

1 **ACCEPTED ARTICLE**

2 **Monitoring of polycyclic aromatic hydrocarbons in edible vegetable oils**  
3 **consumed in Iran**

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5 **ABSTRACT**

6 Concentrations and profiles of 15 environmental protection agency (EPA) priority polycyclic  
7 aromatic hydrocarbons (PAH) of six different edible oils consumed in Iran markets (olive oil,  
8 sesame oil, coconut oil, sunflower oil, frying oil and blend oil) were studied. The evaluated  
9 edible oils in present study have not previously been analyzed concerning their contents of PAH  
10 compounds. PAHs of 207 edible oil samples were determined and quantified by high-  
11 performance liquid chromatography with spectrofluorometric detector (HPLC/FLD). The  
12 results revealed that the highest content of total PAHs was in coconut oil group (46.8  $\mu\text{g}/\text{kg}$ )  
13 which followed by blend oil (22.48  $\mu\text{g}/\text{kg}$ ), frying oil (20.67  $\mu\text{g}/\text{kg}$ ), sesame oil (19.92  $\mu\text{g}/\text{kg}$ ),  
14 olive oil (18.4  $\mu\text{g}/\text{kg}$ ) and sunflower oil (17.6  $\mu\text{g}/\text{kg}$ ). The light PAHs (Naphthalene,  
15 Acenaphtene, Phenantherern, Antrathene and Fluorene) had the highest portion of PAHs  
16 concentration. Benzo[a]pyrene and PAH4 contents (Benz[a]anthracene + Chrysene +  
17 Benzo[b]fluoranthene + Benzo[a]pyrene) were ND-1.32  $\mu\text{g}/\text{kg}$  and 0.14-9.2  $\mu\text{g}/\text{kg}$ ,  
18 respectively; coconut oil had the highest content. In general, the highest values of  
19 Benzo[a]pyrene and PAH4 were not higher than maximum allowable values of 2 and 10 in any  
20 sample, respectively. However, due to the significant content of total PAHs in some vegetable  
21 oils, such as coconut oil, it is necessary to determine the limits and evaluate it in the national  
22 standard and regulations of the country.

23 **KEYWORDS:** Edible oil; PAH; HPLC/FLD method; Iranian market.

24 **Introduction**

25 As a lipophilic organic compound, PAHs (polycyclic aromatic hydrocarbons) contain several  
26 fused aromatic rings; structurally, there are two types of PAHs: A) group of PAHs known as  
27 low molecular weight (LMW) PAHs containing 2 to 3 benzene rings. This group include  
28  
29

30 naphthalene, acenaphthalene, acenaphthylene, fluorene, phenanthrene, anthracene,  
31 fluoranthene, pyrene, benz[a]anthracene, and chrysene. B) There are a number of heavy PAHs  
32 containing 3 or more rings, such as benzo[b]fluoranthene, benzo[k]fluoranthene,  
33 benzo[a]pyrene, benzo[ghi]perylene, indeno[1,2,3-cd]pyrene, and dibenz[a,h]anthracene  
34 (Heshmati *et al.*, 2018). While LMW PAHs are very toxic and not carcinogenic, the heavy  
35 PAHs are less toxic and highly carcinogenic. PAH compounds due to their stability, resistance  
36 and cumulative properties, remain unchanged in the environment or in the body of living  
37 organisms for a very long time and have caused concern in human society. It has been  
38 determined by the Environmental Protection Agency (EPA) that naphthalene, acenaphthene,  
39 acenaphthylene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[a]anthracene,  
40 chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, benzo[ghi]perylene,  
41 indeno[1,2,3-cd]pyrene, and dibenz[a,h]anthracene are listed among the most important  
42 carcinogenic compounds of PAH (US EPA 1984).

43 Contamination by PAH occurs in three ways: respiration, skin contact, and nutrition. The most  
44 important one is through contaminated water and foods. The most vital food sources for the  
45 entry of PAHs into the body are oils and vegetable fats followed by dried fruits, meat products  
46 and smoked fish (Hao *et al.*, 2016). Environmental pollution which contaminates soil and water  
47 as well as technological processing (heating and drying), contact with mineral oils, and  
48 contaminated packaging leave PAH compounds in food, particularly vegetable oils and fats.  
49 Drying and heating of oily seed and fruits are the most important sources of pollution;  
50 depending on temperature and heating time, distance from heat source, type of processing, fuel  
51 type, and fat content in the seed and fruits, the amount of PAH produced in the vegetable oils  
52 could vary significantly. They are also contaminated by PAHs during solvent extraction  
53 process. Adequate oil refining process (neutralization, neutralization and deodorization) under  
54 standard conditions reduce PAHs content to 2  $\mu\text{g}/\text{kg}$  (Sánchez-Arévalo *et al.*, 2020). As noted  
55 earlier, PAHs content of fats and oils is carcinogenically important (Bertoz *et al.*, 2021;  
56 Iwegbue *et al.*, 2020; Hao *et al.*, 2016). Benzo[a]pyrene and PAH4 (benzo[a]pyrene,  
57 benzo[a]anthracene, benzo[b]fluoranthene, and chrysene) considered as the best indicators of  
58 PAHs in food and edible oils by the European Food Safety Authority (EFSA) and the EU  
59 Council Regulation (EU) No. 835/2011 of 19 August 2011 (EFSA 2012; EU 2011a, Singh and  
60 Agarwal., 2018). Furthermore, according to the Institute of Standard and Industrial Research of  
61 Iran, the allowable values of B[a]P and PAH4 in edible oils are 2  $\mu\text{g}/\text{kg}$  and 10  $\mu\text{g}/\text{kg}$ ,  
62 respectively.

63 In Iran, vegetables, crops and fruits are grown in plenty, mainly in the four seasons. In recent  
64 years, some oil-related crops, such as colza, sesame, olive and etc., are commonly cultivated  
65 but not sufficient for our population. In this context, Iran imports more than 80 percent of edible  
66 oil and fats from other countries, refining and packing them locally under a different brand.  
67 Since, there is no comprehensive research about the content of benzo[a]pyrene and heavy PAH4  
68 (as chemical contaminants) of edible fats and oils in Iran, this study aimed to evaluate these  
69 compounds in consumed edible oil and fats by the Iranian population. Although, a  
70 comprehensive research to estimate the total household oil consumption has not been  
71 conducted.

72

## 73 **Materials and Methods**

### 74 **Reagent and Chemicals**

75 Sigma Aldrich (Bellefonte, PA) provided a standard mixture of 16 EPA PAHs (PAH-mix  
76 4S8743), contained naphthalene, acenaphthene, acenaphthylene, anthracene, fluoranthene,  
77 fluorine, phenanthrene, pyrene, benz[a]anthracene (B[ $\alpha$ ]A), benzo[b]fluoranthene (B[b]F),  
78 benzo[k]fluoranthene, benzo[ghi]perylene, benzo[a]pyrene (B[ $\alpha$ ]P), chrysene(CHR),  
79 dibenz[a,h]anthracene, and indeno[1,2,3-cd]pyrene (10 ng/ $\mu$ L in acetonitrile). Merck  
80 (Darmstadt, Germany) provided high-purity acetone, acetonitrile, dichloromethane, hexane,  
81 methanol, and toluene for HPLC analysis. Deionized water was purified using the Milli-Q  
82 system (Millipore, Billerica, MA, USA). Sep-Pak C<sub>18</sub> cartridges were provided by Waters,  
83 Ireland, while Chromabond cartridges were provided by Machery-Nagel, Germany.

84

### 85 **Instruments**

86 A YL 9100 HPLC system was used for the evaluation of samples and standard solutions. This  
87 system included a vacuum degasser YL 9101, a quaternary pump YL 9110, a column  
88 compartment YL 9130, and a fluorescence detector FP-2020 plus running YL clarity software  
89 (Young Lin, Korea). Agilent Technologies, USA; 150 mm by 4.6 mm by 5 m ZORBAX Eclipse  
90 column and C<sub>18</sub> guard column (10 mm by 2.1 mm). Additionally, ultrasonic baths (Elma,  
91 Germany) and tabletop centrifuges (Dynamca, United Kingdom) were used at present work  
92 (VelpScientifica, Italy, and Dynamca, United Kingdom). Vacuum manifolds were used to  
93 prepare, filter, and elute SPE samples (CNW, China). In terms of polymer phase cartridges,  
94 Sep-Pak C<sub>18</sub>, 6 mL, 500 mg LN 034031034B was the one used by Waters, Ireland, and  
95 Chromabond, 3 mL, 500 mg from Macherey-Nagel, Germany was used for Florisil-bonded  
96 phase cartridges.

97 Filters made from nylon (0.45  $\mu\text{m}$ ), micropipettes (200-1000  $\mu\text{L}$ ), centrifuge tubes made from  
98 polypropylene (11 mL), injection vials with screw tops (2.0 mL), and septa and inserts made  
99 from butyl rubber with poly tetra fluoro ethylene (PTFE) coating (200  $\mu\text{L}$ ), and 5 mL syringes  
100 were used as disposables. A Millipore system was equipped with PTFE filters having a pore  
101 size of 1  $\mu\text{m}$ , with an i.d. of 25 mm. These filters were manufactured by Bio-Analytix, Gdansk,  
102 Poland.

### 103 104 **Calibration Standards**

105 For this study, 200 and 50  $\mu\text{g/L}$  in acetonitrile stock and standard solutions of PAHs were  
106 made. The calibration curve standard solution was used to make eight standard solutions of  
107 PAHs in acetonitrile. The solutions were stored in dark place at 4°C. Peaks of calibration curves  
108 represent function of standard PAHs concentration.

### 109 110 **Extraction and Sampling Procedure**

111 The procedure proposed by ISO 15753 (2016) was followed. The polycyclic aromatic  
112 hydrocarbons were extracted with ultrasound-assisted solvent extraction (acetonitrile/acetone  
113 mixture); finally, purified by using reverse-phase C<sub>18</sub> and Florisil-bonded phase cartridges.

### 114 115 **HPLC-FLD Analysis**

116 YL Chromatographic analyses of samples and standard solutions were conducted using an  
117 HPLC-9100 system equipped with a HPLC-FLD fluorescence detector. 30°C was the  
118 isothermal temperature of the column. A volume of 20  $\mu\text{L}$  was injected.

119 According to gradient method, the conditions were: 1.2 mL/min flow rate, acetonitrile (A) as  
120 the mobile phase, and 50/50 acetonitrile/water concentration (B). In order to determine PAHs  
121 by fluorescence detector (FL), the following excitation and emission wavelengths were used  
122 (Ex/Em):

123 270/324 nm (NPH, ACE, FL) at baseline, 248/375 nm (PHE, ANT) at 12.8 min, 280/462 nm  
124 (FT) at 16.8 min, 270/385 nm (PYR, B[a]A, CHR) at 18.1 min, 256/446 nm (B[b]F) at 28 min,  
125 292/410 nm (B[k]F, B[a]P, D[ah]A, B[ghi]P) at 31.2 min, and 270/470 nm (IP) at 38 min (ISO  
126 15753 2016).

### 127 128 **Technique validation**

129 The used technique was validated by ISO 15753 (2016). Repeatability and recovery were  
130 evaluated by spiking blank oil samples with 15 PAHs (1  $\mu\text{g/kg}$ ) and 15 PAHs (5  $\mu\text{g/kg}$ ). Using

131 the same conditions, five analyses were performed on the same day to evaluate reproducibility.  
132 Linearity ( $R^2$ ) was calculated for all 15 PAHs by eight concentrations of 1-200  $\mu\text{g}/\text{kg}$  PAHs  
133 spiked to blank samples. For calculating LOD and LOQ, we multiplied the standard deviation  
134 and mean of the fortified blank samples ( $n=10$ ) by 3.3 and 10 times, respectively, as well as the  
135 slope of the calibration curve.

## 136 137 **Samples**

138 The sampling of edible oils and fats was done according to ISO 5555 (2001) and samples  
139 included as follows: 51 olive oil samples (virgin and refined), 27 sesame oil samples, 45 frying  
140 oil samples, 45 sunflower oil samples, 12 coconut oil samples, and 27 blend oil samples. In  
141 order to conduct the analysis, oil samples (207) were purchased from local supermarkets and  
142 stored at room temperature for further assessment. ,

## 143 144 **Statistical analysis**

145 The experiments were randomly designed. Average of three separate experiments was  
146 reported as result. Statistical analysis was performed using SPSS software (version 22; SPSS  
147 Inc., Chicago, IL, USA); one-way analysis of variance (ANOVA) and Duncan's multiple range  
148 test ( $p\text{-value}<0.05$ ) were used to calculate significance of differences between mean values.

## 149 150 **Results and Discussion**

### 151 **Validation**

152 In the linearity, limits of detection, limits of quantification, and recovery tests were performed  
153 to determine whether the technique was analytically controlled or not. External standard  
154 calibration was used (HPLC/FLD technique) to determine analyte values by eight calibration  
155 solutions containing 1-200  $\mu\text{g}/\text{kg}$  PAHs. According to Table 1, the lowest correlation  
156 coefficient ( $R^2$ ) were related to Naphthalene (0.9851), Acenaphthene (0.9885), and Fluorene  
157 (0.9887), belonging to light PAH. Table 1 illustrated the standard linearity supported by  
158 regression data. It was observed that there was a linear relationship with a satisfactory linear  
159 coefficient in all PAHs ( $R^2>0.9851$ ). LOD ranged from 0.09 to 0.45  $\mu\text{g}/\text{kg}$  and LOQ ranged  
160 from 0.27 to 1.35  $\mu\text{g}/\text{kg}$ . The recoveries varied from 80 to 110%. According to results obtained  
161 by other authors, the limit of quantitation and limit of detection were ranged between 0.6–1 and  
162 0.2–0.3  $\mu\text{g}/\text{kg}$ , respectively. The average recovery was 58.6–90.6% (Liu et al., 2023). Lee et al.  
163 (2019) evaluated the occurrence and risk characterization of polycyclic aromatic hydrocarbons  
164 of edible oils. They also indicated that the LOQ and LOD ranged from 0.06 to 0.44  $\mu\text{g}/\text{kg}$  and

165 0.02 to 0.13  $\mu\text{g}/\text{kg}$  at four types of oil samples, respectively. The relative recoveries of PAH4  
166 were from 79.9 to 112.6% at 10  $\mu\text{g}/\text{kg}$  and from 70.7 to 110.4% at 2  $\mu\text{g}/\text{kg}$  (Lee et al., 2019)

## 167 168 **Total PAHs**

169 The values of some PAHs (naphthalene, acenaphthene, fluorene, phenanthrene, anthracene,  
170 fluoranthene, pyrene, benzo[k]fluoranthene, dibenz[a,h]anthracene, benzo[g,h,i]perylene,  
171 indeno[cd]pyrene) and total PAHs are presented in table 2. The chromatograms of PAHs along  
172 with their retention times in standard and edible oil samples are shown in Fig. 1. According to  
173 Table 2, the concentration of naphthalene in all different edible oils, except for sesame oil and  
174 frying oil, was the highest in comparison to other PAHs. Coconut oil had the highest content of  
175 naphthalene ( $12.97 \pm 5.01 \mu\text{g}/\text{kg}$ ) which was followed by Acenaphthene and fluorene. The sum  
176 of 15 PAHs and PAH4 were  $46.81 \mu\text{g}/\text{kg}$  and  $4.40 \mu\text{g}/\text{kg}$  respectively, indicating that such high  
177 content of PAHs could be referred to low quality of imported coconut oil in Iran. The German  
178 Society for Fat Science (DGF) suggests that PAHs content in edible oils should not exceed from  
179 25  $\mu\text{g}/\text{kg}$ ; FEDIOL (Fédération de l'Industrie d'Huilerie de la Communauté Européenne) also  
180 recommend allowable content of 25  $\mu\text{g}/\text{kg}$  for PAHs in edible fats and oils. Clearly,  
181 concentration of coconut oil was higher than the commended values (EUR-Lex 1989). The 15  
182 PAHs for blended oil, frying oil, sunflower oil, sesame oil, and olive oil, were 22.48  $\mu\text{g}/\text{kg}$ ,  
183 20.19  $\mu\text{g}/\text{kg}$ , 17.60  $\mu\text{g}/\text{kg}$ , 19.92  $\mu\text{g}/\text{kg}$ , and 18.40  $\mu\text{g}/\text{kg}$ , respectively, indicating that other oils  
184 have no more than the specified range except coconut oil. Moreover, other European countries  
185 (Spain, Italy, Portugal and Greece) recommend maximum value of 2  $\mu\text{g}/\text{kg}$  for each individual  
186 PAH and 5  $\mu\text{g}/\text{kg}$  for sum of heavy PAHs listed as follows: benzo[a]anthracene,  
187 benzo[e]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene,  
188 dibenz[a,h]anthracene, benzo[g,h,i]perylene, indeno[1,2,3-c,d]pyrene. In this regard, which  
189 will be discussed below, the concentrations for other PAHs such as benzo[k]fluoranthene,  
190 dibenz[a,h]anthracene, benzo[g,h,i]perylenes and indeno[cd]pyrenes were lower than 2  $\mu\text{g}/\text{kg}$   
191 except for PAH4. Ma et al. (2021) assessed the levels of the 15-priority PAHs in the edible  
192 vegetable oil (canola oil, olive oil, sunflower oil, and corn oil) collected from Egypt. The  
193 authors indicated presence of 15-priority PAHs in all examined oil samples. The highest  
194 residual concentrations of PAHs were reported in Canola oil. Heat treatment of canola oil  
195 resulted in a drastic increase in the formed B[a]P (316.55%), total 4-PAHs (297.42%), total 2-  
196 PAHs (322.47%), total 15-PAHs (443.32%), and total 8-PAHs (285.26%).



197 According to the result of total PAHs regarding to light PAHs, naphthalene had the highest  
198 concentration in all of the different edible oils (specially in coconut oil:  $12.97 \pm 5.01 \mu\text{g}/\text{kg}$ ),  
199 except in sesame oil. Since International Agency for Research on Cancer classified naphthalene  
200 as one of the most important carcinogenic compounds for humans and animals (Group 2B),  
201 edible oils were required to be monitored (Bempelou *et al.*, 2019).

202 As mentioned above, since Iran imports most of the vegetable oil for edible purposes, its  
203 detailed control and monitoring is very important (particularly in coconut oil). The content of  
204 total PAHs in different edible oils varied from 17.6 to  $46.81 \mu\text{g}/\text{kg}$ . The lowest concentration  
205 was belonged to sunflower oil and the highest be related to coconut oil. The higher  
206 concentration of PAH in coconut oil in comparison with other edible oils are also reported by  
207 (Zachara *et al.*, 2017; Silva *et al.*, 2017). The authors announced that higher PAH content of  
208 coconut oil be related to environmental contaminations (water, air and soil) and contamination  
209 during the drying process. Hence, observed high content of PAHs could be referred to low  
210 quality of imported coconut oil in Iran. These findings in relation with the minimum  
211 concentration of total PAHs are in paralleled with the results reported in a similar study  
212 performed in China that developed a method to determine PAHs content of sesame oil, peanut  
213 oil, soybean oil, rapeseed oil, and virgin olive oil, and reported that total PAHs content of  
214 vegetable oils varied from 18.00 to  $639.96 \mu\text{g}/\text{kg}$  (Wang *et al.*, 2014).

215 The PAHs content of different vegetable oils are shown in Table 3. The results of present  
216 study were relatively lower than results reported by other authors (Alomirah *et al.*, 2010; Gharbi  
217 *et al.*, 2017; Iwegbue *et al.*, 2020; Ju *et al.*, 2020; Krajian *et al.*, 2016; Lee *et al.*, 2019; Rascón  
218 *et al.*, 2018; Stenerson *et al.*, 2015; Taghvaei *et al.*, 2015; Zachara *et al.*, 2017). In particular,  
219 Alomirah *et al.* (2010) reported that sum of 16 PAHs content of sunflower oil and olive oil  
220 ranged from 0.42 to  $41.30 \mu\text{g}/\text{kg}$  and 1.09 to  $182.22 \mu\text{g}/\text{kg}$ , respectively. However, the content  
221 of corn and canola oils ranged from 0.30 to 34.49 and 10.29 to  $12.23 \mu\text{g}/\text{kg}$ , respectively, which  
222 are in agreement with the present study. On the contrary, Molle *et al.* (2017) reported that PAHs  
223 content of 69 samples was up to  $13.11 \mu\text{g}/\text{kg}$ . In particular, they reported 13 PAHs content of  
224 canola oil, sunflower oil and corn oil (up to  $31.70 \mu\text{g}/\text{kg}$ ,  $0.65\text{-}17.88 \mu\text{g}/\text{kg}$ ,  $2.61\text{-}38.23 \mu\text{g}/\text{kg}$ ,  
225 respectively). Among PAHs analyzed, B[ $\alpha$ ]P, CHR, B[b]F and B[ $\alpha$ ]A were the most common  
226 PAHs present in three oils studied (99%, 97%, 97% and 96% of the samples, respectively)  
227 (Molle *et al.*, 2017). Liu *et al.* (2023) evaluated the levels and health risk of PAHs in frying oils  
228 and vegetable oils. The profiles and levels of PAH15 in frying oils (after repeated frying by  
229 restaurants) and vegetable oils were analyzed. The authors revealed that vegetable oils are

230 highly contaminated. They also reported that more than 32.4% of vegetable oils from China  
231 exceeding the EU standard limit of the levels of PAHs. Only 6.5% of the oil samples were under  
232 allowable level for BaP (2 µg/kg). The mean concentrations of PAH4 (10.49 µg/kg) and BaP  
233 (2.16 µg/kg) were marginally above the EU maximum permitted levels in oils (Liu et al., 2023).

234

#### 235 **Effect of PAH type and oil type on total PAHs in edible oils**

236 A factorial experiment was used in a completely randomized design to evaluate the effects of  
237 PAH and oil types on PAHs content. After performing the respective variance analysis, the  
238 effects of oil type, PAH type and their interactions were significant. The results of the Duncan  
239 multi-range comparison test (to determine the effects of oil type) showed that the average total  
240 PAHs were categorized into three classes. The first category included coconut oil with the  
241 highest amount of these compounds (Fig. 2a), the second group included other oils, and the  
242 remaining oils settled in a third group that had no significant difference in terms of total PAHs.  
243 In addition, the graph of the effect of PAH type on the total PAHs increasing is shown in Fig.  
244 2b. As can be seen, light PAH compounds, such as naphthalene, acenaphthene, fluorene, etc.,  
245 had the highest effect on increasing the total PAHs.

246

#### 247 **PAH4**

248 The content of PAH4, as an important indicator of chemical contaminants, was obtained from  
249 total contents of four PAHs, B[α]P, CHR, B[b]F and B[α]A. According to Table 2, no  
250 significant difference was observed in B[α]P content between oil samples. B[α]P content ranged  
251 from ND to 1.32 µg/kg. The average B[α]P content in the coconut oil samples was higher than  
252 observed content for other oils. The content of B[α]P in all of the oil samples was less than 2  
253 ppb. It is noteworthy that B[α]P is the most important PAH due to its carcinogenic properties.  
254 There was a significant difference between CHR contents of oil samples so that edible oils were  
255 classified into three separate groups. The first group consisted of sunflower and blended oils,  
256 which had the lowest amount, the second group included olive, sesame and frying oils, and the  
257 third group was coconut oil, containing the highest amount of chrysene. The samples were  
258 categorized into three groups in terms of the B[b]F amount. Coconut oil and olive oil had the  
259 highest and lowest amounts, respectively. The sunflower oil group was placed in the second  
260 group and other groups did not differ from each other significantly. The content of B[α]A was  
261 different among edible oil samples. In coconut, the oil group had a much higher content than  
262 other ones. The lowest content was related to the sunflower oil group. The minimum and  
263 maximum of these heavy PAHs varied from ND to 5.6 µg/kg.



264 The contents of B[ $\alpha$ ]P, CHR, B[b]F and B[ $\alpha$ ]A were different in edible oil samples. The  
265 maximum values of B[ $\alpha$ ]A (5.6  $\mu\text{g}/\text{kg}$ ) and B[b]F (4.32  $\mu\text{g}/\text{kg}$ ) were higher in comparison to  
266 CHR and B[ $\alpha$ ]P that were 1.91  $\mu\text{g}/\text{kg}$  and 1.32  $\mu\text{g}/\text{kg}$ , respectively. The diversity of edible oils  
267 and their various processes of production are the most important reasons for the different values  
268 of these PAHs. The processing steps, such as drying and extraction, are various for different  
269 kinds of oil seeds and fruits. On the other hand, collected samples belong to different  
270 manufacturers, whose technology and their refining methods are likely to be different. This part  
271 of our research are in paralleled with the results of [Amzad-Hossain and Salehuddin \(2012\)](#) who  
272 reported different results for different vegetable oils.

273 These results are in agreement with other studies. Thus, [Molle \*et al.\* \(2017\)](#) reported that  
274 PAH4 content of canola was up to 22.15  $\mu\text{g}/\text{kg}$ , 15.61  $\mu\text{g}/\text{kg}$  in sunflower, and 30.98  $\mu\text{g}/\text{kg}$  in  
275 corn; whereas, [Ingenbleek \*et al.\* \(2019\)](#) reported that PAH4 contained 77% of 13 genotoxic and  
276 carcinogenic PAHs. They also reported that PAH4 content of edible oils (including palm oil  
277 and peanut oil) was higher than maximum 10  $\mu\text{g}/\text{kg}$  in 50% of the cases (12.0  $\mu\text{g}/\text{kg}$  on average  
278 up to 60.6  $\mu\text{g}/\text{kg}$ ).

279 The maximum mean value of PAH4 was related to the coconut oil group. This issue might be  
280 due to the source of oil, coconut as a fruit that has much higher moisture than seed oils. This  
281 difference had led to the application of different drying methods involving different duration  
282 times, which finally resulted in higher amounts of PAH4 in the coconut oil. The results of this  
283 section were in agreement with [Moret and Conte \(2000\)](#) results.

284 In one sample of sesame oil that was produced by the cold pressing method, the amount of  
285 B[ $\alpha$ ]A (5.60  $\mu\text{g}/\text{kg}$ ) and the sum of PAH4 (9.20  $\mu\text{g}/\text{kg}$ ) were higher than observed for other  
286 oils. The reason for this result might be related to the production method in which the refining  
287 process was not used. This result confirms the finding of [Wang \*et al.\* \(2014\)](#), who reported high  
288 PAHs content of sesame oil as a result of high temperature of sesame roasting.

289 It is important to keep in mind that the refining process, especially the bleaching with activated  
290 carbon and deodorization, can be an efficient technique in removing PAHs. The results of other  
291 researches carried out on virgin and cold-pressed oils also confirm this results ([Aliyar-Zanjani  
292 \*et al.\*, 2019](#)). However, the results of this study showed that despite Iran imports more than 90%  
293 of its crude oil from other countries, owing to the refining processes, the amount of B[ $\alpha$ ]P is  
294 less than 2  $\mu\text{g}/\text{kg}$  and the sum of the four PAHs is less than 10  $\mu\text{g}/\text{kg}$  as well.

295 It should be noted that the concentrations of PAH4 reported in this study were relatively lower  
296 than those reported in previous studies ([Niu \*et al.\*, 2021](#); [Yousefi \*et al.\*, 2018](#)). The soybean oil

297 purchased from China showed the distribution of B[ $\alpha$ ]P content ranged from 0.50 to 10.95  
298  $\mu\text{g}/\text{kg}$  (mean 6.26  $\mu\text{g}/\text{kg}$ ), from 0.53 to 11.07  $\mu\text{g}/\text{kg}$  (mean 6.96  $\mu\text{g}/\text{kg}$ ) in peanut oil, and from  
299 0.53 to 9.88  $\mu\text{g}/\text{kg}$  (mean 5.95  $\mu\text{g}/\text{kg}$ ) in colza oil (Niu *et al.*, 2021). Also, Yousefi *et al.*, (2018)  
300 analyzed 40 samples of different edible oils available in Iran (frying oil, blended oil, sunflower  
301 oil, corn oil and canola oil) and reported 0.90 to 11.33  $\mu\text{g}/\text{kg}$  for B[ $\alpha$ ]P, 3.51 to 84.03  $\mu\text{g}/\text{kg}$  for  
302 PAH4, and 129.28 to 19.54  $\mu\text{g}/\text{kg}$  for PAH13. The sesame oils and perilla oils were highly  
303 contaminated with PAHs (Lee *et al.*, 2019). A maximum limit value of 2  $\mu\text{g}/\text{kg}$  the perilla oils  
304 and sesame oils were highly contaminated with PAHs. A maximum limit value of 2  $\mu\text{g}/\text{kg}$  for  
305 BaP was established in edible oils in Korea and EU. The mean concentration of PAHs in 129  
306 sesame oil samples analyzed was 0.18  $\mu\text{g}/\text{kg}$  for BaP, 0.35  $\mu\text{g}/\text{kg}$  for BbF, 0.41  $\mu\text{g}/\text{kg}$  for CHR,  
307 0.41  $\mu\text{g}/\text{kg}$  for BaA, and 1.35  $\mu\text{g}/\text{kg}$  for the sum of 4 PAHs (Lee *et al.*, 2019).

308 Light PAHs account for 65% of all PAHs, while the remaining 35% are heavy PAHs. B[ $\alpha$ ]P  
309 content of 12 samples, according to the Standard Organization of Iran and the European Union,  
310 was higher than the standard value (2  $\mu\text{g}/\text{kg}$ ), whereas PAH4 content of 15 samples was higher  
311 than maximum 10  $\mu\text{g}/\text{kg}$ .

312

### 313 **Effect of PAH type and oil type on PAH4 content in edible oils**

314 A factorial experiment was used by a completely randomized design to evaluate the effects of  
315 PAH compounds and oils on PAH4 content. The results of the Duncan multi-range comparison  
316 test showed that the mean of the PAH4 content in coconut oil differs from other oils (Fig. 3a).  
317 Moreover, the diagram of the effects of four PAH compounds on PAH4 content in coconut oil  
318 are shown in Fig. 3b. As can be seen, Benzo[b]fluoranthene and Chrysene had a significant  
319 effect.

320 Statistically significant differences were observed between different types of oil as well as  
321 different brands. These differences can be attributed to the environmental pollutions; for  
322 example, PAHs content may vary in crops of different regions; moreover, different variables  
323 may be involved in the drying process and oil production/refining processes (Molle *et al.*,  
324 2017). Wen-ting Yin *et al.* (2022) evaluated the effects of microwave pretreatment of sunflower  
325 kernels on the aroma-active composition, tocopherols, lipid oxidation, sensory quality,  
326 heterocyclic amines and polycyclic aromatic hydrocarbons of sunflower oil. The authors  
327 reported that the temperature rise by microwaves was responsible for the gradually increased  
328 PAHs (7.78–109.76  $\mu\text{g}/\text{kg}$ ) and HCAs (7.3–820.6  $\mu\text{g}/\text{kg}$ ) in oils.

329

## 330 **Conclusions**

331 In this study, PAHs as important chemical contaminants were deeply evaluated in edible oils  
332 available in Iran. According to the obtained results, the content of PAH4 was different among  
333 edible oils of this study with a mean PAH4 values of 1.57  $\mu\text{g}/\text{kg}$ . The coconut oil group  
334 exhibited the highest average content (4.40  $\mu\text{g}/\text{kg}$ ) among different kinds of oil and sesame oil  
335 presented the highest value of the sum of PAH4 (1.78  $\mu\text{g}/\text{kg}$ ). According to outcomes of present  
336 work, none of the oil samples exhibited B[a]P and sum of PAH4 contents higher than 2  $\mu\text{g}/\text{kg}$   
337 and 10  $\mu\text{g}/\text{kg}$ , respectively; whereas, 16PAHs content varied significantly. The use of active  
338 carbon in purification processes is therefore recommended, because it efficiently reduces PAHs  
339 content of oils. Moreover, total PAHs factor requires further evaluation and adjustment of  
340 allowable limits.

341

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345

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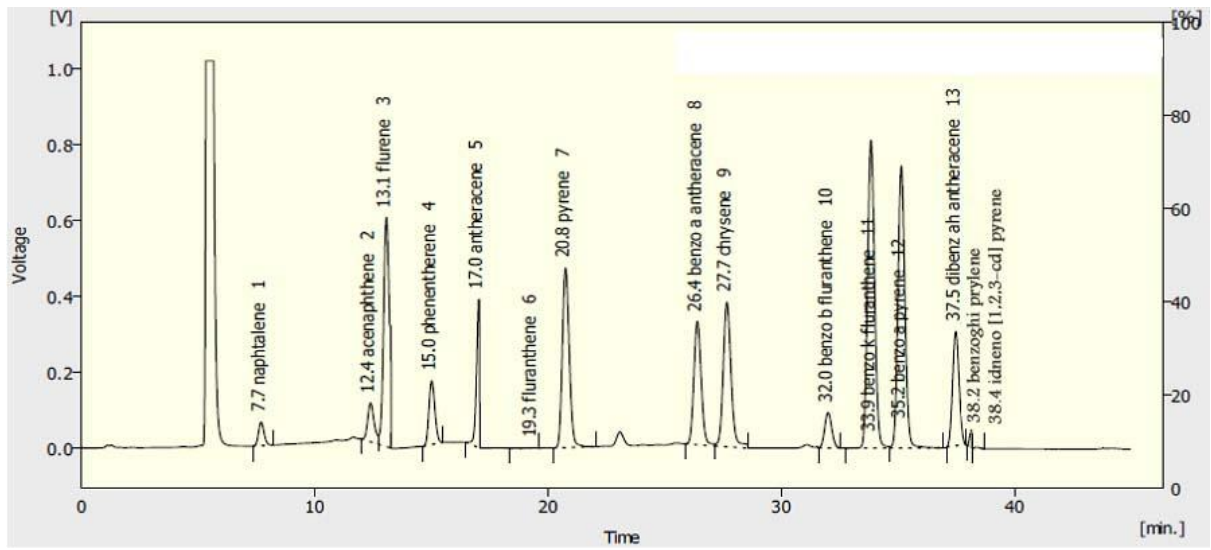
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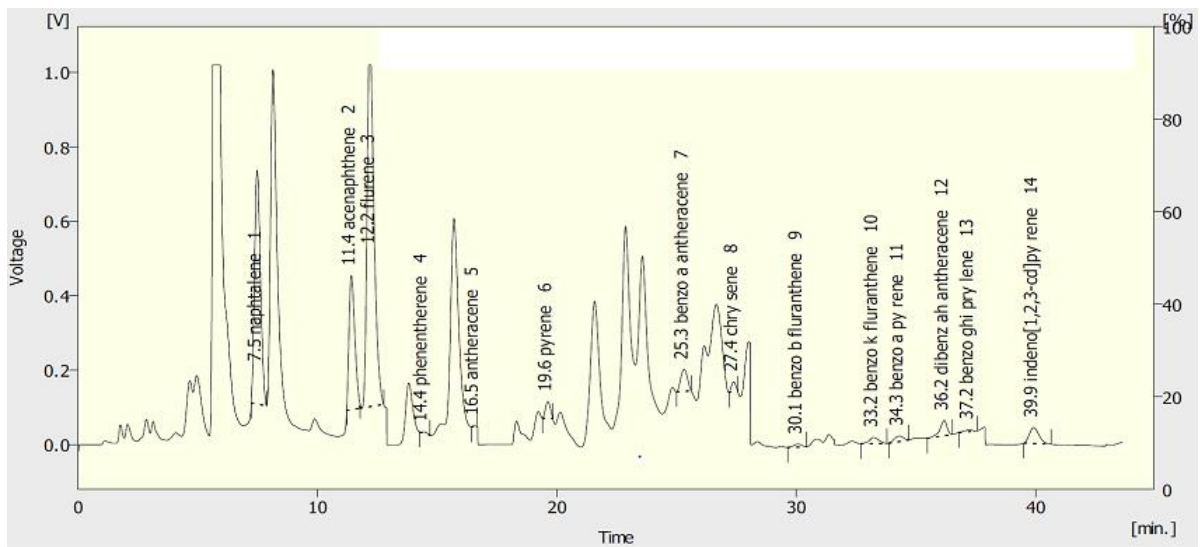
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461 **Figures 1.** Obtained Chromatograms from HPLC/FLD method. (a) Pure standard solution  
462 (15 PAHs: 2 µg/kg); (b) An olive sample.

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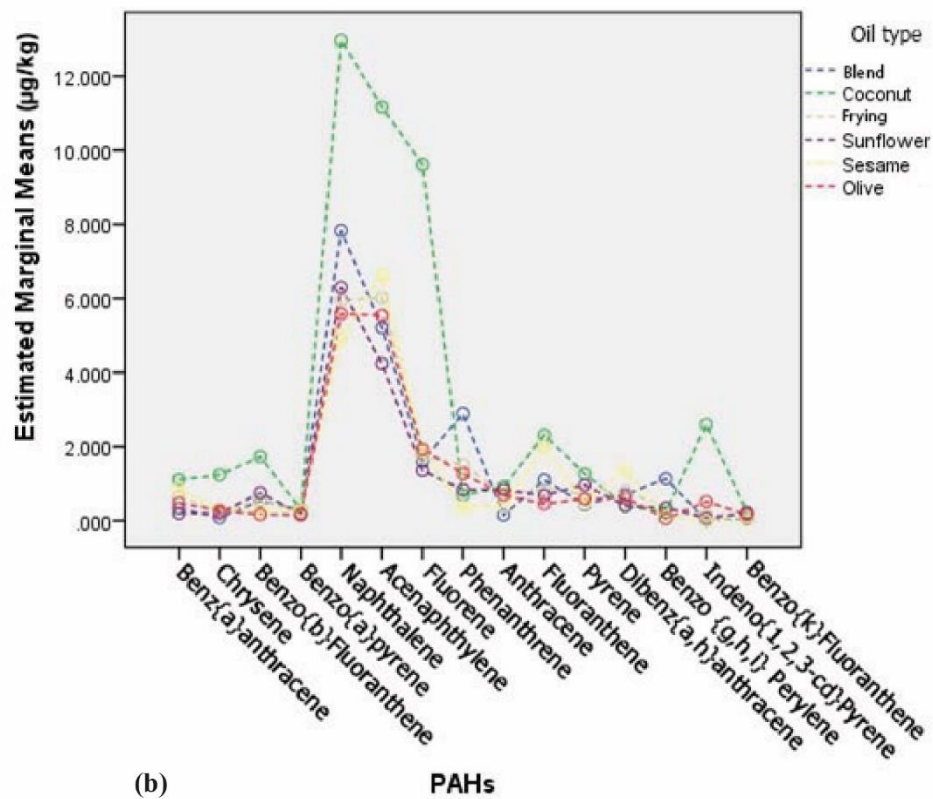
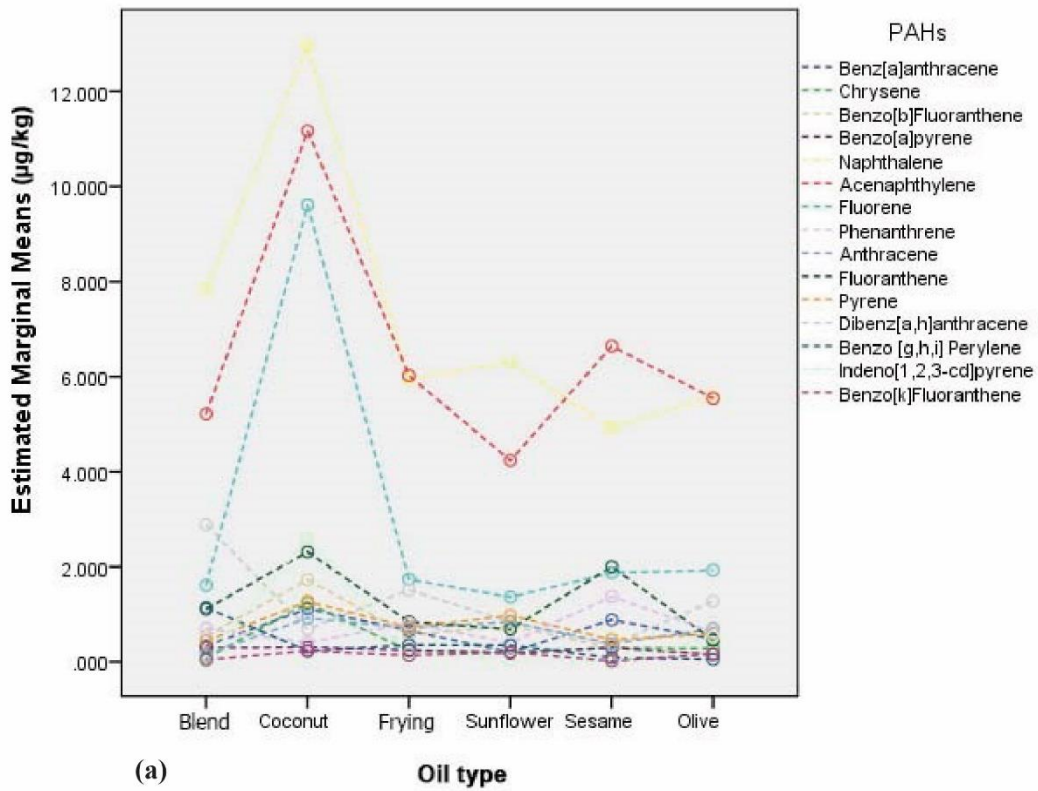
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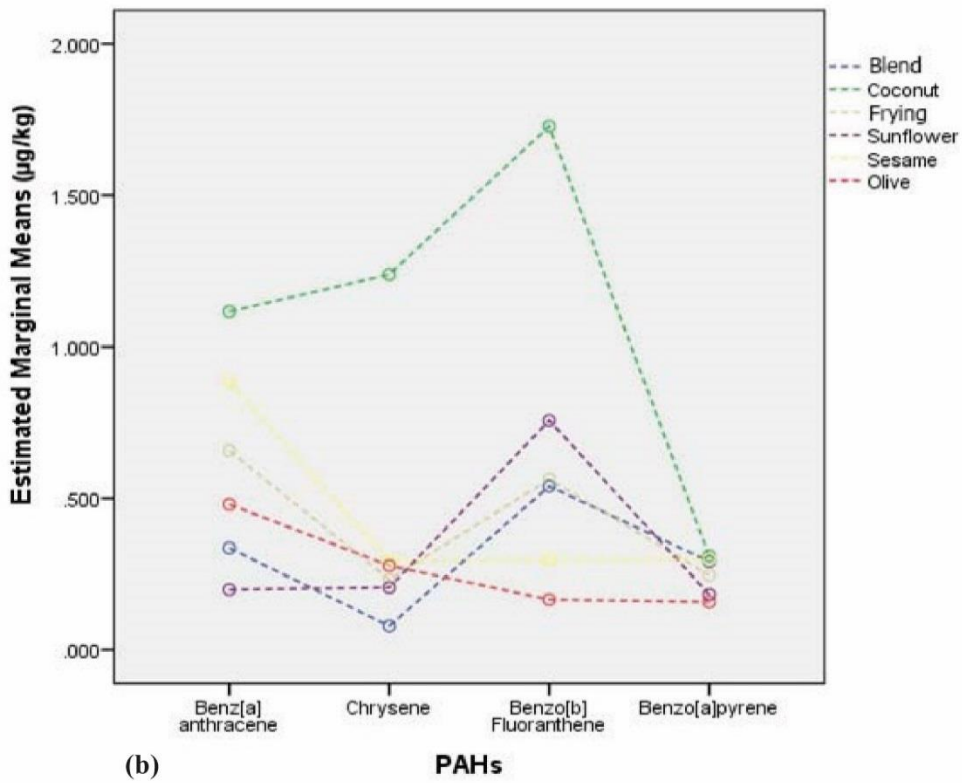
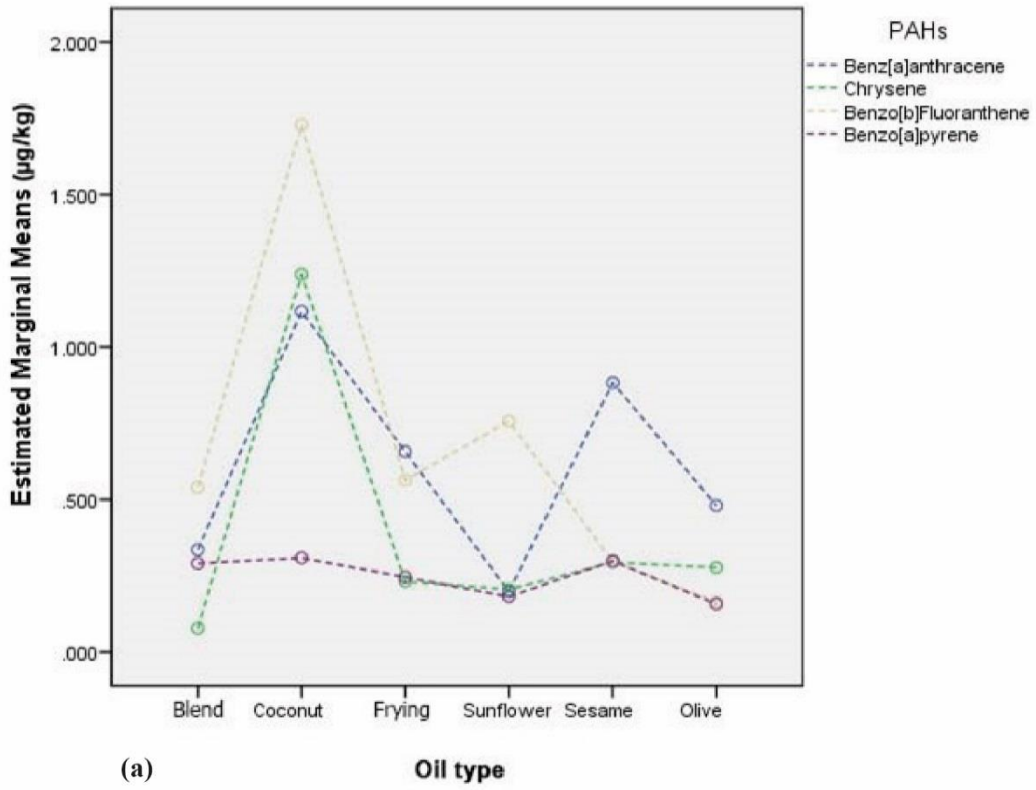
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472 **Figure 2.** The interaction of (a) edible oil type and (b) PAHs type on the average estimate of  
 473 total PAHs.



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**Figure 3.** The interaction of (a) edible oil type and (b) PAHs type on the average estimate of PAH4.

477 **Table 1.** Linear range, correlation coefficient ( $R^2$ ), limit of quantitation (LOQ), limit of  
 478 detection (LOD) and recoveries of PAHs.

Compound	Linear range		$R^2$	LOD ( $\mu\text{g}/\text{kg}$ )	LOQ ( $\mu\text{g}/\text{kg}$ )	Recovery (%)	
	( $\mu\text{g}/\text{kg}$ )					1 $\mu\text{g}/\text{kg}$	5 $\mu\text{g}/\text{kg}$
Naphthalene	1 - 200		0.9851	0.09	0.27	80	83
Acenaphthene	1 - 200		0.9885	0.11	0.33	84	87
Fluorene	1 - 200		0.9887	0.20	0.60	83	96
Anthracene	1 - 200		0.9954	0.18	0.54	87	92
Phenanthrene	1 - 200		0.9981	0.19	0.57	89	101
Fluoranthene	1 - 200		0.9978	0.10	0.30	90	97
Pyrene	1 - 200		0.9993	0.15	0.45	93	95
Benz[a]anthracene	1 - 200		0.9983	0.20	0.60	89	96
Chrysene	1 - 200		0.9989	0.18	0.54	92	105
Benzo[b]fluoranthene	1 - 200		0.9979	0.18	0.54	93	97
Benzo[k]fluoranthene	1 - 200		0.9984	0.20	0.60	91	94
Benzo[a]pyrene	1 - 200		0.9988	0.16	0.36	95	98
Dibenz[a, h]anthracene	1 - 200		0.9985	0.24	0.72	104	106
Benzo[g,h,I]perylene	1 - 200		0.9978	0.26	0.78	106	110
Indeno[c,d]pyrene	2 - 200		0.9975	0.45	1.35	91	94

479

480 **Table 2.** Contents of different contaminants ( $\mu\text{g}/\text{kg}$ ) in different types of edible oil.

Edible oils	Blend		Coconut		Frying		Sunflower		Sesame		Olive	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
<b>PAHs</b>												
<b>Naphthalene</b>	7.83 <sup>b</sup>	1.90	12.97 <sup>c</sup>	5.01	5.95 <sup>ab</sup>	3.72	6.30 <sup>ab</sup>	2.71	4.92 <sup>a</sup>	4.94	5.59 <sup>ab</sup>	4.91
<b>Acenaphthene</b>	5.21 <sup>ab</sup>	3.27	11.17 <sup>c</sup>	3.78	6.02 <sup>ab</sup>	2.57	4.23 <sup>a</sup>	3.53	6.64 <sup>ab</sup>	2.40	5.54 <sup>ab</sup>	4.82
<b>Fluorene</b>	1.60 <sup>a</sup>	1.78	9.61 <sup>b</sup>	3.50	1.73 <sup>a</sup>	2.29	1.36 <sup>a</sup>	1.92	1.87 <sup>a</sup>	1.81	1.92 <sup>a</sup>	2.38
<b>Phenanthrene</b>	2.89 <sup>a</sup>	4.50	0.69 <sup>a</sup>	.58	1.51 <sup>a</sup>	1.43	0.84 <sup>a</sup>	0.90	0.36 <sup>a</sup>	0.40	1.28 <sup>a</sup>	1.47
<b>Anthracene</b>	0.15 <sup>a</sup>	0.15	0.92 <sup>b</sup>	1.43	0.72 <sup>ab</sup>	0.60	0.84 <sup>b</sup>	1.77	0.38 <sup>ab</sup>	0.36	0.69 <sup>ab</sup>	0.82
<b>Fluoranthene</b>	1.12 <sup>a</sup>	0.26	2.31 <sup>b</sup>	0.93	0.84 <sup>a</sup>	0.17	0.69 <sup>a</sup>	0.13	2.00 <sup>b</sup>	0.38	0.45 <sup>a</sup>	0.12
<b>Pyrene</b>	0.51 <sup>a</sup>	0.44	1.64 <sup>b</sup>	1.27	1.25 <sup>ab</sup>	0.73	1.07 <sup>ab</sup>	0.97	0.56 <sup>a</sup>	0.46	0.88 <sup>a</sup>	0.60
<b>Benzo[k]Fluoranthene</b>	0.04 <sup>ab</sup>	0.07	0.23 <sup>c</sup>	0.20	0.14 <sup>abc</sup>	0.40	0.20 <sup>bc</sup>	0.32	0.02 <sup>a</sup>	0.05	0.17 <sup>abc</sup>	0.39
<b>Dibenz[ah]anthracene</b>	0.70 <sup>a</sup>	1.03	0.40 <sup>a</sup>	0.39	0.81 <sup>a</sup>	0.80	0.40 <sup>a</sup>	0.76	1.38 <sup>b</sup>	1.37	0.62 <sup>a</sup>	1.13
<b>Benzo[g,h,I]Perylene</b>	1.14 <sup>b</sup>	1.17	0.22 <sup>a</sup>	0.26	0.35 <sup>a</sup>	0.44	0.34 <sup>a</sup>	0.88	0.09 <sup>a</sup>	0.14	0.05 <sup>a</sup>	0.10
<b>Indeno[cd]pyrene</b>	ND	0.00	2.60 <sup>c</sup>	0.68	0.17 <sup>a</sup>	0.62	0.07 <sup>a</sup>	0.13	ND	0.00	0.52 <sup>b</sup>	0.84
<b>Benzo[a]pyrene</b>	0.29 <sup>a</sup>	0.50	0.31 <sup>a</sup>	0.20	0.25 <sup>a</sup>	0.24	0.18 <sup>a</sup>	0.19	0.30 <sup>a</sup>	0.31	0.16 <sup>a</sup>	0.25
<b>Chrysene</b>	0.08 <sup>a</sup>	0.07	1.24 <sup>c</sup>	0.58	0.23 <sup>b</sup>	0.25	0.21 <sup>ab</sup>	0.16	0.30 <sup>b</sup>	0.22	0.28 <sup>b</sup>	0.23
<b>Benzo[b]fluoranthene</b>	0.54 <sup>ab</sup>	0.98	1.73 <sup>c</sup>	0.61	0.56 <sup>ab</sup>	1.20	0.76 <sup>b</sup>	1.09	0.30 <sup>ab</sup>	0.86	0.17 <sup>a</sup>	0.42
<b>Benz[a]anthracene</b>	0.34 <sup>a</sup>	0.28	1.12 <sup>c</sup>	0.11	0.66 <sup>ab</sup>	0.85	0.20 <sup>a</sup>	0.14	0.88 <sup>bc</sup>	1.68	0.48 <sup>ab</sup>	0.62
<b>PAH4</b>	1.24 <sup>a</sup>	1.62	4.40 <sup>b</sup>	0.81	1.70 <sup>a</sup>	1.61	1.33 <sup>a</sup>	1.09	1.78 <sup>a</sup>	2.70	1.08 <sup>a</sup>	0.68
<b>15PAHs</b>	22.48 <sup>b</sup>	8.84	46.81 <sup>c</sup>	5.67	20.67 <sup>ab</sup>	5.61	17.60 <sup>a</sup>	6.82	19.92 <sup>ab</sup>	6.67	18.40 <sup>ab</sup>	10.62

481 Mean values in row with different superscripts (a, b) are significantly different by Duncan's multiple range test  
 482 at  $p < 0.05$ .

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488 **Table 3.** PAH contents ( $\mu\text{g}/\text{kg}$ ) reported in different vegetable oils from a literature review of  
 489 the last decade.

Oil	Country	Number of samples	EPA 16 PAHs Min–Max (Mean)	PAH4 Min–Max (Mean)	BaP Min–Max (Mean)	References
olive sesame coconut sunflower frying blend	IR	207	1.41–52.25 (21.14)	ND–9.20 1.57	ND–1.32 0.23	This study
olive	TR, SY, IT, ES, PS, TN, LB, CA, SA	21	0.30–182.22 (37.88)	ND	0.06–6.77 (0.53)	Alomirah <i>et al.</i> (2010)
Olive	ES, IT, TR, TN, GR, MA, USA	4	9.9–48.3 (22.5)	ND	<LOQ	Stenerson <i>et al.</i> (2015)
Olive	IR	5	NS (19.05)	NS (1.28)	<LOQ	Taghvaei <i>et al.</i> (2015)
Olive	SY	9	33.4–82.4 (54.8)	0.34–20.2 (7.66)	-	Krajian <i>et al.</i> (2016)
Olive	TN	5	11.4–45.8 (33.7)	0.2–0.6 (0.5)	<LOQ	Gharbi <i>et al.</i> (2017)
rapeseed sunflower olive soybean coconut	PL	3	ND	1.11–3.15 (2.13)	ND–0.25 -	Zachara <i>et al.</i> (2017)
Olive	ES	2	4.42–6.36 (5.4)	0.15–0.32 (0.24)	0.045–0.058 (0.051)	Rascón <i>et al.</i> (2018)
Olive	KR	53	ND	0.42–4.07 (2.05)	ND–1.15 (0.22)	Lee <i>et al.</i> (2019)
Olive	KR	1	ND	2.508	0.481	Ju <i>et al.</i> (2020)
Vegetable oil	NI	6	-	3.97–15.1	0.15–0.80	Iwegbue <i>et al.</i> (2020)

490 ND: not detected, LOD: limit of detection, LOQ: limit of quantification, TR: Turkey, SY: Syria, IT: Italy, ES:  
 491 Spain, PS: Palestine, TN: Tunisia, LB: Lebanon, CA: Canada, SA: Saudi Arabia, GR: Greece, MA: Morocco,  
 492 USA: United States of America, IR: Iran, KR: Korea, PL: Poland, NI: Nigeria.

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