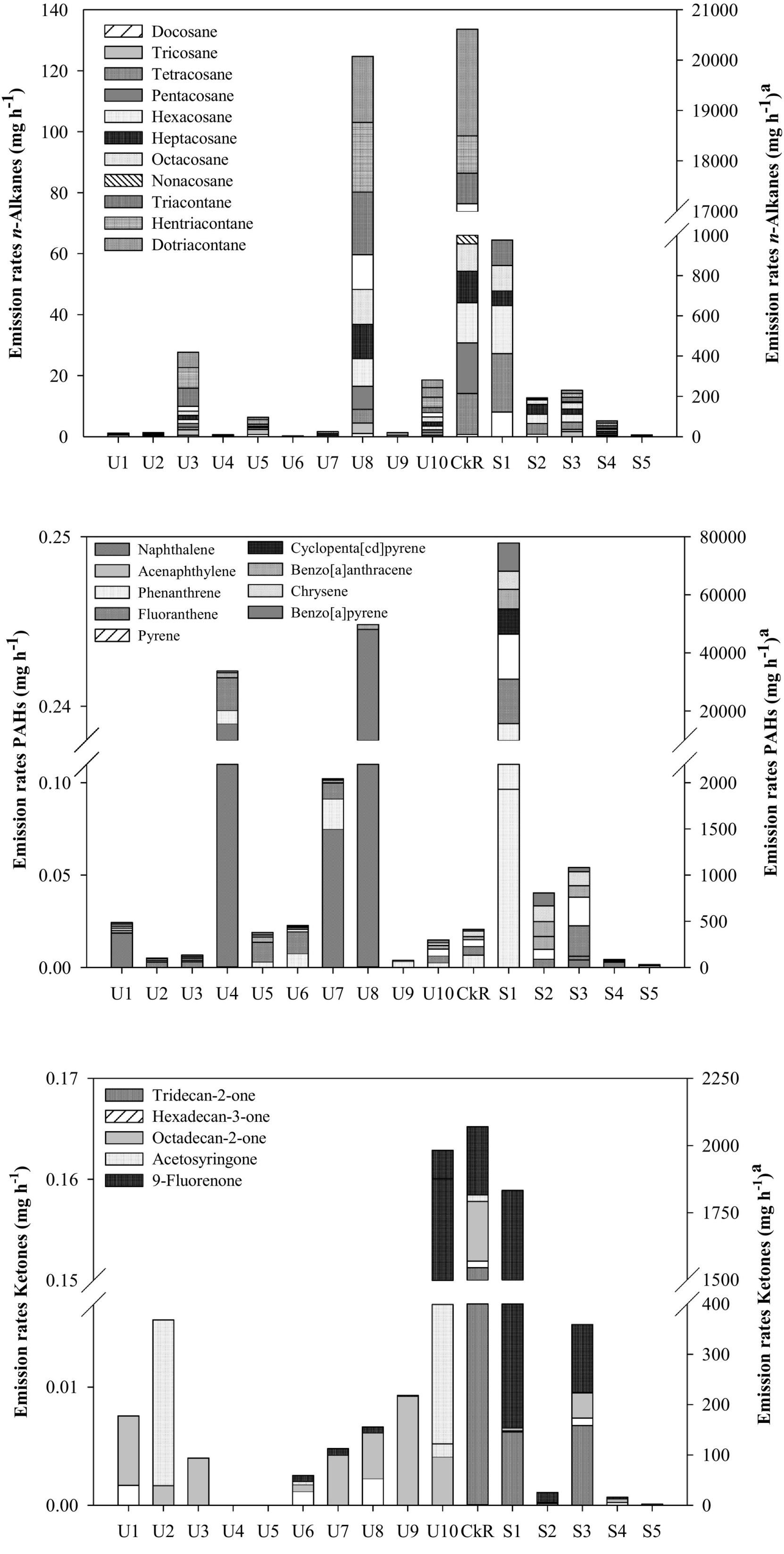
Emission rates n-Alkanes (mg h-1)
Emission rates n-Alkanes (mg h-1)a
0
20
40
60
80
100
120
140
0
200
400
600
800
1000
17000
18000
19000
20000
21000
Docosane
Tricosane
Tetracosane
Pentacosane
Hexacosane
Heptacosane
Octacosane
Nonacosane
Triacontane
Hentriacontane
Dotriacontane
U1
U2
U3
U4
U5
U6
U7
U8
U9
U10
CkR
S1
S2
S3
S4
S5
Emission rates PAHs (mg h-1)
Emission rates PAHs (mg h-1)a
0.00
0.05
0.10
0.24
0.25
0
500
1000
1500
2000
20000
40000
60000
80000
Naphthalene
Acenaphthylene
Phenanthrene
Fluoranthene
Pyrene
Cyclopenta[cd]pyrene
Benzo[a]anthracene
Chrysene
Benzo[a]pyrene
U1
U2
U3
U4
U5
U6
U7
U8
U9
U10
CkR
S1
S2
S3
S4
S5
Emission rates Ketones (mg h-1)
Emission rates Ketones (mg h-1)a
0.00
0.01
0.15
0.16
0.17
0
100
200
300
400
1500
1750
2000
2250
Tridecan-2-one
Hexadecan-3-one
Octadecan-2-one
Acetosyringone
9-Fluorenone
U1
U2
U3
U4
U5
U6
U7
U8
U9
U10
CkR
S1
S2
S3
S4
S5

Figure 2r1

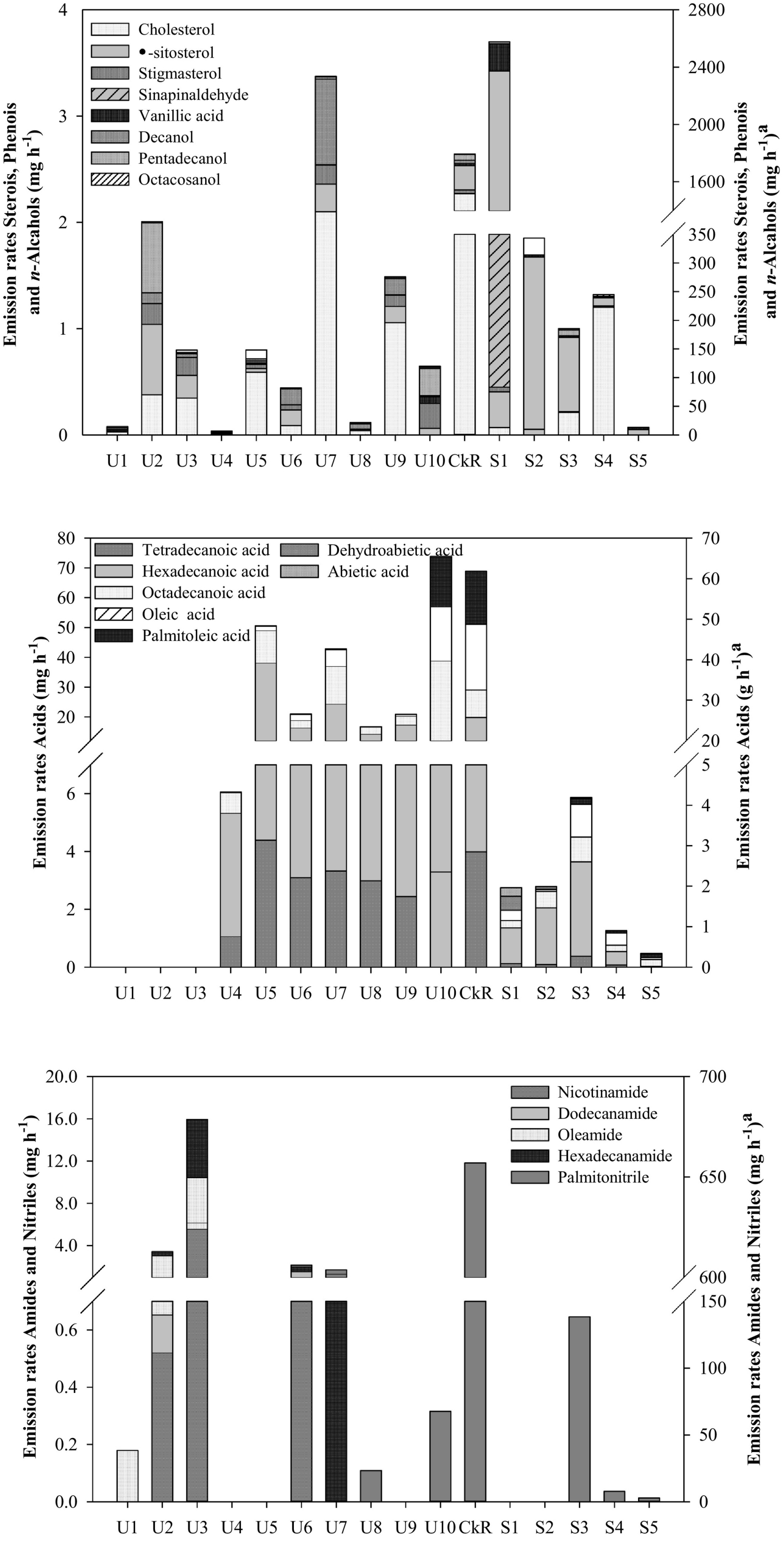
Cholesterol
•-sitosterol
Stigmasterol
Sinapinaldehyde
Vanillic acid
Decanol
Pentadecanol
Octacosanol
Emission rates Sterols, Phenols and *n*-Alcahols (mg h^{-1})
Emission rates Sterols, Phenols and *n*-Alcahols (mg h^{-1})a
0
1
2
3
4
0
50
100
150
200
250
300
350
1600
2000
2400
2800
U1
U2
U3
U4
U5
U6
U7
U8
U9
U10
CkR
S1
S2
S3
S4
S5
Tetradecanoic acid
Hexadecanoic acid
Octadecanoic acid
Oleic acid
Palmitoleic acid
Dehydroabietic acid
Abietic acid
Emission rates Acids (mg h^{-1})
Emission rates Acids (g h^{-1})a
0
2
4
6
20
30
40
50
60
70
80
0
1
2
3
4
5
20
30
40
50
60
70
U1
U2
U3
U4
U5
U6
U7
U8
U9
U10
CkR
S1
S2
S3
S4
S5
Nicotinamide
Dodecanamide
Oleamide
Hexadecanamide
Palmitonitrile
Emission rates Amides and Nitriles (mg h^{-1})
Emission rates Amides and Nitriles (mg h^{-1})a
0.0
0.2
0.4
0.6
4.0
8.0
12.0
16.0
20.0
0
50
100
150
600
650
700
U1
U2
U3
U4
U5
U6
U7
U8
U9
U10
CkR
S1
S2
S3
S4
S5

Figure 2r2

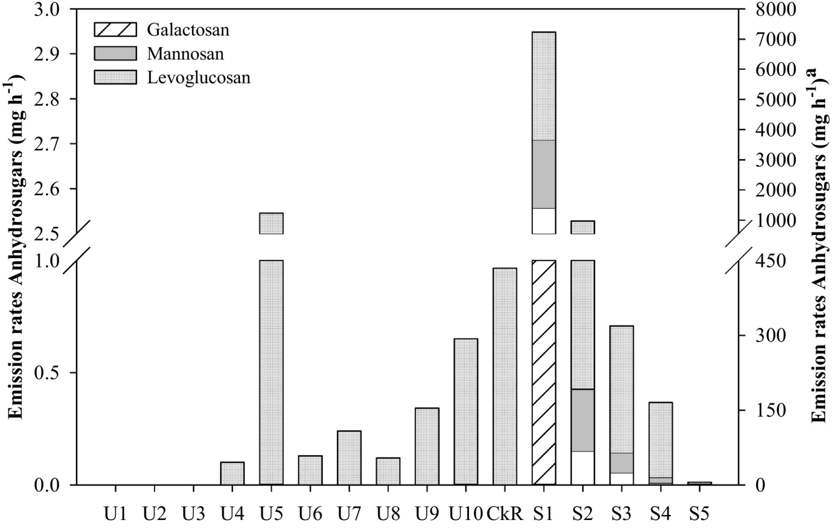
Galactosan
Mannosan
Levoglucosan
Emission rates Anhydrosugars (mg h-1)
Emission rates Anhydrosugars (mg h-1)a
3.0
2.9
2.8
2.7
2.6
2.5
1.0
0.5
0.0
8000
7000
6000
5000
4000
3000
2000
1000
450
300
150
0
U1
U2
U3
U4
U5
U6
U7
U8
U9
U10
CkR
S1
S2
S3
S4
S5

Figure 2r3