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Cooking activities in a domestic kitchen: chemical and toxicological profiling of emissions

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Abstract

To obtain emission factors and cooking-related chemical signatures, a monitoring campaign was carried out in a modern kitchen where different dishes of the Latin cuisine were prepared. Particulate matter (PM₁₀, PM_{2.5} and PM₁) and total volatile organic compounds (TVOCs) were continuously. Passive diffusion tubes for carbonyls and a high volume PM₁₀ sampler were simultaneously used. PM₁₀ filters were analysed for organic and elemental carbon and for multiple organic compounds, including polyaromatic hydrocarbons (PAHs). The toxic potential of PM₁₀ was evaluated using a bioluminescence inhibition bioassay. Acrolein was never detected, while formaldehyde and acetaldehyde levels were comparable to those in the background air. The protection limit for TVOCs was always exceeded. Fine particles comprised more than 86% of the PM₁₀ mass concentrations. PM₁₀ emission rates ranged from 124 to 369 $\mu\text{g min}^{-1}$. Relatively low PAH concentrations were obtained. PM₁₀ encompassed alcohols, acids, plasticisers, alkyl esters, sterols, sugars, polyols, glyceridic compounds, phenolics, among others. Total concentrations of these compounds were 1.9-5.3 times higher during cooking than in the background air but, for some compounds, differences of tens or hundreds of times were registered. PM₁₀ from grilled pork was found to contribute to non-negligible cancer risks and to be very toxic, while samples from other dishes were categorised as toxic.

Keywords: cooking, indoor air quality, PM₁₀ emission rates, organic compounds, bioluminescence inhibition bioassay

1. Introduction

When the COVID-19 pandemic forced global lockdowns, one positive, although temporary, aspect was a decrease in outdoor air pollution (e.g. Baldasano, 2020; Wang et al., 2020). Nevertheless, this focus on outdoor air pollution overlooks the simple fact that most of our exposure takes place indoors, where levels can far exceed those measured outdoors (Alves et al., 2020a). Lockdowns most likely

37 exacerbated this even further. It has long been recognised that cooking is one of the main indoor
38 pollution sources (Abdullahi et al., 2013; Zhao et al., 2019a). It can release unhealthy air contaminants
39 from heating oil, fat and other food ingredients, particularly at high temperatures. Studies have shown
40 that cooking-related pollutants can increase the risk of many diseases (Lee and Gany, 2013; Wang et
41 al., 2017; Zhao et al., 2019a). While there is ample research on the impact of gaseous and particulate
42 matter (PM_{2.5} and PM₁₀) pollution outdoors, much less is known on the health effects of these pollutants
43 when they are generated inside the house. Archer-Nicholls et al. (2016) modelled surface PM_{2.5} fields
44 to assess the impact of residential emissions from cooking and heating activities on the burden of disease
45 in China, estimating that 341,000 premature deaths (159,000 and 182,000 from heating and cooking
46 emissions, respectively) are attributable to residential combustion, which represent about 1/3 of the total
47 deaths due to ambient PM_{2.5} pollution by all causes. These findings stress the need to mitigate emissions
48 from these sources to reduce the health impacts of air pollution.

49 Most households in developing countries without access to clean energy use inefficient combustion
50 appliances and fuels (wood, charcoal, peat, agricultural residues and dung), generally in very poorly
51 ventilated conditions. Due to the extremely high global burden of disease that result from exposure to
52 smoke from household air pollution, research on indoor air quality has been focused on kitchens of low-
53 income countries, mainly considering polycyclic aromatic hydrocarbons (PAHs), because of their
54 documented carcinogenicity (e.g. Chen et al., 2016; Ding et al., 2012; Katiyar and Rastogi, 2014;
55 Munyeza et al., 2020; Taylor and Nakai, 2012). Although more than 2.6 billion people worldwide still
56 do not have access to clean cooking, over 450 million people have changed to modern kitchen
57 appliances since 2010 in India and China, as an outcome of liquefied petroleum gas (LPG) programmes
58 and clean air policies (IEA, 2020). The global household cooking appliance market has been segmented
59 by product into cooktops and cooking ranges, ovens, and specialised appliances. Cooktops and cooking
60 ranges are sub-segmented into gas cooktops, electrical cooktops, induction cooktops, and others. As
61 environmental groups and health authorities have increasingly sounded the alarm over natural gas use
62 in homes, electricity has been promoted as a better option for cooking. The lack of a gas network in
63 many regions, on the one hand, and the ease of cleaning electrical ranges and cooktops, on the other,
64 represent other reasons why the market share of these latter devices has grown. In 2018, across the
65 global market, electrical cooktops represented 33% of the global household cooking appliance market
66 share, while gas cooktops, induction cooktops and others accounted for 40%, 22% and 5%, respectively
67 (OMR, 2019).

68 In addition to the cooking appliance, extraction/ventilation equipment and fuel types, the food
69 preparation techniques, such as frying, roasting and grilling, and the ingredients used can exert a
70 significant impact on pollutant emissions, as demonstrated in bibliographic review articles on the
71 subject (Abdullahi et al., 2013; Kim et al., 2011; Torkmahalleh et al., 2017; Zhao and Zhao, 2018; Zhao
72 et al., 2019a). Cooking ingredients and methods vary widely, reflecting geographical, economic and
73 cultural factors. Thus, it is not possible to generalise emission profiles, being necessary to obtain

74 specific information for each region. A large body of literature has been focused on characterising
75 emissions from Chinese cooking styles and their effects on indoor air quality (Chen et al., 2018; Gao et
76 al., 2013; He et al., 2004a; Hou et al., 2008; Li et al., 2015; Lin et al., 2019; Pei et al., 2016; Peng et al.,
77 2017; Wang et al., 2017; Yao et al., 2015; Zhao et al., 2014, 2018, 2019b; Zhu and Wang, 2003). The
78 determination of new particulate matter emission factors for other types of dishes, more detailed
79 chemical composition profiles, exposure health risk assessments and toxicity testing have been pointed
80 out among the aspects that should be addressed in future researches (Zhao and Zhao, 2018). In order to
81 respond to these challenges and to further develop new cooking-related chemical signatures, which
82 would be beneficial for a quantitative source apportionment, the present study aimed at monitoring
83 indoor air quality in a modern kitchen with clean cooking facilities, equipped with electrical cooktop
84 and range hood, where typical dishes of Latin cuisine were prepared. The study included the
85 determination of particle emission rates, a comprehensive screening of organic compounds and the
86 employment of a toxicity bioassay usually applied to several other matrices but, as far as we know,
87 never to this type of samples.

88

89 **2. Methodologies**

90

91 **2.1. Sampling**

92

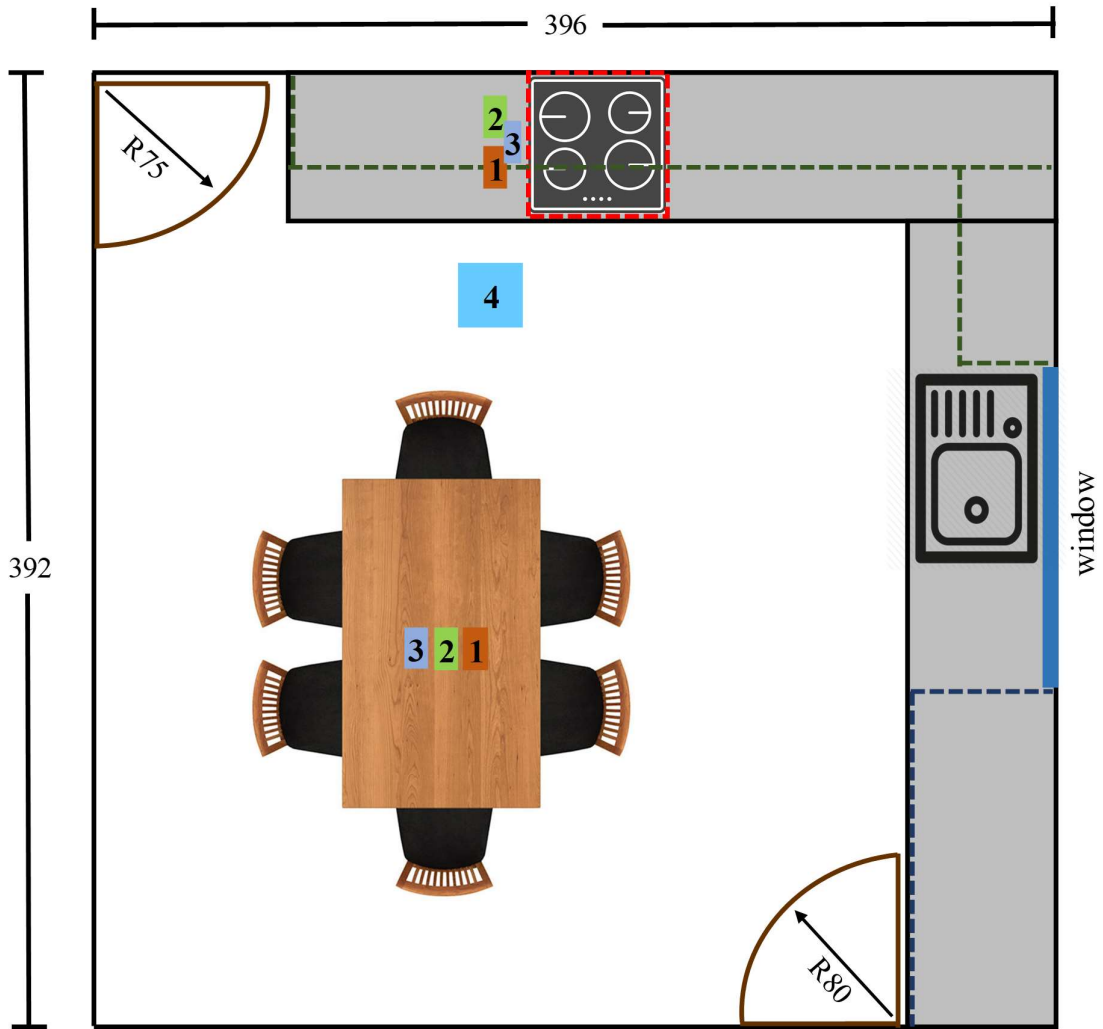
93 The kitchen was equipped with a vitroc ceramic cooktop with four burners and a stainless steel under
94 cabinet range hood with 3 speed settings and maximum suction of $162 \text{ m}^3 \text{ h}^{-1}$. Following the routines
95 of the owner of the house, all the experiments were conducted with the hood turned on, operating at the
96 maximum suction level, and under minimum ventilation (doors and window closed). The 15.5 m^2
97 kitchen was located on the ground floor of a detached house on the outskirts of the city of Aveiro,
98 Portugal. During the experiments, only the cook remained in the kitchen. Aveiro is characterised by a
99 warm summer Mediterranean climate, due to its proximity to the Atlantic Ocean. As typical of
100 Mediterranean climates, summers are dry, and winters are wet. A coastal feature is that frosts are very
101 occasional and never severe. The maritime influence causes a narrow temperature range resulting in
102 summers averaging around $24 \text{ }^\circ\text{C}$ in daytime, substantially lower than inland regions on the same
103 parallel in the Iberian Peninsula. Measurements of cooking emissions took place in September and
104 October 2017. Meteorological information for these two months can be found in Tables S1 and S2.
105 October was exceptionally hot and dry.

106 Real time measurements of temperature, relative humidity, CO_2 , and CO were carried out with
107 indoor air quality probes (Gray Wolf®, WolfSense IQ-610). To monitor CO_2 concentrations outdoors,
108 an IAQ instrument from TSI (model 7575 Q-Trak™) was used. Airborne particulate matter (PM_1 , $\text{PM}_{2.5}$
109 and PM_{10}) mass concentrations were continuously recorded by laser photometric instruments (DustTrak,
110 DRX 8533). These measurements were made in duplicate, simultaneously placing monitors next to the

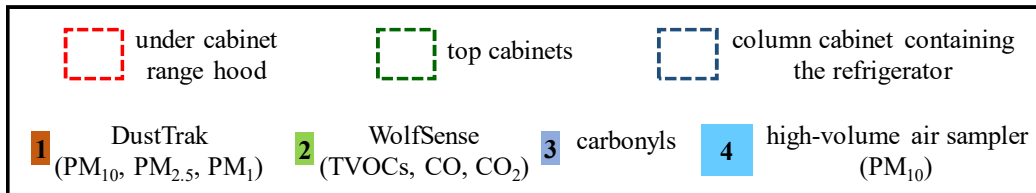
111 cooktop and at about 2.3 m, above the kitchen table (Figure 1). The real time monitors were calibrated
112 by the manufacturers and intercompared with each other at the beginning of the experimental campaign.
113 The DustTrak readings were corrected using the intercomparison line with the gravimetric
114 concentrations obtained in different kitchens, including the measurements of the present study. The
115 main specifications of these automatic instruments can be found in the supplementary material.

116 PM_{10} was collected with a high-volume air sampler (MCV, model CAV-A/mb) equipped with 150
117 mm quartz fibre filters (Pallflex®) and operating at a constant flow of $30 \text{ m}^3 \text{ h}^{-1}$. The filters were
118 preconditioned at 500°C for 6 h. The instrument was positioned between the cooktop and the kitchen
119 table. In order to guarantee enough mass for the subsequent analyses, in some filters it was necessary
120 to accumulate the PM_{10} emitted during the preparation of two or three meals of the same dish. Table 1
121 contains information on what was cooked in the various experiments. The cooking temperature was
122 continuously monitored by a thermocouple immersed in the frying pan or pot. Carbonyls were sampled
123 in parallel with PM_{10} filters using Radiello diffusive passive tubes (Code 165). As for continuous
124 measurements, two sets of passive samplers were used, one next to the electric cooktop, the other on
125 the table. PM_{10} and carbonyl background samples were collected for periods equivalent to the sampling
126 of cooking emissions, after ventilating the kitchen, in the absence of any other activity.

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Figure 1. Layout of the kitchen (dimensions in cm)

Table 1. Details of the preparation of the various dishes

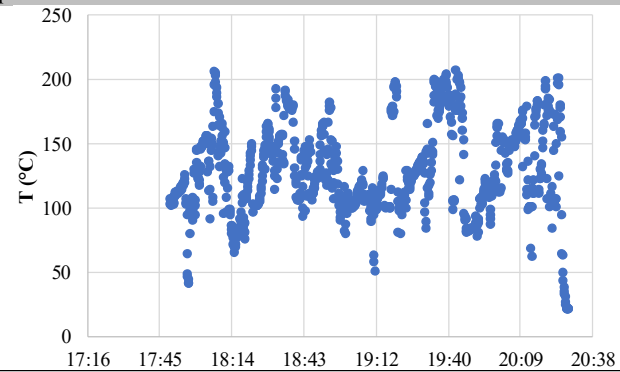
Experience n°	Preparation details	Typical temperature profile
Fried horse mackerel		
# 1	i) 20 little horse mackerels coated with flour + 250 g sunflower oil ii) oil from the frying pan was changed iii) 20 little horse mackerels coated with flour + 250 g sunflower oil Total fish weight = 1630 g Approximate cooking time = 2 h 10 min	
# 2	i), ii) and ii) Idem Total fish weigh = 1273 g Approximate cooking time = 2 h 05 min	
# 3	i), ii) and ii) Idem Total fish weight = 1465 g Approximate cooking time = 2 h 00 min	
Stuffed chicken		
# 1	olive oil 60 g chopped onion and garlic 93 g 1 bay leaf tomato 160 g ½ green bell pepper (<i>Capsicum</i>) 50 g salt + pepper white wine 50 g hot water chicken (with skin partially removed) 1781 g Approximate cooking time = 1 h 50 min	
# 2	olive oil 55 g chopped onion and garlic 104 g 1 bay leaf tomato 185 g ½ green bell pepper (<i>Capsicum</i>) 65 g salt + pepper white wine 115 g hot water chicken (with skin partially removed) 1293 g Approximate cooking time = 2 h 00 min	

3
 olive oil 67 g
 chopped onion and garlic 130 g
 1 bay leaf
 tomato 180 g
 ½ green bell pepper (*Capsicum*) 70 g
 salt + pepper
 white wine 110 g
 hot water
 chicken (with skin partially removed) 1955 g
 Approximate cooking time = 2 h 15 min

Grilled boneless pork strips

1
 3100 g of meat seasoned with lemon juice, salt, pepper and chopped garlic
 Approximate cooking time = 2 h 55 min

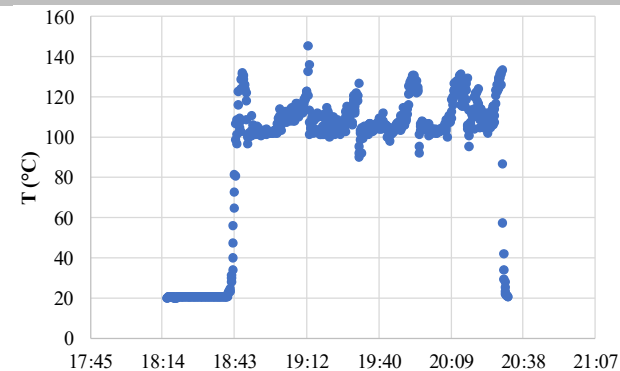
2
 1360 g of meat seasoned with lemon juice, salt, pepper and chopped garlic
 Approximate cooking time = 1 h 55 min



Fried boneless pork strips

1
 2030 g meat marinated for 1 h in wine (200 g), garlic, salt, pepper, bay leaf
 300 mL olive oil
 Approximate cooking time = 2 h 00 min

2
 1595 g meat marinated for 1 h in wine (145 g), garlic, salt, pepper, bay leaf
 300 mL olive oil
 Approximate cooking time = 2 h 20 min



139

140 **2.2. Analytical techniques**

141

142 After gravimetric determination of the mass concentrations, two 9 mm filter punches from each
143 quartz filter were used to analyse the carbonaceous content (organic and elemental carbon, OC and EC)
144 by a thermal optical transmission technique following the EUSAAR2 protocol. A detailed description
145 of the analytical methodology can be found in Vicente et al. (2020). Two filter punches of 47 mm from
146 each sample were extracted, first with dichloromethane (125 mL), and then two times with methanol
147 (25 mL + 25 mL). The total organic extracts were concentrated to a volume lower than 1 mL and dried
148 under a gentle nitrogen stream. The dry extracts were then subjected to silica gel flash chromatography
149 using eluents of increasing polarity to separate organic compounds into different classes. After elution,
150 each organic fraction was vacuum concentrated and dried by nitrogen blow down. Polar compounds
151 with hydroxylic and carboxylic groups were converted into the corresponding trimethylsilyl derivatives.
152 The detailed organic speciation was performed by gas chromatography-mass spectrometry. About two
153 hundred standards were used in the calibration. Standards and sample extracts were both co-injected
154 with internal standards. A full description of the entire procedure for the speciation of organic
155 compounds can be found elsewhere (Alves et al., 2011). Carbonyls were analysed by reverse phase high
156 performance liquid chromatography with UV detection at the Istituti Clinici Scientifici Maugeri (Pavia,
157 Italy) (ICS Maugeri, 2019).

158 The uncertainties associated with the quantification of several organic compounds were estimated
159 following the methodology described by Jiménez et al. (2010). Five main contributions were taken into
160 account: sampling, extraction, clean-up, derivatisation and chromatographic analysis. Depending on the
161 individual compounds, the overall combined uncertainties ranged between 5-12% (PAHs and
162 plasticisers), 10-13% (alcohols), 12-14% (steroids), 6-15% (acids), and 7-11% (sugars).

163

164 **2.3. Particle emission rates**

165

166 Particle emission rates were estimated by a simplified mass balance equation as follows:

$$167 \frac{dC_{in}}{dt} = P\alpha C_{out} + \frac{Q_s}{V} - (\alpha + \kappa)C_{in} \quad (1)$$

168 where C_{in} and C_{out} represent the indoor and outdoor particle concentrations, respectively, P is the
169 penetration efficiency, k is the deposition rate, α is the air exchange rate, Q_s is the indoor particle
170 generation rate, t is time and V is the kitchen volume. The air exchange rates (AER) were estimated
171 based on the CO₂ decay curve (Alves et al., 2020b; see supplementary material), and were as follows:
172 $0.35 \pm 0.29 \text{ h}^{-1}$, $0.51 \pm 0.10 \text{ h}^{-1}$, $0.24 \pm 0.08 \text{ h}^{-1}$ and $0.27 \pm 0.22 \text{ h}^{-1}$ during the preparation of fried horse
173 mackerel, stuffed chicken, fried pork and grilled pork, respectively. The Excepting V , all the parameters
174 in this equation are functions of some other factors and can vary with time. To estimate an average

175 emission rate ($\overline{Q_s}$), equation (1) is simplified by using average values instead of functions. Some further
176 assumptions have been discussed by He et al. (2004b). Thus, the average emission rate can be obtained
177 by equation (2), which ignores the effects of particle dynamics such as condensation, evaporation and
178 coagulation, because these are qualified as being of minor importance, in particular under the conditions
179 usually encountered in residential environments (Buonanno et al., 2009):

$$180 \quad \overline{Q_s} = V \times \left[\frac{C_{in} - C_{in0}}{\Delta t} + \overline{(\alpha + \kappa)} \bar{C}_{in} - \alpha C_{in0} \right] \quad (2)$$

181 where C_{in} and C_{in0} are the peak and initial indoor particle concentrations, respectively, Δt is the time
182 difference between the initial and peak particle concentration, $\overline{(\alpha + \kappa)}$ is the average removal rate and
183 α is the average AER. The particle removal rate is the slope of the line obtained by plotting $\ln(C_{in}(t)/C_{in0})$
184 versus time, where $C_{in}(t)$ are the indoor particle concentrations during the decay period, from the
185 moment the peak concentration is reached, and C_{in0} is the initial indoor concentration (Vicente et al.,
186 2020).

187

188 **2.4. Cancer risk assessment**

189

190 Carcinogenic risks associated with inhalation exposure to PAHs were estimated using the
191 methodology proposed by the United States Environmental Protection Agency and widely described in
192 the literature (Alves et al., 2020b; and references therein). These estimates were made for 2 scenarios:
193 a) a person who spends 4 hours a day cooking, b) a person who spends all day at home, assuming that
194 the background air of the kitchen is representative of the concentrations in the various compartments of
195 the dwelling (4 cooking hours, 20 non-cooking hours). In the latter case, a time-weighted average of
196 the concentrations was calculated. The benzo[a]pyrene equivalent carcinogenicity (BaP_{eq}) was
197 evaluated by multiplying individual PAH concentrations by their toxicity equivalent factor (TEF). The
198 exposure concentration (EC) is given by:

$$199 \quad EC = (CA \times ET \times EF \times ED) / AT \quad (3)$$

200 where CA are BaP_{eq} concentrations (ng m^{-3}), ET is exposure time (h day^{-1}), EF is exposure frequency
201 ($350 \text{ days year}^{-1}$, considering that people spend 15 days on vacation away from home), ED is exposure
202 duration in residential environments (30 years) and AT is average lifetime (613,200 h). The lifetime
203 lung cancer risk (CR) is estimated based on the World Health Organisation inhalation unit risk (IUR)
204 (WHO, 2000) of 8.7 cases per 100,000 people ($8.7 \times 10^{-5} (\text{ng m}^{-3})^{-1}$) and is calculated by multiplying
205 EC by IUR.

206

207 **2.5. Toxicity assessment**

208

209 An ecotoxicity assessment of the bulk material was carried out by the *Vibrio fischeri*
210 bioluminescence inhibition bioassay, as described in the ISO standard 21338 (2010). Being a direct
211 contact test, it simulates a potentially realistic exposure route that occurs between the particles and the
212 recipient biological systems. The repeatability of the kinetic test was assessed in a previous work with
213 the control solution (2% NaCl solution), one sediment sample and a Chromium (VI) solution and was
214 found to be excellent. The coefficient of variation was normally below 1% for 10 replicates of each set
215 (Lappalainen et al., 2001).

216 Filter sample spots of 25 mm in diameter were cut and ground in an agate mortar and then transferred
217 into pre-cleaned 4 mL vials. High-purity water (2 mL) was added to each vial and suspensions were
218 prepared by continuous stirring. The lyophilised bacteria were rehydrated with the reconstitution
219 solution and stabilised at 12°C for 35 min before the luminometer measurements. A duplicate series of
220 eleven two-fold serial dilution was prepared for each sample in 96 well plates. The dilutions were done
221 following the manufacturer protocol with the supplied reagent (2% NaCl solution in water) to ensure
222 optimal osmotic conditions for the bacteria. After addition of the bacterial suspension to the samples,
223 the bioluminescence intensity was continuously recorded for the first 30 s. After the pre-set exposure
224 time (30 min), the luminescence intensity was read again. EC₅₀ (concentration causing 50% of
225 ecological effect, bioluminescence inhibition in this case) was calculated using the Ascent Software
226 provided by Aboatox Co., Finland. The toxicity of each sample was expressed in Toxic Units (TU),
227 which is a dimensionless value given as 100/EC₅₀. Different toxicity classes have been suggested
228 (Chang et al., 2013): TU < 1 non-toxic, 1 < TU < 10 toxic, 10 < TU < 100 very toxic and TU > 100
229 extremely toxic.

230

231 **3. Results and discussion**

232

233 **3.1. Gaseous compounds**

234

235 It has been documented that cooking, especially frying, generates substantial amounts of certain
236 gaseous pollutants such as formaldehyde, acetaldehyde and acrolein (Abdullahi et al., 2013; Katragadda
237 et al., 2010; Peng et al., 2017). In the present study, acrolein, together with butanal, could not be
238 quantified because they were below the detection limit (Table 2). Except next to the cooktop, when
239 horse mackerel was fried, benzaldehyde was also below the detection limit. The levels of formaldehyde
240 and acetaldehyde were generally of the same order of magnitude as those measured in the background
241 air of the kitchen. Formaldehyde never exceeded the protection limit of 100 µg m⁻³ (8-h average)
242 imposed by the Portuguese legislation for indoor air quality (Ordinance n° 353-A/2013). In the case of
243 acetaldehyde, only when boneless pork strips were fried, concentrations were slightly higher (1.2-1.6
244 times) than those registered without cooking activities. Propanal concentrations were up to 15 times
245 higher than in background air. The highest values were recorded for the chicken stew. For isopentanal

246 and pentanal, increases in concentrations in relation to the background levels (1.3-1.8 and 1.3-4.3,
247 respectively) were only observed for frying horse mackerel. In general, there were no marked
248 differences between carbonyls in samples next to the cooktop and those obtained on the kitchen table.
249 All compounds showed concentrations tens of times higher than those recorded outdoors.

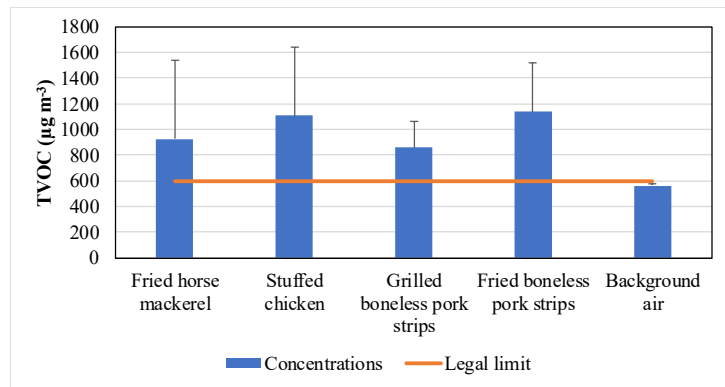
250 As expected, since gas was not used for cooking, CO levels were always less than 2 mg m^{-3} , well
251 below the threshold of 10 mg m^{-3} (8-h average) imposed by the Portuguese legislation. However, the
252 protection limit stipulated for TVOCs was always exceeded (Figure 2). On average, concentrations
253 during cooking were 1.5-2.0 times higher than those recorded without any activity and are within the
254 wide range of values reported for commercial and residential kitchens. Singh et al. (2017) measured
255 TVOC mean concentrations of $2244 \mu\text{g m}^{-3}$ and $932 \mu\text{g m}^{-3}$ in commercial kitchens equipped with LPG
256 stoves with efficient range hoods in north India (Lucknow) and south India (Coimbatore), respectively,
257 while a lower value of $746 \mu\text{g m}^{-3}$ was recorded in control locations (non-kitchen areas). From
258 measurements in China, it was concluded that cooking with electric stoves in commercial kitchens
259 would decrease the concentrations of TVOCs by approximately 44-48% compared to those fuelled by
260 gas (Zhao and Zhao, 2018). Zhao et al. (2014) investigated the effect of exhaust hood while cooking
261 typical dishes of 8 Chinese cuisines. The Chinese researchers concluded that the generation of TVOCs
262 has little to do with the preparation times or the common cooking techniques, but rather with seasonings
263 and ingredients used. When wine was added into a dish, the maximum TVOC concentrations were at
264 least 527-543% higher than those registered during cooking of other dishes. Marinating was pointed
265 out as another reason for high TVOC emissions, even without cooking wine, contributing to peak
266 concentrations that were 80-556% higher than the others. Compared to hood idle mode, the decrease in
267 peak concentrations of TVOCs was 137-4212% in hood working mode. The results of the present study
268 corroborate the conclusions by Zhao et al. (2014), since the highest TVOC concentrations were recorded
269 during the preparation of the dish with addition of wine (stuffed chicken) and for the meat that had
270 previously been marinated (fried pork).

271

Table 2. Carbonyl concentrations during cooking, in the background air of the kitchen and in the outdoor air

Sampling point	Menu	Concentrations ($\mu\text{g m}^{-3}$)								
		Formaldehyde	Acetaldehyde	Acrolein	Propanal	Butanal	Benzaldehyde	Isopentanal	Pentanal	Hexanal
Next to cooktop	Fried horse	33.8	36.6	< 0.02	11.8	< 0.10	2.95	17.4	34.0	156
On the kitchen table	mackerel	33.7	30.5	< 0.02	19.7	< 0.10	< 0.02	12.2	7.59	48.4
Next to cooktop	Stuffed chicken	31.3	40.6	< 0.02	59.9	< 0.10	< 0.02	7.17	< 0.05	27.9
On the kitchen table		33.2	30.1	< 0.02	19.9	< 0.10	< 0.02	11.0	5.42	47.5
Next to cooktop	Grilled boneless	33.6	24.1	< 0.02	7.08	< 0.10	< 0.02	7.07	< 0.05	30.1
On the kitchen table	pork strips	33.4	23.2	< 0.02	6.75	< 0.10	< 0.02	6.80	< 0.05	29.0
Next to cooktop	Fried boneless	36.9	60.5	< 0.02	5.41	< 0.10	< 0.02	6.66	< 0.05	26.4
On the kitchen table	pork strips	33.2	46.9	< 0.02	4.37	< 0.10	< 0.02	5.29	< 0.05	25.1
Background air of the kitchen	-	43.2±0.12	38.0±0.68	< 0.02	4.04±0.12	< 0.10	< 0.02	9.77±0.98	4.88±1.45	36.4±0.40
Outdoor air	-	2.23±0.06	0.75±0.03	< 0.02	0.24	< 0.10	0.25±0.14	0.26±0.07	< 0.05	0.34±0.01

272



273

274 Figure 2. Concentrations of total volatile organic compounds during cooking and protection limit (8-h
275 average) stipulated by the Portuguese legislation of indoor air quality

276

277 3.2. Particle emission rates

278

279 The PM₁₀ emitted by cooking was mainly composed of “high risk” respirable sizes. In fact, fine
280 particles represented a large fraction of the aerosol. On average, PM_{2.5}/PM₁₀ ratios of 0.88, 0.88, 0.95
281 and 0.89 were registered for fried horse mackerel, stuffed chicken, grilled pork and fried pork,
282 respectively. The corresponding ratios of submicron particles (PM₁/PM₁₀) were 0.86, 0.87, 0.94 and
283 0.87. The temporal evolution of the PM₁₀ values measured next to the cooktop and on the table followed
284 the same profile, but the concentration spikes in the latter point were not as pronounced. The global
285 averages recorded near the cooktop were 1.3-1.4 times higher than those obtained on the table. To
286 estimate the PM₁₀ emission rates, the time series obtained at the point closest to the emission source
287 were used.

288 PM₁₀ emission rates did not differ significantly between dishes and were of the same order as those
289 reported for kitchens in Australia, but well below the values described for heating oils and Chinese
290 cooking (Table 3). However, it is necessary to keep in mind that many of the published values were
291 obtained in laboratory simulations with the hood turned off. Chen et al. (2018) reported PM_{2.5} emission
292 rates in the order of thousands of µg min⁻¹ for the preparation of 25 oil-based Chinese recipes in a
293 residential kitchen with range hood off. The repetition of two of the experiments with the exhaust range
294 hood running (233 m³ h⁻¹) showed a reduction of approximately 58% in the PM_{2.5} emission rates. Even
295 so, the emission rates obtained during cooking with the hood on, or those generated during the heating
296 of the pan without ingredients (398 µg min⁻¹), are higher than those of our study. The differences are,
297 at least in part, due to the use of a gas range in the Chinese kitchen. It was also documented that, when
298 a hood was turned on, the peak concentrations of PM_{2.5} suffered a decrease of about 30.8% and 59.6%
299 for oil heating and stir-frying, respectively, with windows and doors closed (Zhao and Zhao, 2018; and
300 references therein). Emission rates of particulate matter have been reported to differ enormously,

301 depending on the type of appliance, cooking method, ingredients, meat/vegetable ratio, among other
 302 factors (Chen et al., 2018). Particulate emissions rates from the oil heating process can also be in a
 303 broad spectrum. Buonanno et al. (2009) found that when grilling, a gas stove produced more particles
 304 than an electric stove. In a compilation of values from the literature, Hu et al. (2012) reported arithmetic
 305 means of 633 ± 1550 and $183\pm817 \mu\text{g min}^{-1}$ for electric and gas ranges, respectively.

306

307 Table 3. Particulate matter emission rates in different cooking experiments

308

Experimental conditions	Food	Emission rates ($\mu\text{g min}^{-1}$)	Reference
PM ₁₀ Residential kitchen Electrical cooktop Hood turned on	Fried horse mackerel Stuffed chicken Grilled pork Fried pork	124-318 250-369 142-225 149-217	This study
Median values of PM _{2.5} Residential kitchens, Brisbane Details not provided	Cooking Cooking pizza Frying Grilling Kettle Microwave Oven	110±990 1590 2680±2180 2780±17,800 670±30 420±30 530±30	He et al. (2004b)
PM _{2.5} Heating oils Measurements in a 0.81 m ³ laboratory hood Oil heated in a glass beaker over a hot plate	Olive Peanut Safflower Soybean Rapeseed Corn Coconut	54×10^3 3600 3000 6000 5400 25.8×10^3 27×10^3	Torkmahalleh et al. (2012)
PM _{2.5} /PM ₁₀ Heating oils Laboratory kitchen Exhaust hood turned off	Rapeseed Soybean Peanut Olive Sunflower Blend	$42.6\times10^3/67.2\times10^3$ $22.8\times10^3/34.8\times10^3$ $66\times10^3/100\times10^3$ $69.6\times10^3/140\times10^3$ $22.2\times10^3/30.6\times10^3$ $24.0\times10^3/40.2\times10^3$	Gao et al. (2013)
PM _{2.5} Chinese residential cooking with range hood off	Chinese cooking Stir-frying, pan-frying, and deep-frying	2056±3034 3352±3358	Chen et al. (2018)
PM _{2.5} Chinese residential cooking with exhaust hood was turned off	Stir-frying Pan-frying Deep-frying Steaming Boiling	2560-2650 1610-1690 320-560 40 80	Zhao et al. (2019b)
Portable kitchen in an 80 m ² open-plan laboratory Gas stove at maximum power when grilling	Cheese Pork meat Bacon Eggplant	9.5×10^3 1.0×10^4 1.2×10^4 5.2×10^2	Buonanno et al. (2009)

Grilling over a gas stove at both minimum and maximum stove power	50 g of bacon	1.3±0.5×10 ⁴ (full power) 5.2±0.2×10 ² (min. power)	Buonnano et al. (2009)
Grilling over a gas stove and an electric stove	50 g of bacon	5.2±0.2×10 ² (gas) 1.5±0.2×10 ³ (electric)	Buonnano et al. (2009)
Gas stove at maximum power for the frying of 50 g of chips	Olive oil	2.8×10 ³	Buonnano et al. (2009)
	Peanut oil	1.8×10 ³	
	Sunflower	1.2×10 ³	
Electric frying pan for frying 50 g of chips and 250 g of cheese	Chips (olive oil)	2.0×10 ²	Buonnano et al. (2009)
	Chips (peanut oil)	6.3×10 ¹	
	Chips (sunflower oil)	3.1×10 ¹	
	Cheese (sunflower oil)	3.3×10 ¹	

309

310

311 3.3. *PM₁₀ levels and carbonaceous constituents*

312

313 The concentrations of PM₁₀ in the filters sampled during cooking were 3 to 6 times higher than that
314 obtained in the background air (Table 4). Bibliographic compilations of particle mass concentrations
315 measured in indoor environments close to cooking activities can be found in Abdullahi et al. (2013),
316 Zhao and Zhao (2018) and Zhao et al. (2019a). Most studies concluded that high-fat foods produce
317 more PM, and frying emit more fine particles than water-based cooking. Concentrations of PM are
318 much higher in gas-fuelled kitchens than those measured in spaces equipped with electrical appliances.
319 In general, PM levels are higher in commercial kitchens than in domestic kitchens mainly due to
320 differences in the amounts cooked. The average mass concentrations of PM_{2.5} released during cooking
321 are within the very wide ranges from 22 to 44,920 µg m⁻³ and from 12 to 4493 µg m⁻³ in commercial
322 and domestic kitchens, respectively. PM₁₀ and PM_{2.5} peak concentrations from heating different types
323 of oil could achieve 7400-30,000 µg m⁻³ and 6500-18,800 µg m⁻³, respectively, under distinct ventilation
324 conditions.

325 Organic carbon accounted for PM₁₀ mass fractions between 17% (fried pork) and 36% (fried horse
326 mackerel), whereas a value of 15% was found in the background air. Regardless of the menu, EC
327 contributed to mass fractions always around 3%, while the background percentage was slightly higher
328 (4.4%). OC/EC ratios in aerosols from cooking were 1.7 to 3.5 times higher than in particulate matter
329 sampled in the absence of activities. Zhao et al. (2019b) collected samples of emissions from a Chinese
330 residential kitchen for the five most common cooking methods. In comparison with our study, higher
331 particulate matter (PM_{2.5}) concentrations were obtained, especially for frying processes: stir-frying
332 0.68-0.99 mg m⁻³, pan-frying 0.29-0.48 mg m⁻³, deep-frying 0.14-0.24 mg m⁻³, steaming 0.04 mg m⁻³,
333 and boiling 0.08 mg m⁻³. In addition to the different cooking styles, the higher concentrations in the
334 Chinese study most likely result from the fact that the fan of the exhaust hood was turned off during the
335 measurement periods. Zhao et al. (2019b) concluded that, in general, the three oil-based cooking
336 methods produced similar chemical profiles, where OC represented the major mass fraction (72-74%)

337 of PM_{2.5}, while the EC contribution was minor (1.0-1.3%). OC mass fractions were lower in samples
 338 from steaming (43.5%) and boiling (59.6%), but the EC proportions were larger compared to the frying
 339 methods (steaming 2.4%, boiling 8.4%). Lower Huboyo et al. (2011) determined the carbonaceous
 340 content of PM_{2.5} collected in a small kitchen of 3 m² in an apartment in Kyoto equipped with exhaust
 341 fan and with a gas-fuelled appliance. OC concentrations ranged from 6.8 µg m⁻³ (chicken boiling) to
 342 43.3 µg m⁻³ (chicken frying), whereas EC varied between 0.78 µg m⁻³ (tofu boiling) and 4.1 µg m⁻³
 343 (chicken frying). Except in PM_{2.5} from chicken boiling, for which an OC mass fraction of 22% was
 344 obtained, the mass fractions for the other 3 samples were around 40%. The EC mass fractions were in
 345 the 3-4% range, irrespective of the menu, in agreement with what was found in the present study. See
 346 and Balasubramanian (2008) reported that the proportions of EC to PM_{2.5} were 9% in boiling and 8-12%
 347 in frying, whilst the respective OC mass fractions were 44% and 52-63%. Since Huboyo et al. (2011)
 348 and See and Balasubramanian (2008) made similar dishes in the same type of appliance, the variations
 349 observed are probably the result of differences in ventilation conditions, given that the latter authors did
 350 not use an exhaust hood. Li et al. (2015) collected PM_{2.5} samples from five different cooking activities
 351 (meat roasting, cafeteria frying, fish roasting, snack-street boiling, and cafeteria boiling) in Ya'an,
 352 China. According to the authors, oils and high-fat raw materials contributed to the high OC contents
 353 (>53% of PM_{2.5}) and OC/EC ratios (>54) in meat roasting and cafeteria frying samples. Contrary to OC,
 354 no significant variation of EC was noticed in all the cooking samples (0.8–2.8% of PM_{2.5}). It was argued
 355 that, in conventional Chinese cooking, the major fuel is natural gas, which has relatively high kindling
 356 temperature and a low EC emission factor. Although clearly higher than those of traffic emissions, very
 357 variable OC/EC ratios have been documented for cooking aerosols in other studies (Li et al., 2015; and
 358 references therein). The wide range arises from differences in the sampling and cooking conditions.

359

360 Table 4. Concentrations of PM₁₀ and carbonaceous constituents during cooking operations and in the
 361 background air of the kitchen

	PM ₁₀ (µg m ⁻³)	OC (µg m ⁻³)	EC (µg m ⁻³)	OC/EC
Fried horse mackerel	71.1	25.7	2.13	12.1
Stuffed chicken	24.3	4.74	0.729	6.5
Grilled boneless pork strips	73.1	25.1	2.39	10.5
Fried boneless pork strips	32.4	5.42	0.918	5.9
Background air	11.7	1.77	0.510	3.5

362

363 3.4. PM₁₀-bound polycyclic aromatic hydrocarbons

364

365 It has been argued that, at high temperature, by pyrosynthesis, organic compounds are partially
 366 cracked to smaller unstable fragments, which recombine and lead to relatively stable PAHs (Zhu and

367 Wang, 2003). Total PAH concentrations of 2.38, 0.920, 2.79, 1.43 and 1.21 ng m⁻³ (Table 5) were found
368 in PM₁₀ samples from fried horse mackerel, stuffed chicken, grilled boneless pork strips, fried boneless
369 pork strips and background air, respectively. The corresponding PM₁₀ mass fractions were 33.5, 37.9,
370 38.2, 44.2 and 104 µg g⁻¹ (Table S3). Contrary to that observed for the other menus, aerosols sampled
371 during the preparation of the chicken presented concentrations lower than those recorded in the
372 background air. This can result from the extraction efficiency of the hood. Chen et al. (2019) conducted
373 a study in a test chamber with an exhaust hood installed 0.7 m above the deep-frying pan operating at
374 flows ratios in the range of those of the present study. The capture efficiencies of the exhaust hood for
375 total PAHs (gaseous and particulate phases) were found to be up to 77%. It is necessary to bear in mind
376 that concentrations of the present study may, in fact, be significantly higher, since an important part of
377 the PAHs, especially the lighter ones, is in the gas phase. The highest PAH concentrations were
378 observed in the aerosol emitted when grilling pork. PAHs are formed when fat and juices from meat
379 drip onto the heated surface of the grill causing charring and smoke. The concentration obtained when
380 frying horse mackerel was about twice that observed during the frying of pork. It should be noted that
381 higher temperatures, up to 200°C, were reached by the sunflower oil used in the first dish, while lower
382 values, up to 140°C, were measured in the olive oil where the second dish was made. It has been reported
383 in previous studies that oil temperatures significantly affect PAH emissions in cooking fumes. Chen
384 and Chen (2003) analysed the smoke emitted during the cooking of chicken legs and concluded that
385 canola oil emitted the highest levels of PAHs, followed by soybean and sunflower oils. Yao et al. (2015)
386 reported that cooking with rapeseed oil produced the highest oil fume PAH concentrations, followed by
387 cooking with olive, peanut and soybean oils.

388 Zhao and Zhao (2018) summarised the PAH levels measured from Chinese cooking in domestic and
389 commercial kitchens. In general, total particle-phase PAH concentrations ranged from 5.6 to 60 ng m⁻³
390 and from 6.7 to 63,050 ng m⁻³ for domestic and commercial kitchens, respectively, which greatly
391 depended on the cooking methods. Jørgensen et al. (2013) measured the gas and particulate phase levels
392 of 32 PAHs in a laboratory kitchen equipped with a canopy hood. Measurements were performed by
393 pan frying fresh and smoked bacon on both electric and gas stoves. Total PAH concentrations ranged
394 between 270 and 300 ng m⁻³. Frying fresh bacon in a gas or in an electric stove was found to produce
395 similar PAH contributions. The highest level of particulate PAHs was obtained when frying smoked
396 bacon. The smoked bacon experiment showed a different PAH profile, in which retene represented 10%
397 of total PAHs, while for the two fresh bacon experiments, it constituted less than 1%. In the present
398 study, retene was also among the compounds with the highest values, accounting for 3.3-8.3% of total
399 PAH concentrations. In the 80's, this alkylated compound was proposed as a marker for softwood
400 combustion and PAH source apportionment but, more recently, it was also detected in coal (Shen et al.,
401 2012) and traffic-related aerosols (Alves et al., 2020c). The presence of retene in cooking aerosols raises
402 strong doubts about its use as a tracer and opens doors for future research on its formation mechanisms.
403 It has been speculated that retene probably has a unique formation process during cooking (Jørgensen

404 et al., 2013). Recently, retene in PM₁₀ from the Amazon region has been proven to cause DNA damage
405 and cell death (Alves et al., 2017).

406 See and Balasubramanian (2008) collected PM_{2.5} samples in the kitchen of an apartment in Singapore
407 while cooking in a gas stove. No exhaust extractor was employed. The mass concentration of the 16
408 priority PAHs was the highest during deep-frying (36.5 ng m⁻³), followed by pan-frying (25.0 ng m⁻³),
409 stir-frying (21.5 ng m⁻³), boiling (14.3 ng m⁻³), and steaming (10.5 ng m⁻³). These concentrations
410 represented a significant enhancement (by a factor of 3-12) over the total PAH loading in the
411 background air (3.10 ng m⁻³). PAH concentrations of the same order of magnitude as those reported by
412 See and Balasubramanian (2008) were found in PM_{2.5} sampled in a Chinese residential kitchen during
413 the preparation of different recipes with the fan of the exhaust hood turned off (Zhao et al., 2019b): stir-
414 frying (58.9-92.1 ng m⁻³), pan-frying (41.2-72.6 ng m⁻³), deep-frying (34.6-50.9 ng m⁻³), steaming (29.5
415 ng m⁻³) and boiling (8.00 ng m⁻³). Concentrations tens or hundreds of times lower in the present work
416 compared to those reported in other studies result not only from different types of cooking, but also
417 from the fact that gas was not used as fuel and the hood was turned on. Much higher PAH levels have
418 been documented for a commercial kitchen in India, equipped with a “low side wall” rangehood
419 chimney, where at least 5-10 frying activities were conducted during indoor air monitoring. In this
420 kitchen, mean individual concentrations up to 18 mg m⁻³ of particulate-phase PAHs were recorded
421 (Singh et al., 2016). See et al. (2006) determined the levels of PM_{2.5}-bound 16 priority PAHs in a
422 university canteen (Chinese, Malay and Indian food stalls). The mass concentrations of PAHs were the
423 highest at the Malay stall (609 ng m⁻³), followed by the Chinese stall (141 ng m⁻³), and the Indian stall
424 (37.9 ng m⁻³). This difference in levels was mainly attributed to the distinctive cooking methods
425 employed at the three food stalls and, to a lesser extent, to the amount of food cooked, and the cooking
426 time. Frying processes, especially deep-frying, release more air pollutants, probably due to the high oil
427 temperatures used in such operations. Moreover, it was observed that deep-frying at the Malay stall or
428 stir-frying at the Chinese stall gave rise to greater abundances of higher molecular weight (HMW) PAHs,
429 while low-temperature cooking, such as simmering at the Indian stall, presented higher concentrations
430 of lower molecular weight (LMW) PAHs. The present study contradicts this finding, since the lowest
431 HMW/LMW ratios were obtained in PM₁₀ samples from fried horse mackerel and fried boneless pork
432 strips. These ratios were 1.0 and 1.7, respectively, whereas higher values were found for stuffed chicken
433 (2.6) and grilled boneless pork strips (4.3). The highest ratios were obtained for very fluctuating
434 temperatures between 100 and 200°C, in the case of grilled pork strips, and for very constant values
435 around 100°C, in the case of chicken. Temperatures in the ranges 150-200°C and 100-140°C were
436 recorded when frying horse mackerel and pork strips, respectively. Thus, it was not possible to observe
437 an unequivocal increase in the concentrations of HMW PAHs with the increase of the cooking
438 temperature. This is in line with the results of Zhao et al. (2019b) who reported higher HMW and LMW
439 abundances for steaming and oil-based methods, respectively, arguing that five- to seven-ring PAHs

440 can be decomposed or destroyed at high cooking temperatures during frying, which facilitates LMW-
 441 PAH formation.

442

443 Table 5. Concentrations of PM₁₀-bound PAHs (pg m⁻³). Empty cells mean not detected or of the same
 444 level of blanks.

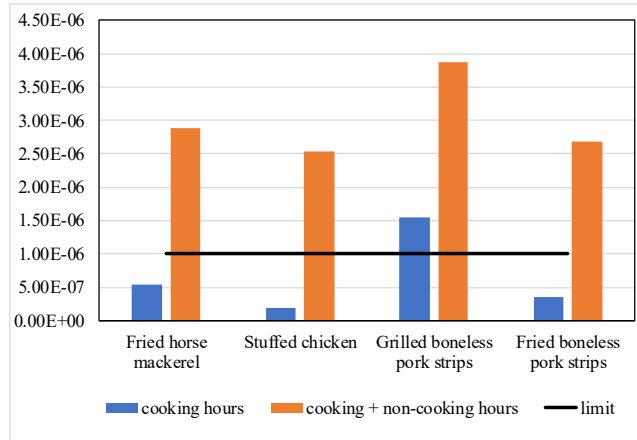
	Fried horse mackerel	Stuffed chicken	Grilled boneless pork strips	Fried boneless pork strips	Background air
Naphthalene	218	2.04	14.0		156
Acenaphthylene	9.13			4.36	4.67
Acenaphthene	11.3	7.48	5.87	3.96	13.4
Fluorene	134	24.7	11.2	91.0	79.8
Phenanthrene	153	43.4	61.6	91.6	27.0
Anthracene	9.62	3.38	6.35	6.45	5.23
Fluoranthene	133	51.7	144	92.8	23.4
Pyrene	341	86.4	185	168	64.9
p-Terphenyl	2.83		3.28		
Retene	198	36.8	90.9	71.4	45.7
Benzo[a]anthracene	232	52.1	212	118	64.7
Chrysene	616	244	864	421	188
Benzo[b]fluoranthene	90.0	142	291	122	169
Benzo[k]fluoranthene	35.9	55.8	194	66.8	
Benzo[e]pyrene	111	132	298	126	202
Benzo[a]pyrene	45.0	1.89	152	21.6	44.6
Perylene					
Indeno[1,2,3-cd]pyrene	20.0	16.7	122	6.76	52.2
Dibenzo[a,h]anthracene			11.0		
Benzo[g,h,i]perylene	22.6	17.9	128	21.6	71.3
SUM	2382	920	2794	1433	1213

445

446 Diagnostic ratios of PAH isomers are not source-specific, since they markedly reflect the efficiency
 447 of combustion and fuel quality. However, these ratios can give some clues about the relative importance
 448 of possible sources (Jørgensen et al., 2013). Most of the ratios were similar between the different
 449 cooking operations. Except for the Phe/(Phe+Ant) ratio (Table S4), for which the values are clearly
 450 higher than those reported for petrogenic sources (0.5 gasoline engines, 0.37-0.70 diesel engines), all
 451 the others overlap with the typical values of traffic exhaust emissions (Saldarriaga-Noreña et al., 2015;
 452 and references therein).

453 The compounds that contributed most to the BaP_{eq} concentrations were benzo[a]pyrene (5.8-58.9%),
 454 followed by benzo[a]anthracene (8.2-55.4%) and benzo[b]fluoranthene (9.8-43.4%). Cancer risks for a
 455 typical behaviour pattern (4 h day⁻¹ cooking, 350 days year⁻¹) are shown in Figure 3). Grilling pork
 456 contributed to risks that exceeded the acceptable level (1 × 10⁻⁶). Although during the preparation of
 457 some dishes the CR values were below the recommended limit, the integrated daily exposure presented

458 non-negligible risks, indicating an increased concern with PAHs in the background air. Higher risks
 459 have been reported in the literature for commercial kitchens and deprived rural areas. See et al. (2006)
 460 estimated cancer risks of 4.08×10^{-3} , 1.21×10^{-3} and 1.07×10^{-3} at Chinese, Malay and Indian stalls,
 461 respectively. Sharma and Jain (2020) measured personal PAH concentrations among three types of
 462 kitchens while using traditional and improved cookstoves in rural settings in India. The overall cancer
 463 risks were higher in enclosed (2.5×10^{-3} , 6.4×10^{-4}) compared to semi-enclosed (8.4×10^{-4} , 1.3×10^{-4})
 464 and open kitchens (2.2×10^{-4} , 4.6×10^{-5}) during use of traditional and improved cookstoves, respectively.
 465



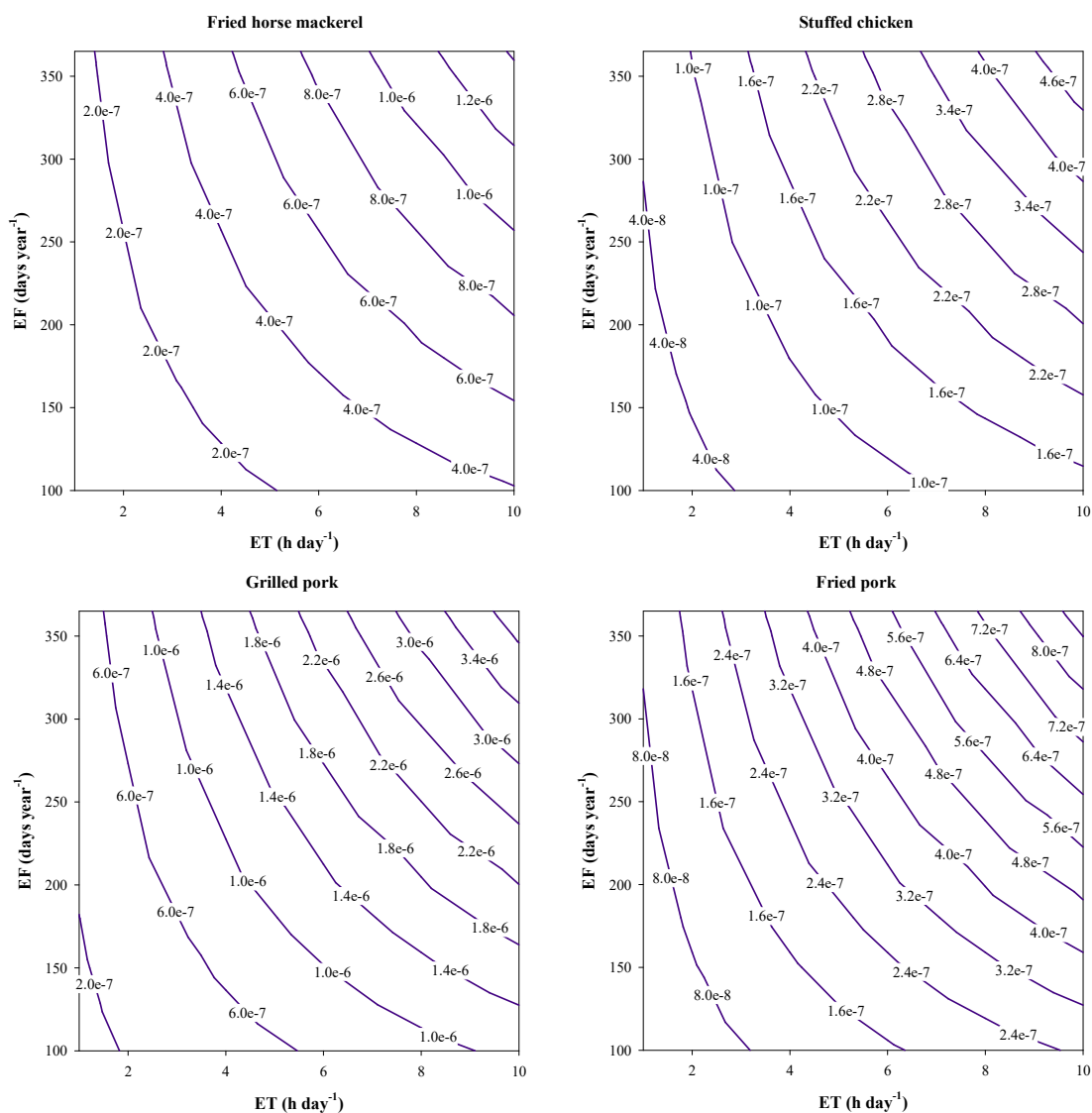
466
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468 Figure 3. Cancer risks from inhalation of PM₁₀-bound PAHs for common kitchen usage patterns (4
 469 hours a day, 350 days a year)
 470

471

472 However, given that the inhaled concentrations, and consequently the cancer risks, depend on the
 473 frequency and duration of the exposure, a sensitivity analysis was made considering several cooking
 474 times per day and variable annual periodicities (Figure 4). It can be observed that, for chicken and fried
 475 pork, regardless of exposure times and frequencies, values are always lower than the carcinogenic target
 476 risk of 1×10^{-6} . Exposure to fumes from frying horse mackerel for more than 7 hours a day or more than
 477 250 days a year leads to the possibility of developing 1 excess cancer case per million people. The PM₁₀
 478 emitted when grilling pork is the one that constitutes the highest risk, exceeding the value recommended
 479 by the WHO when this dish is made every day of the year for periods of exposure greater than 3 hours
 480 a day. If the frequency of cooking is reduced to 180 days a year, the associated risks are considered
 481 unacceptable for periods of exposure that exceed 5 h per day.

481



482 Figure 4. Variation of cancer risks with exposure time and exposure frequency

483

484 3.5. *PM*₁₀-bound oxygenated organic compounds

485

486 About 150 oxygenated constituents were quantified in the particulate matter sampled in the kitchen,
 487 comprising alcohols, several types of acids, plasticisers, alkyl esters, sterols, sugars, polyols, glyceridic
 488 compounds, phenolics and other substances (Figure 5). The full list can be found in the supplementary
 489 material (Tables S6 and S6). Total concentrations of these compounds were globally 1.9 (stuffed
 490 chicken) to 5.3 (grilled pork) times higher during cooking than in the background air. As summarised
 491 by Abdullahi et al. (2013), cooking involves a vast array of chemical reactions. When heated,
 492 disaccharides and polysaccharides or carbohydrates suffer hydrolysis reactions, which break down the
 493 complex molecules into single ring sugars. If sugars are further heated, degradation reactions will take
 494 place with opening of the rings, leading to the formation of new compounds, such as acids. With a

495 sufficiently high temperature rise, recombination of degradation products may occur to form chain-like
496 molecules. Fat in uncooked meat, which occurs as triglycerides (fatty acids esterified to a glycerol
497 backbone), is thermally oxidised or hydrolysed. These reactions result in the formation of free glycerol,
498 free fatty acids and mono and diglycerides. Chemical reactions between proteins and carbohydrates or
499 sugars (Maillard reactions) comprise initial degradation to amino acids and smaller sugars and then
500 production of acids and aldehydes. These latter two classes react with amino acids to form several other
501 new chemicals.

502 Carboxylic acids were always a dominant organic class, constituting 46, 23, 24, 18 and 11% of the
503 total quantified compounds in the PM₁₀ from fried horse mackerel, stuffed chicken, grilled pork, fried
504 pork and background air, respectively. This class encompassed the homologous series of *n*-alkanoic
505 acids from C₆ to C₂₆ and some alkenoic acids. As observed in Chinese cooking aerosols (Lin et al., 2019;
506 Pei et al., 2016; Zhao et al., 2007, 2019a), palmitic, linoleic, stearic and myristic were the most abundant
507 acids. The strong prevalence of even-numbered homologs is highlighted by carbon preference indices
508 (CPI) between 6 and 12. Zhao et al. (2007) reported that concentrations of saturated and unsaturated
509 fatty acids were 14 and 3.2 times higher, respectively, in aerosols from Western-style fast food
510 compared to Chinese cooking, attributing the differences mainly to ingredients and temperatures.
511 Ingredients with higher fat content can produce more fatty acids than low fat-content ingredients under
512 the same cooking condition. Higher temperatures favour the degradation of triglycerides and
513 phospholipids and the release of fatty acids. In our study, the highest concentrations of saturated and
514 unsaturated fatty acids were observed in the PM₁₀ from fried horse mackerel and grilled pork. The
515 highest temperatures, around 200°C, were indeed reached during the preparation of these dishes. The
516 distribution and emission rates of fatty acids in cooking aerosols can also be markedly influenced by
517 the fat content and composition of raw foods. The ratios of the polyunsaturated linoleic acid to the
518 saturated stearic acid (C_{18:2}/C₁₈) was 4 in PM₁₀ from horse mackerel, while values < 1 were found in the
519 remaining samples, according to a much higher content of unsaturated fatty acids in fish.

520 The abundances of dicarboxylic acids were far less than those of saturated and unsaturated fatty
521 acids. Regardless of the menu, nonanedioic acid was always the most abundant diacid, as described for
522 Chinese and Italian cooking styles (Pei et al., 2016; Zhao et al., 2007). However, hexanedioic acid has
523 been depicted as the dominant diacid in meat cooking fumes (Rogge et al., 1991; He et al., 2004a; Zhao
524 et al., 2007). Thus, hexanedioic to nonanedioic ratios > 1 have been pointed out as typical of Western-
525 style fast food, while values < 1 are distinctive of Chinese cooking (Zhao et al., 2007). In our study, this
526 ratio was always below 0.16.

527 Aerosols from cooking also comprised the homologous series of *n*-alkanols from decanol to
528 octacosanol, maximising at C₁₆. CPI values always above 14 reflect the even over odd carbon number
529 predominance. The concentrations of these compounds were 2 to 3 times higher in samples collected
530 during cooking periods compared to the background air. Phenolic compounds were another group of
531 substances with hydroxyl groups present in PM₁₀. They can be classified as synthetic or natural. All

532 parts of plants may contain phenolic compounds such as hydroxycinnamic acid derivatives (e.g.
533 coumaric, caffeic, ferulic) and hydroxybenzoic acid derivatives (e.g. hydroxybenzoic, vanillic, syringic,
534 gallic). All these constituents were detected in PM₁₀ from cooking. Phenolic compounds are used as
535 primary antioxidants to delay or inhibit the initiation step or interrupt the propagation step of lipid
536 oxidation, thus decreasing the formation of volatile decomposition products that cause rancidity
537 (Shahidi and Ambigaipalan, 2015). The most abundant compound was 2,6-di-tert-butylphenol, a
538 synthetic product. Among other applications, 2,6-di-tert-butylphenol is added as antioxidant to plastics.
539 Due to contact with plastic piping, drinking water may contain 2,4-di-tert-butylphenol. It can also be
540 part of machine wash liquids/detergents. Thus, contamination of food by multiple routes can occur. The
541 concentration of this compound was lower in PM₁₀ from stuffed chicken than in the background air.
542 However, in the aerosols of all other foods the levels were 3 times higher, suggesting that its emission
543 may depend not only on the ingredients, but also on the cooking temperatures. Liu et al. (2020)
544 investigated the effects of four cooking methods (boiling, pressure, microwave, slow) and three heating
545 solutions (water, salt, sugar) on the phenolic acids and antioxidant properties of different legumes. It
546 was shown that cooking increased free phenolic acids and lessened bound phenolic acids in some
547 legumes, while decreased both free and bound phenolic acids in others. The effect of cooking methods
548 on the phenolic composition showed significant differences among the four cooking methods. The
549 possible physico-chemical mechanisms by which phenolic compounds are released to form particulate
550 matter are still unclear.

551 Some plasticisers were detected in PM₁₀. These compounds are extensively used in the
552 manufacturing of plastic materials and are now widespread in all environmental compartments. Among
553 the commercially important plasticisers, phthalic acid esters, or phthalates, account for the major share
554 of the market. The migration of phthalates from packaging materials to fatty food stuffs is a well-known
555 source of food contamination (Petersen and Breindahl, 2000). In the last decades, phthalates have been
556 the focus of much attention since several of these compounds are suspected of owning endocrine and
557 disrupting properties either by mimicking or antagonising the effects of sex hormones or by more
558 indirect mechanisms (Bui et al., 2016). Concentrations of plasticisers in aerosols from meat cooking
559 were 1.2-1.6 times higher than the background level, but in PM₁₀ from frying horse mackerel an
560 identical value was obtained. Diethylhexyl phthalate was the most abundant compound. Fierens et al.
561 (2012) analysed 8 phthalate compounds in food products before and after cooking (boiling, steaming,
562 deep-frying and grilling). In general, phthalate concentrations in foods declined after cooking, except
563 in vegetables, where almost no effect was seen. The authors raised the possibility that some phthalates
564 with high vapour pressure can evaporate during processing. After evaporation and condensation, they
565 can partition to the particle-phase. On the other hand, since phthalates are lipophilic, when food products
566 lose fat during processing, they are removed together with the fat phase. Thus, oily fumes can be
567 enriched in plasticiser compounds that were originally present in the lipid phase of foodstuffs.
568 Phthalates can also originate in coatings of non-stick cookware products.

569 Levoglucosan was detected in similar concentrations in background air and in PM₁₀ from stuffed
570 chicken and fried boneless pork strips. However, the levels of these anhydrosugar were 4.6 and 2.2
571 times higher in aerosols from fried horse mackerel and grilled pork compared to the background value,
572 respectively. Although levoglucosan has been widely used as a biomass burning tracer (Vicente and
573 Alves, 2018), it was previously observed in emissions from Chinese cooking (He et al., 2004a; Hou et
574 al., 2008; Zhao et al., 2007). It was hypothesised that the high temperatures reached in cooking
575 processes could lead to thermal degradation of vegetable cellulose and, consequently, to the emission
576 of levoglucosan. Spices such as *Cuminum cyminum* and *Capsicum* (powder of dry vegetables) used as
577 condiments can also decompose to form levoglucosan (Hou et al., 2008). Although spices and/or green
578 bell pepper were used to season the chicken and fried pork, the temperatures necessary to convert
579 cellulose into levoglucosan were possibly not reached. The highest concentrations in horse mackerel
580 PM₁₀ may be associated with the flour used to bread the fish. Most of the fibre present in flour is
581 cellulose.

582 PM₁₀ from grilled boneless pork strips and fried horse mackerel displayed high amounts of sterols,
583 exceeding by 337 and 91 times the concentration recorded in the absence of activities, respectively.
584 Sterols of plant origin are generally referred to as phytosterols, and those of animal origin as zoosterols.
585 Nevertheless, these groups are not mutual exclusive. Cholesterol, a chief zoosterol, can also be found
586 in plant sources, sometimes in large quantities. In the present study, this zoosterol dominated, although
587 the concentration of sitosterol in horse mackerel PM₁₀ also deserves to be highlighted. Sitosterol was
588 detected in Chinese cooking aerosols, but not in emissions from Western-style fast food (Zhao et al.,
589 2007).

590 Several glyceridic compounds were present in the samples of particulate material. Among them, the
591 most abundant were free glycerol and the monoglycerides monomyristin, monostearin and
592 monopalmitin. Globally, the levels of these compounds exceeded by 3 to 40 times the background value.
593 The maximum concentration was achieved in the fumes of grilled pork. The fat in uncooked meat occurs
594 as triglycerides, which consist of three fatty acids esterified to a glycerol backbone. During the cooking
595 process, triglycerides can be hydrolysed or thermally oxidised, yielding, in addition to fatty acids, free
596 glycerol, mono- and diglycerides (Nolte et al., 1999). Triglycerides are also main constituents of
597 vegetable oils. Thus, upon heating, triglycerides of edible oils can form the same reactional products as
598 those observed in meat cooking.

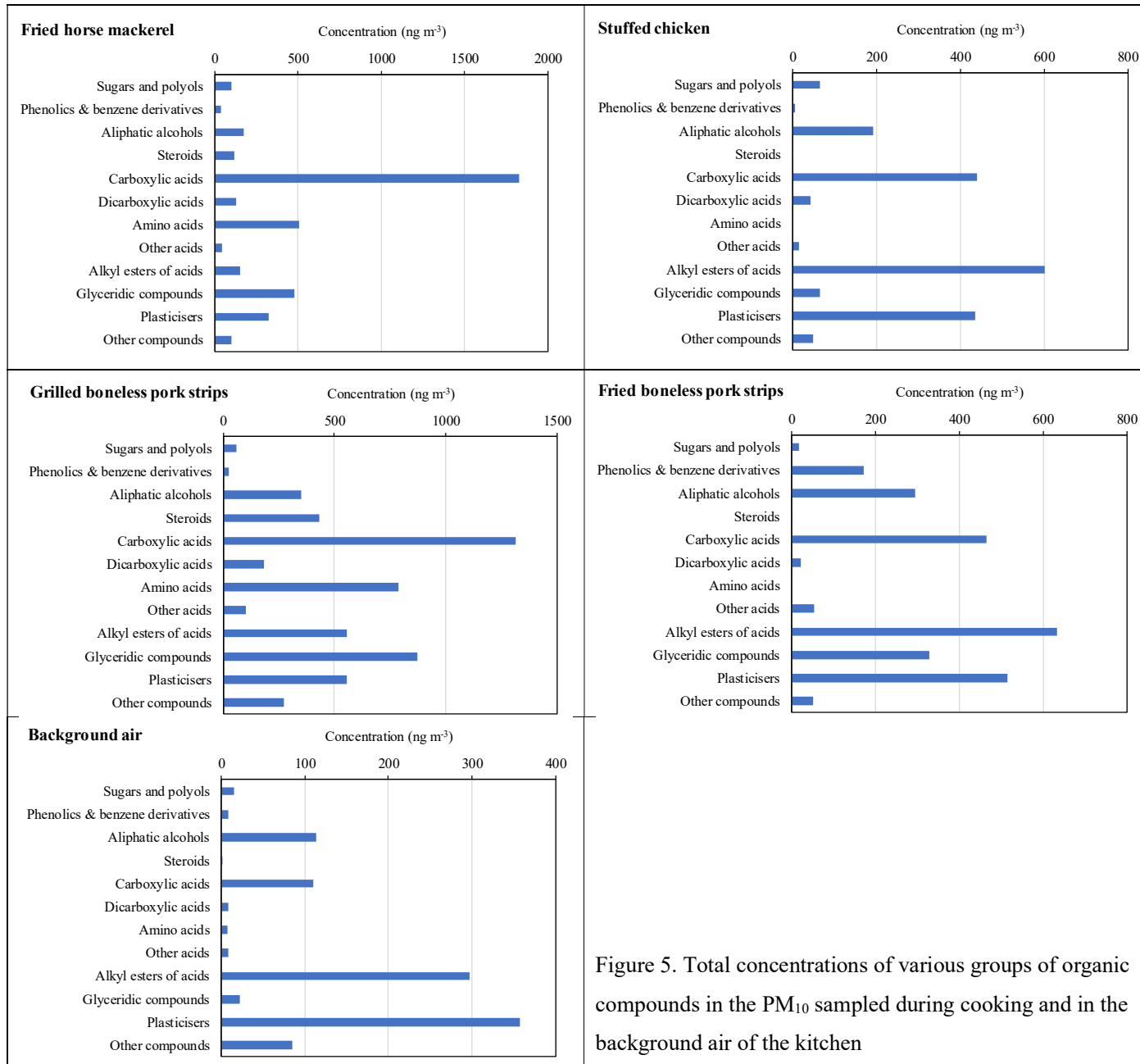


Figure 5. Total concentrations of various groups of organic compounds in the PM₁₀ sampled during cooking and in the background air of the kitchen

599

600 Various fatty acid alkyl esters were detected both in the background air of the kitchen and in the
601 samples collected while cooking. Isopropyl myristate and isopropyl palmitate were the most abundant
602 compounds. These constituents are widespread because they are in the list of ingredients of several
603 cosmetics and personal care products. Fatty acid alkyl esters are also a family of natural neutral lipids
604 present in vegetable oils formed by esterification of free fatty acids with low molecular alcohols.
605 Inappropriate practices during the olive oil extraction process and bad quality of the olive fruits promote
606 their formation. Thus, alkyl esters of fatty acids are direct indicators of degradation or fraudulent
607 adulteration of vegetable oils (Costa et al., 2017). It is also possible that these ester compounds are a
608 secondary product of lipid oxidation, which include destruction of essential fatty acids, during meat
609 cooking (Domínguez et al., 2019). It has been observed that there is a great variability in changes
610 concerning oxidation of individual fatty acids in response to different cooking methods, but it seems
611 that, in general, losses of total lipids increase directly with the cooking time and internal temperature
612 reached by meat (Alfaia et al., 2010). For the three meat dishes of the present study, the total
613 concentrations of fatty acid alkyl esters were approximately double that recorded in the background air.

614 In the PM_{10} emitted during the frying of horse mackerel and the grilling of pork, several amino acids
615 were detected. Their concentrations exceeded by 70 and 108 times, respectively, those obtained in the
616 absence of activities. Although the presence of these compounds, as far as we know, has not been
617 previously described for cooking aerosols, they have been quantified as components of primary
618 biological particles in urban (e.g. Abe et al., 2016; Di Filippo et al., 2014), marine (e.g. Mandalakis et
619 al., 2011) and Arctic (e.g. Rad et al., 2019) environments. Some works on food degradation during
620 cooking have found that the degree of protein denaturation depends on the cooking method, temperature
621 and type of food, demonstrating differences in protein and amino acid contents (e.g. Hodgkinson et al.,
622 2018; Kim et al., 2017).

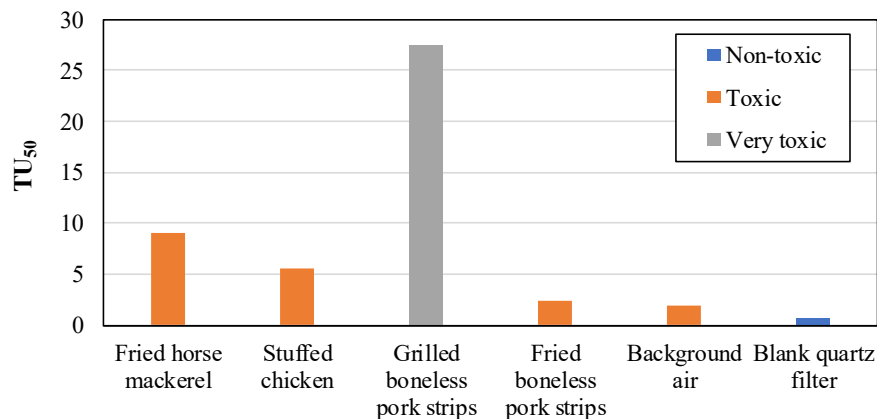
623

624 **3.6. PM_{10} toxicity**

625

626 The ecotoxicity assessment demonstrated that PM_{10} emissions from grilled pork are very toxic, while
627 those obtained during cooking of all other dishes are classified as toxic (Figure 6). Although the TU
628 value for the background air sample was 1.2 to 14 times lower than those of cooking emissions, it still
629 reveals some toxicity of PM_{10} in the absence of activities. Globally, TU values correlated reasonably
630 well with PM_{10} ($r^2 = 0.59$), OC ($r^2 = 0.58$) and EC ($r^2 = 0.68$) concentrations. Ecotoxic effects of cooking-
631 generated aerosols depend directly on the quantity of carbonaceous particles produced, which act as
632 carriers for toxic compounds bound to this particulate material. A moderate relationship was also
633 obtained between TU and total PAH levels ($r^2 = 0.68$). However, when PAHs are split into molecular
634 weight classes, LMW compounds do not show any correlation with TU, while HMW constituents
635 present an excellent association with toxicity values ($r^2 = 0.94$). HMW PAHs are generally less water

636 soluble, have lower vapour pressures and Henry's constants, and partition more readily into organic
637 matter than LMW PAHs. High concentrations of carbonaceous aerosol strongly favour partitioning of
638 semi-volatile toxic species (such as HMW PAHs) to the particulate phase, thus contributing to the
639 enhanced ecotoxicity of aerosol particles. LMW PAHs exist predominantly in the gas phase due to their
640 relatively high vapour pressure.
641



642
643 Figure 6. Toxicity of each sample, expressed in toxic units, based on the results of the *Vibrio*
644 *fischeri* bioluminescence inhibition bioassay
645

646 Significant positive correlations between total concentrations of glyceridic compounds, dicarboxylic
647 acids and amino acids and ecotoxicity were also found (Table S7). Likewise, excellent positive
648 relationships ($r^2 > 0.90$) between TU and levels of some individual PM₁₀-bound constituents were
649 derived. The oxygenated compounds most related to toxicity were mannosan (a stereoisomer of
650 levoglucosan), 4-hydroxycinnamic acid, oleic acid, hexacosanoic acid, some diacids (succinic, malic,
651 adipic and sebacic), glyceric acid, citric acid, several n-alkanols (C₁₈, C₂₂, C₂₃, C₂₅, C₂₇ and C₂₈),
652 cholesterol, monopalmitin and monostearin.

653

654 4. Conclusions

655

656 Given that cooking is recognised as a major air pollution source in indoor environments and bearing
657 in mind that culinary methods, and consequently emissions, greatly depend on geography, a monitoring
658 programme was carried out in a modern domestic kitchen, equipped with electrical cooktop and range
659 hood, where typical dishes of the Latin cuisine were prepared: fried horse mackerel, stuffed chicken,
660 and grilled and fried boneless pork strips. The study included measurements of gaseous compounds,
661 the determination of particle emission rates, a detailed screening of PM₁₀-bound organic constituents,
662 and a direct contact bioassay, which has never been applied to cooking samples, to assess toxicity.

663 Although acrolein has been described as an important carbonyl in cooking emissions, its presence
664 was not observed in the present study. Formaldehyde and acetaldehyde levels were, in general,
665 comparable to those found in the background air of the kitchen, when no activity was being performed.
666 Propanal was enriched in all cooking emissions, whereas isopentanal and pentanal were more abundant
667 in samples obtained while frying horse mackerel. TVOCs were within the wide range of values
668 described for commercial and residential kitchens. The use of marinated meat and the addition of wine
669 during the cooking process seem to contribute to higher concentrations of these compounds.

670 PM₁₀ was mainly composed of high-risk respirable sizes. PM₁₀ emission rates did not vary
671 significantly between dishes and were well below the values documented in previous studies for heating
672 oils and Chinese cooking. Likewise, when compared with many of the values described in the literature,
673 much lower PAH levels were obtained. The differences are justified not only by distinctive ingredients
674 and cooking methods, but also by the fact that, in our study, gas was not used as fuel and an exhaust
675 hood was continuously operated. Retene, an alkylated polyaromatic employed so far as a marker of
676 softwood combustion, was among the most abundant PAHs. The highest concentrations of PAHs were
677 recorded for higher cooking temperatures. In addition to PAHs, about 150 oxygenated organic
678 compounds were quantified, some of which were described for the first time, as far as we know, as
679 constituents of cooking aerosols. The proportions of the various organic classes depended on the menu.

680 Grilling pork was found to emit “very toxic” particles with non-negligible cancer risks, while PM₁₀
681 samples from other dishes have shown to be less hazardous, but still classified as “toxic”. Although the
682 concentrations of most of the compounds were lower in the absence of activities, the background air
683 sample still showed some toxicity. The organic constituents that contributed most to toxicity were
684 HMW PAHs, glyceridic compounds (monopalmitin and monostearin), diacids, amino acids and some
685 individual species, such as cholesterol, oleic acid, mannosan, heavier alkanols, among others.

686 The chemical signatures for cooking emissions in the present study should be seen as representative
687 of some Latin cuisine dishes made in a biomass-free kitchen with modern appliances. Since cooking
688 emissions are unregulated and potentially toxic, it is necessary to conduct additional studies, covering
689 other cuisines and styles. New studies would be beneficial not only to obtain specific emission profiles,
690 but also to investigate which chemical markers induce health effects. The toxicological significance
691 must be further explored through multiple assays (cytotoxicity, mutagenicity, inflammatory processes,
692 oxidative stress, etc.), especially with respect to the exposure to hazardous vapours and particulate
693 constituents. It seems increasingly important to educate the public of the potential risks of cooking
694 emissions so that they may take informed choices about their behaviours, including the adoption of
695 appropriate kitchen exhaust ventilation technologies.

696

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698

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710

711 **CRedit authorship contribution statement**

712

713 **Célia Alves:** Conceptualisation, Supervision, Funding acquisition, Project administration, Formal
714 analysis, Writing - Original draft preparation. **Estela Vicente:** Methodology, Formal analysis, Writing
715 – Review & Editing. **Margarita Evtyugina:** Methodology, Writing – Review & Editing. **Ana Vicente:**
716 Methodology, Writing – Review & Editing. **Tsend-Ayush Sainnokhoi:** Methodology, Writing –
717 Review & Editing. **Nora Kováts:** Supervision, Writing – Review & Editing.

718

719 **Declaration of competing interest**

720

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

723

724 **Appendix A. Supplementary data**

725

726 Supplementary material related to this article can be found, in the online version, at
727 <https://doi.org/10.1016/XXXX.XXXX>

728

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SUPPLEMENTARY MATERIAL

Cooking activities in a domestic kitchen: chemical and toxicological profiling of emissions

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Weather in Aveiro in the period of measurements

Table S1. Weather in September 2017

























Day	Cloudiness	Temperatures (min/max), °C	Precipitation, mm
1		21/26	0
2		22/28	0
3		20/24	0
4		21/24	0
5		21/24	0
6		23/28	0
7		23/28	0
8		18/22	0
9		18/21	0
10		18/21	0
11		19/23	0
12		19/23	0
13		20/24	0
14		19/22	0
15		17/20	0
16		16/20	0
17		17/20	0
18		18/21	0
19		18/22	0
20		18/22	0
21		18/22	0
22		19/23	0
23		17/22	0
24		19/23	0
25		18/22	0
26		18/23	0
27		20/24	0
28		20/25	0
29		19/24	0
30		18/21	0

Table S2. Weather in October 2017

Day	Cloudiness	Temperatures (min/max), °C	Precipitation, mm
1		20/25	0
2		22/29	0
3		19/25	0
4		18/24	0
5		17/23	0
6		17/20	0
7		22/29	0
8		20/28	0
9		20/27	0
10		20/26	0
11		19/24	0
12		20/28	0
13		17/23	0
14		21/28	0
15		25/31	0.1
16		17/23	2.8
17		16/20	14
18		15/18	4.7
19		14/17	0.1
20		18/20	2
21		15/19	0
22		14/19	0
23		15/22	0
24		18/25	0
25		18/25	0
26		18/26	0
27		20/28	0
28		20/25	0
29		18/24	0
30		17/22	0
31		15/23	0

Air exchange rates

In the present study, the air exchange rates per hour (α , h⁻¹) were estimated by the CO₂ concentration decay method. For a well-mixed space, the change in CO₂ concentration with time is given by:

$$C_t = C_{ext} + \frac{q_{CO_2} \times 10^6}{Q} - \left(C_{ext} - C_0 + \frac{q_{CO_2} \times 10^6}{Q} \right) e^{\left(-\frac{Q}{V} t \right)} \quad (1)$$

where C_t is the indoor concentration of CO₂ at time t (ppm), C_{ext} is the outdoor concentration of CO₂ (ppm), C_0 is the concentration of CO₂ in the indoor air at time 0 (ppm), Q is the volume flow rate of air entering the space (m³ s⁻¹), q_{CO_2} is the volumetric indoor emission rate of CO₂ (m³ s⁻¹), V is the volume of the room (m³) and t is the interval since $t = 0$ (s). When the room is unoccupied there is no CO₂ emission from the occupants, and $q_{CO_2} = 0$. Thus, Eq. (1) can be rearranged to give the following expression, which allows the ventilation rate (Q) to be estimated from measured concentration values time t apart:

$$Q = -\frac{V}{t} \times \ln \left(\frac{C_t - C_{ext}}{C_0 - C_{ext}} \right) \quad (2)$$

The air exchange rate is defined as:

$$\alpha = Q/V \quad (3)$$

Main specifications of real-time measurement equipment

- Graywolf (IQ-610 probe, WolfSense Solutions, USA)
 - a) CO₂ (NDIR)

Range: 0 to 10,000 ppm; Accuracy: ± 35 ppm over the IAQ range (350 ppm – 2000 ppm), ± 35 ppm $\pm 3\%$ of reading > 2000 ppm, ± 50 ppm $\pm 3\%$ of reading over the full range
 - b) CO (electrochemical)

Range: 0 to 500 ppm; Accuracy: ± 2 ppm < 50 ppm, $\pm 3\%$ of reading > 50 ppm
 - c) Total volatile organic compounds (PID)

Range: 5 to 20,000 ppb; Resolution: 1 ppb, L.O.D. < 5 ppb

10.6 eV PIDs do not respond to VOCs with ionization potentials > 10.6 , such as ethane, methane or formaldehyde. However, they do respond to the vast majority of VOCs. Standard calibration is to isobutylene.
- TSI (Q-Trak IAQ Monitor, Model 7575, USA)

CO₂ (NDIR)

Range: 0 to 5000 ppm; Accuracy: $\pm 3\%$ of reading or ± 50 ppm, whichever is greater; Resolution: 1 ppm
- DustTrak monitor (8530 model, USA) - light-scattering laser photometer

Measuring range of PM₁₀, PM_{2.5}, and PM₁: 0.001 mg m⁻³ to 400 mg m⁻³; Resolution: $\pm 0.1\%$ of reading or 0.001 mg m⁻³, whichever is greater; Zero stability: ± 0.002 mg m⁻³ per 24 hours at 10 s time constant; Flow rate: 3.0 L min⁻¹; Flow accuracy: $\pm 5\%$ of factory set point, internal flow controlled

Table S3. PM₁₀ mass concentrations of PAHs (µg g⁻¹)

	Fried horse mackerel	Stuffed chicken	Grilled boneless pork strips	Fried boneless pork strips	Background air
Naphthalene	3.07	0.084	0.192	-	13.4
Acenaphthylene	0.128	-	-	0.135	0.399
Acenaphthene	0.158	0.308	0.080	0.122	1.15
Fluorene	1.88	1.02	0.154	2.81	6.82
Phenanthrene	2.15	1.79	0.843	2.83	2.30
Anthracene	0.135	0.139	0.087	0.199	0.447
Fluoranthene	1.87	2.13	1.97	2.86	2.00
Pyrene	4.79	3.56	2.53	5.19	5.55
p-Terphenyl	0.040	-	0.045	-	-
Retene	2.79	1.51	1.24	2.20	3.91
Benzo[a]anthracene	3.27	2.14	2.90	3.63	5.53
Chrysene	8.67	10.1	11.8	13.0	16.1
Benzo[b]fluoranthene	1.27	5.86	3.99	3.77	14.5
Benzo[k]fluoranthene	0.505	2.30	2.65	2.06	-
Benzo[e]pyrene	1.55	5.45	4.08	3.88	17.2
Benzo[a]pyrene	0.634	0.078	2.08	0.666	3.81
Perylene	-	-	-	-	-
Indeno[1,2,3-cd]pyrene	0.282	0.685	1.67	0.209	4.46
Dibenzo[a,h]anthracene	-	-	0.150	-	-
Benzo[g,h,i]perylene	0.317	0.737	1.76	0.668	6.09
SUM	33.5	37.9	38.2	44.2	104

Table S4. Comparison of diagnostic ratios of PAHs

Sample	Location	Type of cooking	Phe/(Phe+Ant)	Flt/(Flt+Pyr)	BaA/(BaA+Chr)	Ind/(Ind+BghiP)	Reference
PM ₁₀	Kitchen (indoor air)	Fried horse mackerel, electric cooktop	0.94	0.28	0.27	0.47	This study
		Stuffed chicken, electric cooktop	0.93	0.37	0.18	0.48	
		Grilled boneless pork strips, electric cooktop	0.91	0.44	0.20	0.49	
		Fried boneless pork strips, electric cooktop	0.93	0.36	0.22	0.24	
TSP and gaseous phase	Kitchen (indoor air)	Fresh bacon, electric stove	0.99	0.53	0.31	0.47	Jørgensen et al. (2013)
		Fresh bacon, gas stove	0.94	0.33	0.33	0.47	
		Smoked bacon, gas stove	0.96	0.30	0.30	0.42	
PM _{2.5}	Kitchen (indoor air)	Stir-frying		0.45	0.31	0.51	Zhao et al. (2019b)
		Pan-frying		0.34	0.25	0.16	
		Seep-frying		0.27	0.16	0.22	
		Steaming		-	0.27	0.47	
		Boiling		-	0.24	0.36	
PM _{2.5}	Kitchen (indoor air)	Steaming	0.96	0.51	0.31	0.54	See and Balasubramanian (2008)
		Boiling	0.96	0.52	0.34	0.52	
		Stir-frying	0.94	0.56	0.28	0.45	
		Pan-frying	0.94	0.55	0.29	0.46	
		Deep-frying	0.95	0.53	0.28	0.44	
PM _{2.5}	Food stall in a	Chinese	0.21	0.32	0.40	0.43	See et al. (2006)
		Malay	0.28	0.38	0.32	0.38	

	university canteen	Indian	0.21	0.43	0.50	0.39	
TSP and gaseous phase	Stacks of restaurants	Chinese	0.86	0.50	0.62	0.63	Li et al. (2003)
		Western	0.86	0.46	0.38	0.63	
		Fast food	0.96	0.60	0.32	0.53	
		Japanese	0.97	0.66	0.13	0.83	
PM _{2.5}	Exit of the restaurants' purification systems	Shanghai-style	0.91	0.53	0.22		Pei et al. (2016)
		Sichuan-style	0.72	0.44	0.43	-	
		Italian-style	0.63	0.46	0.49		
PM _{2.5} and gaseous phase	Under a laboratory fume hood equipped with an electric stove	Deep-frying	0.31	0.16	0.65	0.19	Yao et al. (2015)
		Frying	0.24	0.19	0.88	0	

Table S5. Concentrations of oxygenated compounds (ng m⁻³). Empty cells mean not detected or of the same level of blanks.

	Fried horse mackerel	Stuffed chicken	Grilled boneless pork strips	Fried boneless pork strips	Background air
SUGARS and POLYOLS					
Mannosan	0.644	0.331	3.31	0.284	0.497
Levogluconan	52.3	8.23	25.4	8.10	11.4
Ribitol	2.53	2.37	-	3.69	0.557
Meso-Erythritol	0.910	-	-	-	0.315
Other sugars and polyols	47.2	54.9	29.6	5.01	2.08
PHENOLIC COMPOUNDS & BENZENE DERIVATIVES					
2-Methylphenol (o-cresol)	-	-	-	1.69	0.0278
Benzyl alcohol	11.1	-	-	114	-
Pyrocatechol	-	-	-	0.0314	0.00331
5-Isopropyl-3-methylphenol	0.122	-	-	0.174	0.125

Resorcinol	0.00534	-	0.00407	0.00897	0.000930
4-Methylcatechol	-	-	-	-	-
2,6-Dimethoxyphenol	0.000843	0.000866	-	-	0.00222
Eugenol	0.00277	0.00102	0.00157	0.00494	0.00117
Isoeugenol	0.577	-	0.0117	1.28	-
4-Allyl-2,6-dimethoxyphenol (methoxy eugenol)	-	-	0.00168	0.0171	-
2,4-Di-tert-butylphenol	17.9	1.65	19.7	19.1	6.86
Pyrogallol	0.00279	0.00206	0.00212	0.00798	0.00532
2-methoxy-4-propylphenol	0.00384	0.261	0.00242	20.5	0.00514
4-Phenylphenol	0.206	-	-	-	-
4-Octylphenol	-	-	-	0.374	-
Coniferyl alcohol	0.00597	0.00447	-	0.0102	0.00634
Sinapyl alcohol	-	-	-	-	-
2-Isopropyl-5-methylphenol (Thymol)	1.23	-	0.432	1.14	0.0630
4-tert-Butylphenol	3.72	-	1.74	7.95	0.826
Vanillic acid	0.144	0.0131	0.129	0.0280	0.00775
2,3,4-Trimethoxybenzoic (gallic) acid	-	-	-	-	-
Syringic acid	0.0352	0.00596	0.0453	0.00214	0.00206
4-Hydroxybenzoic acid	-	3.82	-	-	-
Benzoic acid	1.31	0.180	0.178	2.26	0.0734
4-hydroxybenzoic acid	0.684	0.278	0.370	0.0729	0.0201
α -Hexylcinnamaldehyde	-	0.640	1.023	4.01	0.0996
Trans-cinnamic acid	0.635	0.105	0.341	0.150	0.0412
4-hydroxycinnamic (p-coumaric) acid	-	0.0554	0.483	0.0151	0.00156
4-hydroxy-3-methoxycinnamic (ferulic) acid	0.333	0.0119	0.0483	-	0.00778
3,4-Dihydroxy-cinnamic (caffeic) acid	-	-	0.117	-	-
Bisphenol A	0.211	-	1.06	-	0.567
ALIPHATIC ALCOHOLS					
1-Decanol	0.193	-	-	0.316	0.0151
1-Dodecanol	0.657	1.44	-	4.49	0.704

1-Tetradecanol	4.84	5.54	7.92	18.0	3.51
1-Pentadecanol	5.70	5.34	10.1	18.1	4.96
1-Hexadecanol	105	131	226	193	66.9
Unidentified C ₁₆ alcohol	6.13	8.77	12.7	18.4	3.81
C ₁₇ alcohol isomers	1.97	3.41	3.90	1.32	0.624
1-Octadecanol	37.0	33.8	54.0	35.6	27.5
1-Nonadecanol	1.79	0.527	-	-	0.858
1-Eicosanol	5.68	-	10.0	2.13	1.75
1-Docosanol	2.51	1.05	4.73	1.39	0.788
1-Tricosanol	0.159	0.0337	0.801	0.0580	0.0570
iso-Tricosanol	-	-	-	1.08	-
1-Pentacosanol	0.266	0.0432	1.23	0.284	0.0798
1-Heptacosanol	0.0198	0.00376	0.378	-	0.0254
1-Octacosanol	-	0.0998	1.53	-	0.269
1-Tetracosanol	0.364	0.522	11.0	-	-
1-Pentacosanol	-	-	3.84	-	-
1-Hexacosanol	1.49	0.899	-	1.03	0.936
1-Heptacosanol	-	-	0.860	-	-
1-Octacosanol	0.620	0.394	1.80	-	0.233
1-Triacontanol	-	-	0.316	-	0.0830
STEROIDS					
Cholesterol	75.3	2.02	422	0.861	1.08
5-Cholesten-3-ol (dihydrocholesterol)	0.0891	0.0618	0.516	-	-
5-Cholesten-3- β ,7- β -diol	-	-	4.33	-	-
Stigmasterol	6.56	-	2.83	-	-
β -Sitosterol	35.0	0.123	1.86	0.318	0.201
CARBOXYLIC ACIDS					
Hexanoic (caproic) acid	86.7	1.35	4.29	1.97	-
Heptanoic acid	8.99	3.27	6.66	5.90	0.455
Octanoic (caprilic) acid	1.78	0.666	1.41	1.16	0.703

Nonanoic (pelargonic) acid	75.6	33.5	30.1	48.0	0.653
Decanoic (caproic) acid	1.91	1.51	0.624	1.18	4.60
Undecanoic acid	1.43	0.812	1.23	0.288	0.0723
Dodecanoic (lauric) acid	7.17	5.74	7.52	7.50	1.35
Tridecanoic (tridecyllic) acid	2.54	0.971	2.53	1.32	0.159
Tetradecanoic (myristic) acid	216	83.6	89.4	63.3	19.2
C ₁₄ acid isomers	3.00	2.06	-	3.36	0.750
Pentadecanoic acid	29.5	9.98	14.3	7.69	3.47
Hexadecanoic (palmitic) acid	710	205	515	225	60.4
Heptadecanoic acid	20.7	2.28	9.40	1.45	0.385
C ₁₇ acid isomers	24.7	4.69	8.18	5.26	2.69
cis-9,cis-12-Octadecadienoic (linoleic) acid	325	8.52	141	1.45	0.546
Octadecanoic (stearic) acid	78.7	46.1	145	78.9	9.44
Nonadecanoic acid	1.07	1.94	0.436	0.0801	0.0168
Eicosanoic (arachidic) acid	7.38	0.881	2.94	0.578	0.139
Docosanoic (behenic) acid	17.1	2.89	3.65	0.360	0.151
Tetracosanoic acid	8.00	5.74	13.0	-	-
Pentacosanoic acid	1.82	-	-	-	-
Hexacosanoic acid	0.695	0.797	17.8	0.0743	0.0165
Nonenoic acid	58.7	-	5.26	-	-
9-cis-Hexadecenoic (palmitoleic) acid	38.3	0.726	12.4	0.197	0.807
Heptadecenoic acid	8.36	-	6.80	-	-
cis-9-Octadecenoic (oleic) acid	89.9	17.7	265	11.3	3.24
Eicosenoic acid	-	-	9.12	-	0.914
DICARBOXYLIC ACIDS					
1,4-Butanedioic (succinic) acid	12.3	3.65	59.5	1.61	0.428
1,5-Pentanedioic (glutaric) acid	5.35	4.74	2.15	8.99	2.70
2-Hydroxy-butanedioic (malic) acid	6.29	3.07	25.6	2.81	0.677
Hexanedioic (adipic) acid	3.50	2.23	5.95	0.970	0.450
Heptanedioic (pimelic) acid	3.23	0.824	4.03	0.454	0.184

Octanedioic (suberic) acid	8.90	1.32	6.68	0.726	0.301
Nonanedioic (azelaic) acid	90.5	24.7	76.7	6.11	3.04
Decanedioic (sebacic) acid	1.67	0.837	3.54	0.474	0.192
Hexadecanedioic (Thapsic) acid	0.0297	0.0130	0.0513	-	0.00121
AMINO ACIDS					
Alanine	31.5	-	271	-	-
Valine	193	-	106	-	0.437
Leucine	57.4	-	80.0	-	-
iso-Leucine	133	-	56.3	-	-
Thymine	26.3	-	-	-	-
5-Oxo-L-proline	65.0	-	276	-	6.85
OTHER ACIDS					
Laevulic acid	-	-	-	-	2.38
Hydracrylic acid	10.5	5.05	12.4	29.1	2.24
Glyceric acid	20.6	2.40	75.5	9.53	0.684
Pyruvic acid	7.67	-	0.269	-	0.291
3,4-Dihydroxybutanoic acid	4.87	2.25	3.05	7.37	1.06
3-Hydroxybutanoic (hydroxybutyric) acid	0.260	0.156	0.179	0.416	0.139
4-Hydroxy-3-pentenoic acid	-	3.34	-	4.44	-
cis-Pinonic acid	1.11	1.02	2.22	1.28	0.580
Pinic acid	-	1.19	4.15	1.27	0.876
Citric acid	0.490	0.130	1.86	0.139	0.0295
Abietic acid	-	-	0.269	-	-
Dehydroabietic acid	0.835	0.139	0.608	0.107	0.112
Isopimaric	0.592	0.03004	0.785	0.0172	0.00496
ALKYL ESTERS OF ACIDS					
Palmitic acid methyl ester	1.34	8.34	15.6	42.5	14.7
Palmitic acid ethyl ester	0.894	9.10	14.6	26.9	15.7
Palmitic acid isopropyl ester (Isopropyl palmitate)	9.47	94.5	104	125	53.6
Palmitic acid butyl ester	0.283	0.876	1.20	0.288	0.853

Palmitic acid hexadecyl ester	7.94	14.9	3.77	-	0.785
Isopropyl laurate (dodecanoic acid, 1-methylethyl ester)	0.00582	0.0531	0.0258	0.243	0.0229
Dodecanoic acid isooctyl ester	20.5	45.3	34.5	5.58	26.5
Methylisostearate	1.10	9.40	15.9	15.1	12.7
Heptadecanoic acid, 15-methyl, ethyl ester	-	1.03	2.15	1.33	2.42
Octadecanoic acid methyl ester	0.297	10.3	11.7	9.04	11.5
Myristic acid isopropyl ester (Isopropyl myristate)	-	250	202	372	96.4
Myristic acid isobutyl ester (Isobutyl myristate)	5.65	5.93	2.89	-	4.38
Myristic acid tetradecyl ester	14.7	16.2	27.8	-	2.18
Myristic acid hexadecyl ester	5.74	11.9	3.07	-	0.932
Benzoic acid alkyl esters	81.9	102	112	35.6	51.2
Hexanoic acid 2-ethyl-, hexadecyl ester (Hexadecyl 2-ethylexanoate)	2.29	17.9	4.51	-	2.97
Hexadecanoic acid hexadecyl ester	2.62	4.54	1.34	0.0999	0.246
GLYCERIDIC COMPOUNDS					
Glycerol	92.0	26.9	200	293	15.4
1-Monolauroyl-rac-Glycerol	-	-	-	-	-
1-Monooleoylglycerol	69.4	Nd	126	1.13	-
1-Monomyristin	108	0.9	9.15	1.25	0.482
Glycerol monostearate (monostearin)	49.5	13.2	169	14.2	1.72
1-Monopalmitin	156	23.2	359	19.9	1.85
Tetraethylene glycol dimethacrylate	-	-	-	-	2.31
Glycerol tricaprylate	3.73	1.28	9.64	-	0.331
PLASTICISERS					
Phthalic acid	-	-	1.05	-	-
Dimethyl phthalate	0.0117	0.00276	-	0.00786	-
Diethyl phthalate	2.57	5.16	3.12	23.0	1.51
Di-n-butyl phthalate	2.64	0.547	0.674	1.40	-
Benzyl butyl phthalate	5.46	4.57	5.44	3.27	3.51
Bis(2-ethylhexyl) adipate	0.117	0.00622	0.0169	0.0312	0.0147
Bis(2-ethylhexyl)phthalate	14.5	2.63	0.379	0.547	0.182

Diethylhexyl phthalate (DEHP)	204	121	209	-	159
Unidentified phthalates	97.5	303	335	488	193
OTHER COMPOUNDS					
Diethylene glycol	3.254	1.88	3.75	-	2.32
Lupeol	0.651	Nd	0.192	-	0.0696
2,6-Di-tert-butyl-1,4-benzoquinone	0.853	0.366	0.416	4.23	0.437
α-Hexylcinnamaldehyde	-	0.178	0.373	0.971	0.0462
Diethyltoluamide (DEET)	-	2.28	-	6.44	0.575
2-Propanol-1-chloro-,phosphate	11.8	-	-	-	-
7,9-Di-tert-butyl-1-oxaspiro[4.5]deca-6,9-diene-2,8-dione	7.64	10.1	9.67	12.8	7.59
Parsol MCX	21.7	24.0	58.1	23.9	14.0
Galoxolide	-	0.245	1.01	1.33	0.150
Pyrimidine	43.0	-	61.4	-	-
Tributyl acetylcitrate	-	-	-	-	5.53
Piperine	-	-	111	-	-
15-Nonacosanonone	0.911	4.74	-	-	-
Irganox 1906	-	4.44	-	-	33.4
Oxidised irgafos 168	-	-	-	-	18.7
Isophorone	0.253	-	0.0193	-	0.177
Benzothiazole	-	-	-	0.0217	0.0243
Carbazole	0.0284	0.00385	0.00828	-	-
Cholesteryl acetate	12.8	-	12.1	-	-
Cholesteryl esters	0.481	-	5.84	-	-
α-Tocopherol (Vitamin E)	0.316	0.0113	0.0176	0.0177	0.00347
Vitamin E acetate	-	0.992	7.62	1.96	1.36
1,2,3-Hexanetriol	-	-	-	-	0.0116
Methylcyclohexanol	0.0557	-	-	-	-
Myrtenol	-	0.00505	0.0148	0.0194	0.0190
Citronellol	-	-	-	-	-
2,3-Pinandediol	0.0684	0.0218	0.107	0.180	0.0421

Table S6. Particulate mass fractions of oxygenated compounds (ng g⁻¹ PM₁₀). Empty cells mean not detected or of the same level of blanks.

	Fried horse mackerel	Stuffed chicken	Grilled boneless pork strips	Fried boneless pork strips	Background air
SUGARS and POLYOLS					
Mannosan	9.05	13.6	45.3	8.76	42.5
Levoglucozan	736	339	347	250	977
Ribitol	35.5	97.7	-	114	47.6
Meso-Erythritol	12.8	-	-	-	26.9
Other sugars and polyols	665	2261	406	155	178
PHENOLIC COMPOUNDS & BENZENE DERIVATIVES					
2-Methylphenol (o-cresol)	-	-	-	52.0	2.38
Benzyl alcohol	156	-	-	3504	-
Pyrocatechol	-	-	-	0.969	0.283
5-Isopropyl-3-methylphenol	1.71	-	-	5.36	10.7
Resorcinol	0.0751	-	0.056	0.277	0.080
4-Methylcatechol	-	-	-	-	-
2,6-Dimethoxyphenol	0.0119	0.0356	-	-	0.190
Eugenol	0.0390	0.0418	0.0215	0.153	0.100
Isoeugenol	8.12	-	0.160	39.4	-
4-Allyl-2,6-dimethoxyphenol (methoxy eugenol)	-	-	0.0230	0.53	-
2,4-Di-tert-buthylphenol	251	67.7	270	591	586
Pyrogallol	0.0393	0.0850	0.0290	0.246	0.454
2-methoxy-4-propylphenol	0.0540	10.8	0.0332	632	0.439
4-Phenylphenol	2.90	-	-	-	-
4-Octylphenol	-	-	-	11.55	-
Coniferyl alcohol	0.084	0.184	-	0.316	0.542
Sinapyl alcohol	-	-	-	-	-

2-Isopropyl-5-methylphenol (Thymol)	17.3	-	5.91	35.1	5.38
4-tert-Butylphenol	52.3	-	23.8	245	70.6
Vanillic acid	2.02	0.539	1.76	0.863	0.662
2,3,4-Trimethoxybenzoic (gallic) acid	-	-	-	-	-
Syringic acid	0.495	0.246	0.620	0.0662	0.176
4-Hydroxybenzoic acid	-	157	-	-	-
Benzoic acid	18.4	7.41	2.43	69.8	6.27
4-hydroxybenzoic acid	9.63	11.4	5.07	2.25	1.72
α -Hexylcinnamaldehyde	-	26.4	14.0	124	8.51
Trans-cinnamic acid	8.94	4.31	4.66	4.61	3.52
4-hydroxycinnamic (p-coumaric) acid	-	2.28	6.61	0.466	0.133
4-hydroxy-3-methoxycinnamic (ferulic) acid	4.68	0.489	0.661	-	0.665
3,4-Dihydroxy-cinnamic (caffeic) acid	-	-	1.60	-	-
Bisphenol A	2.97	-	14.5	-	48.4
ALIPHATIC ALCOHOLS					
1-Decanol	2.71	-	-	9.73	1.29
1-Dodecanol	9.24	59.4	-	139	60.2
1-Tetradecanol	68.1	228	108	556	300
1-Pentadecanol	80.2	220	138	559	424
1-Hexadecanol	1484	5394	3092	5963	5720
Unidentified C ₁₆ alcohol	86.2	361	173	567	326
C ₁₇ alcohol isomers	27.6	140	53.4	40.9	53.3
1-Octadecanol	520	1390	740	1100	2351
1-Nonadecanol	25.1	21.7	-	-	73.3
1-Eicosanol	79.9	-	137	65.8	150
1-Docosanol	35.3	43.2	64.7	42.9	67.4
1-Tricosanol	2.23	1.39	11.0	1.79	4.87
iso-Tricosanol	-	-	-	33.2	-
1-Pentacosanol	3.75	1.78	16.8	8.75	6.82
1-Heptacosanol	0.279	0.155	5.17	-	2.17

1-Octacosanol	-	4.11	20.9	-	23.0
1-Tetracosanol	5.12	21.5	151	-	-
1-Pentacosanol	-	-	52.6	-	-
1-Hexacosanol	20.9	37.0	-	31.6	80.0
1-Heptacosanol	-	-	11.8	-	-
1-Octacosanol	8.72	16.2	24.6	-	19.9
1-Triacontanol	-	-	4.33	-	7.09
STEROIDS					
Cholesterol	1060	83.2	5778	26.6	92.0
5-Cholesten-3-ol (dihydrocholesterol)	1.25	2.55	7.07	-	-
5-Cholesten-3- β ,7- β -diol	-	-	59.3	-	-
Stigmasterol	92.3	-	38.7	-	-
β -Sitosterol	492	5.04	25.4	9.81	17.2
CARBOXYLIC ACIDS					
Hexanoic (caproic) acid	1220	55.6	58.7	60.6	-
Heptanoic acid	126	134	91.2	182	38.9
Octanoic (caprylic) acid	25.0	27.4	19.3	35.9	60.1
Nonanoic (pelargonic) acid	1063	1379	413	1480	55.8
Decanoic (caproic) acid	26.8	62.2	8.54	36.3	393
Undecanoic acid	20.1	33.4	16.8	8.89	6.18
Dodecanoic (lauric) acid	101	236	103	231	116
Tridecanoic (tridecyllic) acid	35.8	40.0	34.7	40.7	13.6
Tetradecanoic (myristic) acid	3044	3442	1223	1952	1641
C ₁₄ acid isomers	42.2	84.9	-	104	64.1
Pentadecanoic acid	415	411	195	237	296
Hexadecanoic (palmitic) acid	9982	8445	7054	6947	5164
Heptadecanoic acid	291	93.9	129	44.7	32.9
C ₁₇ acid isomers	347	193	112	162	230
cis-9,cis-12-Octadecadienoic (linoleic) acid	4569	351	1928	44.8	46.7
Octadecanoic (stearic) acid	1108	1897	2011	2433	806

Nonadecanoic acid	15.1	79.9	5.97	2.47	1.44
Eicosanoic (arachidic) acid	104	36.3	40.3	17.8	11.9
Docosanoic (behenic) acid	241	119	50.0	11.1	12.9
Tetracosanoic acid	113	236	178	-	-
Pentacosanoic acid	25.6	-	-	-	-
Hexacosanoic acid	9.77	32.8	243	2.29	1.41
Nonenoic acid	826	-	72.0	-	-
9-cis-Hexadecenoic (palmitoleic) acid	538	29.9	170	6.07	69.0
Heptadecenoic acid	118	-	93.1	-	-
cis-9-Octadecenoic (oleic) acid	1265	729	3622	347	276
Eicosenoic acid	-	-	125	-	78.1
DICARBOXYLIC ACIDS					
1,4-Butanedioic (succinic) acid	172	150	814	49.7	36.6
1,5-Pentanedioic (glutaric) acid	75.2	195	29.4	277	231
2-Hydroxy-butanedioic (malic) acid	88.5	127	350	86.5	57.9
Hexanedioic (adipic) acid	49.2	91.7	81.5	29.9	38.4
Heptanedioic (pimelic) acid	45.5	33.9	55.2	14.0	15.7
Octanedioic (suberic) acid	125	54.2	91.4	22.4	25.8
Nonanedioic (azelaic) acid	1272	1018	1050	188	260
Decanedioic (sebacic) acid	23.5	34.4	48.4	14.6	16.4
Hexadecanedioic (Thapsic) acid	0.418	0.536	0.702	-	0.104
AMINO ACIDS					
Alanine	443	-	3714	-	-
Valine	2718	-	1444	-	37.3
Leucine	807	-	1096	-	-
iso-Leucine	1874	-	771	-	-
Thymine	369	-	-	-	-
5-Oxo-L-proline	915	-	3780	-	585
OTHER ACIDS					
Laevulic acid	-	-	-	-	204

Hydracrylic acid	148	208	169	897	192
Glyceric acid	289	98.9	1033	294	58.4
Pyruvic acid	108	-	3.69	-	24.9
3,4-Dihydroxybutanoic acid	68.5	92.5	41.8	227	90.5
3-Hydroxybutanoic (hydroxybutyric) acid	3.65	6.42	2.45	12.8	11.9
4-Hydroxy-3-pentenoic acid	-	137	-	137	-
cis-Pinonic acid	15.6	42.1	30.3	39.5	49.5
Pinic acid	-	49.1	56.8	39.1	74.8
Citric acid	6.90	5.34	25.4	4.28	2.52
Abietic acid	-	-	3.68	-	-
Dehydroabietic acid	11.7	5.72	8.32	3.30	9.56
Isopimaric	8.33	1.25	10.7	0.532	0.423
ALKYL ESTERS OF ACIDS					
Palmitic acid methyl ester	18.8	343	213	1312	1259
Palmitic acid ethyl ester	12.6	375	200	829	1343
Palmitic acid isopropyl ester (Isopropyl palmitate)	133	3890	1421	3843	4584
Palmitic acid butyl ester	3.98	36.1	16.4	8.90	72.8
Palmitic acid hexadecyl ester	112	614	51.6	-	67.1
Isopropyl laurate (dodecanoic acid, 1-methylethyl ester)	0.0818	2.19	0.35	7.50	1.95
Dodecanoic acid isooctyl ester	289	1866	473	172	2260
Methylisostearate	15.4	387	218	466	1083
Heptadecanoic acid, 15-methyl, ethyl ester	-	42.2	29.4	41.1	207
Octadecanoic acid methyl ester	-	426	160	279	982
Myristic acid isopropyl ester (Isopropyl myristate)	-	10309	2760	11463	8236
Myristic acid isobutyl ester (Isobutyl myristate)	79.5	244	39.6	-	374
Myristic acid tetradecyl ester	207	667	381	-	186
Myristic acid hexadecyl ester	80.8	489	42.0	-	79.7
Benzoic acid alkyl esters	1153	4208	1528	1098	4371
Hexanoic acid 2-ethyl-, hexadecyl ester (Hexadecyl 2-ethylexanoate)	32.2	737	61.7	-	254
Hexadecanoic acid hexadecyl ester	36.9	187	18.3	3.08	21.0

GLYCEROL DERIVATIVES

Glycerol	1295	1106	2731	9044	1316
1-Monolauroyl-rac-Glycerol	-	0.0281	0.102	0.221	0.152
1-Monooleoylglycerol	976	-	1720	34.9	-
1-Monomyristin	1515	36.7	125	38.6	41.2
Glycerol monostearate (monostearin)	696	545	2317	439	147
1-Monopalmitin	2198	957	4919	615	158
Tetraethylene glycol dimethacrylate	-	-	-	-	197
Glycerol tricaprylate	52.5	52.8	132	-	28.2

PLASTICISERS

Phthalic acid	-	-	14.4	-	-
Dimethyl phthalate	164	114	-	242	-
Diethyl phthalate	3546	4065	1270	3935	1447
Di-n-butyl phthalate	37116	22533	9230	43347	-
Benzyl butyl phthalate	1610	1052	74.4	101	300
Bis(2-ethylhexyl) adipate	1639	256	232	961	1256
Bis(2-ethylhexyl)phthalate	204230	108215	5186	16866	15566
Diethylhexyl phthalate (DEHP)	2875	4993	2855	-	13591
Unidentified phthalates	1371	12462	4585	15048	16456

OTHER COMPOUNDS

Diethylene glycol	45.8	77.3	51.4	-	198
Lupeol	9.15	-	2.63	-	5.95
2,6-Di-tert-butyl-1,4-benzoquinone	12.0	15.1	5.69	128	37.3
α -Hexylcinnamaldehyde	-	7.32	5.10	30.0	3.95
Diethyltoluamide (DEET)	-	94.0	-	199	49.1
2-Propanol-1-chloro-,phosphate	166	-	-	-	-
7,9-Di-tert-butyl-1-oxaspiro[4.5]deca-6,9-diene-2,8-dione	107	415	132	395	649
Parsol MCX	306	990	796	736	1196
Galoxolide	-	10.1	13.9	41.2	12.8
Pyrimidine	605	-	840	-	-

Tributyl acetylcitrate	-	-	-	-	473
Piperine	-	-	1525	-	-
15-Nonacosanonone	12.8	195	-	-	-
Irganox 1906	-	183	-	-	2857
Oxidised irgafos 168	-	-	-	-	1600
Isophorone	3560	-	265	-	15152
Benzothiazole	-	-	-	669	2078
Carbazole	400	158	113	-	-
Cholesteryl acetate	179	-	166	-	-
Cholesteryl esters	6.76	-	80.0	-	-
α -Tocopherol (Vitamin E)	4.45	0.466	0.240	0.548	0.296
Vitamin E acetate	-	40.9	104	60.4	116
1,2,3-Hexanetriol	-	-	-	-	0.993
Methylcyclohexanol	0.784	-	-	-	-
Myrtenol	-	0.208	0.202	0.598	1.62
Citronellol	-	-	-	-	-
2,3-Pinandediol	0.962	0.897	1.47	5.55	3.60

Table S7. Linear correlations with $r^2 > 0.50$ between total concentrations of groups of compounds, or individual compounds, and toxic units

($y = m x + b$, where y is TU and x represents concentrations in ng m^{-3})

Organic compounds	m	b	r^2
Steroids	0.0563	3.0582	0.982
PAHs	0.0109	-9.7951	0.683
HMW PAHs	0.0158	-9.0431	0.937
Glyceridic compounds	0.0274	-0.3897	0.800
Dicarboxylic acids	0.1255	-0.4395	0.827
Amino acids	0.0263	2.4428	0.831
Other acids	0.2469	-1.868	0.740
Other oxygenated organic compounds	0.1112	-3.1894	0.934
Mannosan	7.9984	1.1861	0.952
Vanillic acid	113.64	1.9754	0.509
Syringic acid	451.24	1.1082	0.768
4-Hydroxycinnamic acid	48.871	3.8981	0.921
Tridecanoic acid	7.4040	-1.8455	0.520
Stearic acid	0.1837	-3.8664	0.753
Oleic acid	0.0951	1.9239	0.985
Hexacosanoic acid	1.3227	4.1626	0.947
Succinic acid	0.4207	2.7709	0.989
Malic acid	1.0301	1.3694	0.987
Adipic acid	4.5885	-2.7318	0.913
Pimelic acid	5.2486	0.1344	0.760
Sebacic acid	7.7450	-1.1083	0.973
Glyceric acid	0.3349	2.0086	0.965
cis-Pinonic acid	15.939	-10.506	0.829
Citric acid	13.771	1.9955	0.987
Benzoic acid	0.2397	-9.0599	0.545
Isopimaric acid	24.789	2.2043	0.767
C ₁₇ alcohol isomers	5.7834	-3.6924	0.571
1-Octadecanol	1.0303	-29.427	0.923
1-Eicosanol	2.4178	-0.1682	0.826
1-Docosanol	6.365	-4.0357	0.941

1-Tricosanol	31.784	2.2422	0.966
1-Pentacosanol	20.739	1.3968	0.907
1-Heptacosanol	62.289	3.9777	0.930
1-Octacosanol	14.916	0.2004	0.981
Cholesterol	0.0574	3.5395	0.980
Parsol MCX	0.5941	-75469	0.924
Benzyl butyl phthalate	7.5321	-24.228	0.543
Monostearin	0.1521	1.7562	0.987
Monopalmitin	0.0685	1.6169	0.957
