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# Evaluation of polycyclic aromatic hydrocarbons (PAHs) in bottled water samples (non-carbonated, mineral, carbonated and carbonated flavored water) in Tehran with MSPE-GC/MS method: a health risk assessment

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## **Abstract**

Polycyclic aromatic hydrocarbons (PAHs) are dangerous environmental compounds that are sometimes found in food. The objective of present study was to measure the level of 16 PAHs in bottled water samples (non-carbonated or drinking, mineral, carbonated and carbonated flavored water) in Tehran by using magnetic solid-phase extraction and gas chromatography–mass spectrometry (MSPE/GC–MS) method. The limit of detections (LOD), limit of quantifications (LOQ) and recovery of PAH compounds were 0.010-0.210, 0.03-0.700 µg/L and 92.5-103.4%, respectively. The results showed that the mean of total PAHs in samples was  $2.98\pm1.63$  µg/L and the mean of Benzo[a]pyrene (BaP) was  $0.08\pm0.03$  µg/L, which were lower than standard level of the US-EPA (0.2 µg/L, BaP in drinking water). Also, our results showed that carbonated flavored water had maximum mean of total PAHs ( $4.95\pm0.8$  µg/L) and mineral water had minimum mean of total PAHs ( $1.24\pm0.8$  µg/L). The Monte Carlo method was applied to calculate the Estimated Daily Intake (EDI) and Incremental Life Cancer Risk (ILCR) indexes. In all samples, the rank order of the estimated CDI values based on the 95 percent percentile was F > B(a)A > Ace > FI > Na > Ph > B(b)F > B(k)F > B(a)P > P > Ac > A. The cancer risk and uncertainty analysis of 95th Percentile for bottled waters studied gave values lower permissible limit of  $10^{-6}$ , indicating not pose a serious concern to humans.

Keywords: Bottled water, Polycyclic aromatic hydrocarbons (PAHs), MSPE-GC/MS, Health risk assessment

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## Introduction

Nowadays, due to various natural disaster and man-made activities, large amounts of organic pollutants may be found in various water sources. Organic chemicals that pollute water are often carcinogenic and toxic and have caused concern around the world [1–4]. Organic Pollutants, especially PAHs, are found in oil, gasoline, coal, wood, natural forest fires, transit trucks, waste incineration, volcanic eruptions, tobacco smoke, industrial



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processes (foundries, steel, aluminum and iron production) in the environment, especially running water [5-8].

PAHs are classified as persistent organic pollutants (POPs). They are a class of stable chemical compounds with rings (2 or further) and are common organic pollutants (xenobiotics) in the environment. So far, more than a hundred PAHs have been discovered in nature, 16 of which [(indeno[1,2,3-cd]pyrene (IP), benzo(b)fluoranthene (BbF), benzo(a)pyrene (BaP), chrysene (Ch), benzo(k)fluoranthene (BkF), naphthalene (NA), pyrene (P), phenanthrene (Pa), benzo(a)anthracene (BaA), acenaphthylene (Ace), acenaphthene (Ac), fluoranthene (Fl), fluorene (F), anthracene (A), benzo[g,h,i]perylene (BgP) and dibenzo[a,h]anthracene (DhA)] have been classified by US-EPA (USA Environmental Protection Agency) as pollutants [9–12].

Human exposure to PAHs is thought to be linked to an augmented risk of a number of cancers (including bladder, lung, stomach and oral cancers) and other disorders (like asthma and heart diseases). Furthermore, these compounds have the ability to suppress the system of immune and are thought to be endocrine disruptive chemicals (EDCs) [1, 13, 14].

The widespread recognition of PAHs in water resources like groundwater seawater and river water is due to the increasing human activities, as well as unregulated and improper disposal of industrial wastes. Many water bodies across the world have been declared unsafe for human consumption owing to high concentrations of PAHs in these [15–17].

Bottled water is one of the most important forms of drinking water, in all over the world. The global usage of bottled water is steadily growing. The market of bottled water raised from fifty-eight to one hundred and forty-four billion liters among 1994 and 2002. Water quality must be assessed permanently as a result of the increased production and use of bottled water [18–21].

For the measurement of PAHs in water samples, several reference techniques have been developed, the most frequent of which are GC-FID, GC-MS, High Performance Liquid Chromatography-Ultraviolet (HPLC-UV), HPLC-flame ionization detection (FLD) and HPLC-Diode-Array Detection (DAD) [1, 5, 21, 22]. EPA techniques and standard methods for the examination of water and wastewater explain the pre-concentration and extraction of PAHs from samples of water by solid-phase extraction (SPE) and liquid-liquid extraction (LLE) that are extensively utilized by many researchers. Other techniques, like liquid-phase microextraction (LPME), stir bar sorptive extraction (SBSE) and solid-phase microextraction (SPME), were developed more newly. The magnetic solid-phase extraction (MSPE) by using magnetic nanoparticles (NPs) have recently emerged as a potential preparation of sample technique [18, 23–28]. Magnetic adsorbents are routinely disseminated into the sample solution directly in the MSPE technique [1, 29, 30].

The health risk estimation specifies potential adverse health impacts of PAH with regular intakes of water [29, 30]. In a specific population, ordinary health risk estimation in water is considered by THQ (Target Hazard Quotient) or EDI (Estimated Daily Intake) and ILCR (Incremental Life Cancer Risk), representing non-carcinogenic and carcinogenic hazards to the health of human, respectively [31, 32].

It should be emphasized that there is no study or very few researches on the presence of PAHs in kinds of bottled water in the world especially in Iran. Therefore, in present research, following objectives were followed: (1) to generate a simple, reliable and effective method for evaluation of PAHs in bottled water samples (non-carbonated or drinking, mineral, carbonated and carbonated flavored water) using the method of MSPE-GC/MS; (2) to compare the PAH concentrations in bottled water with the standard of the US-EPA and other studies (3); use the BaP cancer potency (as a reference) to assess the potential health risk posed by PAHs.

#### Materials and methods

## Reagents and chemical compounds

The reference standards of PAH (QTM PAH-Mix, 2000  $\mu$ g/mL) were bought from Supelco (Bellefonte, PA, USA), The other chemicals and solvents (biphenyl (as internal standard), sodium chloride, hydrochloric acid, potassium hydroxide, acetonitrile, dichloromethane, and methanol) obtained from Merck (Germany) with analytical grade. The multi-walled carbon nanotubes (MWC-NTs) were acquired with specifications of 30–60 nm diameter and 5–30  $\mu$ m length obtained from Nanoshel Co. (Panchkula, India), Our previous study was used to ready the MWCNTs-Fe<sub>3</sub>O<sub>4</sub> combination [1, 3].

## Sample collection

A total of 40 bottled water samples (non-carbonated or drinking, mineral, carbonated and carbonated flavored water) were collected (in duplicate) from marketplaces in Tehran, Iran. The samples were stored in their packaging at laboratory temperature until analysis.

# Preparation of blank sample

The distilled water was chosen as a blank sample, and the usefulness of distilled water was proven by our primary studies [4, 25].

## Preparation of standard

The stock and working mixed standard solution and also internal standard solution were prepared according our previous studies [4, 25]. Solutions of stock and working were maintained at 4 °C and used either diluted or directly on a regular basis.

## Preparation of samples and quality control

The samples preparation procedure were explained in our previous studies [4, 25] that included 3 key phases sample cleanup, analyte adsorption and analyte desorption from the adsorbent. Finally, prepared sample was injected into the GC/MS instrument [1, 25].

The results of studies optimization showed that mentioned procedure is allowed for the repeatable and quantitative PAH compounds extraction from bottled water samples. A mix of identified, certified reference PAHs (QTMPAH-Mix, 2000  $\mu$ g/mL, order number: CRM47930) and internal standard solution, without any sample was prepared and injected to the GC-MS as a quality control sample at begin of phase, middle, and ending of each sample queue. Finally, the mean values were used for quantification and all bottled water samples were evaluated in duplicate.

## Instrumental analysis

For this purpose, the GC device model Agilent 6890 with a detector of mass model 5973 selective quadrupole mass spectrometer was used (Palo Alto, CA, USA). Other conditions (such as type of column, oven and injector temperature, carrier gas etc.) were according our previous studies [1, 25]. The PAH compounds were quantified by the selective ion monitoring (SIM) mode. The

qualification was carried out by comparing the observed mass spectra and retention times to reference retention times and spectra obtained under comparable GC-MS conditions using injecting calibration standards. Each PAH analyte has one quantification and two qualifier ions, according to Table 1.

## Method optimization

Five milliliters of mixed working solution (0.5  $\mu$ g/mL) was combined with the 500 mL of blank sample, and was spiked. For 30 min, the mixture was homogenized (mechanically) and kept at 4 °C for 24 h. After then, it was utilized to optimize the method. Our earlier investigations based on "one factor at a time" tests were used to optimize the method [1, 5].

## Characterization of human health risk

In order to estimate the oral exposure dose of the harmful compound such as PAH, the daily ingestion and ILCR index of indicator PAH via the ingestion of bottled water samples was estimated by Eqs. (1, 2) according our previous studies [8, 33]:

$$BEC = \sum_{i=1}^{n} C_i \times TEF \tag{1}$$

$$EDI = \frac{C \times IRi \times EDi \times EFi}{BW \times AT}$$
 (2)

In this equation estimated daily intake (EDI) is based on the mg/kg, C is the concentration of PAH analyte based

Table 1 Selected ions used for the quantification and qualification of PAH analytes by GC-MS (SIM mode)

lon group	Analyte (PAHs)	Time window (min.)	Confirmation ions (m/z)	Quantification ion (m/z)
1	I.S. (Biphenyl)	8–13	153, 152	154
1	NA (Naphthalene)	6–13	128, 127	128
1	Ace (Acenaphthylene)	8–13	153, 151	152
1	Ac (Acenaphthene)	8–13	154, 152	153
2	F (Fluorene)	13–15	165, 167	166
3	Pa (Phenanthrene)	15–17	179, 176	178
3	A (Anthracene)	15–17	179, 176	178
4	Fl (Fluoranthene)	17–20	203, 101	202
4	P (Pyrene)	17–20	203, 101	202
5	BaA (Benzo[a] anthracene)	20–23	226, 229	228
5	Ch (Chrysene)	20–23	226, 229	228
6	BbF (Benzo[b] fluoranthene)	23–28	253, 126	252
6	BkF (Benzo[k] fluoranthene)	23–28	253, 126	252
6	BaP (Benzo[a]pyrene)	23–28	253, 126	252
7	IP (Indeno[1,2,3-cd]pyrene)	28–31	277, 138	276
7	DhA (Dibenzo[a,h] anthracene)	28-31	279, 139	278
7	BgP (Benzo[g,h,i] perylene)	28-31	277, 138	276

on mg/kg, the definition and description of the above variables are shown in Table 2. PAH concentrations were altered to concentrations of BaP equivalents (BEC;  $\mu$ g/kg) by toxicity equivalency factors (TEFs). The ILCR from exposure to PAH (BaP from group 2A, a probable human carcinogen) through bottled water samples is another approach to evaluate the risk and were calculated using Eq. 3:

$$ILCR = \frac{BEC \times EF \times ED \times SF}{BW \times AT}$$
 (3)

where *SF* denote the oral cancer slope factor of the BaP daily intake (7.3 per mg/kg/d) [34], the definition and description of variables are shown in Table 2.

# Statistical analysis

The study results were presented as mean  $\pm$  standard deviation using the program of SPSS (version 24.0), and the data for PAH concentrations in bottled water samples were checked for normality (Kolmogorov-Smirnov test) and homoscedasticity (Levene's test). Comparisons between various samples were investigated using the Mann–Whitney test (P < 0.05) for nonnormally distributed data. When the PAH analytes were not detected in samples, the mean concentration was calculated using half of LOD (1/2 LOD). A heat map was conducted to ascertain a more accurate distinction between the PAH congener in bottled water samples [3, 39]. Heat map construction (clustering method: average linkage; distance method: Pearson) was used to interpret the association between individuals online at https://biit.cs.ut.ee/clustvis/. The software of Crystal Ball (v. 11.1.2.4.600) was employed to produce simulation predictions [32].

## **Results and discussion**

## Performance validation of the analytical method

In Table 3, the optimum conditions for this investigation are listed. With a correlation coefficient in the range of 0.984-0.996, calibration curves  $(0.005-10 \mu g/L)$  were generated. The LODs was 0.010-0.210 µg/L and LOQs was 0.03-0.700 µg/L for all of the compounds, according to the validation method. The method accuracy was determined by examining the precision of intra-day and inter-day of QC samples. The tested values for repeatability and reproducibility were ranged 6-18 percent and 4.6-10.2 percent (results gathered from 3 different laboratories). The certified reference compounds of PAH (product number: CRM47930, QTM PAH-Mix, 2000 µg/ mL) was used to evaluate the percent of recovery and accuracy of technique in this investigation. The percentages of recovered items were evaluated to be between 92.5 and 103.4. As a result, the reliability and feasibility of the developed technique were approved. By examining 40 bottled water samples, the technique's selectivity was demonstrated. Finally, no interfering peaks were discovered in the region of internal standard and PAH analytes.

## **Evaluation PAHs in bottled water samples**

In Table 4, the statistical analysis of the PAH compounds in bottled water are listed. The results showed that the mean of  $\Sigma PAHs$  was  $2.98\pm1.63~\mu g/L$ . The mean of BaP was  $0.08\pm0.03~\mu g/L$  and varied from not detected (nd) to 0.17 that lower than the USEPA standard level for BaP compound in drinking water (0.2  $\mu g/L$ ). Fl had the maximum level of compounds 1.23  $\mu g/L$  and Ch, D(h) A, B(g)P and I(cd)P were not detected (nd) in all bottled water samples. High levels of PAHs contaminant in bottled water samples can be due to reasons such as primary water pollution (source), secondary pollution such as air pollution, surfaces, devices, bottles and so on [20, 27, 40].

**Table 2** Parameters used in the present study for health exposure assessment in bottled water sample

Exposure p	arameters	Unit	References
SF	Carcinogenic slope factor of oral intake (7.3)	mg/(kg/d)	[34]
C	Concentrations of PAHs	μg/kg	-
EDI	Estimated daily intake	mg/kg	
EF <i>i</i>	Exposure frequency	Days per year	[35]
IR	Average daily intake	kg/day	[36]
ED	Exposure duration	Days	[6]
BEC	Benzo(a) pyrene equivalents concentrations by toxicity equivalency factors (TEFs)	-	[37]
AT	Average time	Days	[38]
BW	Body weight (for children and adults is between 15 and 70)	kg	[32]
TEFs	Toxic equivalent factors	_	Additional file 1: Table S1

**Table 3** Reproducibility relative standard deviation (RSDR; n=6), repeatability relative standard deviation (RSDr; n=6), recoveries, linear range, LOD, LOQ and coefficient of estimation ( $r^2$ )<sup>a</sup>

Target compound	Linear range (μg/L)	Limit of detection (LOD) (μg/L)	Limit of quantification (LOQ) (µg/L)	Coefficient of estimation (r <sup>2</sup> )	Recoveries (%)	Repeatability (RSDr) (%)	Reproducibility (RSDR) (%)
Na	0.005–10	0.01	0.03	0.995	95.3	7.8	7, 11, 13
Ace	0.005-10	0.04	0.12	0.991	96.4	9.5	8, 10, 18
Ac	0.005-10	0.03	0.08	0.984	100.2	5.3	7, 9,14
F	0.005-10	0.06	0.20	0.989	92.5	4.6	6, 9, 10
Ph	0.005-10	0.02	0.06	0.993	98.1	7.7	11, 13, 15
Α	0.005-10	0.02	0.07	0.996	103.4	10.2	8, 11, 14
FI	0.005-10	0.05	0.18	0.990	101.6	7.8	8, 10, 16
P	0.005-10	0.02	0.07	0.986	98.7	8.3	11, 13, 17
B(a)A	0.005-10	0.10	0.32	0.984	99.2	6.6	10, 12, 14
Ch	0.005-10	0.19	0.64	0.994	101.4	8.6	7, 10, 15
B(b)F	0.005-10	0.13	0.43	0.996	98.8	9.3	9, 11, 16
B(k)F	0.005-10	0.11	0.35	0.989	99.2	6.9	7, 10, 14
B(a)P	0.005-10	0.15	0.50	0.987	99.1	9.5	11, 13, 17
D(h)A	0.005-10	0.14	0.46	0.995	100.3	10	9, 12, 18
B(g)P	0.005-10	0.21	0.70	0.994	102.4	9.9	10, 13, 15
I(cd)P	0.005-10	0.09	0.30	0.990	101.3	9.6	11, 14, 17

 $<sup>^{</sup>a}$  RSDR of 1  $\mu$ g/L, 5  $\mu$ g/L, and 10  $\mu$ g/L standard value (n = 6)

**Table 4** Statistical analysis of PAH compounds in bottled water  $(\mu g/L)$ 

Analyte	Min.	Max.	Mean	SD
Na	nd	0.76	0.29	0.23
Ace	nd	0.9	0.45	0.27
Ac	nd	0.32	0.06	0.1
F	nd	0.98	0.57	0.33
Ph	nd	0.3	0.13	0.11
Α	nd	0.11	0.04	0.04
FI	nd	1.23	0.37	0.36
Р	nd	0.5	0.07	0.13
B(a)A	nd	1.04	0.46	0.4
Ch	nd	nd	-	_
B(b)F	nd	0.15	0.08	0.03
B(k)F	nd	0.15	0.08	0.04
B(a)P	nd	0.17	0.08	0.03
D(h)A	nd	nd	-	-
B(g)P	nd	nd	-	_
I(cd)P	nd	nd	-	-
Total	0.69	5.84	2.98	1.63

Karyab et al. mesoured the mean levels of total PAHs in mineral bottled water and bottled drinking water in Iran and reported that the mean of total PAHs was 20.54 and 32.20 ng/L, respectively, that was less than present results [41]. Güler measured the level of PAHs in kinds of water samples and reported that the mean of total PAHs in

processed drinking water, drinking water, natural mineral water and natural spring water were ND, ND,  $3\pm5$  and  $6\pm7~\mu g/L$ , respectively that was higher than our results [20]. Aygun et al. measured PAHs in samples of drinking water in Turkey and showed that the mean of total PAHs were range from  $1.08\pm0.62$  to  $5.85\pm3.82$  ng/L (lower than our results) and the mean of BaP was  $0.11\pm0.08$ to  $0.97 \pm 0.75$  ng/L, which was lower than this research [42]. Guart et al. assessed the level of PAHs in bottled water samples in Spain and reported of all the PAHs compounds, only naphthalene (0.005–0.202 µg/L) should be observed in the samples, that was a little lower than this research [19]. Vega et al. evaluated PAHs in bottled drinking water samples in Mexico and reported the mean of total PAHs was ranged from 12.78 to 20.15 ng/L, which was lower than this research [21]. Zhang et al. evaluated PAHs in drinking water samples in China and reported the mean of 16 PAHs was  $56.25 \pm 48.53~\mu g/L$  (lower than our finding) and the mean of BaP was  $1.49 \pm 1.98$  ng/L, which was lower than this research [43]. Ambade et al. evaluated concentration of PAHs in drinking water in India and showed that the mean of total PAHs were ranged from  $9.41 \pm 8.63$  to  $21.5 \pm 14.8$  ng/L (lower than our results) and the mean of BaP was ranged from  $0.08\pm0.11$  to  $0.22\pm0.05$  ng/L, that was lower than present research [44]. Cardoso et al. measured concentration of PAHs in drinking water samples and showed that the PAH compounds in all samples were assessed less than the limits proposed by the Portuguese legislation [limits

the total concentration and four PAHs (IcdP, BghiP, BkF and BbF) to 0.10 µg/L; and BaP limited to the max level of 0.010 µg/L], which was lower than this research [45]. Froehner et al. evaluated concentration of PAHs in water in Brazil and reported that the mean of total PAHs was 51.20–162.37  $\mu$ g/L, which was higher than this study [22]. In 2021, Ciemniak et al. measured PAHs in water samples and reported that the BaP in all samples was ranged nd to 0.01 µg/kg, which was somewhat similar to our results [46]. Sarria-Villa et al. assessed the level of PAHs in Cauca River (Colombia) and reported that the BaP was not detected in all samples (somewhat similar to the our study) and the mean concentration of total 16 PAHs were ranged from  $0.688 \pm 0.544$  to  $4.47 \pm 3.95$  µg/L, which was higher than this research [47]. Kafilzadeh et al. evaluated 16 PAH compounds in Kor River (Iran) and reported the mean level of  $\Sigma PAHs$  were varied from 51.42 to 291.4 ng/L (lower than our finding) and the mean level of BaP were ranged from 1.22 to 7.18 ng/L, which was lower than this research [40]. Essumang measured the level of PAHs in water in Ghana and showed that the mean concentration of ΣPAHs were varied 6.3–26.3 μg/L (higher than our findings) and the BaP compounds was not detected in all samples, which was somewhat similar to this study [48].

Higher or lower levels of PAH contaminants can be due to reasons such as the distance or proximity of water sources to environmental pollutants such as factories, highways, urban centers, treatment plants, municipal and industrial wastewater. In addition, there is the possibility of water contamination with secondary factors such as contamination of water bottles, contamination of packaging and processing equipment, air pollution and contamination transmitted from the factory personnel (such as clothes, body etc.) [20, 21, 27, 40, 43].

## **Evaluation PAHs in kinds of bottled water samples**

Statistical analysis of PAHs in kinds of bottled water (non-carbonated, mineral, carbonated and carbonated flavored) are shown in Table 5. Our results showed carbonated flavored water had maximum mean of total PAHs  $(4.95\pm0.8 \mu g/L)$  and mineral water had minimum mean of total PAHs  $(1.24 \pm 0.8 \mu g/L)$  that can due to addition of gas, contaminated flavorings and secondary contaminants such as contaminants to surfaces and devices to PAH compounds. The BaP compound not detected in non-carbonated and mineral water, and the mean of this compound was  $0.09 \pm 0.8 \,\mu\text{g/L}$  in carbonated and carbonated flavored water, which was lower than the USEPA standard level (0.2 µg/L). The mean level of total PAHs in samples of bottled mineral water and non-carbonated bottled water samples in Iran and reported the mean concentration of total PAHs was 20.54 and 32.20 ng/L, respectively, which was lower than our results [41]. In 2007, Güler measured PAHs in kinds of water samples and reported that the mean concentration of total PAHs in water of natural mineral, water of natural spring, drinking and processed drinking water were  $3 \pm 5$ ,

**Table 5** Statistical analysis of PAHs in kinds of bottled water (µg/L)

Analyte	Non-c	arbona	ted		Miner	al			Carbo	nated			Carbo	nated f	avored		P value
	Min.	Max.	Mean	SD	Min.	Max.	Mean	SD	Min.	Max.	Mean	SD	Min.	Max.	Mean	SD	
Na	nd	0.37	0.18	0.14	nd	0.12	0.05	0.06	nd	0.55	0.4	0.1	nd	0.76	0.56	0.18	0
Ace	nd	0.6	0.38	0.23	nd	0.39	0.16	0.19	nd	0.71	0.56	0.14	nd	0.9	0.71	0.19	0.01
Ac	nd	0.28	0.07	0.12	nd	0.02	0.02	0	nd	0.32	0.08	0.14	nd	0.27	0.07	0.11	0.77
F	nd	0.72	0.56	0.3	nd	0.68	0.28	0.35	nd	0.77	0.59	0.32	nd	0.98	0.85	0.12	0.02
Ph	nd	0.13	0.05	0.06	nd	0.2	0.05	0.08	nd	0.23	0.17	0.09	nd	0.3	0.24	0.05	0.01
Α	nd	0.06	0.02	0.02	nd	0.01	0.01	0	nd	0.11	0.06	0.04	nd	0.09	0.05	0.04	0.04
FI	nd	0.56	0.24	0.29	nd	0.27	0.07	0.11	nd	0.61	0.35	0.3	nd	1.23	0.81	0.26	0.01
Р	nd	0.01	0.01	0	nd	0.01	0.01	0	nd	0.22	0.07	0.09	nd	0.5	0.17	0.23	0.19
B(a)A	nd	0.34	0.15	0.14	nd	0.21	0.08	0.07	nd	0.98	0.69	0.27	nd	1.04	0.9	0.23	0
Ch	nd	nd	-	-	nd	nd	-	-	nd	nd	-	-	nd	nd	-	-	-
B(b)F	nd	nd	_	-	nd	nd	_	-	nd	0.14	0.08	0.03	nd	0.15	0.1	0.04	0.24
B(k)F	nd	nd	_	-	nd	nd	_	-	nd	0.13	0.1	0.04	nd	0.15	0.1	0.05	0.05
B(a)P	nd	nd	_	-	nd	nd	_	-	nd	0.17	0.09	0.04	nd	0.16	0.09	0.04	0.55
D(h)A	nd	nd	-	-	nd	nd	-	-	nd	nd	-	-	nd	nd	-	-	-
B(g)P	nd	nd	-	-	nd	nd	-	-	nd	nd	-	-	nd	nd	_	-	-
I(cd)P	nd	nd	-	-	nd	nd	-	-	nd	nd	-	_	nd	nd	_	-	-
Total	1.31	3.46	2.17	0.82	0.69	2.42	1.24	0.8	2.75	4.87	3.55	0.9	4.11	5.84	4.95	0.8	0

 Table 6
 Statistical analysis of PAHs in different brands of bottled water (µg/L)

Analyte	Brand A	A		Brand B	_ 		Brand C	U		Brand D	٥		Brand E	ш		P value
	Min.	Мах.	Mean ± SD	Min.	Мах.	Mean±SD	Min.	Max.	Mean ± SD	Min.	Max.	Mean ± SD	Min.	Max.	Mean ± SD	
Na	pu	0.53	0.24 ± 0.23	pu	69:0	0.36±0.25	pu	0.76	0.45 ± 0.27	pu	0.51	0.24 ± 0.27	pu	0.3	0.18±0.14	0.686
Ace	pu	0.7	$0.4 \pm 0.29$	pu	0.88	$0.6 \pm 0.23$	pu	6.0	$0.65 \pm 0.21$	pu	0.58	$0.28 \pm 0.30$	pu	0.52	$0.32 \pm 0.23$	0.686
Ac	pu	pu	I	pu	0.27	$0.08 \pm 0.13$	pu	0.32	$0.16 \pm 0.17$	pu	pu	I	pu	pu	I	<b>.</b>
ш	pu	0.87	$0.58 \pm 0.37$	pu	0.93	$0.76 \pm 0.12$	pu	0.98	$0.79 \pm 0.14$	pu	92.0	$0.21 \pm 0.37$	pu	0.7	$0.52 \pm 0.33$	0.486
R	pu	0.24	$0.11 \pm 0.12$	pu	0.27	$0.13 \pm 0.14$	pu	0.3	$0.22 \pm 0.07$	pu	0.2	$0.13 \pm 0.09$	pu	0.19	$0.06 \pm 0.09$	0.686
⋖	pu	0.1	$0.05 \pm 0.05$	pu	0.07	$0.05 \pm 0.03$	pu	pu	I	pu	0.03	$0.02 \pm 0.01$	pu	0.11	$0.05 \pm 0.05$	<b>.</b>
正	pu	0.64	$0.18\pm0.31$	pu	0.88	$0.37 \pm 0.42$	pu	1.23	$0.67 \pm 0.40$	pu	0.57	$0.42 \pm 0.27$	pu	0.71	$0.2 \pm 0.34$	0.886
Ь	pu	0.3	$0.11 \pm 0.14$	pu	0.22	$0.06 \pm 0.11$	pu	pu	ı	pu	pu	I	pu	0.5	$0.13 \pm 0.25$	0.886
B(a)A	pu	1.02	$0.53 \pm 0.55$	pu	66.0	$0.47 \pm 0.39$	pu	1.04	$0.63 \pm 0.44$	pu	0.98	$0.39 \pm 0.44$	pu	0.49	$0.27 \pm 0.25$	0.686
Ch	pu	pu	I	pu	pu	ı	pu	pu	I	pu	pu	I	pu	pu	I	ı
B(b)F	pu	0.15	$0.09 \pm 0.04$	pu	pu	ı	pu	0.14	$0.08 \pm 0.04$	pu	pu	I	pu	0.14	$0.08 \pm 0.04$	0.886
B(k)F	pu	0.12	$0.09 \pm 0.04$	pu	0.15	$0.1 \pm 0.05$	pu	pu	ı	pu	pu	ı	pu	0.14	$0.09 \pm 0.04$	0.886
B(a)P	pu	pu	ı	pu	pu	ı	pu	0.17	$0.12 \pm 0.05$	pu	pu	ı	pu	pu	ı	I
D(h)A	pu	pu	I	pu	pu	ı	pu	pu	I	pu	pu	I	pu	pu	ı	ı
B(g)P	pu	pu	I	pu	pu	I	pu	pu	I	pu	pu	I	pu	pu	1	ı
l(cd)P	pu	pu	ı	pu	pu	ı	pu	pu	ı	pu	pu	I	pu	pu	ı	ı
Total	69.0	5.07	2.78土1.88	1.74	9.9	$3.43 \pm 1.73$	2.42	5.84	4.15±1.51	69:0	4.15	$2.22 \pm 1.55$	69.0	4.11	2.31 ± 1.46	0.686

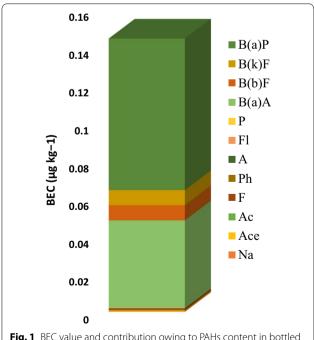
Percentiles/	Adults				Children					
analyte	5%	50%	75%	95%	5%	50%	75%	95%		
NA	5.12E-7	7.81E-7	9.27E-7	1.17E-6	2.39E-6	3.65E-6	4.27E-6	5.55E-6		
ACE	8.24E-7	1.22E-6	1.46E6	1.82E-6	3.80E-6	5.71E-6	6.79E-6	8.54E-6		
AC	1.09E-7	1.66E-7	1.92E-7	2.47E-7	5.01E-7	7.51E-7	8.87E-7	1.14E-6		
F	1.06E-6	1.55E-6	1.78E-6	2.35E-6	4.78E-6	7.22E-6	8.55E-6	1.07E-5		
PH	2.36E-7	3.54E-7	4.10E-7	5.30E-7	1.10E-6	1.64E6	1.94E-6	2.46E-6		
Α	7.30E-8	1.08E-7	1.28E-7	1.63E-7	3.49E-7	5.16E-7	6.00E-7	7.61E-7		
FL	6.71E-7	1.01E-6	1.19E-6	1.49E-6	3.21E-6	4.66E-6	5.47E-6	6.77E-6		
Р	1.27E-7	1.92E-7	2.22E-7	2.81E-7	5.94E-7	8.98E-7	1.07E-6	1.31E-6		
B(A)A	8.41E-7	1.27E-6	1.48E-6	1.89E-6	3.94E-6	5.77E-6	6.74E-6	8.55E-6		
B(B)F	1.44E-7	2.19E-7	2.58E-7	3.34E-7	6.81E-7	1.03E-6	1.21E-6	1.52E-6		
B(K)F	1.44E-7	2.17E-7	2.55E-7	3.33E-7	6.76E-7	1.02E-6	1.19E-6	1.54E-6		
B(A)P	1.45E-7	2.17E-7	2.55E-7	3.29E-7	6.68E-7	1.00E-6	1.18E-6	1.53E-6		

Table 7 Uncertainly analysis for the daily intake (µg/kg bw/day) of PAHs in bottled water samples

 $6\pm7,$  ND and ND  $\mu g/L,$  respectively that was higher than our results [20].

# Evaluation of PAHs in different brands of bottled water samples

In Table 6, statistical analysis of PAHs compounds in different brands of bottled water samples are presented. The results showed brand C had maximum mean level of total PAHs ( $4.15\pm1.51~\mu g/L$ ) and brand B had minimum



**Fig. 1** BEC value and contribution owing to PAHs content in bottled water samples

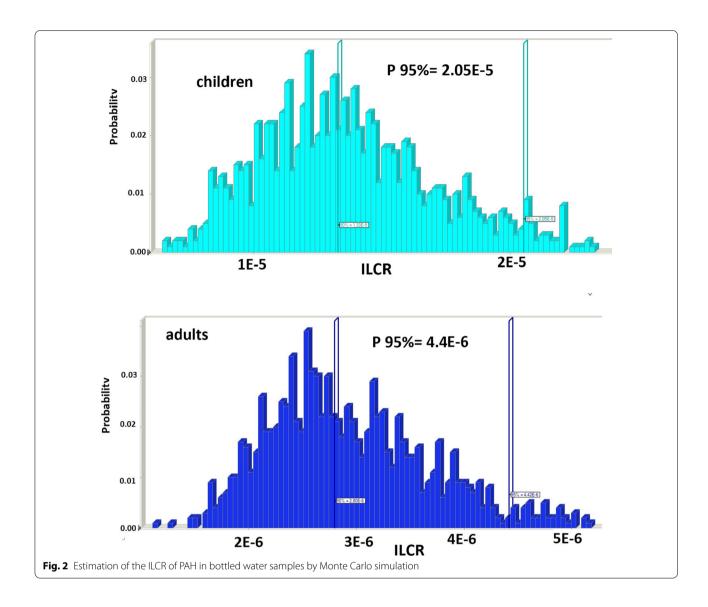
mean level of total PAHs ( $2.22\pm1.55~\mu g/L$ ). The BaP was detected in brand C with mean of  $0.12\pm0.05~\mu g/L$ , which was lower than the US-EPA standard level ( $0.2~\mu g/L$ ). Higher levels of contaminants can be due to primary contamination of water (source), secondary contamination such as contamination of surfaces, devices, bottles, etc.

## Human health risk assessment

The practical models such as EDI and ILCR indexes indicate the carcinogenic and non-carcinogenic health hazards due to long-term oral exposure of PAH mixtures contaminated food. As the EPA guidelines recommend, a Monte-Carlo was applied in the probabilistic risk evaluations to decrease of risk uncertainties with probability position for each variable to elude overestimation or underestimation [29, 30, 32, 35]. Several investigations have been conducted on the probabilistic health hazard estimation by Monte Carlo Simulation (MCS) for PAHs in ground water in Indian [49], PAHs commercial coffee and tea in Iran [6], acrylamide level in commercial nuggets [50], PAHs in edible mushrooms [8], toxin elements and sulfur compounds in raisins [31]. The rank order of the estimated daily intake (95th percentile) was:>B(a) A > Ace > Fl > Na > Ph > B(b)F > B(k)F > B(a)P > P > Ac > Aas shown in Table 7.

The types of PAHs studied in food vary by region and product type. According to the guidelines recommend EPA, exposure of BaP over 200 ng/L bw/day through diet has been suggested as a potential danger to health of human. Among all samples, EDI values were below accepted value; consequently, bottled water samples was not dangerous due to PAH to the public's health.

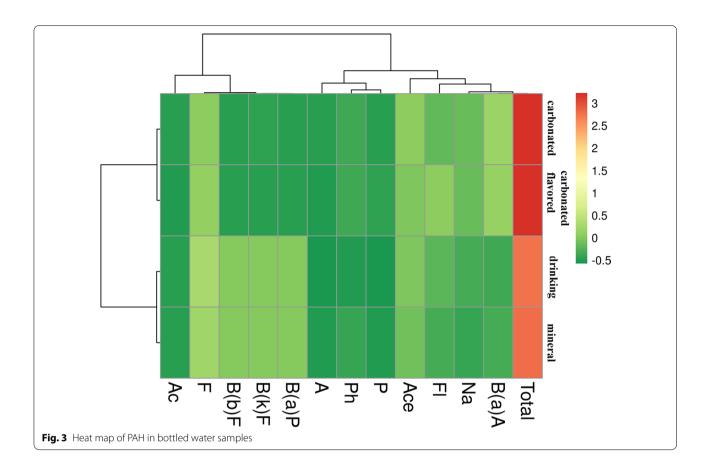
Figure 1 shows that the BaP and BaA are two principal contributors to the total BEC ( $\mu g/kg$ ), and whiles the



other PAHs included have a contribution of lower than 14 percent. According to the MCS results, the ILCR indexes (percentile 95 percent) in the bottled water samples for adults and children was 2.05E-5 and 4.4E-6, respectively. The probabilistic distributions and simulation histogram of BaP risk for the bottled water is shown in Fig. 2. The qualitative classification of carcinogenic risk terms can describe in three forms; the ILCR indexes with value less than  $10^{-6}$  is the safe zone; the ILCR indexes with value higher than  $10^{-4}$  is the limit of threshold risk; the ILCR indexes higher than  $10^{-3}$  is the zone of significant danger. In similar study, Wu et al. showed carcinogenic risks of PAH compounds owing to the drinking water sample ingestion were accepted [43]. Hence, they recommended a more comprehensive survey on carcinogenic PAHs (especially BaP, DahA) in China's drinking water to provide drinking water safety. The acquired results can be a useful reference for organizations like the health and agriculture ministry.

# Multivariate analysis

The heat map involves comprehending the PAH congener profiles associations in different bottled water samples. Additionally, classification rows and columns of similar parameters, a heat map visualization gives a comprehensive pattern of the highest and least variables in the generating model. Moreover, heat maps showed which the bottled water samples (non-carbonated, mineral, carbonated and carbonated flavored) were independent variables in the PAH compounds congener clustering. The heat map clustered samples of bottled water into two major clusters (Fig. 3). The first cluster includes Ac, F, BbF, BkF and BaP, second cluster includes, A, P, Ace,



Fl, Na, BaA, Ph and total PAH. ClustVis was employed to visualize the clustering of related data. The Ac, BbF, BkF and BaP groups were the closers, showing that the frequency variations of these PAH compounds had a similar trend in various samples.

# **Supplementary Information**

The online version contains supplementary material available at https://doi.org/10.1186/s13765-022-00696-9.

Additional file 1: Table S1. PAHs and their toxic equivalent factors (TEFs).

# Acknowledgements

This research has been supported by the Shahid Beheshti University of Medical Sciences (SBMU).

## **Author contributions**

NS: conceptualization, supervision, design of study, writing—reviewing and editing. SS: design of study, writing—reviewing and editing. AM: data curation, writing—reviewing and editing. MA: visualization, investigation, software, methodology. Software, validation, AMM: methodology. Software, validation, AMM: methodology. Software, validation, MM: data curation, writing—original draft preparation, GS: design of study, writing—reviewing and editing. All authors read and approved the final manuscript.

## Availability of data and materials

The datasets used and/or analyzed during the present study are available from the corresponding author.

## **Declarations**

## Ethics approval and consent to participate

Not applicable.

## **Consent for publication**

Not applicable.

## **Competing interests**

There is no competing interests declared by the authors.

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Received: 2 March 2022 Accepted: 5 April 2022 Published online: 19 May 2022

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