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Estimating human health risks associated with heavy metal exposure from bottled water using Monte Carlo simulation

Nezam Mirzaei [a], Safa Kalteh [b], Hakime Zamani-Badi [c], Heshmatallah Moradpour [a], Zeinab Parmoozeh [a], Mansour Baziar [d,*]

[a] *Department of Environmental Health Engineering, Social Determinants of Health (SDH) Research Center, Kashan University of Medical Sciences, Kashan, Iran*
[b] *Department of Environmental Health Engineering, School of Public Health, Tehran University of Medical Sciences, Tehran, Iran*
[c] *Department of Health, Safety and Environment Management, Faculty of Health, Kashan University of Medical Sciences, Kashan, Iran*
[d] *Department of Environmental Health Engineering, Ferdows Faculty of Medical Sciences, Birjand University of Medical Sciences, Birjand, Iran*

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ABSTRACT

Water is the most important non-organic compound for living cells, and the life of all living organisms depends on it. Water is not found purely in nature, but it always contains some solutes, suspended matters and soluble gases. In this study, 11 bottled water brands (500 mL) were sampled across the Kashan city market to determine the concentration of selected heavy metals (Cr, Cd, Pb, Ni and As) and evaluated their potential risks following consumption. The concentration range of Cr, Cd, Pb, Ni and As were 5–34 μg/L, 1.5–7 μg/L, 1–7 μg/L, 2–29 μg/L, <LOD – 3.5 μg/L, respectively. The heavy metals (HMs) concentration in this study was compared with other national and international studies on bottled water and other drinking water sources. Additionally, the results of non-carcinogenic health risk assessments using point estimations and Monte Carlo Simulations (MCS) showed that the concentration of HMs in none of the samples pose adverse health effects. However, it was found that drinking the studied bottled water may potentially lead to carcinogenicity over the lifetime. Lack of including spring water and larger-sized (1.5 L) bottled water samples, though less consumed than the samples of this study and lack of study on children's exposure level can be mentioned as potential limitations.

1. Introduction

The utilization of bottled water has steadily risen over the last half century due to its ease of access, low price and low levels of impurities [1,2]. Basically, bottled water is a purchasable packaged water supplied from various resources such as spring, well or tap water [3]. According to regulations, no chemicals must be present in the bottled water, except for antibacterial agents [3]. However, they may not always be safe and their resources may be contaminated with various contaminants [4,5]. Many researchers have identified the presence of high levels of certain substances in the bottled water, especially those that are toxic, such as radionuclides and trace elements [1,2,6]. Trace elements can be divided into two groups, the first group comprised of elements such as cobalt, copper, iron, molybdenum, selenium and zinc, which are essential for human life as well as for the growth and reproduction of organisms [2,7].

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[*] Corresponding author.
E-mail address: Mansour.baziar@bums.ac.ir (M. Baziar).

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The second group, however, is characterized by their toxic properties for humans and organisms such as silver, aluminum, arsenic, cadmium, mercury, lead and nickel, generally known as heavy metals (HMs) [8–10]. In past years, there has been many reports regarding the pollution of water resources by HMs that made them a potential global concern [4,11,12]. HMs, even at minuscule amounts can be toxic to humans. They are known to be highly stable and biologically undegradable elements in the water and wastewater treatment processes [13,14]. The entry of HMs along with their accumulation into the human body and living organisms can cause health issues such as neurological disorders, disturbance of hormonal balance, anemia, osteoporosis, skin and respiratory disorders and finally, carcinogenesis [15,16].

Studying the literature shows that there has been a direct correlation between some of the adverse health effects, such as cardiovascular disorders, kidney diseases, and several types of cancers and the occurrence of HMs in the water [17–19]. Generally, the most important source of HMs in water resources is chemical weathering of rocks. In addition, human activities such as discharge of untreated industrial wastewater, runoff from agricultural fields, leachates and seepages from municipal landfill and heavily polluted areas like mines are the major culprits as well [20,21]. According to the definition of the United States Environmental Agency (USEPA), health risk assessment is a process used to assess the nature and probability of creating harmful health effects on people due to their exposure with a polluted environment in the present and/or future [22]. In last decades, non-carcinogenic and carcinogenic health risk assessment of heavy metals has become an essential process due to the incidence of clinical signs such as kidney and heart disorders as a result of HM exposure even at concentrations lower than previously thought safe levels [4,23,24]. Therefore, stringent national and international limits were set to prevent adverse public health outcomes due to dietary exposure to HM. In Iran, such limitations are set by Institute of Standards and Industrial Research of Iran (ISIRI), which normally adapts World Health Organization (WHO) standard levels if deemed adequate. For instance, the permissible limits of WHO and ISIRI are as follows:Cr (0.05 mg/L), Cd (0.003 mg/L), Pb (I0.01 mg/L), As (0.01 mg/L) and Ni (0.07 mg/L) [25].

Numerous studies have been carried out to determine the level of heavy metal in drinking water in Iran [26,27]. However, most of these studies focused on the comparison of heavy metal content with corresponding standard level. Meanwhile, a comprehensive evaluation of non-carcinogenic and carcinogenic health risk assessment based on the obtained data can be important to enhance the public awareness. Also, it can help decision makers regarding the occurrence of heavy metals in the drinking water. In addition, since nearly 70% of Iran's land has arid climate, so water tensions and water problems are nearly the same in many cities located in the arid region. Kashan, an industrially developed city located at central Iran is in the crossroads to other major cities such as Isfahan, Ghom, and Tehran [28–30]. The objectives of current study are: (a) determination of five heavy metals (Cr, Cd, Pb, Ni and As) concentration in the bottled waters marketed in Kashan city, located in northern parts of Isfahan province of Iran; (b) compare the determined levels with WHO and ISIRI standards as well as comparing them with other similar studies domestic and abroad (c) conducting a human health risk assessment the (both carcinogenic and non-carcinogenic) for the adult population of Kashan city and compare the findings with the results of other studies.

2. Materials and methods

2.1. Study area

Kashan is a historical city and the capital of Kashan County. The city is located in the center of Iran in northeast of Isfahan province

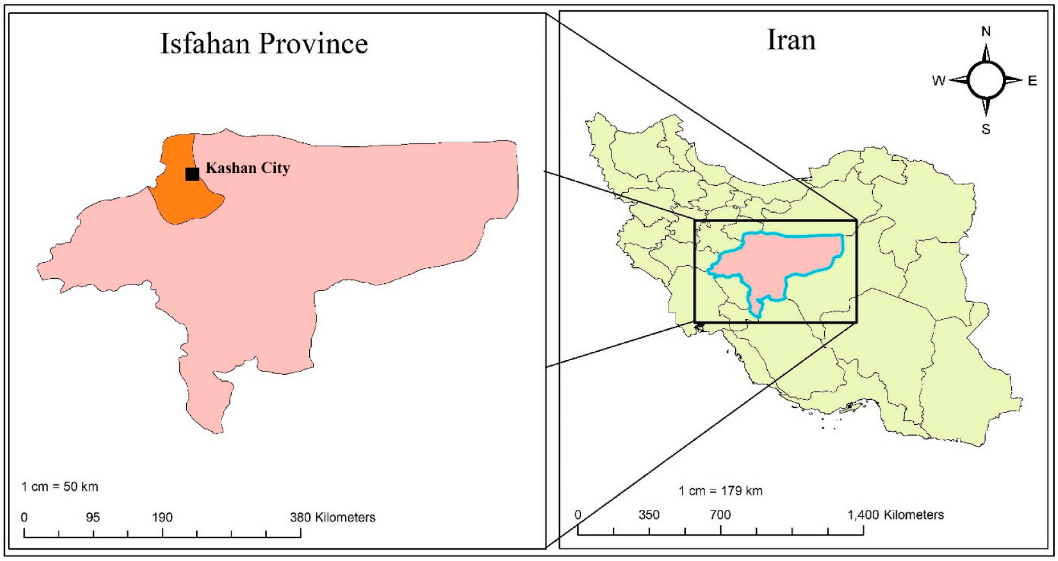

Fig. 1. The studied area location map (Kashan, Iran).

in proximity of the industrial and transport arteries of central Iran (Fig. 1). It has a population of 396,987 people according to the latest census. The altitude of the city from sea level is 982 m and has warm and desert climate with an average annual precipitation equal to 128 mm which typically occurs in winter and autumn. The city latitude and longitude are 34°05′N and 51°30′E respectively. More than 80 % of Kashan's drinking water is supplied from underground resources [1,15]. In recent years, water shortages have become more frequent in Kashan and other cities in Iran as nearly 70 % of the Iranian land has an arid climate. Like other cities with similar characteristics, the bottled water is supplied from various sources [28–31].

2.2. Sample collection, preparation and heavy metals analysis

A total of 33 bottled waters (500 mL volume) from 11 brands (3 samples from each brand) were purchased from local shops in Kashan city. The selected brands, with price range between 10 and 15 cents, were filled with tap water and were of the most consumed commercial bottled water products. The samples were transported to the laboratory in sealed condition after they were coded from A to K. In addition, each brand tag was assessed to specify the source of supply (All brands were supplied from spring water, except brands A, B and F which were supplied from wells). The analysis was performed in Environmental and Chemistry Laboratory of Kashan University of Medical Sciences (Kashan, Iran) using a PerkinElmer, AAS-PEA- 700 model, Graphite furnace atomic absorption spectroscopy (GFAAS). 100 mL of bottled water samples were acidified using 20 mL of nitric acid and then digested for 1 h at 100 °C in a special fume cupboard until the generation of a transparent solution. Then, the volume of the samples was decreased to 20 mL and the obtained solutions were dipped into a volumetric flask with volume of 100 mL and diluted using distilled water. Next, the samples were filtered using a filter paper and analyzed to determine the concentrations of Cr, Cd, Pb, Ni and As [32]. In addition, method validation parameters such as limit of detection (LOD), limit o quantification (LOQ) and accuracy procedures were performed according to Hadiani et al. (2014) work [33] and are shown in Tables 1 and 2.

2.3. Human health risk assessment

We determined the non-carcinogenic health risks associated with exposure to chrome, cadmium, lead, nickel, and arsenic in bottled waters by calculating the risk ratio, based on the equations provided by U.S. Environmental Protection Agency (USEPA). The non-carcinogenic health risk was assessed by obtaining the Hazard quotient (HQ) value. The obtained value is interpreted as follows: a HQ value less than 1 indicates minor potential risk of non-carcinogenic in the exposed population while an HQ value greater than 1 indicates the presence of potential adverse health effects [15,34].

$$HQ = \frac{CDI}{RfD} \tag{1}$$

in equation (1), CDI is chronic daily intake (mg/kg/day) which calculated based on equation (2) and Rfd is a reference dose for a given heavy metal.

$$CDI = \frac{C_w \times DI \times EF \times ED}{BW \times AT} \tag{2}$$

in equation (2), CW is metal concentration (Cr, Cd, Pb, Ni and As mg/L); DI is the daily intake water (L/day); ED represents the exposure duration (year); EF indicates the exposure frequency which (days/year); BW is body weight for adults (kg); AT demonstrates the average exposure time for non-carcinogenic effects (day).

The HQ value was calculated for all of the studied heavy metals (Cr, Cd, Pb, Ni and As) and their total sum value was also calculated using equation (3) to obtain value the hazard index (HI), which is an index for showing the non-cancer effects of heavy metals.

$$HI = \sum_{k=1}^{n} HQ = HQ_{As} + HQ_{Pb} + HQ_{Cd} + HQ_{Cr} + HQ_{Ni} \tag{3}$$

If the amount of HI is less than 1, then there would be no significant effect of non-carcinogenicity, and if this value is higher than 1, a significant health effect [25]. Additionally, to determine the risk of carcinogenic effects of the studied heavy metals, cancer risk was calculated using USEPA proposed formula [27].

$$CR = CDI \times CSF \tag{4}$$

in equation (4), CSF denotes the cancer slope factor (mg kg^{-1} d^{-1}). In the present study, the estimated risk of cancer is compared with the maximum acceptable risk proposed by USEPA which is a value between the 10-e6 and 10-e4.

Table 1
Quality control factors (ng ml^{-1}).

Parameter	Cr	Cd	Pb	Ni	As
Linear range	0.5–20	0.–10	3–20	5–50	3–20
Limit of Detection	0.1	0.2	1	1	1
Limit of Quantification	0.3	0.6	3	3	3

Table 2
Average recovery of Cr, Cd, Pb, Ni and As from bottled water samples within 3 days.

Element/Concentration	Cr			Cd			Pb			Ni			As		
Spike level (ng g^{-1})	0.5	1	2	1.5	3	4.5	5	10	15	10	20	40	5	10	15
Mean result (ng g^{-1})	0.51	0.99	1.9	1.3	2.5	4.2	5.2	9.6	14.5	11	23	37.1	5.3	10.4	14.5
Mean recovery (%)	103	99	96	90	85	94	104	96	97	110	115	92.7	106	104	97

To estimate the risk of heavy metals, both carcinogenic and non-carcinogenic effects via bottled drinking water ingestion (oral path), different assumptions were made. Table 3 displays the assumptions and parameters used to calculate the human health risk assessment in this work. Each parameter can differ for individuals, which might result in varied levels of individual exposure to the contaminant of interest, the actual extent of the risk can diverge. Monte Carlo Simulation (MCS) is a useful probabilistic method to reduce uncertainty levels in human health risk assessment [32].

In addition to obtaining the point estimates of the carcinogenic and non-carcinogenic health risks, the MCS was run with 10000 iterations to obtain an upper bound 95 percentile estimate of the dietary risks.

3. Results

3.1. *Concentrations of heavy metals in bottled water*

The average levels of Cr, Cd, As, Pb, and Ni in each brand of bottled waters along with their corresponding national and WHO standards and the results of the analysis are presented in Table 4. Among the brands, the highest (0.034 mg/L) and lowest (0.005 mg/L) concentration of Cr was found in brands D and I, respectively. The highest (0.0025 mg/L) and lowest (0.0015 mg/L) concentration of Cd was found in brands C and F, respectively. The highest (0.007 mg/L) and lowest (0.001) concentration of Pb was found in brands F and B, respectively. Further, the highest (0.029 mg/L) and lowest (0.002 mg/L) concentration of Ni was found in brands J and K, respectively. The highest (0.0035 mg/L) and lowest (0.001 mg/L) concentration of As was observed in brands K and A. It should be noted that As was not detected in brands C, H, and J.

3.2. *Human health risk assessment*

In this study, non-carcinogenic and carcinogenic risk assessments were performed using the obtained information regarding the heavy metal concentration in bottled waters of Kashan. For evaluating the human health risk regarding the presence of heavy metals in bottled water and chronic exposure by them in adult consumers, the hazard quotient (HQ) and hazard index (HI) values were estimated for Cr, Cd, As, Pb, and Ni. Table 5 illustrates the values of HQ and HI for the investigated heavy metals in each brand. The obtained point estimate and probabilistic HQ values in each studied brand and for each studied HM were all below 1, indicating the safety of the bottled water brands in terms of dietary HM exposure. In addition, the obtained HI also shows that no adverse health effects can be expected in overall as the sum of the estimated HQs in all brands, both in point estimates and MCS analysis were below 1 as well. However, the highest HI value was observed in brand D and the lowest was found in brand H. This was also true for HI values obtained from MCS.

4. Discussion

The concentration of HMs in the water differs depending on the supply source (e.g., groundwater or surface water) [15]. The investigation of HMs in the drinking water sources is very important for the health of community and the decision-makers because of their high toxicity and resistance in the natural environments as well as their adverse health outcomes due to accumulation in the human body [35]. Drinking water can be one of the important sources of human exposure to HMs. Especially, in areas where the concentration of metals in the water is higher than the level determined by the WHO [10]. The investigation on the adverse health effects of HMs in the water is extensive. However, human exposure to various types of heavy metals has been an ongoing problem,

Table 3
Assumptions and parameters used to assess the risk of heavy metals exposure via bottled drinking water ingestion for adult population.

Parameter	Unit	Pb	Cr	Cd	As	Ni	Ref.
Ingestion rate (IR)	Lit/days	2.2	2.2	2.2	2.2	2.2	[25,35]
Exposure frequency (EF)	days	365	365	365	365	365	[25,35]
Exposure duration (ED) for CR	Year	70	70	70	70	70	[25,35]
Exposure duration (ED) for HQ	Year	35	35	35	35	35	[25,35]
Body weight (BW)	Kg	70	70	70	70	70	[25,35]
Average time (AT) for CR	days	25550	25550	25550	25550	25550	[25,35]
Average time (AT) for HQ	days	12775	12775	12775	12775	12775	[25,35]
Oral Reference dose (RfD)	mg kg^{-1} d^{-1}	0.0035	0.003	0.0005	0.0003	0.02	[10,15,36]
Oral Cancer slope factor (CSF)	mg kg^{-1} d^{-1}	0.0085	0.5	6.1	1.5	–	[10,15,36]

Table 4
Concentrations (mean ± SD) of the studied heavy metals in the samples bottled water brands.

Heavy Metals		Cr	Cd	Pb	Ni	As
WHO and ISIRI limits		0.05	0.003	0.01	0.01	0.07
Brands	A	0.027 ± 0.001	0.0019 ± 0.00021	0.0027 ± 0.0013	0.022 ± 0.001	0.001 ± 0.00013
	B	0.031 ± 0.0082	0.0019 ± 0.00022	0.001 ± 0.0010	0.025 ± 0.002	0.002 ± 0.00010
	C	0.027 ± 0.0074	0.0025 ± 0.0001	0.007 ± 0.0015	0.003 ± 0.001	0
	D	0.034 ± 0.0095	0.0019 ± 0.00024	0.0025 ± 0.0013	0.024 ± 0.003	0.003 ± 0.00105
	E	0.032 ± 0.0080	0.002 ± 0.0001	0.001 ± 0.0012	0.0023 ± 0.0001	0.002 ± 0.00011
	F	0.024 ± 0.0066	0.0015 ± 0.0003	0.007 ± 0.0014	0.0025 ± 0.0002	0.002 ± 0.00010
	G	0.031 ± 0.0082	0.0017 ± 0.00022	0.002 ± 0.0011	0.0021 ± 0.0001	0.002 ± 0.00011
	H	0.025 ± 0.0072	0.0019 ± 0.00024	0.0055 ± 0.0010	0.017 ± 0.001	<LOD
	I	0.005 ± 0.0009	0.0018 ± 0.00026	0.0065 ± 0.0016	0.025 ± 0.0023	0.0025 ± 0.00011
	J	0.029 ± 0.0072	0.0019 ± 0.00027	0.003 ± 0.0011	0.029 ± 0.001	0
	K	0.025 ± 0.0062	0.0021 ± 0.0002	0.004 ± 0.0012	0.002 ± 0.0012	0.0035 ± 0.00010

Table 5
Estimated values for carcinogenic and non-carcinogenic human health risk assessment.

Brands	HMs	A	B	C	D	E	F	G	H	I	J	K
HQ	Cr	0.283	0.325	0.283	0.356	0.335	0.251	0.325	0.262	0.052	0.304	0.262
	Cd	0.119	0.119	0.157	0.119	0.126	0.094	0.107	0.119	0.113	0.119	0.132
	Pb	0.024	0.009	0.063	0.022	0.009	0.063	0.018	0.049	0.058	0.027	0.036
	Ni	0.035	0.039	0.005	0.038	0.004	0.004	0.003	0.027	0.039	0.046	0.003
	As	0.105	0.210	–	0.314	0.210	0.210	0.210	–	0.262	–	0.367
HI		0.57	0.70	0.51	0.85	0.68	0.62	0.66	0.46	0.53	0.50	0.80
HQ-P95	Cr	0.199	0.224	0.193	0.246	0.230	0.173	0.223	0.180	0.036	0.210	0.180
	Cd	0.083	0.083	0.108	0.082	0.087	0.065	0.073	0.082	0.078	0.083	0.091
	Pb	0.017	0.006	0.043	0.016	0.006	0.043	0.012	0.034	0.040	0.019	0.025
	Ni	0.024	0.028	0.003	0.027	0.003	0.003	0.002	0.019	0.028	0.032	0.002
	As	0.072	0.145	–	0.217	0.144	0.144	0.144	–	0.180	–	0.254
HI (95)		0.39	0.49	0.35	0.59	0.47	0.43	0.45	0.31	0.36	0.34	0.55
CR	Cr	1.70E-03	1.95E-03	1.70E-03	2.14E-03	2.01E-03	1.51E-03	1.95E-03	1.57E-03	3.14E-04	1.82E-03	1.57E-03
	Cd	3.98E-06	3.98E-06	5.24E-06	3.98E-06	4.19E-06	3.14E-06	3.56E-06	3.98E-06	3.77E-06	3.98E-06	4.40E-06
	Pb	9.98E-03	3.70E-03	2.59E-02	9.24E-03	3.70E-03	2.59E-02	7.39E-03	2.03E-02	2.40E-02	1.11E-02	1.48E-02
	Ni	–	–	–	–	–	–	–	–	–	–	–
	As	2.10E-05	4.19E-05	–	6.29E-05	4.19E-05	4.19E-05	4.19E-05	–	5.24E-05	–	7.33E-05
CR-P95	Cr	1.20E-03	1.34E-03	1.17E-03	1.46E-03	1.38E-03	1.04E-03	1.34E-03	1.08E-03	2.16E-04	1.25E-03	1.07E-03
	Cd	1.20E-03	1.34E-03	1.17E-03	1.46E-03	1.38E-03	1.04E-03	1.34E-03	1.08E-03	2.16E-04	1.25E-03	1.07E-03
	Pb	2.75E-06	2.75E-06	3.59E-06	2.73E-06	2.88E-06	2.17E-06	2.46E-06	2.75E-06	2.58E-06	2.74E-06	3.04E-06
	Ni	6.86E-03	2.56E-03	1.77E-02	6.34E-03	2.55E-03	1.78E-02	5.06E-03	1.39E-02	1.67E-02	7.64E-03	1.02E-02
	As	1.43E-05	2.88E-05	–	4.32E-05	2.89E-05	2.87E-05	2.86E-05	–	3.60E-05	–	5.01E-05

particularly in developing countries [28]. Health issues related to the human exposure and the presence of HMs in drinking water can be resulted from (i) a severe contamination of drinking water sources (due to accidental pollution) or (ii) from the long-term exposure of drinking water consumers with low concentrations. A good understanding regarding the quality of drinking water and monitoring the levels of contaminants is essential in protecting public health [15].

The levels of Cr in this study was higher than the findings of Ghaderpoori et al. (2018) in Khorramabad city, Iran (min = 0.0006, max = 0.0106 mg/L) [28], study of Eslami et al. (2022) (min = 0.00102, max = 0.01053 mg/L) in Rafsanjan city, Iran [29] and the findings of Jafarzadeh et al. (2022) (min = 0.00049, max = 0.02 mg/L) in Saravan county, Iran [37]. Additionally, the Cr levels found in this study were higher than the concentration range reported by Maleki and Jari (2021) sampled in three cities and their surrounding rural areas in Kurdistan province of Iran during wet and dry seasons (min = 0.0001, max = 0.053 mg/L) [25].

From the obtained results in analysis of eleven brands of bottled water, it was demonstrated that the studied heavy metal concentrations in all brands were lower than the WHO and national standards (Table 4). However, due to their accumulation properties, there is the possibility of adverse health effects by heavy metals for consumers of the studied drinking water brands. Children, due to their rapid metabolism and growth rate, are considered as the most vulnerable group in exposure to Pb [15]. Cd is an element that is found naturally in the ambient air, fresh and saline water, and in soil. Owing to the harmful effects of Cd to human, the WHO recommended that the levels of it always be controlled in water and food [29]. Cd can accumulate in the given organs after entering the human body. It can create disorders in the brain, liver, lung as well as nervous system [29,37]. In the case of chromium, like other HMs discussed, the least health threat can be expected as the concentration of chromium did not exceed the standard level in any of the studied brands. It should be mentioned that Nickel, like other HMs, can lead to health-related hazards in humans. The HM found in bottled water can be traced back to the earlier pollution of the tap water either by means of dissolution from metallic pipes used for distribution or during filling in the factory. This is the case for Pb contamination [38]. Contamination to As can be traced back even further at the source. As mentioned in the literature, a ubiquitous source of exposure to inorganic As is via drinking water supplied from groundwater resources, springs and streams [25,39]. Contamination via metal pipes, external leakage in supply and production chain such as Antimony (Sb) from the bottle, geological properties and potential contamination of at the source are among the frequently cited origins for the occurrence of HMs in the bottled water [28,40]. Groundwater resources are the main source of water supply in Iran and some regions of the country, such as rural areas of Kurdistan province, are known for their contamination to a specific trace element which can variate over seasons [25].

Table 6 displays other similar studies on the HMs in drinking water sources. The mean concentrations of Cr in this study was lower than the range reported by studies of Ungureanu et al. (2022) in Romania [41], Maxwell et al. (2018) in Nigeria and Annibaldi et al. (2018) [42] in Italy conducted on the samples of marketed bottled water. However, the concentration range of Cr in this study was higher than the levels reported in the study of Ristić et al. (2011) [38] in Serbia and within the concentration range reported for bottled water samples by Zazouli et al. (2015) [40] in Sari city, Iran; Singla et al. (2014) [43] in India; Bamuwamye et al. (2022) [44] in Uganda. Moreover, the Cr levels was also in range with the Cr levels reported for tap water and groundwater supply samples of rural areas of Kurdistan province [25], Khorram-Abad city [28], Rafsanjan city [29] and Saravan county [37] in Iran. The concentration range of Cd in this work was higher than the range in bottled water samples reported for by the studies conducted in Romania [41], Italy [42], Iran (Sari city) [40], Serbia [38], and samples taken from other water resources in other regions of Iran such as tap water in Khorramabad city [28], and groundwater samples Rafsanjan city [29] and Kurdistan province [25]. Also, the Cd levels found in this study were within the concentration range reported in the study of Hadiani et al. (2015) [33] on bottled water but lower than the levels reported for bottled water samples in the studies of Maxwell el al (2018) [30] in Nigeria, Fakhri et al. (2015) [45] and Jafarzadeh et al. (2022) in Saravan county [37] in Bandar Abbas city and Saravan county in Iran, respectively. The levels of Pb in this study was lower than the levels reported in the studies of Annibaldi et al. (2018) [42] in Italy, Bakidere et al. (2013) [39] in Turkey, Ristić et al. (2011) [38] in Serbia. Moreover, the Pb levels of our study were consistent with the levels reported in the studies of Ungureanu et al. (2018) [41] in Romania, Hadiani et al. (2015) [33] in Iran, tap water samples in Bakırdere et al. (2013) [39] in Turkey and Ghaderpoori et al.

Table 6
Comparison of the heavy metal levels in this study with other national and international studies.

Country	Studied water source	Brands	Sample count	Heavy metal Concentration range (μg/L)					Ref.
				Cr	Cd	Pb	Ni	As	
Iran (this study)	Bottled water (tap)	11	33	5–34	1.5–2.5	1–7	2–29	0–3.5	–
Romania (2022)	Bottled water (mineral)	50	50	<0.10–4.02	<0.09–<0.09	<0.07–6.0	0.16–3.77	–	[41]
Nigeria (2018)	Bottled water,	20	N/A	ND	<LOD – 72.1	<LOD – 348.1	<LOD – 12.4	–	[30]
Italy (2018)	Spring, mineral, tap water	N/A	46	–	0.0008–0.0024	0.006–0.104	–	–	[42]
Iran (2015)	Bottled water (mineral & tap)	42	128	–	0.6–1.5	3–7.6	–	3.1–526.6	[33]
Sari, Iran (2015)	Bottled water (tap)	15	60	10–49	<LOD – 0.1	<LOD – 13.7	<LOD – 31	<LOD – 0.5	[40]
Bandar Abbas, Iran (2015)	Bottled water (tap)	8	432	–	0–7.1	–	–	–	[45]
Turkey (2013)	Bottled water	17	N/A	–	ND	ND – 0.013	–	ND – 0.011	[39]
	Tap water	–	78	–	ND	ND – 4.82	–	ND	
Dehli, India (2014)	Sachet water (tap)	N/A	16	ND	–	30–329	–	–	[43]
	Bottled water (tap)	N/A	4	<LOD - 16	–	<LOD - 58	–	–	
Nepal (2020)	Bottled water (tap)	N/A	100	–	ND	ND	0.8–23		[46]
Kampala, Uganda (2017)	Tap water	8	24	ND – 34	ND	17–310	ND - 170	5–14	[44]
	Spring water	12	36	ND – 98	ND	105–412	ND - 373	8–14	
	Bottled water (tap)	9	27	ND – 107	ND		ND	ND	
Serbia (2011)	Bottled water (mineral)	16	16	<0.21–1.51	<0.01–0.18	<0.04–1.06	<0.30–8.38	<0.20–6.32	[38]
	Bottled water (spring)	7	7	<0.21–1.57	<0.01–0.36	0.21–0.91	<0.30–3.52	<0.20–6.06	
	Tap water	4	4	0.25–0.58	0.08–1.19	0.33–2.89	0.90–1.82	2.20–6.41	
Khorram-Abad, Iran (2018)	Tap water	45	45	0.6–10.6	0.02–1.49	0.35–8.27	–	–	[28]
Rafsanjan, Iran (2021)	Well and tap water	23	23	1.02–10.53	0.51–1.66	4.12–33.42	–	7.02–102.22	[29]
Saravan, Iran (2021)	Well	89	89	0.49–20	0.11–12.8	0.1–36.02	–	–	[37]
Kurdistan province, Iran (2021)	Well	138	138	0.1–53	0.1–0.2	0.2–14.3	0.1–5.3	1–54	[25]
Yazd, Iran (2017)	Bottled water (tap)	2	2	–	<0.2 (Brand 1) <0.2 (Brand 2)	3.2 (Brand 1) 3.1 (Brand 2)	1.3 (Brand 1) 4.1 (Brand 2)	–	[31]

(2018) [28] in Khorramabad city in Iran. However, the Pb levels found in this work were higher than the levels reported by Maxwell et al. (2018) [30] in Nigeria, Zazouli et al. (2015) [40] in Sari city in Iran, Singla et al. (2014) in India, Bamuwamye et al. (2022) [44] in Uganda, Eslami et al. (2022) [29], Jafarzadeh et al. (2022) [37] and Maleki and Jari (2021) [25] in Iran. The observed levels for Ni in this study were higher than the levels reported in the studies of Ungureanu et al. (2022) in Romania, Maxwell et al. (2018) in Nigeria, Ristić et al. (2011) in Serbia and Maleki and Jari (2021) [25] in Iran; within the mean concentration range reported in the studies of Zazouli et al. (2015) [40] in Iran and Gautam (2020) [46] in Nepal. On the other hand, the Ni levels of this study were lower than the levels reported for samples of tap and ground water in the study of Bamuwamye el al (2022) [44] in Uganda. In this study, the levels of As was higher than the levels reported in the study of Zazouli et al. (2015) [40] and Bakırdere et al. (2013) [39]. It was also consistent with the levels of Ristić et al. (2011) [38] but lower than the levels reported by Hadiani et al. (2015) [33], Bamuwamye et al. (2022), Eslami et al. (2022) [29] and Jafarzadeh et al. (2022) [37].

In this study, a human health risk assessment (HHRA) was conducted according to USEPA guidelines based on the observed HM concentration in the samples (Table 3). The obtained point estimate HQ and estimated HQ at P95 for All of the studied HMs were below 1 in all the of the eleven studied samples indicating that the bottled water samples pose no adverse health effects at the determined concentrations. The findings of this study were in line with the results of Jafarzadeh et al. (2021) work on groundwater sources in Saravan, Iran. In their study, the HQ values were lower than one. This was explained by the low industrial activity in proximity of supply wells [29].Maleki et al. (2021) found similar results in terms of HI and HQ estimations for adults, except for As, the value of which was higher than one suggesting that consumers may experience health problems due to the exposure [25]. Similar findings (HQ > 1) were reported for As and Pb in the study of Eslami et al. (2021) [28]. Since this study has not investigated health risk for other age groups such as children, further research might be required to ascertain bottled water's safety for children in terms of non-carcinogenic risks as estimations for children may show higher risks due to their lower body weight [30]. In Ungureanu et al. (2022) study in Romania, from 50 samples taken from mineral bottled water, 30 % had HI values greater than 1. Also, HI index for groundwater fed tap water, spring and bottled water were all above 10 in the study of Bamuwamye et al. (2017) in Uganda, demonstrating the alarming rate of health risks.

However, the results of the carcinogenic health risk assessment showed opposite results. In this study, the estimations of lifetime cancer risk (LCR) for Cr levels showed that in 10 out of 11 samples, the risk level is higher than the maximum acceptable value (i.e. between 10-e6 and 10-e4) proposed by USEPA and only one sample (Sample I) the estimated value fell into the acceptable range. Additionally, the estimated LCRs for Cd and As were in the acceptable range. The highest estimated LCR values were obtained for Pb and in all samples, the LCR values were greater than the acceptable range (between 10e-6 and 10e-4) of USEPA. The MCS charts for each of the analyzed samples are shown in the supplementary material. In the study of Ungureanu et al. (2022) on mineral bottled water in Romania, only 28 % of the samples had estimated LCR values in the acceptable range [37]. In the study of Bamuwamye et al. (2017) in Uganda on spring, tap and bottled water samples, it was found that bottled water had significantly lower LCR values compared to other drinking water sources which was explained by the presence of very low levels of trace elements such as As [42]. Despite LCR levels of this study were determined to be in an acceptable range, it must be noted that this calculation is done to estimate cancer in the lifetime. Therefore, avoiding early life exposure to carcinogenic elements and chemicals is crucial due to incremental probability of carcinogenesis.

Another finding was that the point estimate values were greater than MCS values highlighting the importance of conducting MCS analysis to prevent overestimation caused by varying parameters.

5. Conclusions

From the obtained results in analysis of eleven brands of bottled water, it was demonstrated that the studied heavy metal concentrations in all brands were lower than the WHO and ISIRI. However, due to their accumulation properties, there is the possibility of adverse health effects by heavy metals for consumers of the studied drinking water brands. The determined HM concentration in this study was compared with other national and international studies on bottled water and other drinking water sources. Additionally, the results of non-carcinogenic health risk assessments using point estimations and MCS showed that the concentration of HMs in none of the samples pose adverse health effects. However, it was found that drinking the studied bottled water may potentially lead to carcinogenicity over the lifetime. This study is not without limitations. For example, investigating water samples from other supply sources such as municipal drinking water distribution or bottled water products filled by spring water have been missed which could provide a comparison between the sources. Additionally, this study only focused on medium sized bottled water products (500 mL) while larger sized bottled water packaging (1.5 L) was also available, though less consumed than the selected size in this study. Also, lack of individual human health risk assessment for different age groups and sexes were of other noted limitations of this study. A more comprehensive or a complimentary follow up study in the future can resolve the shortcomings of this study. In addition, it is suggested to use advanced treatment techniques such as reverse osmosis to be used in factories to further remove such contaminants from bottled water.

Author contribution statement

Nezam Mirzaei: Conceived and designed the experiments, analyzed and interpreted the data, materials, contributed reagents, materials, analysis tools or data, wrote the paper.

Safa Kalteh: Analyzed and interpreted the data, wrote the paper.

Hakime Zamani-Badi: Analyzed and interpreted the data, wrote the paper.

Heshmatallah Moradpour: Analyzed and interpreted the data, wrote the paper.

Zeinab Parmoozeh: Analyzed and interpreted the data, wrote the paper.

Mansour Baziar: Conceived and designed the experiments, analyzed and interpreted the data, materials, contributed reagents, materials, analysis tools or data, wrote the paper.

Data availability statement

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2023.e20647.

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