JOURNAL OF NUTRITION FASTING AND HEALTH

The Occurrence and Levels of Polycyclic Aromatic Hydrocarbons in Vegetables, Fruits, Tea Samples Marketed Iran: A Systematic Review

Seyedeh Belin Tavakoly Sany [1]

1. Department of Health Education and Health Promotion, Faculty of Health, Mashhad University of Medical Sciences, Mashhad, Iran.

ARTICLEINFO

Article type:
Review Article

Article History:
Received: 04 Dec 2021
Accepted: 18 Dec 2021
Published: 27 Dec 2021

Keywords:
PAHs
Health risk assessment
Pollution
Food safety
Public health

ABSTRACT

Cultivation of vegetables and fruits is a main food sources for human population. However, these products can be polluted by different types of contaminates like polycyclic aromatic hydrocarbons (PAHs). Thus, this systematic review served as a comprehensive report on the occurrence, levels, and health risk effects of PAHs in vegetables and fruits samples marketed in Iran. The Preferred Reporting Items for Systematic Reviews and Meta-Analysis (PRISMA) were used to develop this systematic review, and four databases (Web of Knowledge, Scopus, Scientific Information Database (SID), and Google scholar) were searched from inception until November 2021. A total of 12 studies with data on 1447 samples were met inclusion criteria and included in the final report. The average total PAHs recorded in various samples showed that PAHs concentration in most of the samples was above the standard limits and the mean CRs ranged from low to very high levels of health risks for both children and adults, which is associated with harmful epidemiological and environmental effects. Therefore, there is the need to take up the bodies or institutions involved in policy implementation and environmental management to reduce and prevent PAHs pollution agriculture products in Iran. Furthermore, gaps in literature have been addressed to provide a basis for future studies.

► Please cite this paper as:
Tavakoly Sany SB. Carcinogenic Polycyclic Aromatic Hydrocarbons in Vegetables Marketed in Mashhad: Levels, Dietary Intakes, and Health Risk. J Nutr Fast Health. 2021; 9(4): 263-271. DOI: 10.22038/JNFH.2021.62033.1369.

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a commonly known group of persistent organic pollutants that are made up of two or more fused benzene rings[1, 2]. The production of PAHs is usually produced by anthropogenic activities such as pyrolysis of organic compounds, incomplete combustion of carbon, local food processing practices, the use of leaded petrol, intense oil exploration, and poor attitude and knowledge toward burning and dumping of toxic waste[3, 4]. Based on the number and molecular weight of benzene rings, it is classified into heavy PAHs (HPAHs, more than 4 rings) and light PAHs (LPAHs, 2–4 rings)[5, 6]. PAHs contamination is common contaminates in all parts of the environment and various food products and have mutagenicity, carcinogenicity, teratogenicity, and posing a critical threat to human health. Because of the adverse effects of PAHs on human health, the United States Environmental Protection Agency (US EPA) has provided a reference standard to examine the degree of PAH's contamination based on 16 US EPA priority PAHs[2, 7]. These 16 PAHs have been widely used as acceptable standards to monitor PAHs in many fields, especially in food and environment studies[8]. These 16 US EPA priority PAHs in the food include urine (FLO), naphthalene (NAP), acenaphthylene (ACY), phenanthrene (PHE), fluoranthene (FLA), acenaphthene (ACE), benz[a]anthracene (BaA), anthracene (ANT), benzo[b] uoranthene (BbF), pyrene (PYR), chrysene (CHR), benzo[g,h,i]perylene (BghiP), benzo[a]pyrene (BaP), benzo[k]uoranthene (BkF), dibenzo[a,h] anthracene (DahA), and indeno[1,2,3-cd] Pyrene (IcdP). Among these PAHs, BaP , BaA, and DhA are classified as PAHs with carcinogenic effects on humans (Group 2A) by the IARC[9-11].

Notably, ingestion, inhalation, and contact of dermal are the main pathways of contact to PAHs as well as a critical concern related to PAHs compounds occurring via ingestion routes[11]. The great effect of food on the non-occupational exposure to PAHs is well documented. For example, in non-smokers, 70% of PAH exposure is attributed to food[12, 13]. Thus, a study on the status of PAHs compounds in food needs more attention. Likewise, PAHs can migrate from the

* *Corresponding author:* Seyedeh Belin Tavakoly Sany, Department of Health Education and Health Promotion, Faculty of Health, Mashhad University of Medical Sciences, Mashhad, Iran. Tel: +989150060122, E-mail: belintavakoli332@gmail.com.

environment media (soil, air, and water) to raw food (such as vegetables and fruits) and crops[14, 15]. Furthermore, verity factors influence the accumulation rate of PAHs in raw food including the concentration of PAHs in environmental, physiological properties of fruits and vegetables, and soil characteristics[9, 11].

In Iran, several studies examined the occurrence and sources of PAHs in environmental samples and their risk that affect public health[4, 9]; but these researches are still disconnected because these studies have not been well organized as a comprehensive report and general view on the situation of PAH contamination in Iran. In consequence of the health risks and adverse effects posed by PAHs compounds, there are more concerns for public health to be well-informed about PAH's levels and their possible health risks presented in foods products, especially, raw food such as vegetables and fruits[3, 8]. Likewise, providing comprehensive data on PAH's contamination levels and distribution is very essential to improve management programs related to food safety. It seems essential to develop a general report on the PAH-related surveys in food conducted in Iran. Thus, this systematic review served as a comprehensive report on the occurrence, levels, and health risk effects of PAHs in vegetables and fruits samples marketed in Iran.

Methods

Study Design

We used the Preferred Reporting Items for Systematic Reviews and Meta-Analysis (PRISMA) to develop this systematic review [16]. The main outcome of interest for the review were occurrence and levels of PAHs compounds in vegetables and fruits and the level of risk originated from the effects of PAHs exposures to human's health.

Search Strategy

In this study, four databases were searched, including the Web of Knowledge, Scopus, Scientific Information Database (SID), and Google scholar, from inception until November 2021. The search was independently conducted for each database, and references of studies were cross-checked for identification of literature that related to PAHs exposure and their adverse effects on humans. The reference list of all eligible studies was also hand-searched to find other relevant studies that may have been ignored by the search process. We used keywords related to the term "polycyclic aromatic hydrocarbons" and "PAHs". Then, these keywords were combined with the terms "vegetables", "fruit", "health risk", "Iran.

Table 1. Quality assessments of the included studies based on Newcastle–Ottawa scal

Code	Authors (years)	Standard sampling protocol indicated	Period of sampling indicated	QA/QC conducted	All objectives achieved	Report on PAH sources	Report on mean PAH levels	Overall quality score
1	Mojtaba Yousefi et al 2018	Yes	No	Yes	Yes	No	Yes	4/6
2	Fariba Khaliliet et al 2019	Yes	yes	Yes	Yes	No	Yes	5/6
3	Seyedeh Faezeh Taghizadeh 2021	Yes	yes	Yes	Yes	No	No	4/6
4	Maryam Fahimdanesh 2011	No	No	Yes	Yes	No	No	2/6
5	Nabi Shariatifar et al 2021	Yes	yes	Yes	Yes	No	Yes	5/6
6	Zohreh Taghvaee et al 2016	Yes	No	Yes	Yes	No	No	3/6
7	Safa Kalteh 2020	Yes	yes	Yes	Yes	Yes	Yes	6/6
8	Soghra Bahrami et al 2019	yes	yes	Yes	Yes	Yes	Yes	6/6
9	Arash Asfaram 2020	No	yes	Yes	Yes	No	No	3/6
10	Seyedeh Faezeh Taghizadeha 2021	Yes	yes	Yes	Yes	No	Yes	5/6
11	Aliakbar Roudbari 2020	Yes	yes	Yes	Yes	No	NO	4/6
12	Mehdi Khiadani 2020	No	yes	Yes	Yes	No	Yes	4/6

Inclusion and Exclusion Criteria

We included studies in this review that carried out in Iran, published in the Persian and English language, reported concentration of PAHs compound or level of health risk in vegetables and fruits in Iran, and focused on subjects such as the exposure to PAHs and public health effects of PAHs in Iran. The study selection for inclusion eligibility was conducted by scanning the titles, abstracts, and full texts of retrieved articles. All review studies and duplicate researches were excluded.

Table 2. Characteristics of included studies

Code	Study area	sample/ sample size	Extraction	Analytical method	Type of PAHs	PAHs concentration (µg/ kg)	Outcome
1	Iran´s market (Tehran)[19]	Vegetable oils: frying, blended, sunflower, corn (n=23)	ISO 15753 2nd	HPLC-FLD	16 PAHS	from 12.63 to 129.30	Most samples were above the standard limits ; highest concentration related to frying> blended > sunflower> corn
2	Tehran[7]	32 different vegetables and fruits (n=192)	MSPE	GC-MS	16 PAHS	Fruits: from 123.2 to 252.4/ vegetable: 104.7 to 314.9	Most samples were above the standard limits:Mean concentration of the NAP and ACE in all vegetable and fruit samples had the highest level; the highest concentration observed in cucumber and tomato
3	Gorgan,Rijab, Rudbar,Semnan, Shiraz, Tarom[18]	Iranian olive from six different cultivation zones (n=1800)	UE method 20mLof DCM	GC-MS	16 PAHS	From 0.1 to 0.923	All samples were lower than standard limits; Highest concentration related to Bap and Chr
4	Iran´s market[23]	Olive oil (n=7)	UE method 50mL DCM	HPLC	13 PAHS	0.12 to 4.63	Highest concentration related to Phe, Bap, Chr, and ACE; All samples were lower than standard limits.
5	Tehran[13]	edible mushrooms (n= 120)	MSPE and gas	GC-MS	16 PAHS	from 0.82 to 6.25 (average: 3.85 ± 0.24	All samples were lower than standard limits; Ace had the maximum mean level
6	Tehran[24]	olive oil	UALL	HPLC-FLD	15 PAHS	Average: 19.05	Most samples were above the standard limits; The highest concentration related to NPH and ACE
7	Tehran[3]	leafy vegetables: lettuce, cabbage, celery and spinach (n= 24)	MSPE	GC-MS	16 PAHS	Ranged from 9.98 to 51.6	Most samples were above the standard limits; LPAHs was dominance in all vegetables; The rank order: lettuce (51.61 µg/kg) > cabbage (28.13 µg/kg)> spinach (24.85 µg/kg) > celery (9.98 µg/kg)
8	Ahvaz[4]	26 different vegetable (n= 105)	EPA method 3630C	GC-MS	16 PAHS	For spinach, parsley, and dill is 49.8, 496.4, and 442	Most samples were above the standard limits; Nap, Ace and AceP have high concentration;
9	local markets in Yasouj[25]	vegetables and Fruits (n= 12)	MD-µ SPE	HPLC	4 PAHs (Nap, Ant Pyr, Phe)	Fruits: from 113.2 to 2022.4 / vegetable: 142 to 187	All samples were above the standard limits
10	Abadan, Bam, Dashtestan , Jahrom, Minab, and Saravan[20]	Samples of date fruits(n= 900)	MSPE	GC–MS	16 PAHS	Ranged from 2.26 to 4.3	Most samples were lower than the standard limits; Bap has highest concentration.
11	Iran main Market[21]	Commercial tea and coffee (n=64)	MSPE	GC–MS	16 PAHS	From 4.77 ± 1.01 to 13.75 ± 2	Most samples were lower than the standard limits; NPH had the highest mean in the samples
12	Iran main Market[21, 22]	eight brands of black tea	MSPE	GC–MS	16 PAHS	139 to 2082	Most samples were higher than the standard limits; PAHs with 5 to 6 rings were not found in the teas samples. Four rings PAHs compounds composed 46% of the total PAHs compounds and were the most dominant compounds

MSPE: Magnetic solid-phase extraction, **UALL:** modified ultrasound-assisted liquid–liquid; **MD-µSPE:** magnetic dispersive micro-solid phase extraction, **HPLC:** liquid chromatography, GC–MS: chromatography-mass spectrometry, **LPAHs:** Dominance of low molecular weight PAHs, 10 µg/kg set for PAHs compound as standard limit.

Data extraction and quality assessment

In this study, a set of basic data were extracted as follow: the publication year/author, study objective, study area, period of sampling, sample size type(s) of samples, quality control/quality assurance, analytical technique, number of PAHs and PAH concentration, and potential pollution sources. The quality of the selected studies was examined via Ofori and Cobbina adapted the Newcastle–Ottawa Scale (NOS)[17]. This scale includes six items that were provided with either a "no" or "yes" response to make a general score for each study (Table 1).

Table 3. PAHs Health risk situation in different samples

Code	samples	Health risk level (ILCR)	Other
1	Vegetable oils: frying, blended, sunflower, corn (n=23)	Child: from 3.17E-5 to 3.76E-5, Adult: 4.17E-6, 5.20E^{-6}	The rank order of edible oil based on CR: frying> blended > sunflower> corn
2	32 different vegetables and fruits (n=192)	Vegetables: 5.2×10-5; fruits is 7.7×10-5	Highest ILCR was related to cucumber (5.1×10-4) and tomato (4.3×10-4)
3	Iranian olive from six different cultivation zones (n=1800)	CR: from 4.90×10−14 to 3.30×10−13	No risk.
5	edible mushrooms (n= 120)	Cr for adult: 6.85E-08; Child: 3.47E-07	No risk
7	leafy vegetables: lettuce, cabbage, celery and spinach (n= 24)	Cr: 3.88E-5/ HQ<1	BaPeq and body weight as the most influential parameters.
8	26 different vegetable(n= 105)	CR is 3.1×10^{-5} to 1.5×10^{-4}, 6.3×10^{-5} to 3.6×10^{-4}, and 1.3×10^{-4} to 9.5×10^{-4}, for spinach, parsley and dill, respectively	local population is high cancer risk through consumption of PAH contaminated vegetables
10	date fruits (n= 900)	Cr: 5.39 × 10− 12 to 8.76× 10− 12	No risk.
11	Commercial tea and coffee (n=64)	CR for ranged adult: 1.82E-8 to 9.29E-9T CR for child ranged from 8.77E-8 and 4.25E-8; HQ for both adult and child <1	No risk.

HQ: target hazard quotient, less than 1 indicates acceptable hazard risk; **CR:** incremental lifetime cancer risk, 10^{-6} is acceptable cancer risk level and indicates acceptable or negligible risk.

Results

Search Outcome and quality assessment

The inclusion and exclusion criteria were implemented to select eligible studies (Figure 1). A total of 32 published articles were selected for evaluation from which 12 studies with data on 1447 samples were met inclusion criteria and included for the final report. All information was extracted from 11 different cities of Iran of which Tehran (capital of Iran) represented the highest number of studies (Table 2).

For the quality assessment of eligible articles, about 42% of the studies showed that appropriate quality and ranged from 5 to 6 out of 6[3, 4, 7, 13, 18]. All these studies used a standard protocol for sampling, valid method for exposure measurement compounds, and provided a valid and reliable way for outcome measurement. The quality of 4 (33 %) studies was moderate with an average score of 4 [19-22], and about 25% of the articles showed low quality (from 2 to 3) [23-25] and their major reasons were: a lack of standard protocol for sampling, non-indication of mean concertation of PAH levels, PAH sources, and period of sampling (Table 1).

Study samples characteristics

Of 12 included articles, 5 (47%) studies included vegetables and fruits samples[3, 4, 13, 22], 3 (25 %) studies included edible vegetable oils[19, 23, 24], and other studies (33%) included black tea[21, 22], coffee[22], date fruits [20], and olive[23] (Table 1). Most of the studies have measured 16 PAHs (75%) compounds[3, 4, 7, 13, 18, 19, 21, 22], but there are three studies focused on measuring the 13 PAHs[23], 4 PAHs[24], and 15 PAHs[25]. In this review, 4 (33%) and 8 (67 %) studies measured PAHs using liquid chromatography (HPLC)[7, 19, 24, 25] and chromatography-mass spectrometry (GC–MS)[3, 4, 13, 18, 21-23], respectively. Our findings showed that chromatography-mass spectrometry (GC–MS) was the most common valid method used to measure PAHs compound in Iran, while other tools were rarely implemented. Most studies (6: 50%) extracted PAHs compound using the magnetic solid-phase extraction (MSPE)[3, 7, 13, 20-22], and 2 (17 %) studies extracted PAHs using the UE method with 50mL DCM[20, 23], and other studies used different methods for extraction[4, 24, 25]. Based on results, 47% of the studies were conducted in

Tehran[3, 7, 13, 21, 22, 24] and the sample size was varied from 7 to 900. The first articles related to measuring PAHs compound in vegetable and fruit samples were published in 2018 (Table 2).

Levels of PAHs in samples

Based on results in this review, PAH concentration ranged from 0.1 to 2082 µg/ kg. Of the 12 studies that measured PAHs concentration in vegetables, fruit, olive and tea, 8 studies (67%) reported that PAHs concentration in most of their samples were above the standard limits (10 µg/kg)[3, 4, 7, 19, 22, 24, 25] which set by the European Union and the Standard Organization of Iran[9]. Most implicated PAHs included NAP, ACE, Bap, Chr, Nap, Ace, and AceP.

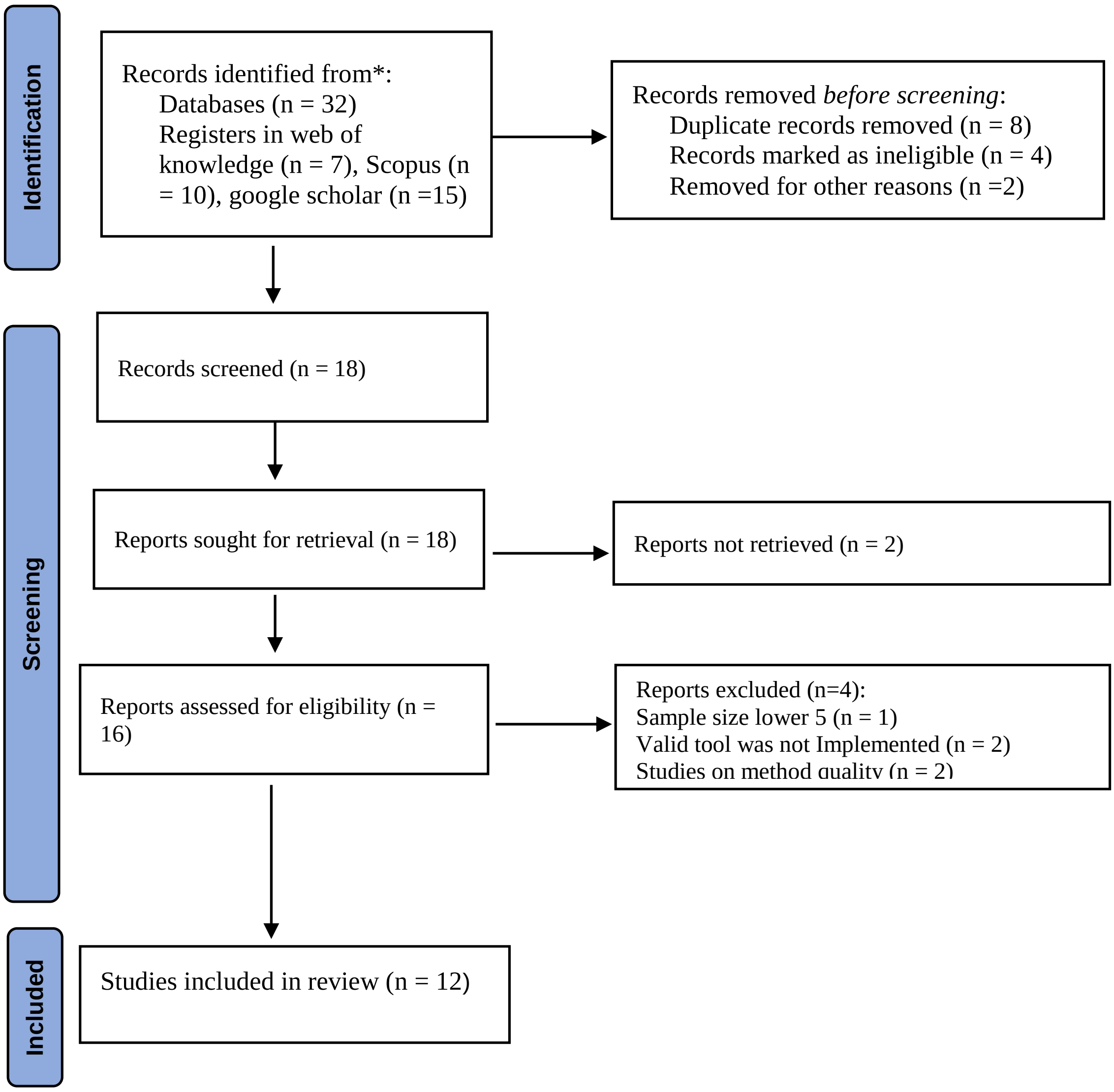

Figure 1. PRISMA flowchart for studies selection

Risk Assessment of PAHs

Of the 12 studies included in a systematic review, 8 studies assess health risks and cancer risk (CR) in vegetables and fruit samples [3, 4, 7, 13, 19-21] (Table 3). Based on risk results, 3 included studies reported moderate risk levels (HQ< 1, 10^{-6} <CR <10^{-4}) via consumption of edible vegetable oils and different vegetables and fruits [3, 7, 19], and only one study indicated a high level of risk

(HQ> 1, 10^{-4} <CR) [4] through consumption of vegetables that were contaminated with PAHs compounds. Others studies indicated acceptable or negligible risk (HQ< 1, CR <10^{-6}).

Discussion

The main objective of this systematic review was to summarize the results of studies conducted on PAH levels and health risk assessments of PAHs in vegetables and fruits samples marketed in Iran. In this review, 12 published studies were identified that measured PAH levels in vegetables and fruits samples marketed Iran; among them, 8 studies assessed the health risk effect of PAHs exposure on human health [3, 4, 7, 13, 19-21]. As our finding showed, most studies on vegetables and fruits samples reported that PAHs concentration in most of their samples was above the standard limits set by the European Union and the Standard Organization of Iran [3, 7, 19]. PAHs level ranging from 12.63 to 129.30 μg/kg. The high concertation of PAH in fruit and vegetables in marketed Iran may occur via different pathways. The main pathway of PAHs is related to environmental media, which is related to the presence of PAH in air, soil, and water[3, 8]. Several studies in Iran show the high level of PAHs in environmental media especially cultivation soils[4, 5, 15]. Therefore, PAHs contamination in vegetables and fruits could be a growing national concern in Iran, like results from some PAHs researches that were conducted on the food products in other countries such as Africa, China, and Europe [17, 26]. Jia et al. measured the PAHs concentration in vegetable samples collected near the industrial area of Shanghai, China[27]. Concentrations of PAH was ranged from 65.7 to 458.0 μg/kg in leafy vegetables (Shanghai green cabbage, chines cabbage, and romaine lettuce), pod and seed vegetables, stem vegetables, and rhizome vegetables[27]. They highlighted heavily contaminate the cultivation land of vegetables. likewise, Mohammed et al. investigate the PAHs concentration in leafy vegetables cultivated within the city center of the Accra Metropolitan[28]. They observed that BaA, BaF, BaP, Acenaphthylene, and Acenaphthene were the most frequent hydrocarbon in most samples and the total concentration of PAHs ranged from 0.037 to 16.2 μg/kg[28].

In our study, 4 studies were performed on edible vegetable oil [19, 23, 24], of which 3 studies reported PAHs concentration in all of the samples were above the standard limits and PAHs level ranging from 12.63 to 129.30 μg/kg [19, 23, 24]. Phenanthrene, ACE, and Chr, Bap were the most frequent hydrocarbon in these oil samples. This result is in agreement with several studies in the world that investigated the PAHs level types of commercial vegetable oil[11, 26]. Pandey et al. [96] examined the level of PAHs in different vegetable oil samples marketed in India[29]. They indicated that refined vegetable oils and extra-virgin olive oil displayed the lowest and highest PAH levels, respectively. Likewise, they reported that 90% of coconut oil samples and all safflower and linseed oil samples were contaminated with PAHs compounds. Chr (20.7 μg/kg) and Bap (2.6 μg/kg) were the most common hydrocarbons in most samples[29]. Ciecierska and Obiedzinski examined PAH levels in nonconventional vegetable oils (safflower, coconut, linseed oils, and evening primrose) in Poland[30]. They indicated concentrations of PAHs compounds ranging from 23.41 to 234.30 μg/kg, with phenanthrene and BaP being the most frequent hydrocarbon. Silva et al. measured PAHs contamination level in nonconventional vegetable oils in Brazil[30]. They reported that PAHs levels ranging from undetectable to 14.99 μg/kg in 96% of samples and Chr considered as the most frequent contaminants in these samples. Based on the literature, it seems that the high levels of PAHs contamination to be common in vegetable edible oils[26, 30]. This could be due to the lipophilic characteristics of PAHs which facilitate the rate of pollution of oils. However, several routes affect PAH contamination in vegetables edible oils including environmental contamination (contaminated air, soil, water, and atmospheric deposition), oilseeds drying with combustion smoke, the solvent used for oil extraction, and materials of packaging[27, 30].

The average concentration of the PAHs in tea and coffee showed that 4 rings of PAHs were the most dominant compounds (around 46% of the total PAHs)[7, 21]. In one study that conducted on eight brands of black tea, PAHs concentration in all of the samples were above the standard limits (ranging from 139 to 2082 μg kg^{-1} μg/kg) and had the highest concentration[21], which was consistent with the study of Londoño VAG et al.[31], they reported that the total 16 PAHs in black tea was ranged from 509.7 to 2746.5 μg kg^{-1} on a dry mass. In the study of web CM et al.

(2015), the concentrations of 16 PAHs in the tea and coffee ranged from and 5.2 to 913.1 and 38.7 to 593.1 µg kg $^{-1}$, respectively[11]. In 2020, Lan-Anh Phan Thi et al. indicated that the average of 15 PAH in samples of black dry and green teas was from 75.3 to 387.0 µg kg $^{-1}$dry weight[32]. In the study of Singh Groveretal, the concentration of PAH16 in Nescafe, Nescafe Classic, and Suncare were ranged from 831.7 to 1589.7µg kg[30]. Higher PAHs contamination in tea of coffee is probably due to the amount of caffeine or other raw materials, use of higher temperatures during processing, storage conditions, and packaging[4, 7].

Health risk evaluation quantifies the effect of exposure, hazard, and risk level based on the potential toxicity of chemical pollutants and exposure intensity. The risk assessment uses exposure assessment, hazard characterization, risk characterization, and hazard identification to examine the level of risk–benefit and safe concentrations. Based on our finding on health risk assessment for PAHs, 3 included studies reported moderate risk levels (HQ< 1, 10^{-6} <CR < 10^{-4}) via consumption of edible vegetable oils and different vegetables and fruits [3, 7, 19], and only one study indicated a high level of risk (HQ> 1, 10^{-4} <CR) through consumption of vegetables that were contaminated with PAHs compounds[4]. In recent decades, several studies investigated PAHs contamination in raw foods to understand whether a potential risk exists when certain foods are consumed.

Conclusion

This study is the first systematic review that was conducted on levels and health risk assessments of PAHs in vegetables and fruits samples marketed Iran. This review presented relevant information that is essential to better understand the situation of PAHs contamination in a national context. With appropriate presentation and summary of the researches on the very high to low concentration of PAHs compounds recorded in vegetables and fruits samples in Iranian markets, the public is awarded of the common activities that cause the situation of PAHs contamination in their natural environment and food sources.

The mean total PAHs recorded in the different studies conducted in various samples showed that PAHs concentration in most of the samples was above the standard limits defined by the European Union and the Standard Organization of Iran. Likewise, the mean CRs reported in the various studies indicated low to very high levels of health risks for both children and adults, which is associated with harmful epidemiological and environmental effects. Therefore, the high exposure to PAHs contamination is certainly a cause for concern because it is not devoid of PAHs implications and reduces an individual's quality of life. Studies conducted in Iran mainly focused on examining PAHs concentration and a few studies effectively assessed the adverse health effects and potential carcinogenic risks associated with PAHs contamination. It is essential for more researches to be conducted to investigate the potential health risks posed by PAHs or other persistent organic pollutants. There is thus the need to take up the bodies or institutions involved in policy implementation and environmental management to serve as a program for improving and controlling PAHs contamination. In addition, we address gaps in the literature and provide a basis for studies in the future.

Acknowledgements

We would like to thank all the vice president of research in Mashhad University of Medical Sciences.

Conflict of Interest

There is no conflicts of interest in this work.

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