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1 **Chemical profiles of PM<sub>2.5</sub> emitted from various anthropogenic sources of**  
2 **the Eastern Mediterranean: cooking, wood burning, and diesel generators**

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11 Abstract

12 The chemical profiles of PM<sub>2.5</sub> emitted from a non-road diesel generator, wood burning and  
13 cooking activities including chicken and beef charcoal grilling and general cooking activities  
14 were determined. The characterization included the carbonaceous fraction (OC/EC), water-  
15 soluble ions, elements, and organic species comprising n-alkanes, polycyclic aromatic  
16 hydrocarbons, carboxylic acids, levoglucosan, dioxins, furans, and dioxin-like polychlorinated  
17 biphenyls. The main component in the PM<sub>2.5</sub> from the different sources was carbonaceous  
18 matter with a mass contribution to PM<sub>2.5</sub> of 49% for cooking activities, 53% for wood burning,  
19 66% for beef grilling, 72% for chicken grilling, and 74% for diesel generator with different  
20 OC/EC concentration ratios. The analysis of organic compounds contents using diagnostic  
21 ratios and indexes showed differences between the sources and revealed specific source  
22 markers. The water-soluble ions had the highest contribution in the cooking activities profile  
23 with 17% of PM<sub>2.5</sub> and the least in the chicken grilling profile (1.1%). Additionally, 29 analyzed  
24 elements were identified, and their contribution varied with the sources (ranging from 1% to  
25 11% of PM<sub>2.5</sub>). These findings could be used to differentiate these sources and could assist in  
26 the use of source apportionment methods.

27 Keywords: source profiles, PM<sub>2.5</sub>, organic species, inorganic species, cooking, diesel  
28 generator, combustion.

29

## 30 Introduction

31 Source profiles or chemical fingerprints refer to the average relative chemical composition of  
32 the PM deriving from a pollution source, expressed as the mass ratio between each species to  
33 the total PM or the total identified mass (Belis et al., 2019; Pernigotti et al., 2016). These  
34 profiles are commonly used as input data for some receptor modeling approaches such as  
35 Chemical Mass Balance (CMB) where the knowledge of the composition of the emissions is  
36 crucial (Viana et al., 2008). They are also used to attribute output factors to specific sources in  
37 multivariate factor analysis methods such as Positive Matrix Factorization (PMF) (Bove et al.,  
38 2018). Additionally, source profiles contribute to the chemical speciation of emission  
39 inventories whether to estimate emissions of hazardous and toxic pollutants or in source  
40 oriented modeling for policy makers (Chow et al., 2004).

41 Several source profile databases have been created across the globe. Source profiles have been  
42 gathered since 1988 in the United States Environmental Protection Agency (USEPA)  
43 SPECIATE database (Simon et al., 2010). To date, the database contains more than 6700 PM,  
44 gas and other profiles covering more than 2800 unique species. Recently, a European database,  
45 SPECIEUROPE, was published in 2015 by the European Joint Research Centre consisting of  
46 more than 280 profiles and reaching 250 species (Pernigotti et al., 2016). In 2017, researchers  
47 from the Chinese Research Academy of Environmental Sciences (CRAES) developed a new  
48 database of emission source profiles for PM in Chinese environments (Liu et al., 2017). The  
49 China Source Profile Shared Service contains more than 500 profiles in its first version (CSPSS  
50 1.0) (Liu et al., 2017). These databases were built following the shortage of local source profiles  
51 that represented a true challenge for receptor modeling studies in terms of source identification  
52 and comparability between studies (Pernigotti et al., 2016). Numerous source apportionment  
53 studies have been carried out using the American, European, and Chinese databases not only in  
54 their countries of development but also for most of the source apportionment studies in  
55 developing countries despite the divergence in national circumstances (Chelani et al., 2008;  
56 Gupta et al., 2007). Indeed, the chemical speciation of emissions from certain sources largely  
57 depend on parameters such as geological substrate, local practices, emission control  
58 technologies, as well as the fuel quality in the case of combustion sources and the combustion  
59 efficiency, etc. which leads to higher uncertainty in source apportionment studies (Bove et al.,  
60 2018; Colombi et al., 2010; Salameh et al., 2014; Weber et al., 2019). Hopke (2016) stressed  
61 on the idea that no efforts were deployed to develop source profiles especially for stationary

62 sources and for many others in developing countries. The main obstacle to achieve effective  
63 and accurate source apportionment remains in the lack of regional source profiles.

64 The Middle East is a vast region encompassing around 300 million inhabitants, different  
65 countries including some of the East Mediterranean. Most of these countries are considered as  
66 developing, with several cultures, traditions, and languages but mainly share common practices  
67 such as some cooking techniques, lack of law enforcement in many cases, etc. Very few source  
68 profiles were established in this region and presented chemical profiles from vehicular  
69 emissions (Daher et al., 2013; Gaga and Ari, 2019). However, other important sources of  
70 pollution can be of interest among which cooking, power generation, and wood burning. The  
71 composition of PM<sub>2.5</sub> emitted from these typical anthropogenic sources might differ from those  
72 observed in other regions due to local traditions observed in the East-Mediterranean-Middle  
73 Eastern region. The variety of products, seasoning, and cooking techniques make the  
74 gastronomical rituals different in the Middle East from other regions. It is well established that  
75 cooking plays a major role in the Middle Eastern culture, unlike anywhere else (Essid, 2012)  
76 especially during family gatherings on weekends, holidays, and special occasions with outdoor  
77 barbecue and several other cooking activities. Most of the literature studies related to emissions  
78 from cooking focused on Indian (Bano et al., 2018), Chinese (He et al., 2004; Zhang et al.,  
79 2017; Zhao et al., 2019), and fast food (Rogge et al., 1991) that have different cooking practices  
80 than the Middle East and focusing on certain species and classes of compounds. Additionally,  
81 it has been demonstrated that the inhalation of PM<sub>2.5</sub> emitted from different cooking styles could  
82 result in adverse human health effects (Bandowe et al., 2021).

83 On the other hand, diesel generators are reciprocating engines widely used in the world in  
84 temporary projects or as stand-by power in developed countries at power sensitive facilities or  
85 even as continuous backup power in developing countries due to power supply instability  
86 (Klimont et al., 2016). The literature mostly present PM<sub>2.5</sub> profiles for regulated diesel engines  
87 (Liang et al., 2005; Yilmaz and Davis, 2016) like Tier 1 to 4 in North America, Stage 1 and  
88 above in Europe, etc. Even with these lean combustion conditions, experimental and  
89 computational evidence showed important emission of pollutants from the fuel burning such as  
90 PAHs (Yilmaz and Donaldson, 2007). However, many countries especially developing ones do  
91 not always effectively enforce the law when it comes to stack emissions (Waked et al., 2012),  
92 do not have regulations on the import of regulated engines, and mostly allow the installation of  
93 the generators in heavily populated areas without any pollution abatement.

94 Another combustion source commonly known in the region is the wood burning due to forest  
95 fires but also used for cooking, residential heating, and outdoor activities such as camping. The  
96 emission characterization of these sources in the region is still limited (Alves et al., 2010;  
97 Kostenidou et al., 2013) and more focus should be given due to their impacts on air quality.

98 The objective of this paper is to determine, for the first time in the Middle Eastern region, PM<sub>2.5</sub>  
99 chemical source profiles for different anthropogenic sources such as cooking, wood burning,  
100 and emissions from unregulated non-road diesel generator. While most of PM<sub>2.5</sub> chemical  
101 profiles found in the literature focus on specific chemical fractions or species, this study  
102 presents for the first time, to the best of our knowledge, a full chemical characterization  
103 including carbonaceous fraction, ionic, elemental, and organic species. The organic speciation  
104 includes n-alkanes, Polycyclic Aromatic Hydrocarbons (PAHs), carboxylic acids,  
105 levoglucosan, dioxins (PCDD), furans (PCDF), and dioxin-like polychlorinated biphenyls (DL-  
106 PCBs). These exhaustive chemical profiles will be of utmost importance for future source  
107 apportionment studies in the region and will add additional components to the pre-existing  
108 source profiles such as carboxylic acids, sugars, PCDD/Fs and DL-PCBs. Furthermore, source  
109 markers, diagnostic ratios, and indexes are presented in order to be used in source identification.  
110 All of these results will be compared to available database in order to conclude whether the  
111 differences are significant. Additionally, these results will provide valuable information for  
112 source apportionment applications, speciated emission inventories, as well as for health impact  
113 assessment studies.

114

## 115 1. Materials and methods

### 116 1.1 Sample collection

117 PM<sub>2.5</sub> samples were collected onto 150 mm pure quartz microfibre filters (Fiorini, France)  
118 using a high-volume sampler (CAV-A/mb, MCV S.A., Spain) operating at 30 m<sup>3</sup>/h in different  
119 locations in Lebanon. Filters were pre-heated for 12 hours at 550°C before sampling to decrease  
120 the level of organic impurities. The collected filters were sealed in aluminum foil and stored at  
121 -20°C until analysis. It is worth noting that for combustion sources, the sampling was made at  
122 a sufficient distance from the combustion process to let the plume cool down and to collect the  
123 condensed fraction in the particulate phase.

124

## 125 1.2 Experimental design

126 The operating conditions, under which the sampling for the different anthropogenic sources  
127 was conducted, were detailed in the sections below and were summarized in **Table 1**. All the  
128 methods described below are representative of common practices in the East Mediterranean  
129 region. Despite the limited number of samples, this paper will give an insight on the chemical  
130 composition of PM<sub>2.5</sub> emitted from these sources, especially for some species that are scarcely  
131 presented in the literature. Variations between the replicates of the same source are related to  
132 the fact that combustion and cooking sampling were done under real world conditions (variation  
133 in humidity content, instantaneous variation of reciprocating engine load, etc.). High standard  
134 deviations were generally observed between the replicates of the same source (Akbari et al.,  
135 2021; Chantara et al., 2019; Salameh et al., 2014).

136

### 1.2.1 Wood burning (WB)

137 Hardwood combustion from different kinds of broad-leaved trees (oak, maple, beech trees, etc.)  
138 is commonly observed in the East Mediterranean for heating and outdoor activities. The wood  
139 combustion (open fire) experiment was conducted outdoor away from trees and other plant  
140 materials. Dry wood branches of hardwood broad-leaved trees (oak and fruit bearing trees) were  
141 added into the firepit in the shape of a pyramid, ignited using a lighter and burned with free air  
142 supply.

143 The sampler was placed directly in the smoke plume and 3 filters (sample label: WB) were  
144 exposed during the flaming combustion phase. Throughout the sampling, new pieces of wood  
145 were added to keep the fire going. The sampling duration varied between 10 and 15 minutes  
146 for each sample.

147

### 1.2.2 Diesel generator (DG)

148 The generator, with an engine capacity of 100 kVA, is placed in the basement of a residential  
149 building. This type is representative of the most used category (below 250 kVA) of diesel  
150 generators producing electricity for households in the region. The fuel type used is automotive  
151 diesel oil. The generator has an extended exhaust pipe of about 18 m to release emissions above  
152 the rooftop level. The sampler was placed at the same level as the top of the stack in order to  
153 collect the particulate sample with minimal influence by ambient PM. No sample was taken

154 during the cold-start phase (the first 20 min of the engine operation). Four diesel generator  
 155 samples (DG) were collected under the same operating conditions for 30 min each.

156

### 1.2.3 Cooking

157 In order to have exhaustive cooking source chemical profiles, 7 samples were collected from  
 158 different cooking experiments: 2 for charcoal grilling and 5 for general cooking activities. The  
 159 plumes emitted from the different cooking activities regroup mixed emissions released by food  
 160 and the corresponding fuel used.

161

162 **Table 1:** Operating conditions under which the different anthropogenic samples were obtained

	<b>Wood burning</b>	<b>Diesel generator</b>	<b>Cooking</b>		
			<b>Beef grilling</b>	<b>Chicken grilling</b>	<b>Cooking activities</b>
<b>Abbreviation</b>	<b>WB</b>	<b>DG</b>	<b>BG</b>	<b>CG</b>	<b>GCA</b>
<b>Main contents</b>	Wood branches of broad- leaved trees (oak and fruit bearing trees)	Volvo Penta TAD531GE diesel generator, 100 kVA, four cylinders at 1500 rpm	Small pieces of beef grilled on both sides	Large pieces of skinned chicken grilled on both sides	Burger grilling, french fries frying, use of vegetables, daily dishes served
<b>Combustible</b>	Biomass	Diesel fuel	Charcoal	Charcoal	Natural gas liquid (LPG)
<b>Sampling method</b>	High volume sampler for PM <sub>2.5</sub>				
<b>Filter used</b>	Quartz filters – 150 mm diameter				
<b>Number of samples</b>	4	3	1	1	5
<b>Sampling time</b>	10-15 min	30 min	20 min	20 min	75 min

163

#### 1.2.3.1 Beef and chicken charcoaled experiments

165 The Mediterranean region is known for its tradition of charcoal grilling especially during  
 166 weekends and holidays. The most commonly observed charcoal-grilled food in the region is  
 167 mainly beef and chicken. The charcoal is made of oak wood. It was piled in a pyramid shape in  
 168 the grill to help increase the contact between the charcoal and make the fire spread. A fire starter  
 169 cube was inserted into the pyramid and lighted. After 20 min, the coal was covered in ash. It  
 170 was then laid evenly in the grill and was ready to use. Two PM<sub>2.5</sub> samples were taken during  
 171 this procedure: the first one was sampled during beef grilling (BG) and the second one for

172 chicken (CG). The sampling duration was 20 min for each sample. First, the meat (BG) was  
173 cooked over charcoal for 20 min and turned several times during the total cooking time. The  
174 distance between the meat and the charcoal was approximately 10 cm. The sampler was placed  
175 directly in the grill smoke plume. Then, the chicken (CG) was grilled.

176

#### 177 1.2.3.2 General Cooking activities (GCA)

178 The sampling was carried out in a university kitchen cafeteria during the peak hour of work for  
179 5 consecutive days. The cooking equipment mainly consists of gas deep frying machines, free  
180 standing griddles, and steam tables for the daily dishes. The main cooking technique is gas-  
181 grilling activity to make burgers or sandwiches (meat, chicken, etc.), and frying of French fries,  
182 etc. The frying oils used are Canola and Soybean seed. There was no major outdoor ventilation  
183 leading to the assumption that the PM<sub>2.5</sub> mass collected was mainly due to the cooking activities.  
184 The sampler was placed next to the steam tables, deep-frying machine, and grill and 5 samples  
185 (GCA) were taken for 75 min each.

186

### 187 1.3 Chemical analysis

188 The chemical characterization included the analysis of the carbonaceous matter (OC, EC),  
189 water-soluble ions, major and trace elements, and organic species for the collected samples and  
190 for blank filters.

#### 1.3.1 Carbonaceous fraction

191 The organic carbon (OC) and the elemental carbon (EC) were quantified on a punch of the  
192 quartz filter (1 cm<sup>2</sup>) by a thermal optical transmission technique. It consists of a Sunset  
193 Laboratory OC/EC analyzer implementing the EUSAAR2 protocol (Cavalli et al., 2010) with  
194 a transmittance optical correction for pyrolysis.

195

#### 1.3.2 Organic compounds

196 The organic compounds were quantified by gas chromatography (ISQ 7000, Thermo Scientific,  
197 United States of America) coupled to a mass spectrometer (GC-MS) following the method  
198 described in previous studies (Fadel et al., 2021; Waked et al., 2013; 2014). In brief, one quarter  
199 of each filter was extracted with acetone and dichloromethane (1:1, v/v) by sonication for 30  
200 min at 50°C after spiking it with two internal standards (bornylacetate and ketopinac acid). The



201 extracts were concentrated to 200  $\mu\text{L}$  under a gentle flow of nitrogen gas. An aliquot of the  
202 sample is injected directly in the GC-MS to quantify non-polar compounds such as n-alkanes  
203 and PAHs while another aliquot was derivatized before injection to identify levoglucosan and  
204 carboxylic acids. The detection limits (D.L) were evaluated for the different species and  
205 correspond to the blank filter value plus 3 times the standard deviation calculated over 3  
206 measurements. D.L. varied between 0.0003 and 0.25  $\text{ng}/\text{m}^3$  except for stearic acid with higher  
207 detection limit (2.3  $\text{ng}/\text{m}^3$ ). Additionally, recoveries were determined by spiking blank filters  
208 by standard solutions. The recovery values were estimated to be between 80 and 92% for the  
209 different organic species.

210

### 1.3.3 Elements

211 Elements were analyzed by inductively coupled plasma (More detailed information on the  
212 analysis are available in previous publications (Kfoury et al., 2016; Ledoux et al., 2006a). In  
213 brief, for each sample, a 47 mm punch of the quartz filter was digested by an acid mixture of 4  
214 mL nitric acid ( $\text{HNO}_3$ ), 1 mL of hydrofluoric acid (HF), and 0.5 mL of perchloric acid ( $\text{HClO}_4$ )  
215 in a PTFE flask at 120°C overnight. Then, the acid evaporation was made at 170°C to the last  
216 drop followed by the addition of few milliliters of ultrapure water. The obtained solution was  
217 then diluted to 15 mL by ultrapure water, acidified to 2% by  $\text{HNO}_3$  and filtered on 0.45  $\mu\text{m}$   
218 cellulose acetate filter prior to analysis. Mg, Mn, Al, Na, Ba, Ca, Fe, K, Ni, P, Pb, S, Sr, Ti, and  
219 Zn were quantified by Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES,  
220 iCAP 6000 series, Thermo Scientific, United Kingdom). V, Cr, Co, Cu, As, Rb, Nb, Ag, Cd,  
221 Sn, Sb, La, Ce, and Bi were quantified by ICP-Mass Spectrometry (ICP-MS, Agilent 7900,  
222 United States of America). The analytical procedure by ICP-AES and ICP-MS was validated  
223 by considering standard reference material NIST-SRM 1648a (National Institute of Standards  
224 and Technology, United States of America). Recovery rates for all the elements varied between  
225 89% and 103% with an exception for Cr (76%). The detection limits for the elements analyzed  
226 by ICP-AES and ICP-MS corresponds to blank (water and acid blanks) values plus 3 times the  
227 standard deviation calculated over 10 measurements. D.L. for the elements analyzed by ICP-  
228 AES varied between 0.004 and 0.45  $\text{ng}/\text{m}^3$ . As for the trace elements analyzed by ICP-MS, it  
229 varied between 0.0003 and 0.01  $\text{ng}/\text{m}^3$ .

230

#### 1.3.4 Water-soluble ions (WSI)

231 The protocol for water-soluble analysis was detailed elsewhere (Kfoury et al., 2016; Ledoux et  
232 al., 2006b). Briefly, three filter punches of 19 mm diameter each for every sample were  
233 ultrasonically extracted by few milliliters of ultrapure water for 20 min. The obtained solution  
234 is collected using a syringe, then filtered through a 0.45  $\mu\text{m}$  porosity cellulose acetate  
235 membrane previously rinsed with ultrapure water. The leachate containing the ions was placed  
236 in a polyethylene flask of 20 mL. The filters were submerged with ultrapure water prior to 2  
237 additional ultrasonic extractions and filtrations and then stored at  $-20^{\circ}\text{C}$  until analysis. The  
238 samples were analyzed by liquid ion chromatography (Dionex<sup>TM</sup> ICS-900, Thermo Scientific,  
239 United Kingdom) for the quantification of  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{Na}^+$ , and  $\text{NH}_4^+$ .  
240 D.L. varied between 0.17 and 0.78  $\text{ng}/\text{m}^3$  for the different WSI.

241

#### 1.3.5 Dioxins, furans and dioxin-like polychlorobiphenyls

242 Dioxins, furans and dioxin-like polychlorobiphenyls were quantified by MicroPolluants  
243 Technology SA (Saint Julien Les Metz, France). The PCDD/PCDF and DL-PCB analysis  
244 method was based on the US EPA method 1613 and 1688 respectively using a high-resolution  
245 gas chromatography coupled to a high-resolution mass spectrometer (HRGC/HRMS). Detailed  
246 information are found in Borgie et al. (2015).

247

#### 248 1.4 Mass reconstruction of $\text{PM}_{2.5}$

249 The reconstructed mass of  $\text{PM}_{2.5}$  is calculated by summing the inorganic ions, elemental carbon,  
250 geological minerals, organic matter (OM) and other elements calculated as follows (Eq.1):

$$251 \quad PM = OM + EC + [\textit{geological minerals}] + [\textit{inorganic ions}] + [\textit{other elements}] \quad (\text{Eq. 1})$$

252

253 Organic matter (OM) is calculated by multiplying OC with an empirical coefficient (OM/OC)  
254 to account the mass of unmeasured oxygen and hydrogen in the organic matter (Turpin and  
255 Lim, 2001). In this study, the OM/OC ratio was chosen to be 1.7 for the wood burning  
256 experiment (Reff et al., 2009; Reid et al., 2005), 1.32 for beef grilling, 1.36 for chicken grilling,  
257 1.42 for general cooking activities (Mohr et al., 2009; Zhao et al., 2007a), and 1.2 for diesel  
258 generator emissions (Petzold et al., 2008; Sothea and Kim Oanh, 2019). Elemental carbon arises  
259 from primary combustion sources and therefore no multiplication factor is needed, its mass is  
260 used unchanged (Harrison et al., 2003). For the inorganic ions, a hydration factor of 1.29 was

261 applied to convert the concentration of these species ( $\text{Na}^+$ ,  $\text{Cl}^-$ ,  $\text{Mg}^{2+}$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{NH}_4^+$ )  
262 into hydrated ones to consider the bound water (Harrison et al., 2003; Sciare et al., 2005). The  
263 geological minerals concentration was estimated by assuming that these elements exist as  
264 oxides:  $\text{Al}_2\text{O}_3$ ,  $\text{SiO}_2$ ,  $\text{K}_2\text{O}$ ,  $\text{FeO}$  and  $\text{Fe}_2\text{O}_3$  in equal amounts, and  $\text{TiO}_2$ . The formula used to  
265 calculate the geological minerals concentration (Chow et al., 2015):

$$266 \quad [\textit{geological minerals}] = 2.2 \times [\textit{Al}] + 2.49 \times [\textit{Si}] + 1.63 \times [\textit{Ca}] + 2.42 \times [\textit{Fe}] + 1.94 \times [\textit{Ti}]$$

267 Elemental Si was not analyzed by the ICP method used, it was estimated using the (Si/Al) ratio  
268 derived from Si and Al concentrations derived from field measurements near sources in other  
269 studies. The Si/Al ratio used was 2.4 for wood burning (Sun et al., 2019), 0.5 for beef and  
270 chicken grilling (Wang et al., 2020), 0.9 for general cooking activities (Sun et al., 2020), and  
271 1.0 for diesel generators (Yayong et al., 2018). Other elements correspond to the sum of the  
272 remaining elements.

273

## 274 2 Results and discussions

275 The chemical relative abundance of the different species in the carbonaceous, ionic, and  
276 elemental fractions were calculated by dividing their mass by the reconstructed  $\text{PM}_{2.5}$  mass.  
277 They were presented in **Fig. 1**, and in details in **Table S1** and **Figure S1**. The organic species  
278 were reported to the OM mass and were given in **Table S2**.

### 279 2.1 Carbonaceous fraction

280 In the different chemical profiles of  $\text{PM}_{2.5}$ , OC was the main component, ranging from 40% of  
281  $\text{PM}_{2.5}$  for WB to 72% for CG (**Fig. 1**). EC ranged from 0.1% for CG to 20% for DG which  
282 presents the second most abundant share in WB and DG profiles.

283 High differences in the distribution of the carbonaceous fraction (OC+EC) occur between  
284 charcoal and general cooking activities profiles. GCA releases less carbonaceous matter (49%  
285 of  $\text{PM}_{2.5}$ ) compared to BG (66%) and CG (72%). These differences were mainly attributed to  
286 the cooking process and ingredients (Abdullahi et al., 2013). Many studies have compared  
287 cooking styles and reported different chemical profiles for the emitted particulate matter.

288 The OC/EC ratio is an essential tool to differentiate combustion sources. This study determined  
289 a value of 3.4 for wood burning, comparable to the findings reported by Sun et al. (2019) for  
290 wood branches (2.69). As for cooking profiles, the highest ratio was found for CG (622),  
291 followed by BG (45) and GCA (18). This large difference between CG and BG could be

292 attributed to the difference in fat content between the two types of food. Foods containing high  
293 percentage of fat usually generate higher OC emission rates than those with a lower fat  
294 percentage (Abdullahi et al., 2013).

295 The ratio for GCA was close to the value of 19.3 reported for samples collected from the exhaust  
296 stacks of a university canteen in Portugal (Alves et al., 2014). The lowest ratio was found for  
297 the diesel generator (2.9), slightly higher than 2.61 reported by Sothea and Kim Oanh (2019)  
298 for a 92 kW backup generator in Cambodia. For comparison, values of 0.37 were reported for  
299 tunnel studies (diesel and gasoline vehicles), 2.2-6.2 for urban areas, and 0.64 for diesel trucks  
300 (El Haddad et al., 2009; Waked et al., 2014).

301

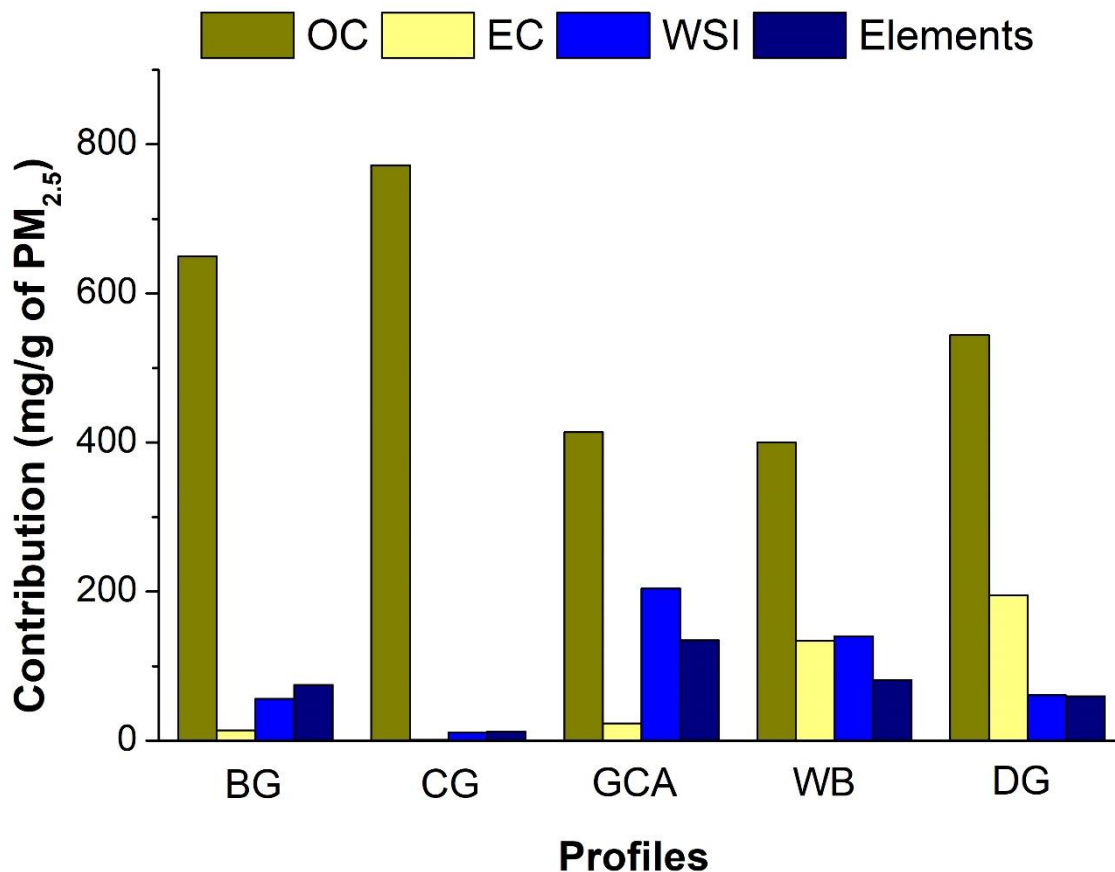
## 302 2.2 Organic compounds

### 2.2.1 N-alkanes

303 C<sub>13</sub>-C<sub>32</sub> homologue series are presented for the different profiles in **(Fig. 2)**. Total C<sub>13</sub>-C<sub>32</sub>  
304 average contribution to OM were 11%, 0.8%, 0.6%, 0.6%, and 0.3% for DG, BG, WB, GCA,  
305 and CG, respectively. The high contribution of n-alkanes in the DG profile is mainly due to the  
306 composition of the diesel fuel that comprises a large fraction of n-alkanes (Liang et al., 2005).

307 Different methods can be used to characterize the alkanes profiles (Fadel et al., 2021). Briefly,  
308 the overall carbon preference index (Overall CPI) for C<sub>13</sub>-C<sub>31</sub> is a measure of odd to even carbon  
309 predominance with a value close to 1 for petrogenic sources, 2 to 5 for biomass and coal burning  
310 and values higher than 6 for biogenic emissions. The wax ratio (Wax%) can be used to assess  
311 the contribution of wax n-alkanes from plants. The n-alkanes distribution pattern can be used  
312 to identify alkanes with the highest concentrations. For wood burning, the profile pattern shows  
313 a predominance of high molecular weight n-alkanes especially C<sub>27</sub>, C<sub>29</sub>, and C<sub>31</sub> with a  
314 maximum at C<sub>29</sub> **(Fig. 2)**. This pattern is considered as a plant epicuticular wax signature  
315 leading to a high Wax% of 58%. C<sub>27</sub>, C<sub>29</sub>, and C<sub>31</sub> are considered as n-alkanes tracers for  
316 biomass burning (Simoneit, 2002). The CPI value for WB (CPI=4) is in the range reported for  
317 biomass activities of 2 to 5 (Simoneit, 2002). A distinctive pattern was observed for DG where  
318 the n-alkane homologues partitioning exhibited a smooth hump-like distribution with the  
319 highest contribution for C<sub>20</sub>-C<sub>21</sub> **(Fig. 2)**. A similar profile was observed for diesel powered  
320 engines (Rogge et al., 1993).

321



322

323 **Fig. 1:** Contribution of OC, EC, water-soluble ions (WSI), and elements to reconstructed  $PM_{2.5}$   
 324 emitted from the different sources (BG: beef grilling, CG: chicken grilling, GCA: general  
 325 cooking activities, WB: wood burning, DG: diesel generator).

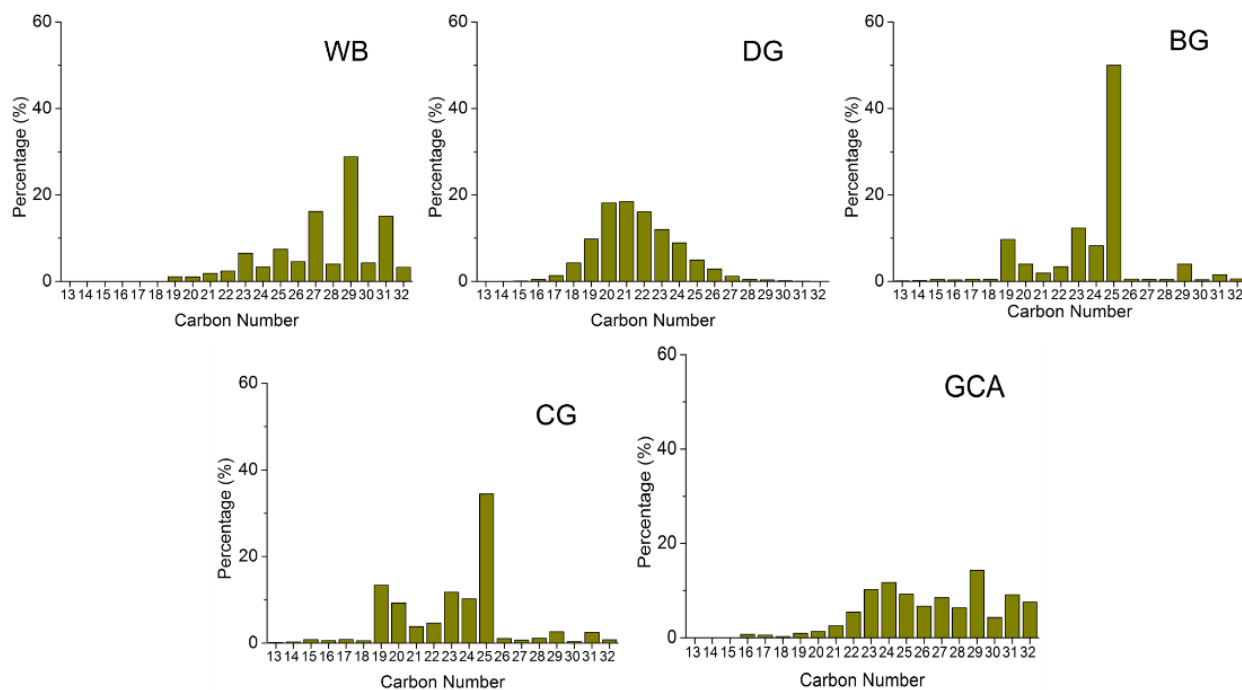
326

327 A CPI value (0.9) close to unity usually attributed for petrogenic sources and a very negligible  
 328 Wax% characterizes the diesel emissions from non-road generators.

329 For charcoal cooking (BG and CG), the profiles are much alike with a maximum contribution  
 330 for  $C_{25}$ . This observation can lead to the assumption that the n-alkanes are not predominantly  
 331 emitted from the food itself but from the charcoal combustion. This hypothesis can be supported  
 332 by CPI values of 2.5 and 4.5 for CG and BG respectively that are in the range of coal  
 333 combustion. However, a different pattern was observed for GCA. The CPI value for this GCA  
 334 profile (1.3) and the Wax% (22%) were relatively low compared to various Chinese cooking  
 335 restaurants with calculated CPI and Wax% in the ranges of 2.3-4.6 and 40-64%, respectively  
 336 (He et al., 2004; Zhao et al., 2007a). These differences may be attributed to the different cooking

337 methods and frying techniques without forgetting the large consumption of vegetables in the  
338 Chinese cooking causing higher CPI and Wax% ratios (Zhao et al., 2007b).

339



340  
341 **Fig. 2:** N-alkanes profile patterns associated with PM<sub>2.5</sub> for the different sources (WB: wood  
342 burning, BG: beef grilling, CG: chicken grilling, DG: diesel generator, GCA: general cooking  
343 activities).

344

### 2.2.2 Polycyclic Aromatic Hydrocarbons

345 The relative concentration profiles of the different PAHs are presented in **Table S2** with the  
346 share (in mg/g of OM) of each one. Additionally, the distribution of these species with aromatic  
347 ring number was also shown in **Fig. 3** to further characterize them in the profiles.

348 The wood burning experiment emitted at least 5 times more PAHs normalized to OM than the  
349 other sources (2.64 mg/g for WB against 0.54 for BG, 0.35 for CG, 0.14 for GCA, and 0.44 for  
350 DG). The higher PAHs share for WB compared to CG and BG can be attributed to the more  
351 complete combustion of charcoal compared to wood. Furthermore, the volatilization and the  
352 pyrolysis of the volatile matter during the production of charcoal, and its low water content  
353 could also reduce the aromatic compounds formation (Oanh et al., 1999; Shen et al., 2011).

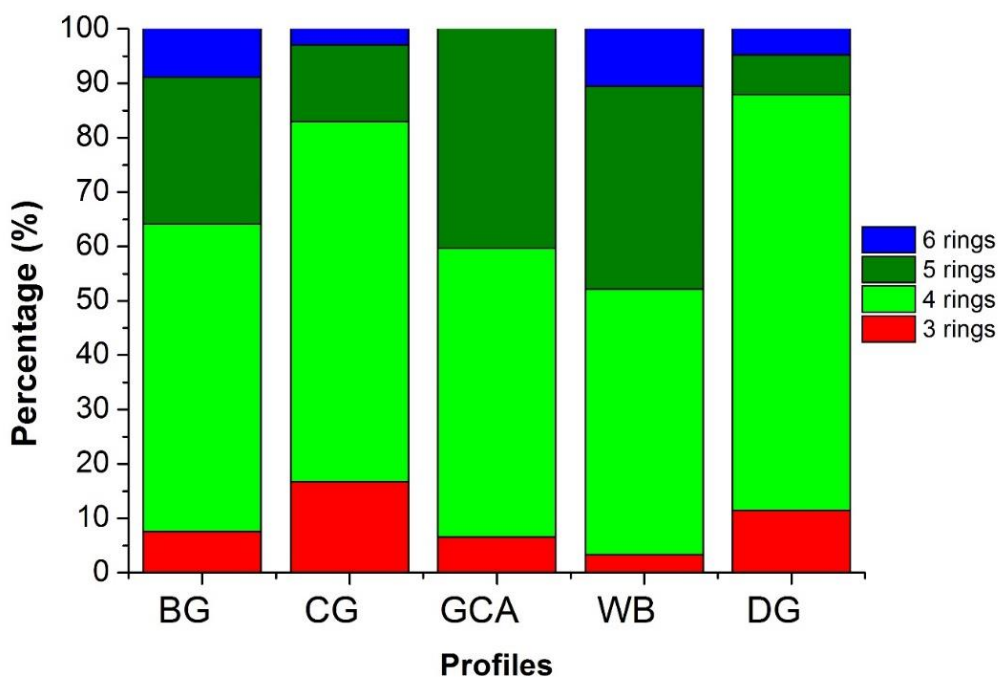
354 Low PAHs shares were observed for the different cooking profiles. McDonald et al. (2003)  
355 suggested that the PAHs emitted during grilling activities were formed from the interaction  
356 between the lipidic material in food and the cooking appliance since the temperature reached  
357 on the surface of food was relatively low to form PAHs (maximum PAHs production occurs at  
358 750 °C) and PAHs compounds were absent in the food chemical analysis. Furthermore, the  
359 abundance of 4-rings PAHs in BG and CG profiles might be related to the coal combustion.  
360 According to Vicente et al. (2018), fly ashes from charcoal combustion are rich in 4-rings PAHs  
361 with fluoranthene and pyrene as the most abundant aromatics whereas raw charcoal is mainly  
362 rich in 3-rings PAHs. As for GCA, several studies showed that 6-rings PAHs were rarely  
363 generated during cooking processes especially during deep frying activities because high  
364 temperatures can decompose high molecular weight PAHs (Sun et al., 2020; Zhao et al., 2019).  
365 The most abundant PAHs for GCA were benzo[b]fluoranthene (B[b]Fl), pyrene (Pyr), and  
366 chrysene (Chr).

367 Like BG, CG, and GCA, 4-rings PAHs predominated the total detected PM<sub>2.5</sub>-bound PAHs  
368 emitted from WB with Fluoranthene (Fla), Pyr, benzo[a]anthracene (B[a]An) as the most  
369 abundant compounds. This distribution pattern along with the dominant PAHs were consistent  
370 with the data reported for the combustion of different types of biomass fuels (maize straw,  
371 wheat straw, and wood branches) (Sun et al., 2019). Pyrene was the most abundant PAH in the  
372 diesel generator profile, accounting for more than 50% of the sum of PAHs and highly  
373 contributing to the 4-rings category that is the most abundant.

374 PAHs diagnostic ratios were usually used for source apportionment in different environmental  
375 samples (Tobiszewski and Namieśnik, 2012). For every ratio, the species used should have  
376 similar reactivity in the atmosphere: Fla and Pyr (Molecular Weight =202 g/mol), B[a]An and  
377 Chr (MW = 228 g/mol), indeno[1,2,3-c,d]pyrene (InPy) and benzo[g,h,i]perylene (B[ghi]Pe)  
378 (MW= 278 g/mol), and anthracene (Anth) and phenanthrene (Phe) (MW= 178 g/mol). These  
379 ratios could serve as markers for the different source profiles presented in this paper and were  
380 reported in **Table 2**.

381 The values obtained for wood burning (0.49 for Fla/(Fla+Pyr), 0.57 for B[a]An/(B[a]An+Chr),  
382 0.45 for InPy/(InPy+B[ghi]Pe), and 0.7 for Anth/(Anth+Phe)) are mostly in the ranges of ratios  
383 reported in other studies for biomass burning (Gong et al., 2019; Tobiszewski and Namieśnik,  
384 2012; Vicente et al., 2012).

385



386

387 **Fig. 3:** Distribution of PAHs for the different sources (BG: beef grilling, CG: chicken grilling,  
 388 GCA: general cooking activities, WB: wood burning, DG: diesel generator) according to the  
 389 number of aromatic rings in mass percentages.

390

391 As for grilling activities, the similarity in the ratios and the closeness to the values reported in  
 392 the literature imply that these PAHs are mainly emitted from the charcoal combustion in the  
 393 barbecue grills (Gong et al., 2019; Vicente et al., 2018). For GCA, Fla/(Fla+Pyr) and  
 394 B[a]An/(B[a]An+Chr) ratio values are close to those reported for deep-frying activities and  
 395 Chinese gas cooking (See and Balasubramanian, 2008; Wei See et al., 2006). A particular  
 396 attention was given to the ratios determined for diesel generators emissions. The values present  
 397 in the literature describe mostly diesel emissions from on-road vehicles (Gong et al., 2019;  
 398 Ravindra et al., 2008; Riffault et al., 2015; Shi et al., 2014). Although many similarities exist,  
 399 the modes of operation are different. Non-road diesel generators run at a certain rpm  
 400 (revolutions per minute) and voltage and the amperage they produce can vary with the load  
 401 while on-road diesel vehicles operate in transient conditions (Liang et al., 2005; Shah et al.,  
 402 2006). These differences in the operation mode can potentially result in different emission  
 403 characteristics and different diagnostic ratios for PAHs. On the other hand, Liang et al. (2005)  
 404 studied the organic composition of diesel particulate matter from a non-road diesel generator  
 405 and showed Fla/(Fla+Pyr) and B[a]An/(B[a]An+Chr) ratio values (0.28 and 0.5, respectively)



406 close to the values obtained in this study (0.24 and 0.41, respectively) while values for  
407 InPy/(InPy+B[ghi]Pe) and Anth/(Anth+Phe) were different (**Table 2**).

408

### 2.2.3 Carboxylic acids

409 According to **Table S2**, carboxylic acids were the most abundant organic species in the  
410 chemical profiles of cooking activities. The highest contribution was observed for GCA  
411 explaining 24% of the OM mass followed by BG with 19% and CG with 11%. During high  
412 temperature treatment of food, several chemical processes occur such as sugar degradation,  
413 pyrolysis of proteins and amino-acids, and the degradation of fats. These mechanisms lead to  
414 the production of free fatty acids, glycerol, and glycerides (Abdullahi et al., 2013). Additionally,  
415 the use of oils in cafeteria cooking operations such as deep frying will increase fatty acids  
416 emissions being the major constituents of triglycerides in seed oils (Schauer et al., 2002; Zhao  
417 et al., 2007a). From the saturated fatty acids presented in this paper, the most abundant are  
418 palmitic (C<sub>16</sub>) and stearic (C<sub>18</sub>) acids accounting for more than 80% of the total mass of fatty  
419 acids in the three cooking profiles. These compounds are usually used as organic markers for  
420 cooking activities in different source apportionment studies (Abdullahi et al., 2013). Carboxylic  
421 acids were also found in biomass burning events and contribute to 5% of the OM mass. The  
422 highest concentration was observed for palmitic acid (C<sub>16</sub>) accounting for 33% of the sum of  
423 carboxylic acids. These findings were in accordance with the results obtained from wildfires  
424 organic speciation in central Portugal (Vicente et al., 2012) and with the combustion of  
425 hardwoods (Oros and Simoneit, 2001). Long-chain fatty acids are mainly considered as basic  
426 units of plant fats, oils, and phospholipids while short-chain species (C<16) are reported as  
427 minor species of fungi and insects. As for the diesel generator, carboxylic acids contribute to  
428 8% of the OM mass. Liang et al. (2005) determined the abundance of fatty acids in the engine  
429 oil and the diesel particulate matter and found a strong similarity, bringing the conclusion that  
430 fatty acids in diesel particles are not emitted from the combustion process itself but from the  
431 evaporation of engine oil.

432

### 2.2.4 Levoglucosan

433 Levoglucosan or 1,6-anhydro-β-D-glucopyranose originates from the pyrolysis of cellulose and  
434 hemicellulose at high temperature (Giannoni et al., 2012). It is considered as an organic  
435 molecular marker for biomass burning due to its stability and its relatively high share in biomass

436 smoke (Chantara et al., 2019). This compound is the most abundant organic species in the WB  
437 profile accounting for 17% of the OM mass. Levoglucosan/OC ratio is commonly used to  
438 differentiate softwood and hardwood combustion. The value obtained in our study of 0.3 is in  
439 the range of hardwood combustion (0.25-0.4) reported by Fine et al. (2004) compared to lower  
440 ratios for softwood ranging between 0.1 and 0.23.

441

#### 2.2.5 Dioxins, furans, and DL-PCBs

442 **Table S3** shows congeners contribution to OM of PCDD/F and DL-PCBs for each source. The  
443 BG and CG were combined in one factor and will be presented as “charcoal grilling”.  
444 1,2,3,4,6,7,8 HpCDD was identified in all emissions and was the only PCDD detected in GCA,  
445 while OCDD contributed the most to the other profiles (73% of PCDD in WB, 76% in DG, and  
446 70% in charcoal grilling, i.e., BG and CG). The DG emissions are 4.5, 3.7, and 2 more  
447 concentrated in PCDD than GCA, charcoal grilling, and WB, respectively. However, charcoal  
448 grilling showed the highest value when calculating the 2,3,7,8-TCDD equivalent (calculated by  
449 summing the multiplication results of each congener by its corresponding toxic equivalent  
450 factor- WHO-TEF(Van den Berg et al., 2006). As for furans, 6 species out of 10 were identified  
451 in the grilling sample mainly HxCDF and HpCDF.

452 The highest concentrations were recorded for PCDF emitted from the diesel generator with also  
453 the highest 2,3,7,8-TCDD equivalent. The PCDD/PCDF ratio is generally used to identify  
454 emission sources. It was calculated for the different profiles and values are presented in **Table**  
455 **2**.

456 A similar observation was made by Lee et al. (2005) showing higher PCDD/PCDF emission  
457 factor ratio for domestic biomass burning (4.7) compared to coal combustion (2.1). Up to our  
458 knowledge, there is no PCDD/F data reported for oil cooking activities. The ratio in our study  
459 showed that the oil cooking source emits 2.4 times more PCDD than PCDF. The most abundant  
460 DL-PCB was PCB 118 accounting for 46%, 51%, 52%, and 52% of the total PCB in charcoal  
461 grilling, WB, GCA, and DG respectively followed by PCB 105. The total TEQ value expressed  
462 in pg/g of OM was the highest for DG, followed by charcoal grilling, GCA, and WB.

463

464

465

466 **Table 2:** Experimental source diagnostics ratios and comparison with the literature (in brackets)  
 467 when available

	Wood burning	Diesel generator	Cooking		
			Beef grilling	Chicken grilling	General cooking activities
<b>OC/EC ratio</b>	3.4	2.9	45	621	18
<b>n-alkanes diagnostic indexes</b>					
Overall CPI	4.00	0.94	4.48	2.53	1.34
Wax%	57.5%	1.4%	64.7%	46.1%	22.1%
<b>PAHs diagnostic ratios</b>					
Fla/(Fla+Pyr)	0.49 (0.2-0.7) <sup>a</sup>	0.24 (0.28) <sup>b</sup>	0.51 (0.47) <sup>a</sup>	0.56 (0.47) <sup>a</sup>	0.47 (0.53) <sup>c</sup>
B[a]An/(B[a]An+Chr)	0.57 (>0.37) <sup>e</sup>	0.41 (0.5) <sup>b</sup>	0.46 (0.47) <sup>a</sup>	0.43 (0.47) <sup>a</sup>	0.44 (0.28; 0.4) <sup>c,f</sup>
InPy/(InPy+B[ghi]Pe)	0.45 (>0.5) <sup>d</sup>	0.18 (0.68) <sup>b</sup>	0.56 (0.50) <sup>a</sup>	0.45 (0.50) <sup>a</sup>	-
Anth/(Anth+Phe)	0.70 (>0.1) <sup>e</sup>	0.85 (0.05) <sup>b</sup>	0.75 (>0.1) <sup>e</sup>	0.73 (>0.1) <sup>e</sup>	-
<b>PCDD/PCDF ratio</b>	3.27	1.16	1.95		2.44
<b>Water-soluble ions ratios</b>					
K <sup>+</sup> /EC	0.36	0.01	1.32	3.61	0.12
K <sup>+</sup> /OC	0.12	0.003	0.03	0.01	0.007
Cl <sup>-</sup> /SO <sub>4</sub> <sup>2-</sup>	3.96	-	1.34	0.4	-

468 <sup>a</sup>(Vicente et al., 2012), <sup>b</sup>(Liang et al., 2005), <sup>c</sup>(See and Balasubramanian, 2008), <sup>d</sup>(Tobiszewski  
 469 and Namieśnik, 2012), <sup>e</sup>(Gong et al., 2019), <sup>f</sup>(Wei See et al., 2006)

470

### 471 2.3 Water-soluble ions (WSI) composition

472 Water-soluble ions have the highest share of PM<sub>2.5</sub> in the GCA profile (17%), then WB (14%),  
 473 then DG and BG (6%), and finally CG (1%) (**Fig. 2**).

474 GCA profile showed more than 85% of the ionic mass for SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>. This result  
 475 was also observed by See and Balasubramanian (2008) for oil deep-frying activities. Although  
 476 WSI contributed more to the total PM<sub>2.5</sub> mass in BG compared to CG, they have a similar  
 477 distribution in ions with K<sup>+</sup>, SO<sub>4</sub><sup>2-</sup>, and Cl<sup>-</sup> having the highest contribution and accounting for  
 478 more than 70% of the total WSI mass. McDonald et al. (2003) also reported these ions as the  
 479 most abundant in the fumes of meat charbroiling. The high amounts of K<sup>+</sup> and Cl<sup>-</sup> in both

480 charcoal and wood burning are mainly due to their abundance in the fluid of plants ; they  
481 volatilize during the combustion process and, after condensation or nucleation reaction (Alves  
482 et al., 2010), become in the particulate phase. In the three cooking profiles,  $\text{Ca}^{2+}$  ions could  
483 mainly be emitted from the cooking utensils and raw materials (Sun et al., 2020).

484 As for wood burning,  $\text{K}^+$  and  $\text{Cl}^-$  have the highest share of WSI (74%) followed by  $\text{SO}_4^{2-}$  and  
485  $\text{Ca}^{2+}$ . These ions account for nearly 92% of the total soluble ions in the profile. Our results are  
486 consistent with those reported for other biomass burning experiments (86-94%) (Chantara et  
487 al., 2019; Sun et al., 2019).

488 The main water-soluble ions in diesel particulate matter were  $\text{Ca}^{2+}$ ,  $\text{SO}_4^{2-}$ , and  $\text{NO}_3^-$ .  $\text{SO}_4^{2-}$  only  
489 accounted for 27% of the total WSI mass which is lower than the 80% value reported by Sothea  
490 and Kim Oanh (2019) for a 25 kVA and a 110 kVA diesel backup generators. However, the  
491 main difference between the two experiments remains in the diesel sulfur content. The diesel  
492 used by the latter study was assumed to have the same quality as the diesel oil used for  
493 transportation and thus a sulfur content of 0.2% (2,000 ppm). On the other hand, since 2017,  
494 Lebanon implemented stricter laws on the quality of the imported automotive diesel by  
495 decreasing the sulfur content to 10 ppm and allowing higher sulfur content diesel to be delivered  
496 only to the big industries (ex. cement plants, fertilizer industry, etc.) and power plants. Despite  
497 that law enforcement is not always guaranteed, the sulfur content in automotive diesel did not  
498 exceed the 200-300 ppm since 2014-2015. This difference in the fuel oil sulfur content might  
499 explain the chemical composition differences since it has been established that sulfate in diesel  
500 particulate matter is function of the engine operating mode and the fuel sulfur content  
501 (Saiyasitpanich et al., 2005).

502 Different diagnostic ratios including water-soluble ions are used to distinguish emission  
503 sources. For instance,  $\text{K}^+/\text{EC}$  and  $\text{K}^+/\text{OC}$  ratios can be used to distinguish biomass burning from  
504 fossil fuel combustion. The ratios calculated from this study can be used as marker values for  
505 the sources and are reported in **Table 2**.

506 DG exhibited low  $\text{K}^+/\text{EC}$  and  $\text{K}^+/\text{OC}$  (0.01 and 0.003, respectively) compared to WB (0.36 and  
507 0.12, respectively). These findings were also reported by Benetello et al. (2017) that assigned  
508 a range of  $\text{K}^+/\text{EC}$  and  $\text{K}^+/\text{OC}$  at the source of 0.025-0.09 and 0.01 for fuel burning, respectively  
509 and 0.21-0.46 and 0.1 for biomass burning, respectively. Additionally, the  $\text{Cl}^-/\text{SO}_4^{2-}$  ratio might  
510 be used to differentiate wood and coal combustion. In this study, higher values were observed

511 for biomass burning (3.96) compared to coal combustion (0.4-1.34) and indicate that  $\text{SO}_4^{2-}$  is  
512 less emitted from WB compared to CG and BG.

513

#### 514 2.4 Elemental composition

515 Twenty-nine analyzed elements in the profiles present different fractions of  $\text{PM}_{2.5}$ : the lowest  
516 total contribution of the considered elements was observed for CG which accounted to 1.2%,  
517 followed by DG (6%), then BG (7%), WB (8%), and GCA (11%) (**Figure S1**).

518 Ca is the most abundant element in the DG profile contributing to 56% of the sum of elements  
519 and is classified as the third most abundant species after OC and EC contributing to 3.3% of  
520  $\text{PM}_{2.5}$ . Other elements such as Fe, Al, Mg, K, and Zn were also abundant in the profile sharing  
521 25% of their sum. These results are consistent with the ones from the composition of particles  
522 emitted by a lab-designed engine (Mg, Na, Al, K, Ca, Fe, Zn) (Kerminen et al., 1997), and in  
523 diesel PM composition reported by Yang et al. (1998) including Fe, Al, Ca, Na, Cr, Ba, Ni, Mg,  
524 and K. However, some differences exist between the three profiles that might be caused by the  
525 presence of organometallic additives in the diesel, the lubricating oil quality, and the operating  
526 conditions. For instance, elements such as Ca and Zn can be used as markers of total lubrication  
527 oil consumption and their contribution to the total mass of elements may vary depending on its  
528 quality (Sippula et al., 2014).

529 In the WB profile, K shares 50% of the mass of the 29 analyzed elements and is identified as a  
530 major metal in biomass burning since it is a major constituent of plants (Adam et al., 2021).  
531 The other abundant elements in descending order were Ca, Al, S, Na, Fe, and Mg. These  
532 findings were also observed during biomass burning events and in ash samples in a simulated  
533 open combustion chamber (Akbari et al., 2021). Fe is considered as an important component in  
534 vegetation and intervenes in physiological processes while Mg and Na play a major role in the  
535 metabolism and the structure of the plant respectively (Alves, 2008; Brownell, 1979). Other  
536 elements accumulated by trees were also present in WB such as Cu, Pb, Ti, Zn, and Mn.

537 Even though the total mass fraction of elements differs between BG, CG, and GCA, the most  
538 abundant elements observed are S, Ca, Na, K, Al, Mg, and Fe. They account for about 98% of  
539 the measured elements despite the cooking method used. Similar observations were made by  
540 See and Balasubramanian (2008) showing high content in Ca, Na, Fe, Mg, and Al for different  
541 cooking styles. This study showed sulfur as the dominant element for GCA in agreement with  
542 other residential cooking experiments (Wang et al., 2015; Zhao et al., 2019). This might be due

543 to the addition of organic sulfur compounds such as ethyl mercaptan to liquid natural gas used  
544 for the detection of leak.

545 As for the other elements, Na is attributed to the salts and seasoning added to the food while Ca  
546 which has an important contribution in the three cooking profiles could be released from raw  
547 materials (Zhang et al., 2017). K showed a considerable abundance in BG and CG and is mainly  
548 associated with charcoal combustion (Zhao et al., 2019). Although their contribution is  
549 negligible to the total mass of the profile, trace elements are known to have harmful effects on  
550 the human health. The GCA profile shows the presence of Pb and Mn that could be emitted  
551 from cooking oil (peanut and canola oil) (Zhao et al., 2019). Other trace elements such as Cr,  
552 Ni, Cu along with Fe are released from the interaction between the heating source and the  
553 stainless steel wok used as cooking utensils in the different cooking profiles (Sun et al., 2020).

554

## 555 2.5 Organic and inorganic source markers

556 The knowledge of chemical profiles is an important tool to be used by receptor model users in  
557 the source apportionment exercise. According to Galvão et al. (2020), for a better separation  
558 between sources, the chemical speciation should include carbonaceous fraction, ions, elements,  
559 and organic compounds with a focus on chemical markers. The latter are considered as  
560 fingerprints that link the chemical profiles to the sources.

561 The markers of hardwood burning were OC, EC (with an OC/EC ratio of 3.4), potassium in its  
562 ionic and elemental form, and  $\text{Cl}^-$ , accounting for 68% of the  $\text{PM}_{2.5}$ . Levoglucosan was the  
563 major contributor to organic compounds along with the predominance of high molecular weight  
564 n-alkanes, higher PAHs mass per gram of OM compared to the other profiles especially for  
565 fluoranthene, pyrene, and benz[a]anthracene. The values of the characteristic PAHs ratios were  
566 0.49 for fluoranthene/ (fluoranthene + pyrene) and 0.57 for benz[a]anthracene  
567 / (benz[a]anthracene + chrysene). The  $\text{K}^+/\text{EC}$  and  $\text{K}^+/\text{OC}$  ratios were used to differentiate wood  
568 from fuel combustion and were reported as 0.36 and 0.12 for wood burning, respectively. The  
569 ratio between levoglucosan and OC for the hardwood combustion was equal to 0.3 in this study.

570 The major contributors to diesel generators profile were OC and EC accounting for more than  
571 74% of  $\text{PM}_{2.5}$  with an OC/EC ratio of 2.9, followed by  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , Ca,  $\text{Ca}^{2+}$ , and S. N-alkanes  
572 showed a high contribution to OC in this profile with a profile pattern maximizing at  $\text{C}_{20}\text{-C}_{21}$ ,  
573 a carbon preference index value close to unity (0.9) and very low wax percentage (1.4%).  
574 Pyrene was the most abundant compound between PAHs. The PAHs ratios reported for the

575 diesel combustion in our study were 0.24 for fluoranthene/(fluoranthene + pyrene), 0.41 for  
576 benz[a]anthracene /(benz[a]anthracene +chrysene), and 0.18 for indeno[1,2,3-c,d]pyrene /  
577 (indeno[1,2,3-c,d]pyrene + benzo[g,h,i]perylene). The profile also recorded the highest relative  
578 share of PCDD/F and DL-PCBs compared to the other profiles. The  $K^+/EC$  and  $K^+/OC$  ratios  
579 were 0.01 and 0.003, respectively.

580 The three cooking profiles showed similarities with a high contribution of carboxylic acids,  
581 especially palmitic and stearic acids, high OC contribution and low EC share. The presence of  
582 food with different fat content and composition might increase the share of OC in the chemical  
583 profile. Chicken grilling profile recorded the highest OC contribution of 72% of  $PM_{2.5}$  with an  
584 OC/EC ratio of 622. Beef grilling profile markers were OC, EC (with an OC/EC ratio of 45),  
585  $Cl^-$ , K,  $NO_3^-$ , and  $SO_4^{2-}$ . For both profiles,  $C_{25}$  having the highest contribution to n-alkanes,  
586 carbon preference index values between 2 and 5, and high abundance of 4-rings PAHs with  
587 fluoranthene/(fluoranthene + pyrene), benz[a]anthracene /(benz[a]anthracene +chrysene), and  
588 (indeno[1,2,3-c,d]pyrene + benzo[g,h,i]perylene) close to 0.5 were attributed to the charcoal  
589 combustion. The general cooking activity profile presented an OC/EC ratio of 18, an important  
590 contribution of  $SO_4^{2-}$ ,  $NO_3^-$ , and  $NH_4^+$  (85% of WSI), and a high abundance of Al, Na, and Ca.  
591 The n-alkanes pattern maximized at  $C_{29}$ , and the major emitted PAHs were  
592 benzo[b]fluoranthene, pyrene, and chrysene.

593

### 594 3 Conclusions

595 This study is a first in the East Mediterranean and Middle East region presenting a full chemical  
596 characterization of 5  $PM_{2.5}$  sources in the East Mediterranean region: hardwood burning, diesel  
597 generator, beef and chicken charcoal grilling and general cooking activities. This  
598 characterization included the analysis of the carbonaceous fraction, ions, elements, and organic  
599 species comprising n-alkanes, polycyclic aromatic hydrocarbons, carboxylic acids,  
600 levoglucosan, dioxins, furans, and dioxin-like polychlorobiphenyls. The markers for the  
601 profiles along with the different ratios will be of utmost importance for future source  
602 apportionment and modeling studies. It would be also important for further studies to consider  
603 other sources such as waste incineration, tire burning, etc. The results of this work will  
604 contribute to further broaden our knowledge of the PM sources in the region by expanding the  
605 database for the East Mediterranean - Middle Eastern region. It might be also interesting to  
606 evaluate the impact of using these local profiles on the policy making in the region.

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