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Analysis and health risk assessment of priority gaseous polycyclic aromatic hydrocarbons during meat frying: A mathematical modelling

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ABSTRACT

Introduction: Emissions from cooking activities are among the major sources of indoor and ambient air pollution.

Materials and methods: This experimental research aims to explore the levels of 16 gaseous Polycyclic Aromatic Hydrocarbons (PAHs) during calf meat frying in laboratory, utilizing different frying temperatures (i.e 150, 190, and 240 °C) and oils (non-frying oil and, frying oil). Furthermore, non-cancer and cancer risks were also assessed. For the purpose of the study, 36 air samples were taken during meat frying and analyzed by a Gas chromatography mass spectrometry (Agilent GC8890, USA) equipped with a Flame Ionization Detector (FID) for 16 PAH compounds.

Results: The concentration of ∑16PAHs during meat frying using sunflower oil and frying oil use varied from 5.037-10.025 µg/m^3 and 3.978-8.075 µg/m^3, respectively. Hazard Quotients (HQs) associated with PAHs exposure during meat frying using frying oil for cooks, adults and children were in the range of 0.440-1.338 (0.769), 0.503-1.527 (0.879) and 0.504-1.531 (0.881), respectively. For frying oil, HQ values were in the ranges of 0.32-1.19 (0.69), 0.37-1.36 (0.79), and 0.37-1.36 (0.79) for cooks, adults, and children, respectively. The inhalation cancer risk values through exposure to meat using sunflower oil for cooks, adults and children were 1.4E-04-4.2E-04 (2.4E-04), 2.8E-05-8.6E-05 (4.9E-05), and 7.7E-06-2.3E-05 (1.3E-05), respectively. For frying oil, the cancer risk values were as: 1.0E-04-3.7E-04 (2.2E-04) for cooks, 2.1E-05-7.6E-05 (4.4E-05) for adults and 5.63E-06-2.08E-05 (1.2E-05) for children.

Conclusion: This study showed high levels of PAHs during meat frying indicating health risks for children and adults. The research's results have practical use for public health professionals and policy makers, for regular monitoring of indoor PAHs during cooking and the development of policies to reduce exposure to these air pollutants in enclosed spaces.

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Introduction

The investigation of the quality of indoor air is essential due the presence of different sources of emission inside, and to the fact that nowadays most of individuals spend more than seventy percent of their time in indoor spaces either at home or at their work places [1-3]. Normal daily activities performed indoors could release numerous air pollutants [4, 5]. Cooking, which is considered as the key common indoor activity, can be one of the main sources of air pollutants indoors. On a global scale, annually more than 4 million premature deaths including 0.5 million children is attributed to air pollution related to cooking emissions [6]. PAHs are a group of aromatic hydrocarbons with 2, 3 or more benzene rings in their structure [7]. PAHs are toxic compounds emitting into the indoor and outdoor air, soil, water and food by high-temperature reactions during incomplete combustion of organic matters and pyrolysis of fossil fuels [8]. PAHs are ubiquitous in environment. They are generated when organic compounds burn incompletely during both natural processes and human activities [9]. Generally, above 10,000 types of PAHs are currently identified, of which above 100 can be analyzed [10].

PAHs are of great concern for the public and are classified as a critical subset of persistent organic pollutants (POPs) due to their hydrophobic nature and high stability/persistency in the environment [11]. There are numerous (hundreds) of PAHs in environment. Hence, the United States Environmental Protection Agency (USEPA) has considered 16 of PAHs as the priority pollutants mainly because of their potential unique characteristics including carcinogenicity, immunotoxicity, mutagenicity, teratogenic and endocrine-disrupting properties in some lab animals and humans [12-14]. Human exposure to PAHs may occur through three main pathways, including inhalation of polluted air by respiratory tract, intake of contaminated food or water by digestive tract, and skin contact to polycyclic aromatic hydrocarbons. Therefore, PAHs may be present in different human organs and have also been found in the urine, blood, and hair samples of the exposed individuals [15]. Many scientists around the world have investigated the amounts and types of PAHs released during food cooking [16-21]. PAHs are very toxic, which negatively affects human wellbeing as well as the animals thus causing cancer (particularly lung, skin, and bladder cancer), skin problems, lung and respiratory problems, adverse effects on reproduction and development system, and may also affect behavior, and neurons [22-24]. PAHs may cross the placental barrier, exposing fetuses to these toxic compounds during period of critical windows of development and cause numerous developmental issues, such as lower birth weight (weight at birth of < 2500 grams), microcephaly, and neurological impairment [25]. For example, in a previous study, among the sixteen types of PAHs tested, thirteen were found in both maternal and baby samples [11]. PAH exposure is a public health concern, more especially for children because their organs are in developing stages [26].

Emissions from cooking and smoking activities could be the main source of PAHs, particulate matters and nitrogen oxides etc. [16, 27]. Generally, cooking at high temperatures for example, grilling, frying, and roasting activities may significantly emit PAHs into the indoor spaces, particularly when putting the meat directly over the fire, as the high temperatures cause fat to drip and release PAHs [28]. Moreover, the type of fuel used for cooking greatly affects PAH concentrations in food and indoor air of kitchens. Accordingly, it is reported that wood and kerosene burning produce more PAHs than gas [6, 29-31]. Cooking pollutants may arise from burning of fuels (solid fuels, coal, natural gas, kerosene), as well as from the vaporization and heat pyrolysis of oil in foods or organics in foods during cooking activities [32].

Frying is a food cooking technology and is usually used in many homes, restaurants, and food industries [33, 34]. Food frying process in an oil bath is a common method that is becoming progressively popular in homes, restaurants, and industries in many nations, mainly because of the increase in the consumption of ready-made/pre-cooked foods [35, 36]. These foods are routinely used because their preparations are easy and quick and also they have favorite smell, taste, aroma, texture characteristics which make them more attractive to the people [37]. Traditionally, many foods are deep-fried using fats and oils [38]. It is found that deep-fat frying method provides more browning and thicker crusts compared to those produced by hot air frying method [39]. Large variations in the release of PAHs are expected due to different cooking styles in different countries and dietary compositions. In almost all cities of Iran, natural gas is the primary energy source that populations primarily rely on for food cooking and heating indoors. To the best of our knowledge, representative emission profiles of meat frying are lacking. Therefore, investigating PAHs emission during cooking activities is crucial due to their presence, and detrimental health effects, prompting the need to identify their sources. In this study in order to assess the health impact associated with the inhalation of PAHs emitted from meat frying, indicators of risk assessment proposed by the US EPA including Hazard Quotient (HQ) and the cancer risk (CR) were employed. The present study increases our understanding regarding the potential hazards of PAHs aiming to fill this gap and provide useful information and insights for environmentalists, health professionals and policymakers to improve kitchen indoor air conditions and shape health policies for the proper management of indoor air quality.

Materials and methods

Air sampling and analysis of samples

In the present study, 3 important influencing parameters, such as frying technique, frying temperatures (i.e 150, 190, and 240 °C), and type of cooking oils commonly sold in Iran’s grocery stores (i.e sunflower oil was used as non-frying oil and, sunflower oil + canola oil was used as frying oil), were investigated. Calf’s meat was cooked in oil by deep-frying technique. In Iran, during meat cooking for example for Kebab, tail fat 24-36 % is commonly added to meat. Fat adding causes the cooked meat become more fatty and delicious. Meat in this study contained 20-30 % tail fat. All cooking tests in this study were performed by the use of natural gas. The study scheme was a gas stove using natural gas, a cooker, a sampling cartridge (adsorbent) and a sampling pump for gas. Samples of gaseous PAHs were collected at 1.5 m above floor surface and 30 cm away from the gas stove. In the first step, a clean pan was heated up and then the selected oil was carefully poured heated at the desired temperature. After that, some meat was added to the pan. The air containing PAHs released during frying was collected by the use of standard adsorbent tubes. Each experiment was conducted for 3 times at 3 desired temperatures (i.e 150, 190 and 240 °C). In total, 36 samples (for two oils, 3 temperatures with repetition) were collected and tested for PAH compounds. During the sampling of PAHs in the lab air, indoor air temperature was in the range of 22.5-24.5 °C.

In this research, all windows and doors in the laboratory were closed to avoid the entrance of pollutants from ambient air and subsequent exit of indoor air. Moreover, ventilation also was not used. The collection, preparation and measurement of the samples for PAHs followed the described procedure given by NIOSH method No. 5515, issue 2 [40]. Prior to collection of air samples, the sampling pump was calibrated using a standard flow meter. After that, one tube was connected to an adsorbent cartridge, and another one was connected to a sampling pump. The PAHs samples were taken by a sampler [sorbent (washed XAD-2, 100 mg/50 mg] at a flow rate of 2 L/min using XAD-2 adsorbent. The sampling time of for experiments was approximately 10-20 min during the frying of meat. Firstly, sampling pump was calibrated and then switch on it at flow of at 2 L/min. After sampling, the sorbent tubes were capped and then wrapped in aluminum foil to prevent PAH degradation from UV radiation and then refrigerated upon receipt at laboratory and analysis. For PAHs desorption from sorbent, each tube was broken at score line and then front glass wool plug and front sorbent section were transferred to a culture tube. After that, sorbent section was transferred back and the middle glass wool was plugged to a second culture tube. Then, 5 mL of toluene was added into each culture tube, capped and mixed occasionally. After 30 min, the samples were sieved through a 0.45 µm membrane filter. After extraction, PAHs in the samples were analyzed using a Gas chromatography-mass spectrometry (Agilent GC8890, USA) equipped with a Flame Ionization Detector (FID) using a capillary column (30 m × 0.32-mm ID, fused silica capillary,1 µm DB-5). Temperatures of injector, detector and program were 200, 250 and 120-290 °C, respectively. The injector volume was 4 µL. The Limit of Detection (LOD) and Limit of Quantitation (LOQ) for measured gases were in the ranges of 0.032-0.231 and 0.09-0.7 $\mu g/m^3$, respectively. The recovery rates for polycyclic aromatic hydrocarbons on XAD-2 tube were above 93 %. The accuracy of the method, considering 3 replicates was found to be above 97 % ($R^2 > 0.97$). The temperature of ion source, injection port, and transfer line was maintained at 320 °C and temperature of quadrupole mass analyzer was fixed at 150 °C. The calibration of GC-MS/FID was done by standard solutions containing all sixteen selected PAHs at concentrations in the range of 0.5-50 pg/mL (Standard for PAHs). Second order calibration curves were adjusted to the obtained data by employing the least-squares method. All the instruments used for the sampling in this study were calibrated before use according to the specifications given by their manufacturers.

In the current research, 16 EPA priority PAHs as given in Table 1 were measured. The analytical standards for determination of each PAH considered in the present study were obtained from Sigma Aldrich Company with high purity (above 99%). All used solvents and chemicals in this study were of analytical reagent grade obtained from the Merck, and all the needed reagents were prepared with double distilled water.

Risk assessment through PAHs inhalation

People's health risks associated with the respiration of PAHs were assessed using methods recommended by the United States Environmental Protection Agency (US EPA) [42-44]. In this study, both non-cancer and cancer risks from the inhalation of sixteen priority PAHs during meat frying were estimated. For non-cancer risk, hazard quotients (HQs) PAHs was calculated [45, 46]. For the estimation of the cancer risk (CR) from exposure to PAHs, sum of Benzo a pyrene equivalent (BaPeq) was calculated [47-51]. ∑BaPeq was calculated by considering each PAH utilizing the following equation:

$$\sum \mathrm{BaPeq} = \sum_{i=1}^{n} (C_{PAH} \times \mathrm{TEFs}) \quad (1)$$

Where C_{PAH} denotes the level of each PAH expressed in unit of $\mu g/m^3$. TEF is toxic equivalent factor of each PAH (Table 1). BaP has been used by numerous studies across the world for investigation of levels of carcinogenic PAHs [49, 50, 52-55].

Table 1. PAHs considered in the present study with their number of rings, molecular weight (MW), and TEF [56-58]

No	PAH	Abbreviation	MW(g/mol)	Number of rings	TEF
1	Naphthalene	Naph	128.15	2	0.001
2	Acenaphthylene	Acy	152.19	3	0.001
3	Acenaphthene	Ace	154.08	3	0.001
4	Fluorene	Flu	166.22	3	0.001
5	Phenanthrene	Phen	178.23	3	0.001
6	Anthracene	Anth	178.23	3	0.01
7	Fluoranthene	Flt	202.26	4	0.001
8	Pyrene	Pyr	202.25	4	0.001
9	Benz(a)anthracene	BaA	228.09	4	0.1
10	Chrysene	Chr	228.09	4	0.01
11	Benzo(b)fluoranthene	BbF	252.09	5	0.1
12	Benzo(k)fluoranthene	BkF	252.09	5	0.1
13	Benzo(a)pyrene	BaP	252.09	5	1
14	Dibenz(a.h)anthracene	DahA	278.10	5	1
15	Benzo(g.h.i)perylene	BghiP	276.09	6	0.01
16	Indeno(1.2.3-cd)pyrene	Ind	276.09	6	0.1

In this study, HQs and CRs from exposure to PAHs during meat frying was quantified for different age groups including cooks (at workplace), adults (at home) and children (at home) using following Eqs [2, 49, 59-62]:

$$\text{EDI}_{\text{Inhalation}} = (\sum \text{BaPeq} \times \text{InhRa} \times \text{F} \times \text{EP} \times 10^{-3})/(\text{BW} \times \text{AT}) \quad (2)$$

$$\text{EC}=(\sum \text{BaPeq} \times \text{F} \times \text{EP} \times 10^{-3})/\text{AT} \quad (3)$$

$$\text{HQ} = \text{EDI}/\text{RfD} \text{ or } \text{HQ} = \text{EC}/\text{RfC} \quad (4)$$

$$\text{RfD} = (\text{RfC}, 2 \times 10^{-6} \frac{\text{mg}}{\text{m}^3}) \times \frac{\text{InhR}}{\text{BW}} \quad (5)$$

$$\text{CR} = (\text{EDI} \times \text{CSF}_{\text{inh}}) \quad (6)$$

Where, EDI is the estimated daily intake of polycyclic aromatic expressed in mg/kg/day and EC denotes exposed level of pollutant in indoor air in mg/m^3. In Table 2 all the required factors for the estimation of human health risks via PAHs exposure during meat frying are given.

Table 2. Details of the factors and their values utilized in human risk assessment equations in the present study

Abbreviation	Description	Unit	Age group			Reference
			Cooks	Adults	Children	
F	Exposure frequency	days/years	121	15.2	15.2	[63]
CSF_{inh}	Inhalation cancer slope factor	mg/kg/day	3.85	3.85	3.85	[50, 64, 65]
BW	Body weight	kg	80	80	15	[49, 52, 60, 66]
EP	Exposure period	years	30	49	5	[28, 49, 67]
InhRa	Air inhalation rate	m^3/day	20	20	10	[50, 60, 68]
AT	Averaging timing of carcinogenic exposure	days	25550	25550	25550	[50, 69]
AT	Averaging timing of non-carcinogenic exposure	days	8 h/24 h*365= 121.6	1 h/24 h*365= 15.2	AT	[49]
C_{PAH}	Concentration of PAHs	μg/m^3				
10^{-3}	Conversion factor	mg/μg	0.001	0.001	0.001	
RfC	Inhalation reference concentration of BaP	mg/m^3	0.000002	0.000002	0.000002	[70]

For non-cancer risk descriptions, HQs ≥ 1 signifies the probability of occurrence for hazardous non- carcinogenic effects of the exposed person from a specific pollutant [46, 71-73]. CR shows the possible occurrence of cancer in exposed individuals, resulting from a lifetime exposure to carcinogenic compounds. For cancer risk description, CR < 10^{-6} (the risk of developing cancer during a human lifetime is less than 1 in 1,000,000) represents negligible risk to individuals; 10^{-6} < CR < 10^{-4} indicates a probable carcinogenic effect of PAHs to exposed individuals; and CR < 10^{-4} shows high cancer risk in exposed person via exposure to PAHs [66, 74-77].

Results and discussion

Quantification of 16 PAHs during meat frying

Clean indoor air quality is of great importance as it directly relates to the health and comfort of inhabitants in these micro environments. Cooking activities releases numerous air pollutants that may result in significant exposure and health hazards within indoor spaces [78]. The popularity of fried foodstuffs among world consumers is largely attributed to its rich nutritional profile, easy preparation and inexpensiveness. Therefore, PAH emissions from food frying is an important issue, especially considering the increasing consumption rates of these types of foods. However, the release of different PAH species during frying into foods or indoor air remains uncertain. This study was undertaken to help to analyze and understand the distribution of indoor air PAHs and their associated health risks due to Iranian style cooking. It is reported that deep frying method produces 6 times higher amount of total gaseous PAHs than the steaming method [16]. In this study deep frying in oil was used for meat cooking. Tables 3 and 4 outline 16 gaseous PAHs released during meat frying using sunflower oil and frying oil. In the present study, levels of PAHs ranged from 0.032 to 2.060 (0.579), 0.032 to 1.830 (0.521) and 0.032 to 2.10 (0.668) μg/m^3 during meat frying using sunflower oil at 150, 190 and 240 °C, respectively. But in the case of frying oil usage, levels of PAHs ranged from 0.032 to 1.013 (0.384), 0.032 to 1.531 (0.494) and 0.032 to 2.015 (0.715) μg/m^3, respectively. These results clearly show that less amounts of PAHs are emitted when frying oil is used. Distribution of each PAH emission during meat frying using, sunflower oil and frying oil is displayed in Fig. 1 (A and B). As can be seen in the figure, the highest amount of gaseous PAH emission belongs to BaP and then Naph.

Table 3. 16 Gaseous PAHs released during meat frying using non frying oil in μg/m^3 in the present study

Items	150 °C			190 °C			240 °C		
	Replication			Replication			Replication		
	1	2	3	1	2	3	1	2	3
Minimum	0.032	0.032	0.065	0.032	0.032	0.065	0.032	0.032	0.065
Mean	0.267	0.318	0.378	0.290	0.379	0.192	0.385	0.437	0.299
Maximum	0.950	1.110	2.060	1.100	1.830	0.773	1.434	2.100	1.232
∑16PAHs	5.037	6.153	7.426	6.600	8.271	5.288	8.891	10.025	5.988

Table 3. Continued

Items	150 °C			190 °C			240 °C		
	Replication			Replication			Replication		
	1	2	3	1	2	3	1	2	3
∑CarPAHs*	1.821	2.396	2.140	2.647	3.504	2.501	3.473	4.290	2.006
∑LMWPAHs**	2.010	2.217	3.614	2.421	2.917	1.861	3.463	3.407	2.456
∑MMWPAHs***	0.322	0.610	0.426	0.582	0.901	0.632	0.955	1.318	0.423
∑HMWPAHs****	2.706	3.326	3.385	3.597	4.452	2.796	4.473	5.300	3.109

*CarPAHs= carcinogenic PAHs, **LMWPAHs= low molecular weight PAHs, ***MMWPAHs= medium m weight PAHs, ****HMWPAHs= high molecular weight PAHs.

Table 4. 16 gaseous PAHs released during meat frying using frying oil (sunflower oil + canola oil) in µg/m^3

Items	150 °C			190 °C			240 °C		
	Replication			Replication			Replication		
	1	2	3	1	2	3	1	2	3
Minimum	0.032	0.032	0.065	0.032	0.032	0.065	0.032	0.032	0.129
Mean	0.210	0.245	0.220	0.222	0.324	0.204	0.317	0.354	0.262
Maximum	0.735	0.900	1.013	0.745	1.531	1.295	1.390	2.015	1.904
∑CarPAHs*	3.978	4.838	6.604	4.808	6.813	4.720	6.968	8.078	7.850
∑LMWPAHs**	1.377	1.925	2.612	1.816	2.908	3.085	2.843	3.669	2.265
∑MMWPAHs***	1.688	1.665	2.304	2.016	2.536	1.238	2.566	2.734	3.862
∑HMWPAHs****	0.272	0.412	0.806	0.430	0.753	0.529	0.732	0.944	1.219
∑CarPAHs*	2.018	2.761	3.494	2.362	3.523	2.953	3.670	4.400	2.769

*CarPAHs= carcinogenic PAHs, **LMWPAHs= low molecular weight PAHs, ***MMWPAHs= medium molula weigh PAHs, ****HMWPAHs= high molecular weight PAHs.

In Africa, the PAHs level was found in the range of 0.00226-29.95 71 μg/m^3 with a mean level of 14.71 μg/m^3 [29, 79, 80]. In the EU and the American countries, PAHs levels were relatively low,

in the range 0.00075-1.58 in Europe and in the range 0.011-0.327 in the Americas, with mean levels of 0.39 μg/m^3 and 0.0916 μg/m^3, respectively [6]. The relatively lower PAHs in the Europe and Americas compared to the current study is due to the economic and technological advancements in these countries, where natural gas and electricity are predominantly being used, and efficient ventilation systems are commonly used during cooking activities. In a previous study in Kenyan houses, light PAHs (Naph-Anth) contributed to about 85% of gas phase PAHs. Mean gaseous PAH levels per household were higher in rural homes (0.81-6.09 μg/m^3) compared to urban homes (0-2.59 μg/m^3). The peak levels of PAHs was detected in homes burning wood [29]. It was reported that cooking activities are the main sources of indoor air pollution [81], and the mean levels of PAHs in cooking settings recorded in Asia and Africa were significantly higher than those in Europe and America. The highest average PAHs level in cooking settings was detected in Africa at 14.74 μg/m³ [6].

The total concentrations of ∑16PAHs released during meat frying using sunflower oil ranged from 5.037- 7.426 (6.205), 5.288- 8.271 (6.720) and 5.988- 10.025 (8.301) μg/m^3 at 150, 190 and 240 °C, and varied significantly among the samples. Similarly, concentrations of ∑16PAHs during meat frying using frying oil ranged from 3.978-6.604 (5.140), 4.808-6.813 (5.447) and 6.968-8.078 (7.632) μg/m^3 at 150, 190 and 240 °C, and varied significantly among the samples. The sum of carcinogenic PAHs (∑CarPAHs) released during meat frying using sunflower oil were in the range from 1.821 to 2.396 (2.119), 2.501 to 3.504 (2.884) and 2 to 4.290 (3.256) μg/m^3 at 150, 190 and 240 °C, respectively. Levels of ∑CarPAHs during meat frying using frying oil ranged from 1.377 to 2.612 (1.972), 1.816 to 3.085 (2.603) and 2.265 to 3.669 (2.926) μg/m^3 at 150, 190 and 240 °C, respectively. In Kenya's study, the use of wood burning devices in rural houses exposed occupants indoors to the high amount of carcinogenic gaseous PAHs (total averages of 46.23 μg/m^3) which is much higher than those in this study [29]. Table 5 outlines a summary of recently published studies in the field of PAHs emission during cooking.

In 2002, the European Scientific Committee on Food (SCF) has considered BaP as an indicator for the presence of carcinogenic PAHs in foodstuffs [82]. It is reported that that approximately thirty percent of food samples had low BaP concentrations but high concentrations of other potential carcinogenic compounds [83]. The European Union (EU) has introduced PAH2 (BaP, Chr), PAH4, and PAH8 as new markers. Recently, the EU proposed maximum permissible limits for BaP and PAH4 compounds in thermally treated meat and meat products in human nutrition as 5.00 and 30.0 μg/kg, respectively [84].

As summarized in Tables 3 and 4, levels of total low molecular weight PAHs (∑LMWPAHs) were in the ranges of 2.01-3.614 (2.613), 1.861-2.917 (2.400) and 2.456-3.463 (3.109) μg/m^3 during meat frying using sunflower oil at 150, 190 and 240 °C, respectively. For frying oil, the values were in the ranges of 1.688-2.304 (1.886), 1.238-2.536 (1.930) and 2.566-3.862 (3.054) μg/m^3, respectively. Levels of total medium molecular weight PAHs (∑MMWPAHs) ranged from 0.322 to 0.610 (0.453), 0.582 to 0.901 (0.705) and 0.423 to 1.318 (0.899) μg/m^3 during meat frying using sunflower oil at 150, 190 and 240 °C, respectively. For frying oil, the values were in the ranges from 0.272 to 0.806 (0.497), 0.430 to 0.753 (0.571), 0.732 to 1.219 (0.965) μg/m^3, respectively. Concentrations of total high molecular weight PAHs (∑HMWPAHs were in the ranges of 2.706-3.385 (3.139), 2.796-4.452 (3.615) and 3.109-5.30 (4.294) $\mu g/m^3$ during meat frying using sunflower oil at 150, 190 and 240 °C, respectively. During meat frying using frying oil, the concentrations were in the ranges of 2.018-3.494 (2.758), 2.362-3.523 (2.946) and 2.769-4.40 (3.613) $\mu g/m^3$, respectively.

Distributions of 16 PAHs emitted into indoor air during meat frying are presented in Fig. 2 (A & B). From the figure, it can be concluded that the contribution of high molecular and carcinogenic compounds to the overall PAHs increased by temperature increase. In total, sunflower oil (non-frying oil) released more of PAH levels. Amounts of LMW, MMW and HMW PAHs in the published literature reviews are as: 0.210-41.5, 0.111-13.5, 0.179-1.92 $\mu g/m^3$ in household kitchens and 51.5-76.1, 3.06-17.6, 0.975-3.47 $\mu g/m^3$ in restaurants [85]. ∑LMWPAHs were 13.19, 16.30, 52.74 $\mu g/m^3$ in Chinese, Western, and barbeque restaurants. Furthermore, ∑MMWPAHs were 0.61, 0.81, 0.84 $\mu g/m^3$ and ∑HMWPAHs were 7.19, 4.37 and 5.22 $\mu g/m^3$ [86].

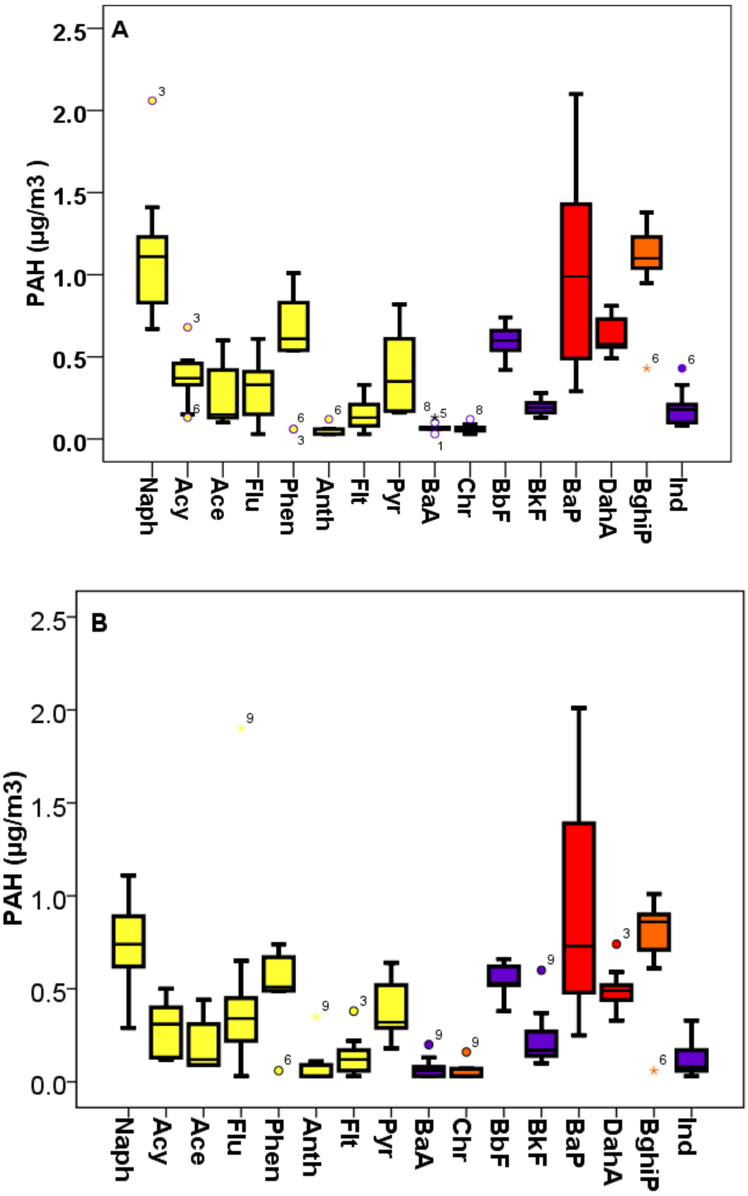


Fig. 1. Distribution of PAHs emitted during meat frying using, (A) sunflower oil and, (B) frying oil (sunflower oil + canola oil)

Table 5. A summary of recently published studies in the field of PAHs emission during cooking

PAHs	Setting	Levels (μg/m^3)	Reference
∑ PAHs	Household kitchens	1.44-56.9	[87]
∑16 PAHs	Western fast food kitchens	2.371-4.699	[88]
	Chinese cafeteria kitchens	1.648-5.342	
	Street food carts	1.183-147.585	
PAHs	Night markets	23.4 to 44.2	[89]
∑PAHs	Oil particles during rape-seed oil, corn oil, peanut oil	1.08-22.8	[90]
∑PAHs	Chinese restaurants	20.99	[86]
	Barbeque restaurants	58.81	
	Western restaurants	21.47	
∑PAHs	Western	92.9	[85]
	Chinese	80.1	
	Fast food	63.3	
	Japanese	55.5	
Highest levels of PAHs	Chinese cooking	0.141	[58]
	Malay cooking	0.609	
	Indian cooking	0.0379	
∑PAHs	Commercial kitchens	17	[91]
	Domestic kitchens	7.6	
Mean gaseous PAHs	Rural houses	0.81-6.09	[29]
	Urban houses	0-2.59	
∑PAHs	Residential houses	1.26	[92]
∑16 PAHs	cooking fuels	10.82 – 14.06	
∑16 PAHs	Emission from meat frying using sunflower oil	5.037-10.025	Present study
	Emission from meat frying using frying oil	3.978-8.078	

The different levels of PAHs observed in the published literature for PAHs may be due to fuel type used for cooking, oil type, fat levels, cooking temperature, cooking style, sampling and analysis technique, etc [58].

For example, Anwarul Hasan detected 18 PAHs in dust samples from sixty indoor kitchens from households using wood, kerosene, and gas stoves. The total 18 PAHs were in the range from 8.7 to 36.8 μg/kg for kerosene stoves, from 4.3 to 61.5 μg/kg for wood stoves and from 8.9 to 32.2 μg/kg for gas stoves [28].

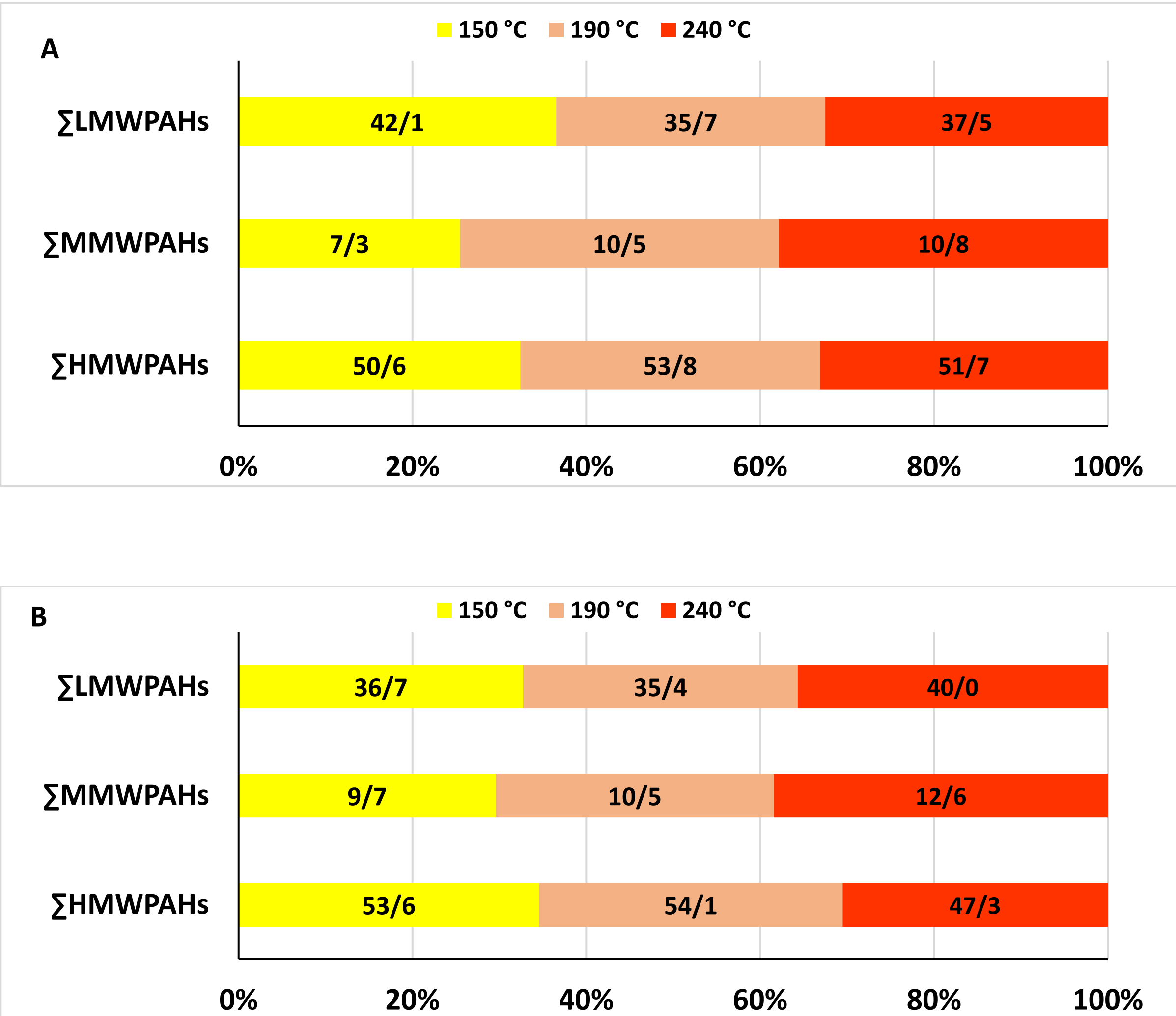


Fig. 2. Molecular distributions of 16 PAHs emitted into indoor air during meat frying using, (A) sunflower oil and, (B) frying oil based on mean values

Influencing factors in the PAHs emission

Generally important parameters that affect indoor air quality due to cooking activities in kitchens are the type foods/ingredients used, cooking styles, temperature, dishes being cooked, type of fuels, and the ventilation conditions [93, 94]. These could be the primary reasons why cooking in different studies produce different concentrations and types of pollutants. Cooking time and temperature are parameters which both affect the release of PAHs in food [95]. The results of the present study showed PAHs increase with temperature. These results is consistent with a those in literature [91, 96, 97]. It is reported that PAH concentrations increased from 3.48 to 7.92 μg/kg in meat by a heating duration increase from 15 to 30 min at 80 ◦C [98]. Scientists have reported that LMWPAHs predominantly form at moderate cooking temperatures (from 200 to 300 ◦C) via the degradation of organic compounds including proteins, fats, and carbohydrates in foodstuffs [99, 100]. Cooking at high-temperatures like those in pan-frying, deep-frying, grilling, and stir-frying, increase temperatures near to 300 ◦C, increase the pyrolysis of fats and also intensify the deterioration rate of organic materials in foods leading to more PAHs formation. It is reported that levels of PAHs in these cooking methods can reach to sixteen times greater than those when steaming or boiling methods are used. It is worthy to note that, these cooking conditions generate higher proportion of HMWPAHs, sometimes up to 30.97% [6, 101].

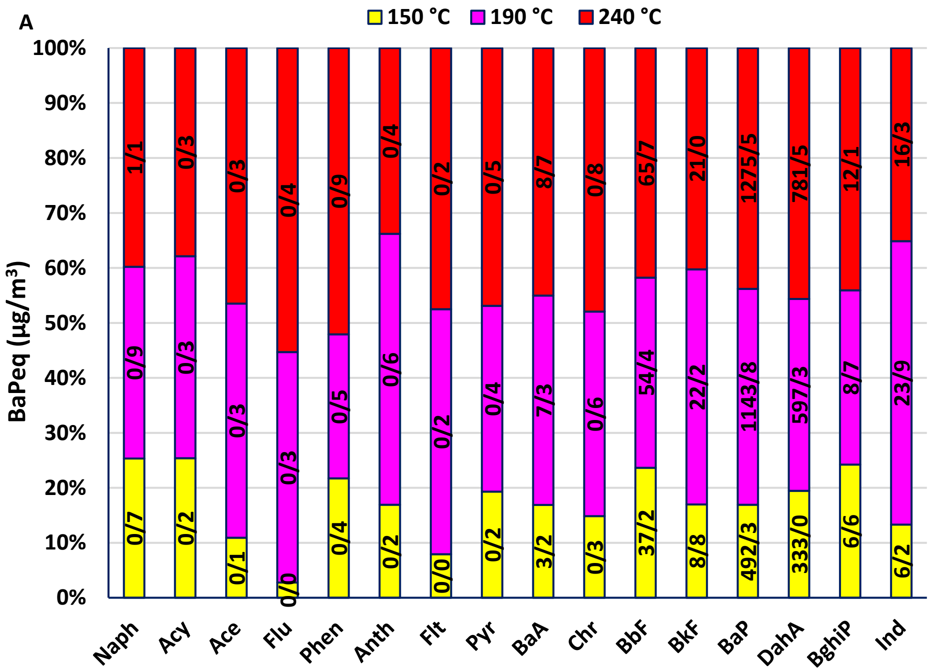
Moreover, direct flame cooking provides higher temperatures exceeding 500 ◦C compared to indirect flame, leading to more PAHs emission [102]. The higher amount of PAHs is due to the close proximity of foods to a flame source, inducing higher temperatures, and smoke and tar particles resulted from oil droplets lead to the formation of more levels of polycyclic aromatic hydrocarbon [103]. Generally, higher content of fat in food results in higher amount of PAHs formation during cooking [91, 104-106]. For example, pork meat grilling have shown higher (1.5-1.8 times) concentrations of PAHs compared to beef meat [107]. Levels of fat in the beef in this study were in the range of 25-30 % and meat was fried using indirect- fire deep frying method. In a study, PAHs production during meat and fish grilling was explored. They reported that the PAHs level released by meat during grilling process was approximately 0.420 μg/m^3 which was two times higher than that produced during fish grilling. The higher PAHs production was due to the higher amount of unsaturated and saturated fatty acids in meat, use more oil and seasonings in meat grilling compared to fish, and higher surface area of the meat (meat is almost cut into small pieces but fish is commonly grilled whole) [91].

Type and amount of oil is another important factor in PAHs formation. Researchers have reported levels of particulate matter release rates from some cooking oils (peanut, soybean, canola oils and lard) utilized for cooking at various temperatures. Their findings showed that when deep-frying was done at a frying temperature equal to 180 ◦C, olive oil and lard releases the highest concentrations of aerosols [108]. In another study, high carcinogenic levels of BaA, BaP, and DahA were found in the smoke when lard, soybean oil, and peanut oil were used at temperature 250 °C for 30 min[109]. Other researchers explored the PAHs levels generated during cooking with different oils. The order of PAHs levels in their study were as: rapeseed oil > olive oil> peanut oil> soybean oil [97]. It is reported that in general, gas stoves releases more amounts of pollutants when compared to electric stoves [110]. In another study, exposure of kitchen staffs in 4 different types of restaurants in Sweden (Large scale, European, Fast food, and Asian) was comprehensively investigated. The results showed that workers in the Asian kitchens exposed to higher levels of total PAHs

[111]. By the comparing the related studies in literature, it can be concluded that significant differences in PAHs levels in cooking settings in nations was found mainly because of differences in the stages of economic development, extent of resource availability, and cultural traits and customs.

Distribution of BaPeq during meat frying

Distributions of BaPeq during meat frying are depicted in Fig. 3. During meat frying using sunflower oil, the BaPeq values in the present study (µg/m^3) ranged from 0.000032 to 0.992 (mean 0.120) at 150 °C, 0.000065 to 1.83 µg/m^3 (mean 0.19) at 190 °C and 0.00012 to 2.1 µg/m^3 (mean 0.22) at 240 °C, respectively. Moreover, ∑ BaPeq values were in the ranges 1.005-1.60, 1.366-2.511 and 1.196-3.055 µg/m^3 at 150, 190 and 240 °C, respectively. During meat frying using frying oil, the values of BaPeq obtained in this work (µg/m^3) were 0.000032-0.744 (mean 0.11) at 150 °C, 0.000064-1.531 µg/m^3 (mean 0.177) at 190 °C and 0.00012-2.015 µg/m^3 (mean 0.188) at 240 °C, respectively. For frying oil, ∑ BaPeq were in the ranges 0.738-1.372, 1.133-2.119 and 0.870-2.720 µg/m^3 at 150, 190 and 240 °C, respectively. Previous studies have reported ranges of 0.001-0.372 µg/m^3 and 0.041-0.233 µg/m^3 for BaPeq from family kitchens [112] and commercial cooking workplaces [88], respectively. In another study, BaPeq in Chinese, Western, fast food and Japanese restaurants ranged from 2.95 to 5.20 (mean 4.07), 3.01 to 6.70 (mean 4.86), 0.484 to 0.715 (mean 0.60) and 0.314 to 0.598 (mean 0.486) µg/m^3, respectively [85]. Furthermore, values of ∑BaPeq for twenty one PAHs in exhaust stack of Chinese, western and barbeque restaurants were 1.82. 0.86 and 0.59 µg/m^3, respectively [86].



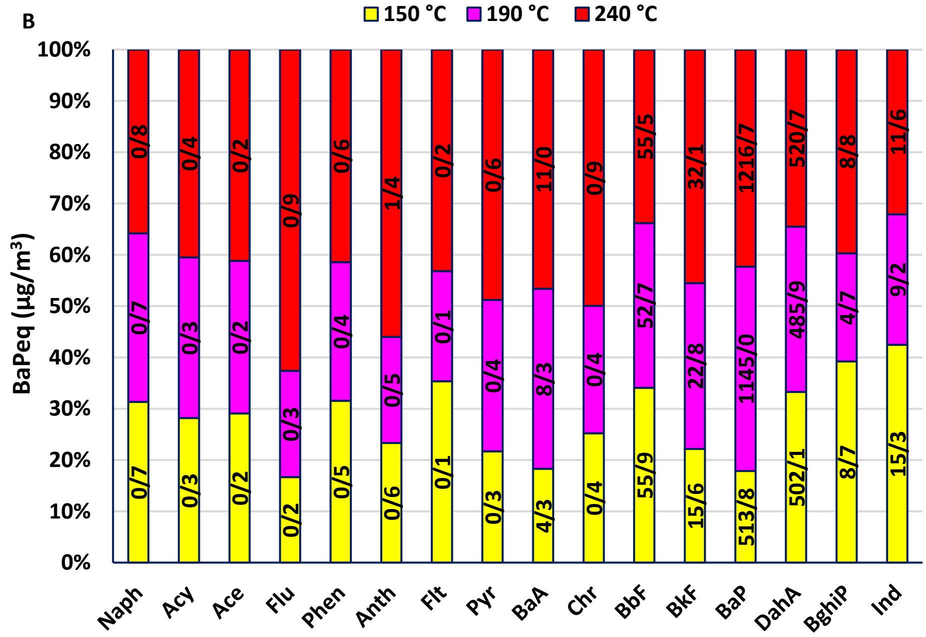


Fig. 3. Distribution of BaPeq during meat frying using, (A) sunflower oil and, (B) frying oil (based on average values) in this study

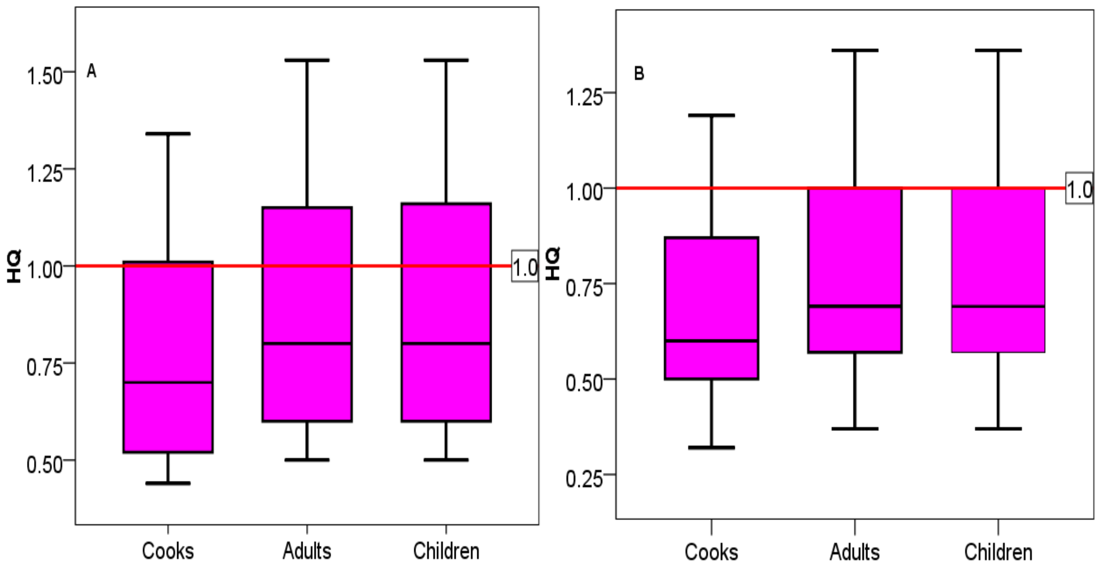


Fig. 4. Non-cancer risk through the inhalation of PAHs released during meat frying using, (A) sunflower oil and, (B) frying oil for cooks, adults and children

Health risk assessment

Non-cancer risk estimation

The non-cancer risk from PAHs exposure was estimated for inhalation route. Non-cancer risk from inhalation of PAHs using, sunflower oil and, frying oil for cooks, adults and children are provided in Fig. 4. HQs associated with PAHs exposure during meat frying using frying oil for cooks, adults and children were in the ranges of 0.440-1.338 (0.769), 0.503-1.527 (0.879) and 0.504-1.531 (0.881), respectively. For frying oil, HQ values were in the ranges of 0.32-1.19 (0.69), 0.37-1.36 (0.79), and 0.37-1.36 (0.79) for cooks, adults, and children, respectively. In this study, exposure to PAH compounds through inhalation for the different age groups were estimated as: children > adults > cooks. The results showed that meat frying with non-frying oil emitted more PAHs, consequently more HQs. The HQs values were seen in both unsafe (HQ>1) and safe ranges (HQ<1) for all the groups, indicating that there is the possibility of considerable non-cancer risk.

Cancer risk of PAHs

Kitchens are places where cooks, workers routinely exposed to numerous gaseous and particulate pollutants released during the processing of foodstuffs using different cooking techniques. In 2010, the International Agency for Research in Cancer (IARC) considered the pollutants from high-temperature cooking in the category of probable carcinogenic compounds to people [113]. Previous works have linked a potential relationship between exposure to PAHs and increased cancer incidence in human [114]. For example, it is reported that exposure to high levels of BaP compound can cause pulmonary tumors, stomach tumors, and lung tumors [95]. Deep frying, a universally popular cooking technique, presents unique conditions that increase the production and release of PAHs. In the present study, the EPA's risk assessment equations (equations 1-6) were employed to estimate the cancer risk of PAHs emissions during meat cooking. Cancer risk from PAHs exposure in indoor air during meat frying is shown in Fig. 5. The inhalation CR values through exposure to meat frying cooking emissions using sunflower oil in the present study for cooks, adults and children were 1.4E-04-4.2E-04 (2.4E-04), 2.8E-05-8.6E-05 (4.9E-05), and 7.7E-06-2.3E-05 (1.3E-05), respectively. For frying oil, the CR values were as: 1.0E-04-3.7E-04 (2.2E-04) for cooks, 2.1E-05-7.6E-05 (4.4E-05) for adults and 5.63E-06-2.08E-05 (1.2E-05) for children. Based on the estimations in this study and considering US EPA cancer risk classification, in long-term exposure basis it may high incidence of cancer risk be seen in cooks. But the cancer risk for adults and children is probable mainly due shorted exposure periods compared to cooks. The cancer risk in the study age groups in the present study decreased in the order of cooks > adults > children. Children exhibited the lowest cancer risk and despite their higher inhalation rates, their cancer risk from PAH compounds was relatively low mainly because they participate less in food cooking and frying and have smaller body size and body surface area, which in general diminishes their exposure extent to contaminants released into indoor air from cooking activities. In total, meat frying by commercial sunflower oil exhibited more cancer risk.

Cancer risk in previous studies in literature were in the ranges from 6.15×10^{-8} to 1.40×10^{-5} in household kitchens in Taiwan for men and women [112]; 2.6×10^{-6} to 31.3×10^{-6}, 1.5×10^{-6} to 14.8×10^{-6}, and 1.31×10^{-6} to 12.2×10^{-6} for Chinese, Western, and barbeque restaurants, respectively through inhalation, skin contact and ingestion pathways [86]. These results are similar to the results in the current study. In an investigation conducted by See et al., the CR values were 4.08×10^{-3}, 1.21×10^{-2} and 1.07×10^{-3} at Chinese, Malay and Indian stalls, respectively, indicating cancer risk for the exposed individual. These values are much greater than the limit of 10^{-6} for cancer risk proposed by the US EPA [58] which is also greater than those obtained in the present study. In a recent review, the cancer risks due to PAHs emission from food cooking across the world were in the ranges of 2.23E-11-8.07E-6, 2.05E-11-8.07E-6 and 7.0E-12-2.54E-6 for males, females and children, respectively. In total, female group demonstrated relatively higher cancer risk estimations, which can be due to their more participation in cooking activities and subsequently higher exposure in cooking settings. Some factors including social and cultural habits may cause more cooking activities by women, resulting in increased and continuous exposure to PAHs, therefore elevating the risk of cancer development [6].

In study in villages of China, effect of solid fuel cooking on PAHs formation was investigated. The results showed that the highest cancer risk was from ambient air PAHs exposure and within the main study sites of neighboring buildings was above the recommended limit of 1×10^{-6} [115]. In continental scale comparison, the total cancer risk has been reported higher in Africa and Asia while the risk is relatively lower in the Americas and Europe which was attributed to differences in development status, types cooking fuels, and ventilation conditions. Developed countries usually use cleaner energy sources and more efficient ventilation techniques [6]. Respiratory exposure frequency and PAH concentrations during food cooking may differ significantly among individuals and nations due to the differences in types of cooking techniques, fuel types, meat types, etc. In this study, the carcinogenic risk estimated for cooks exceeded safe threshold. In general in this study, PAHs emitted by meat frying pose a significant threat to cooks, with long-term inhalation of these indoor air pollutants can significantly increase the risk of cancer development. This finding was also consistent with the result of a previous research. Due to greater PAH formation with fat contents of meat, it is suggested to reduce the contents of fat prior to frying via public education.

This work provides essential information indoor air emissions of PAHs during meat frying. However, the findings of this study might somewhat underestimate/overestimate the actual risk and cannot be generalized to all kitchens and commercial cooking settings because the analyses were limited to a laboratory study in which the primary cooking method was frying of meats. Furtherer studies should be done in real cooking places for obtaining more reliable and comprehensive results since many factors affect on the release of PAHs during cooking. Additionally, there are also uncertainties associated with health risk assessment especially in the use of exposure factors and reference values.

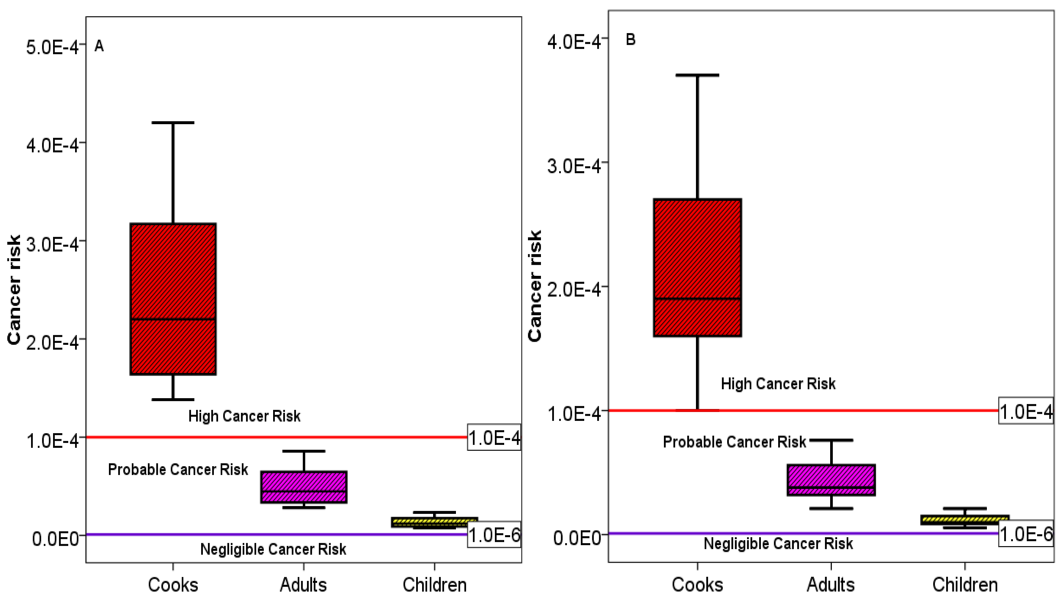


Fig. 5. Cancer risk of PAHs from exposure to PAHs during meat frying using, (A) sunflower oil and, (B) frying oil for cooks, adults and children

Conclusion

This study undertakes an exploration of the levels of PAHs and their possible health risks from inhalation of these air pollutants during meat frying. In the present study, levels of PAHs ranged from 0.032 to 2.060 (0.579), 0.032 to 1.830 (0.521) and 0.032 to 2.10 (0.668) µg/m^3 during meat frying using sunflower oil at 150, 190 and 240 °C, respectively. But in the case of frying oil usage, levels of PAHs ranged from 0.032 to 1.013 (0.384), 0.032 to 1.531 (0.494) and 0.032 to 2.015 (0.715) µg/m^3, respectively. These results clearly showed less amounts of PAHs are emitted when frying oil was used. Based on the estimations in this study and considering US EPA cancer risk classification, exposure of cooks in long-term basis can cause high cancer risk. But the cancer risk for adults and children is probable. The cancer risk in the study age groups in the present study decreased in the order of cooks > adults > children. In conclusion, meat frying by commercial sunflower oil exhibited more cancer risk. The results of this study, which show the health risks associated with PAH emission from meat frying, emphasize the urgent need for targeted interventions to take requisite steps and appropriate measures to mitigate and control harmful human health effects to upgrade indoor air quality and public health.

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Competing interests

All the authors of the present study conclude that they have no any competing financial interests or personal relationships that could affect the findings provided in this manuscript.

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Ethical considerations

This research was approved by Gonabad University of Medical Sciences, Iran (Ethical code: IR.GMU.REC.1398.074). Ethical issues (Including plagiarism, Informed Consent, misconduct, data fabrication and/or falsification, double publication and/or submission, redundancy, etc) have been completely observed by the authors.

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