POLYCYCLIC AROMATIC HYDROCARBONS (PAHS) IN COMMERCIAL EDIBLE OILS AND HEALTH RISK ASSESSMENT IN MALAYSIA

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ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) is a group of hazardous organic substance, which also categorized as persistent organic pollutants (POPs) has widely distributed in the atmosphere that able to trigger the environment and human health. In this study, the concentration of PAHs in three different commercial edible oils and brands were evaluated. The liquid-liquid extraction and solid-phase extraction were used for clean-up before being examined by high-performance liquid chromatography coupled with a fluorescence detector. The results showed the sunflower oil contained the highest PAH amount among 27 edible oil samples with the mean value of 4.28 µg/kg. The mean concentrations of the extra virgin olive oil, olive oil, and sunflower oil for benzo(a)pyrene were below 2 µg/kg and complied with the European Union standard. There was no significant difference for PAH concentrations in a different types of edible oils except chrysene as well as in different brands of edible oils. The estimation of health risk assessment for PAHs was carried out using the incremental lifetime cancer risk (ILCR). The calculation revealed the ILCR value were less than 1.0×10^{-6} for all the children and both male and female adults. These values pointed out a low potential risk of carcinogenic to the populations exposed through consumption. The implementation and maintenance of regular monitoring from time to time on the concentration of PAHs in edible oils is necessary due to the lower probability of risk of cancer in order to increase food security.

Keywords: edible oil, PAHs, liquid-liquid extraction, HPLC, health risk assessment

1.0 INTRODUCTION

Polycyclic aromatic hydrocarbons, shorts as PAHs are a group of organic compounds that are typically colorless to white, or pale yellow in solids. From the chemical point of view, the structure of PAHs comprised of many aromatic rings of carbon and hydrogen atoms fused in linear, angular, and cluster arrangement, which ranged from two rings (naphthalene, $C^{10}H^{8}$) to seven rings (coronene, $C^{24}H^{12}$**)** (El-Mekawy et al., 2016). PAHs introduced from natural and anthropogenic sources have received great attention as they are considered to be both environmental and food contaminants (Bansal & Kim, 2015). The single benzo(a)pyrene and sum of four specific PAHs (benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene, and chrysene) are useful to indicate PAHs in food (Wu et al., 2016). Therefore, the European Union (Commission Regulation No. 2015/1125) has set a standard for edible oils intended for direct consumption or use as ingredients in food (Commission Directive (EU), 2015). Based on the Regulation as mentioned earlier, the maximum level for benzo(a)pyrene has been set to 2 µg/kg, while the oils and fats can contain a maximum of 10 µg/kg of a sum of the four individual PAHs. Moreover, the United States Environmental Protection Agency (US EPA) has stated the reference dose for oral exposure (RfD) in the Integrated Risk Information System (IRIS) Program. The oral reference dose for fluorine and pyrene are 6×10^{-2} and 3×10^{-3} mg/kg/day individually whereas no reference dose for oral exposure on chrysene because there is a lack of information as it was not assessed in the IRIS Program. However, there is no establishment of standard governing the PAHs content of the food in Malaysia Food Regulations 1985.

In the literature, the fatty matrices were typically extracted using liquid-liquid extraction (LLE) and solid-phase extraction (SPE) with silica or C_{18} as a sorbent (Molle et al., 2017; Lee et al., 2019) for sample clean-up procedures prior to high-performance liquid chromatography coupled with a fluorescence detection analysis of PAHs. Recently, PAHs in edible oils were mostly analyzed by high-performance liquid chromatography (HPLC) with ultra-violet (UV) and fluorescence detectors (FLD) (Hao et al., 2016; Zachara et al., 2017; Yousefi et al., 2018) due to their sensitivity and selectivity in the analysis of PAHs (Wang et al., 2019)**.** The exposure to PAHs-contaminated oil may involuntarily develop risk to human health. PAHs may enter into the human body system through different routes of exposure, including inhalation, dermal absorption, and ingestion (Hao et al., 2016)**.** Besides, the ingestion of food has been a significant route of PAH exposure to humans (Zhu et al., 2019)**.** This situation then increases the possibility of risk to human health that consumed the oil. Thus, the incremental lifetime cancer risk (ILCR) is the appropriate tool to assess the health risk arising from the presence of PAHs in food, which are both genotoxic and carcinogenic (USEPA, 2002).

Malaysia is known as the second-largest exporter of palm oil in terms of volume in the world and was in the ranking 28th in the world for 3,600,000,000 barrels of oil consumption in 2016. In Malaysia, every consumer uses edible oils for different reasons. Instead of cooking, it is worth stating that edible oils are used for health and nutrition purposes as vital proof for the intake of fats (Boateng et al., 2016). This occurrence may trigger human health even though it consumes a long time to develop deleterious impacts. However, there is has limited study conducted regarding the contamination of PAHs in edible oils in Malaysia. There are also limited pieces of evidence in the literature regarding risk assessment on health in edible oils. The study aims to determine and compare the concentration of PAHs in different commercial edible oils and to perform a health risk assessment of PAHs in edible oils based on the ILCR.

2.0 METHODOLOGY

2.1 Sampling Design and Collection

The samples of edible oils were randomly collected from a local supermarket in Puncak Alam City, Selangor. A total of nine samples representing extra virgin olive oils (*n=3*), olive oils (*n=3*), and sunflower oils (*n=3*) indicated three diverse brands of available commercial edible oils to test for evidence of PAHs contamination. The types of edible oils were chosen because they were commonly sold for residents in Malaysia. The samples were stored in a dark room at room temperature. The sample size was adapted from a previous study since there were no consuming data for different edible oils in Malaysia National Health and Nutrition Examination Survey.

2.2 Analytical Procedure

2.2.1 Sample Preparation for PAHs Determination

The preparation of the sample began by dissolving 0.5 g oil in 5 mL of n-hexane and transferred it to a separator funnel. PAHs were extracted twice with 5 ml of dimethylformamide: water (9:1, v/v). The combined extracts were diluted with 17 ml of water at the proportion of 1:2 (v/v) dimethylformamide: water and the solution were submitted to a clean-up step using SPE cartridge with the C-18 sorbent. The cartridge was preconditioned with 5 ml of methanol and 5 ml of dimethylformamide: water (1:2, v/v). The solution was passed through the cartridge, followed by washing with 10 ml of dimethylformamide: water (1:2, v/v) and 10 ml of water. After drying the SPE using the SPE block under vacuum and a concentrator at 1,400 rpm for 25 minutes, the analytes were eluted with n-hexane and concentrated under nitrogen gas. The residue was dissolved in 500 µL acetonitrile and then passes through a 0.20-µm membrane filter and transfers into vials for the HPLC analysis (Secretariat, 2011).

2.2.2 Preparation of Standard Solution

The individual standards (fluorine, chrysene, benzo(a)pyrene and pyrene) were used for the identification and quantification of PAHs. To achieve a concentration of 1 µg/ml for a normal working solution, a certified standard solution of PAHs was used. A 1000 µg/ml of PAHs stock solution was stored in the dark (-20˚C) and the intermediate 1 µg/ml dilutions were prepared with sufficient acetonitrile.

2.2.3 Preparation of Calibration Curve

The calibration curves corresponding to PAHs compounds were completed using the standard working solution, in triplicates, of five levels of concentrations of the PAHs standards, ranging from 0.1 to 1 µg/ml through the external calibration method. A linear dependence (y = ax) of the measured signal (y) in the function of the peak area of the standard substance (x) was evaluated to plot the calibration curves by calculating the correlation coefficient (r^2). The r^2 of the calibration curves for PAHs standard varied from 0.9985 to 0.9999.

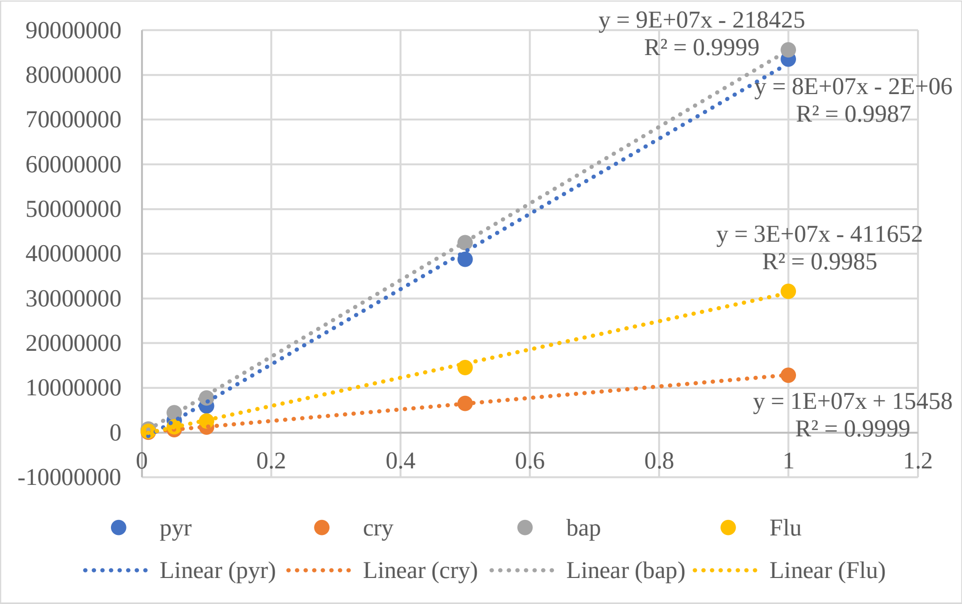

Figure 2.1. Calibration curves for PAHs standard

Table 2.1. Regression equations and correlation coefficients (r^2) of individual PAHs

PAHs	**Regression equations**	**Correlation coefficient (r^2)**
Fluorine	y = 3E + 07x - 411652	0.9985
Pyrene	y = 8E + 07x - 2E + 06	0.9987
Chrysene	y = 1E + 07x + 15458	0.9999
Benzo(a)pyrene	y = 9E + 07x - 218425	0.9999

2.2.4 HPLC Analysis

The chromatographic analysis was carried out by HPLC-FD using a Shimadzu system. The system of Shimadzu equipped with LC-20AT quaternary pump, DGU-20A5 on-line degasser, SIL-20A autosampler (30 µL injection volume), CTO-20A column oven (30˚C), and RF-10AXL fluorescence detector. The acquired data was processed with LC solution software. The use of a C18 column and a gradient, mobile phase of acetonitrile and water at a flow rate of 1 mL/min came for the reason to separate the peak. Gradient elution program started with a 20 minutes linear gradient from 70% to 75% acetonitrile, followed by a linear gradient from 75% to 100% acetonitrile in 15 minutes, and maintained until 55 minutes for 100% acetonitrile isocratic when it finally returned to the initial conditions and the column re-equilibrated for 15 minutes with the composition of the initial mobile phase. Excitation and emission (Ex/Em) wavelength were used for the detection of PAHs which; 224/320 nm for fluorine, 268/398 nm for benzo(a)pyrene, and 238/398 nm for chrysene and pyrene as presented in Table 2.2.

Table 2.2. Wavelength program and retention time for PAHs detection

Compound	**Excitation (nm)**	**Emission (nm)**	**Retention time (min)**
Fluorine	224	320	10.32
Pyrene	238	398	16.40
Chrysene	238	398	20.37
Benzo(a)pyrene	268	398	25.08

2.3 Estimation of Health Risk Assessment

Benzo(a)pyrene, a carcinogen PAHs was used as the indicator of PAHs exposure to human health. BaP-equivalent toxicity was utilized to assess the carcinogenic risk of PAHs. Table 2.3 showed the TEFs value of individual PAHs. The concentration of TEQ_{BaP} completed through the multiplication of every single concentration of PAHs with the toxic equivalent factors (TEFs) is calculated based on Equation (1),

$$\mathbf{TEQ_{BaP} = \sum_{i=1}^{n} Ci \times TEFi} \qquad \mathbf{(1)}$$

$\mathbf{C_i}$ = the quantity of detected individual PAHs in the samples

$\mathbf{TEF_i}$ = the toxicity equivalency factor of congener (i) in food product

Table 2.3. Toxicity equivalency factor of individual PAHs

PAHs	**Toxicity Equivalency Factors (TEFs)**
Nap, Ace, Fle, Phe, Flu, Pyr	0.001
Ant, Chr, BghiP	0.01
BaA, BbF, BkF, IcdP	0.1
BaP, DahA	1

The incremental lifetime cancer risk (ILCR) was useful in estimating health risks associated with the ingestion of carcinogenic toxicants, including PAHs. The ILCR of PAHs in edible oils was measured based on the following Equation (2),

$$\mathbf{ILCR = TEQ_{BaP} \times IR \times EF \times ED \times SF \times CF / (BW \times AT)} \qquad \mathbf{(2)}$$

IR = the daily intake of edible oils (0.05 kg/d), the obtained data from the 2003 Malaysian Adult Nutrition Survey;

EF = the frequency of exposure (350 days/year);

ED = the exposure duration (70 and 6 years for adults and children);

BW = the average body weight (70 and 15 kg for adults and children);

SF = the oral cancer slope factor of BaP (geometric mean of 7.3 mg/kg/day^{-1});

CF = the conversion factor (10^{-6} mg/ng);

BW = the average body weight (70 and 15 kg for adults and children);

AT = the average exposure time (equal to 76 years for carcinogens, 27, 740 days).

3.0 RESULT AND DISCUSSION

3.1 Comparison of PAHs Concentrations in Different Type of Edible Oils

The average concentrations (µg/kg) of PAHs in the extra virgin olive oil were 0.71, 0.18, 0.09, and 1.29 for fluorine, chrysene, benzo(a)pyrene, and pyrene, respectively as presented in Figure 3.1(a). In the extra virgin olive oil, the highest number of pyrene was detected while the content of benzo(a)pyrene was found at the lower concentration. However, in a study by Krajian and Odeh (2018), they stated that the mean value of benzo(a)pyrene was higher in the concentration. Differences in PAH content in the extra virgin olive oils are commonly influenced by either direct (during food processing) or indirect (environmental pollutants such as dust) contamination to the olive fruit (Elaridi et al., 2020). Figure 3.1(b) shows the concentration of PAHs in the olive oil with the mean value of pyrene (0.70 µg/kg) was identified at a higher concentration of PAHs, followed by fluorine (0.34 µg/kg) and chrysene (0.10 µg/kg). On the contrary, the benzo(a)pyrene was very low in concentration with a mean value of 0.98 µg/kg. Previously, Ju et al. 2020 revealed the detection of benzo(a)pyrene in the olive oil even though their amounts were not quantified in the study. The presence of PAHs in olive oil may be coming from the surrounding environment. As the skin layer of the olive had a waxy surface, thus PAHs had a possibility to adsorb onto the fruit's surface (Zelinkova & Wenzl, 2015). In this study, the sunflower oil samples generated the least amount of benzo(a)pyrene and the highest amount of pyrene, with the average value of 0.18 and 2.08 µg/kg, individually as displayed in Figure 3.1(c). The drying process of the oilseeds with the combustion gases may lead to the occurrence of PAH contamination in this edible oil (Silva et al., 2017).

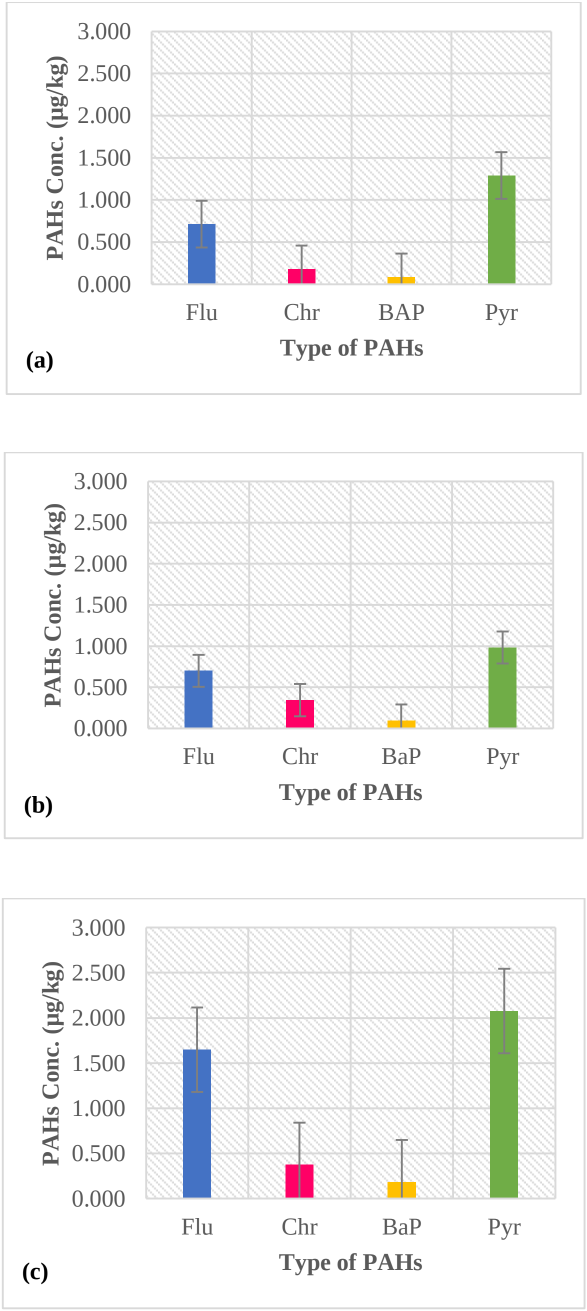

Figure 3.1. Average concentrations of PAHs in (a) extra virgin olive oil; (b) olive oil; (c) sunflower oil

Table 3.1. Mean concentration (μg/kg) of PAHs in commercial edible oils

PAHs	Extra virgin olive oil	Olive oil	Sunflower oil	p-value
Flu	0.71 ± 0.01	0.70 ± 0.05	1.65 ± 0.18	0.072
Pyr	1.29 ± 0.06	0.98 ± 0.03	2.08 ± 0.06	0.172
Chr	0.18 ± 0.01	0.34 ± 0.01	0.37 ± 0.01	0.002
BaP	0.09 ± 0.01	0.10 ± 0.01	0.18 ± 0.01	0.196

Table 3.1 shows the mean concentrations of fluorine, chrysene, benzo(a)pyrene, and pyrene in the commercial edible oil samples from a supermarket in Puncak Alam City, Selangor. The results were expressed as the means ± standard deviations. Among 27 samples of commercial edible oils investigated in this study, a wide range of PAHs contents were observed and varied based on the category of edible oils. The most abundant PAHs were pyrene and followed by fluorine and chrysene. Even though the concentrations for both fluorine and pyrene were higher in all three edible oils, but their content has not exceeded the reference dose for oral exposure. Meanwhile, benzo(a)pyrene was the least found in the edible oils with a concentration of less than 2.0 μg/kg. In this study, the concentrations of PAHs except for chrysene, were not significantly different ($p > 0.05$, $F = 0.816$) between the extra virgin olive oil, olive oil, and sunflower oil samples. The sunflower oil was highly contaminated by PAHs for the current study. Instead of oilseeds drying, the contamination may be also due to the accumulation of PAHs in the oilseeds as

a result of the polluted soil by these persistent chemicals in the surrounding environment (Shi et al., 2016). Although this oil contained higher PAHs amounts, it is safe to eat as the concentration complied with the US EPA and EU legal standards.

3.2 Comparison of Individual PAHs Concentration in Different Brands

Every edible oil has different content of PAHs since it came from various brands and undergoes different processes in food production. Figure 4.3.2 presents the concentration of individual PAHs in three brands of commercial edible oils purchased from a supermarket in Puncak Alam City, Selangor. In this study, the highest concentration of fluorine was detected in the sunflower oil of Brand A, which was 1.95 µg/kg. The high amount of fluorine was then followed by the sunflower oil in Brand B and Brand C, which were 1.67 and 1.32 µg/kg, respectively. In Brand A, the extra virgin olive oil contained more fluorine than the olive oil whereas the concentrations of fluorine in the same edible oils were vice versa in Brand B. Meanwhile, Brand C has the least contamination of fluorine in all different edible oils; the extra virgin olive oil, olive oil, and sunflower oil at the correspondent level of 0.56, 0.52, and 1.32 µg/kg. Regarding the US EPA standard, the concentrations of fluorine in different edible oils for Brand A, B, and C in the current study did not exceed the limit dose for the oral route. Nevertheless, there were limited studies that concerned the fluorine in commercial edible oils. The reason was that it has been categorized in the non-carcinogenic group of PAHs as stated by the US EPA.

Benzo(a)pyrene or BaP was commonly used as an indicator for PAH contamination in food. The concentrations of BaP in the edible oils for various brands were demonstrated in Figure 4.3.2. The BaP level in the olive oil from Brand C was the lowest among other edible oils, which was 0.06 µg/kg. There were four edible oils from two different brands that have the same BaP concentration of 0.09 µg/kg, including the extra virgin olive oil and olive oil from Brand B, and also the extra virgin olive oil and sunflower oil from Brand C. Additionally, the sunflower oil of Brand A exhibited the highest BaP with the amount of 0.28 µg/kg. On the other hand, the level of BaP in every edible oil from the selected brands have complied with the EU standard as their concentrations were not more than the maximum level of 2 µg/kg for BaP.

Concerning the contents of chrysene identified, the olive oil sample of Brand A has the largest amounts of the mentioned PAH, which was 0.8 µg/kg, and followed by the sunflower oil from the same brand at the level of 0.6 µg/kg. On a contrary, the number of chrysene in the extra virgin olive oil was the least for Brand A as displayed in Figure 4.3.2. Besides, in Brand B, the concentration of chrysene in these three different edible oils were in the range of 0.16 µg/kg to 0.25 µg/kg. Meanwhile, the chrysene was in the least concentration of 0.07 µg/kg in the olive oil from Brand C. Nonetheless, the concentrations of chrysene in these edible oil samples were not able to identify and compare with the maximum limit. This was due to the absence of reference dose for consumption in the US EPA legal standard even the chrysene was grouped as B2, the probable human carcinogen PAH.

Figure 3.2 revealed the mean concentrations of pyrene in the edible oils of Brand C were in the range of 1.52 µg/kg to 1.82 µg/kg. Furthermore, the content of pyrene in the sunflower oil was doubled the amount of pyrene in the extra virgin olive oil and olive oil originated from Brand A, which the level set as 2.74 µg/kg. The number of pyrene in the olive oil of Brand B was the least among those three commercial edible oil with a concentration of 0.63 µg/kg. As a whole, the concentrations of pyrene in all edible oils were complying with the oral reference dose set by the US EPA.

From this study, there was no statistically significant difference between individual PAH concentrations in different brands of commercial edible oil ($p > 0.05$, $F = 0.160$). The differences observed in PAHs concentration between the brands of commercial edible oils could be attributed to different technological processes during the food preparation such as direct fire drying, at which the combustion products may come into contact with oil seeds or oils (Abdel-Shafy & Mansour, 2016). The edible oils contained PAHs can also decrease significantly through the refining process (Hua et al., 2016). Since there was no standard governing the PAHs content of the food established in Malaysia Food Regulations 1985, the European Union (Commission Regulation No. 2015/1125) has been used as a reference. Based on the Regulation as mentioned earlier, identified PAH values were lower than the maximum acceptable level of 2 µg/kg for benzo(a)pyrene in the edible oils according to the EU limit. The standard of US EPA also has been referred for the oral reference dose for fluorine and pyrene concentrations in the commercial edible oils.

Figure 3.2. Comparison of individual PAHs concentration in different brands

3.3 Health Risk Assessment of PAHs Exposure From Consumption of Commercial Edible Oils

The value of ILCR in the case of benzo(a)pyrene for different population groups in Selangor, Malaysia was demonstrated in Table 3.2. The ILCR approach was applied to evaluate the threat of PAH exposure to human health. According to the standard set by US EPA, the risk level is considered acceptable or inconsequential when a one in a million chance of additional human cancer over a 70 year lifetime (ILCR = 1.0×10^{-6}). Meanwhile, the level of risk is considered serious when an additional lifetime cancer risk of one in ten thousand or greater (ILCR = 1.0 × 10^{-4}), and there is a high priority to pay attention to such health problems. In this study, the ILCR values were calculated to be 6.71 × 10^{-9}, 2.28 × 10^{-8}, and 1.94 × 10^{-8} for children, male and female adults of the population groups, individually. According to the calculation, it was found that the values of ILCR were lower than the acceptable risk level of 1.0 × 10^{-6} indicating a significant potential of carcinogenic risk. Besides, they are also all lower than the priority risk level of 1.0 × 10^{-4}, showing no significant cancer risk related to commercial edible oil consumption for the groups of population in Selangor, Malaysia. Among the population groups, the ILCR value for children was the lowest due to lower level of exposure through ingestion and short duration of exposure compared to the adults (Wu et al., 2016). To compare with other literature, the result for the health risk assessment showing similarities to this study. The ILCR value for dietary exposure was lower for the population in the Shandong Province of China (Jiang et al., 2015).

Table 3.2. ILCR values for different age groups among Selangor, Malaysia

Population groups	**Incremental Lifetime Cancer Risk (ILCR)**
Children	6.71 × 10^{-9}
Adults (Female)	1.94 × 10^{-8}
Adults (Male)	2.28 × 10^{-8}

4.0 CONCLUSION

In the present study, the concentration of PAHs in commercial edible oils collected from a supermarket in Puncak Alam City, Selangor were identified. The established method was applied in determining the PAH levels in nine samples of commercial edible oils. Among all the edible oil samples, the sunflower oil contained a high number of PAHs. It is possible to conclude that it is a challenge to reduce the content of PAHs due to PAHs contamination in the environment and food preparation process. However, the amounts of PAHs in those three different oil and brand samples did not exceed 2 µg/kg, the standard of the maximum acceptable level of the European Union. There was no significant difference for PAH concentrations in a different types of edible oils except chrysene. In addition, PAH concentrations in different brands of edible oils were also not significantly different.

In order to improve the findings of PAHs in the future, this study would recommend using the PAH4 indicator instead of single benzo(a)pyrene only. It is because the combination of benzo(a)pyrene and PAH4 are useful to indicate the level of PAHs in food including edible oils. Besides, since there is no standard for PAHs level in edible oil stated in the food legislation of Malaysia, it would be essential to implement and maintain the regular monitoring from time to time on the concentration of PAHs in the edible oils in order to increase food safety. To summarize, the current study filled the information gap of PAHs contamination through dietary exposure of commercial edible oil in Malaysia.

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