#### **RESEARCH ARTICLE**



# Deterministic and probabilistic human health risk assessment approach of exposure to heavy metals in drinking water sources: A case study of a semi-arid region in the west of Iran

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## Abstract

In the current study, the concentration of heavy metals (Ba, Mn, Pb, and Cd) in drinking water resources of 328 villages in Hamadan Province were measured using ICP-OES apparatus during two dry (September 2018) and wet (April 2019) seasons. The assessment of the non-carcinogenic risk of selected heavy metals was conducted based on the recommendations of the USEPA. Also, sensitivity analysis and uncertainty of the effective variables were performed using Monte-Carlo simulations. Based on the results, Mn level in drinking water samples ranged  $0.08-25.63 \mu g/L$  and  $0.08-20.03 \mu g/L$  in dry and wet seasons, respectively. Similarly, Ba levels in water samples ranged  $0.15-70.13 \mu g/L$  and  $0.84-65 \mu g/L$ . Also, Cd and Pb concentrations in all sampling sites were below the limits of detection (LOD) of the ICP-OES apparatus. The hazard index (HI) values for adult and children were  $2.17 \times 10^{-3}$  and  $3.29 \times 10^{-3}$ , respectively, which show a lack of non-carcinogenic risk for the examined heavy metals (Mn and Ba) to the local inhabitants. The results of the sensitivity analyses for adults and children revealed that two variables including metal concentration and ingestion rate of drinking water (IR) had the highest positive effects on the non-carcinogenic risk for the local residents in the studied area due to drinking water consumption.

Keywords Heavy metals · Sensitivity analysis · Hamadan Province · Non-carcinogenic risk

# Introduction

Drinking water sources are essential for human survival, but the existing problems such as water pollution and scarcity can be a threat to human health and the sustainable development

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of society [1]. The safety of drinking water has recently been of great importance because various anthropogenic activities in agriculture, industry, and geogenic agents like volcanic activities, bedrock erosion, and ore deposits may threaten its quality [2–5]. Among different contaminants which can affect the safety of drinking water, heavy metals are considered as one of the most serious problems to human health due to their high toxicity, bioaccumulation, and high persistence in the environment [6-8]. Therefore, consumption of drinking water containing noticeable amounts of heavy metals may cause adverse human health implications including hearing loss, reading and learning disabilities, cardiovascular diseases, lung problems, liver diseases, attention problems, and several types of cancer [9-11]. For example, high levels of Pb can cause adverse health effects including headaches, stomach cancer, abdominal pain, kidney damage, lung cancer and high blood pressure. Prolonged exposure to Cd causes acute and chronic effects on human health such as cancer and anemia. Similarly, maximum levels of Mn result in mental health problems such as Alzheimer's and manganism [12, 13]. Therefore,

determining and monitoring the concentrations of heavy metals in drinking water supplies has become one of the essential problems for health researchers due to the harmful effects of these elements. Consequently, for the quantitative assessment of the potential risks and study of the effects of exposure to trace elements, the human health risk assessment methods have been utilized [10]. The risk assessment method is an effective approach for assessing health risks due to various carcinogenic and non-carcinogenic pollutants; the method is obtained by calculating the daily consumption of contaminants through drinking water and the Hazard Quotient (HQ) [14].

In recent years, several research studies have examined the status of heavy metals pollution in different areas of Hamadan Province. For instance, Aleseyyed et al. reported the concentration of heavy metals (Hg, As, Cr, Cu, Pb, Cd, and Zn) in 60 samples of drinking water resources in urban and rural parts of Hamadan Province [15]. The non-carcinogenic health risk assessment of heavy metals: Pb, Cr, and Zn in drinking water resources in the city of Hamadan was evaluated by Farokhneshat et al. [16]. In another study, the contaminations of As, Zn, Pb and Cu in 20 groundwater samples during two seasons of spring and summer in the Qahavand Plain located in Hamadan were reported by Ardakani et al. [17].

Despite numerous studies related to the status of heavy metals in drinking water resources of Hamadan Province, there is limited information from the literature that reports the seasonal variations of heavy metal concentration in rural drinking water sources and the potential health risk related to these pollutants. Also, no comprehensive study has been conducted to determine the concentration of heavy metals in the drinking water sources of the rural areas of this province.

With the above background, the present study was designed to investigate the concentrations of heavy metals in drinking water resources of rural areas of Hamadan Province in two dry and wet seasons, as well as potential noncarcinogenic health risks associated with exposure to these metals. Additionally, uncertainty and sensitivity analyses for the selected heavy metals were conducted using the Monte Carlo-simulation approach in both age groups (adults and children) to recognize the critical input parameters and quantify uncertainties during the risk assessment method.

# Materials and methods

#### The study area description

-30 °C on some days of the year. This area has a semi-arid climate with an area of approximately 19,546 km<sup>2</sup>. The average annual precipitation in the studied area is reported to be 234.7 mm with approximately 70% falling throughout the rainy season [18]. Moreover, domestic and agricultural water demands in the studied area are mainly supplied by groundwater sources.

#### Sample collection, preparation, and analysis

Totally, 328 samples of drinking water were collected in polyethylene plastic bottles from the springs, wells, and distribution system network of the studied area during two dry (September 2018) and wet (April 2019) seasons. The drinking water samples were transported to the laboratory for the measurement of selected heavy metals (Pb, Cd, Mn, and Ba), followed by the addition of 65% HNO<sub>3</sub>. Then, the acidified drinking water samples were preserved in refrigerator. Heavy metal contents in the samples were measured using an ICP-OES apparatus (Model Spectro Arcos, Germany). It should be informed that the sampling process, preservation of samples, and analysis protocols were performed according to the procedures suggested by Standard Methods for the Examination of Water and Wastewater [19].

#### Quality control and quality assurance (QC/QA)

Heavy metals content in the samples was measured using an ICP-OES apparatus (Model Spectro Arcos, Germany). In cases where the concentrations of heavy metals were lower than the limits of detection (LOD), half of the LOD was considered as a representative value for statistical analysis [20]. In order to assess the accuracy of the data, the heavy metal concentration in the drinking water samples, standard spiked solutions (standard stock solution Fluka-51,844 and Fluka-54,704, Sigma-Aldrich, Switzerland), and blank samples were analyzed in triplicate. It should be informed that the relative standard deviations of repeated measurements were 5-10%. In the present study, to evaluate the efficiency of the method used in the analysis of the heavy metals, a standardized curve was drawn in the ranges of specified concentrations. Finally, a linear relationship was achieved between signal intensity and heavy metal concentrations (with the regression coefficients  $R^2 > 0.999$ ). Also, the ICP-OES apparatus was calibrated before the start of the measurement process and also during the analysis of the samples.

## Human health risk assessment

The health risk evaluation process is an important method for understanding the adverse health effects of exposure to contaminants in polluted media. This process is the first step in protecting safety and health. Human exposure to contaminants

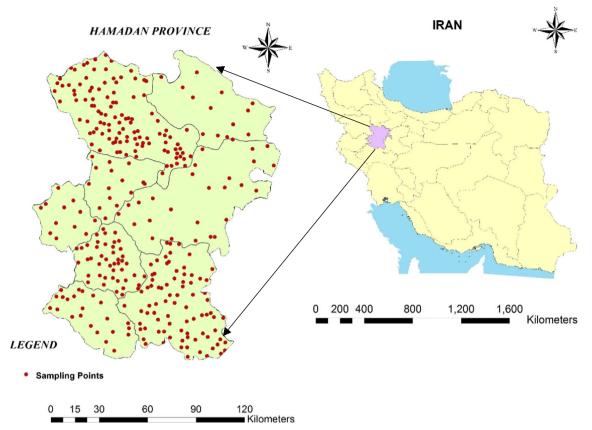


Fig. 1 Location map of the studied area and sampling sites

occurs through the three main pathways including inhalation, ingestion, and dermal absorption, which drinking water consumption is considered as the main route of exposure [21]. In the current study, the models proposed by the United State Environmental Protection Agency (USEPA) were utilized to estimate the non-carcinogenic effects of contaminant exposure [22]. For this purpose, the average daily dose (ADD) of a single parameter through the ingestion route for two subpopulation including adults and children can be calculated by the following equation (Eq. 1) [23].

$$ADD_{ing} = \frac{C_W \times IR \times ED \times EF}{BW \times AT}$$
(1)

Where,  $ADD_{ing}$  is the estimated ADD (mg kg<sup>-1</sup> day<sup>-1</sup>) of chemical constituent exposure through the oral pathway. Other parameters including C<sub>w</sub>, EF, BW, IR, ED, and AT represent the average contaminants levels in drinking water (mg L<sup>-1</sup>), exposure frequency (day yr<sup>-1</sup>), body weight (kg), ingestion rate (L day<sup>-1</sup>), exposure duration (yr), and average life time (day), respectively [23].

To quantify the risk characterization, a Hazard Quotient index (HQ) was used reflecting the potential non-carcinogenic risks (Eq. 2) [24].

$$HQ = \frac{ADD_{ing}}{RfD}$$
(2)

Where, RFD (mg kg<sup>-1</sup> day<sup>-1</sup>) represents the reference dose of selected heavy metals e.g. 0.0005, 0.0035, 0.14, and 0.2 for Cd, Pb, Mn, and Ba [9, 10, 25]. Hazard Index (HI) was defined as the sum of all the HQ values of different intake routes including inhalation, ingestion, and dermal absorption. However, the ingestion route is considered as the main pathway of exposure to contaminants in the present study. Therefore, the HI values were considered equally to HQ values for ingestion. It is worth noting that the HI values more than 1 have major health impacts, but HI values less than 1 do not have significant non-carcinogenic impact on consumers [26]. Therefore, the HI values are calculated using the following equation (Eq. 3) [9].

$$HI = \sum HQ \tag{3}$$

The assumption values of all the above input parameters used for calculating the risk assessment due to exposure selected heavy metals for both age groups (adults and children) have been depicted in Table 1.

# Uncertainty analysis using Monte-Carlo simulation method

There is a high degree of uncertainty in the deterministic approach of risk calculation due to variations in individual 
 Table 1
 Input assumption

 variables to risk assessment
 calculation

Parameters	Units	Representative value	References	
		Children	Adult	
BW	kg	Lognormal (32±3.2)	Lognormal (76±7.6)	[27]
ED	years	10	70	[27]
EF	days year <sup>-1</sup>	365	365	[24]
IR	$L day^{-1}$	Lognormal (1.25±0.57)	Lognormal (1.95±0.64)	[28]
AT	days	3650	25,550	[29]

characteristics and environmental differences [14]. To overcome this defect, the Monte-Carlo simulation approach was employed using the software Crystal Ball presented by Oracle Company (Oracle® Crystal Ball software). In the current study, the Monte-Carlo technique with 10,000 iterations was used to evaluate the non-carcinogenic risk of exposure to heavy metals (Ba and Mn) for children and adults. The sensitivity analyses were achieved using the Monte-Carlo technique to recognize the input parameters that have a greatest impact on the output of the risk assessment model.

# **Results and disscution**

# Distribution of heavy metals concentrations in drinking water samples

Descriptive statistics of trace elements including Pb, Mn, and Ba at 328 sampling sites with average, minimum, maximum, and standard deviation in two dry and wet seasons have been presented in Table 2. Because the concentration of Cd in all sampling sites was less than the LOD of the ICP-OES apparatus, its concentration has not been reported in Table 2. Also, in order to assess the suitability of water sources for drinking purposes, the concentration of each parameter was analyzed according to the standards of the World Health Organization (WHO). According to Table 2, the Mn concentration in drinking water samples ranged  $0.08-25.63 \mu g/L$  and 0.08- $20.03 \mu g/L$ , with mean values of  $0.97 \mu g/L$  and  $1.10 \mu g/L$ in both dry and wet seasons, respectively. Similarly, the Ba levels in water samples ranged  $0.15-70.13 \mu g/L$  and 0.8465  $\mu$ g/L, with mean values of 16.29 and 14.64  $\mu$ g/L in two dry and wet seasons, respectively. According to the results, the maximum concentration of Mn in the dry and wet seasons were 25.63 and 20.03 µg/L, respectively. Similarly, the maximum concentration of Ba in the dry and wet seasons were 70.13 µg/L and 65 µg/L, respectively. Therefore, it can be concluded that due to high precipitation and snow melts, the concentration of heavy metals in the wet season is lower than that of the dry season [7]. However, as can be seen in Table 2, the concentrations of Mn and Ba in all the drinking water samples in both dry and wet seasons were much lower than the maximum allowed concentrations recommended by the WHO. In the case of Pb, except for a limited number of samples that had a concentration lower than the recommended standard, the Pb levels in most samples were less than the limits of detection (LOD) of the ICP-OES apparatus. Based on a general conclusion, the levels of heavy metals examined in the current study were observed to be much lower than the recommended limits (Table 2). Therefore, according to these criteria, drinking water consumption in the studied area had no significant health risks for the local residents. However, some researchers believe that, in order to estimate the adverse health effects associated with heavy metals, it is not enough to just pay attention to the concentration of trace elements, and water resources should be evaluated by other indices as well [10].

The levels of heavy metals in drinking water sources in a region of western Iran (Shabestar province) was investigated by Barzegar et al. In their study, 29 samples of groundwater sources were gathered from different parts of the area, and the concentrations of heavy metals: Ni, Zn, As, Cr, Fe, Al, Cu, Mn, and Pb were estimated. The findings of their study

Table 2 Statistical characteristics of measured heavy metals (µg L<sup>-1</sup>) in drinking water samples

Element	Dry season			Wet season				$\begin{array}{c} \text{MCLWHO} \\ (\text{mg } \text{L}^{-1}) \end{array}$	
	Min	Max	Mean	S.D	Min	Max	Mean	S.D	(ing L )
Mn	0.08	25.63	0.97	2.35	0.08	20.03	1.10	2.25	0.4
Ba	0.15	70.13	16.29	13.94	0.84	65	14.64	11.57	0.7
Pb	1.08	5.17	1.10	0.26	1.08	5.17	1.11	0.33	0.01

presented that the concentrations of some heavy metals such as As, Zn, and Pb in several samples were higher than the standards recommended by the WHO [23].

In a study, Rasool et al. surveyed the concentration of heavy metals (As, Cd, Cu, Mn, Fe, Cr, Zn, Ni, and Pb) for 44 tube well drinking water samples in Punjab, Pakistan. Their results revealed that average concentrations of trace elements such as Fe, Cd, Pb, As exceeded the permitted limits recommended by the WHO. Untreated industrial wastewater, domestic wastewater, and extensive agricultural activities in the region have been mentioned as the main reasons for the contamination of water sources with Cd and Pb [30].

Moreover, human health risk assessment of exposure to heavy metals (As, Cd, Cr, Ni, Pb, B, Al, Hg, Mn, Zn, Cu, Fe, Se, and Ba) in drinking water sources of the city of Zahedan was evaluated by Dashtizadeh et al. In this study, the levels of the heavy metals in 155 drinking water samples were analyzed using an ICP-OES apparatus. The findings of their study showed that the average concentration of total heavy metals were much lower than the limits recommended by the WHO and USEPA. They also stated that, according to these criteria, drinking water consumption in the area lacks health risks [10].

In another study, the levels of Cd in groundwater sources in 39 villages of Bajestan and Gonabad (eastern Iran) were examined by Qasemi et al. The average concentration of Cd obtained in the studied rural areas for Gonabad ranged from 0.087 to 14.32  $\mu$ g/L, while in Bajestan, the mean levels of Cd ranged from 0.417 to 18.36  $\mu$ g/L. Based on the outcomes of this study, the researchers recommended that appropriate treatment methods should be used to remove Cd in contaminated rural areas [13].

### Human health risk assessment

# Non-carcinogenic human health risk assessment of exposure to heavy metals

In the current study, the measured concentration of heavy metals (Mn and Ba) was performed to evaluate the human health risk through ingestion of drinking water in both age groups of children and adults. It is worth noting that the concentration of Cd and Pb in most of the sampling sites was less than the limits of detection; therefore, the levels of these metals were not considered in the calculation of non-carcinogenic health risk. The mean values of HQ and HI for Ba and Mn in the two dry and wet seasons for both age groups of children and adults have been given in Table 3. According to the findings, in dry season, the HI values for children and adults were  $3.54 \times 10^{-3}$  and  $2.27 \times 10^{-3}$ , respectively. Also, in wet season the HI values for children and adults were  $3.17 \times 10^{-3}$  and  $2.08 \times 10^{-3}$ , respectively. As a result, due to lower body weights and shorter lifespans in children compared to

adults, the average non-carcinogenic risk of heavy metals (Mn and Ba) for children is more than the value of noncarcinogenic risk for adults [31]. In general, the HI values less than 1 indicate that there is no significant non-carcinogenic risk for the examined heavy metals (Mn and Ba), but routine monitoring due to the possibility of unpredictable contamination in the future is necessary.

In their study, Barzegar et al. estimated the human health risks of heavy metals (Fe, Ni, Mn, Co, Cr, Al, Cd, Zn, Pb, and As) in drinking water sources of the city of Khoy in north-western Iran. Based on the results of the health risk assessment presented in this study, Cr and As with HQ values of  $1 \times 10^{-4}$  and 11.55 had the lowest and highest effects of non-carcinogenic health risk on children and adults, respectively. Also, these high-risk sampling sites were situated in the southwest and northeast of the studied area where the drinking water was saline [32].

Heavy metal contamination by Pb, Mn, Cd, Ni, Cu, Cr, Zn, and Fe in 129 sources of drinking groundwater in the Vehari area of Pakistan and their health risk assessment were investigated by Khalid et al. The findings of their study displayed that the levels of metals such as Pb, Cd, and Fe in 93, 68, and 100% of the samples were higher than the permissible limits of the WHO, respectively. Also, the mean hazard quotient (HQ) values for all the reported metals were lower than 1, while Pb showed an HQ value higher than 1 envisaging non-carcinogenic risk with the ingestion of drinking groundwater. In this study, the highest and lowest HQ values were related to Pb (10.3) and Mn (0.02), respectively [33].

In another study, heavy metal sources (Pb, Cd, Zn, Fe, and Ni) and their pollution level in groundwater of Ghaziabad district, India, was examined by Chabukdhara et al. Among the heavy metals reported, the concentrations of Pb, Cd, Zn, and Fe were higher than the limits recommended by the WHO. The HQ values for Pb (2.4) and Cd (2.1) in children in the dry season were higher than the safe level (HQ = 1), while in the wet season, a high HQ value was reported only for Pb (1.23). Also, the HQ values for adults in both the dry and wet seasons were well within the safe limits [3].

#### Monte-Carlo simulation technique and sensitivity analysis

In the current study, the Monte-Carlo technique was performed to calculate the uncertainty of the exposure to Mn and Ba in the drinking water samples. The distribution of the HQ values for both age groups (adults and children) have been displayed in Fig. 2. According to Fig. 2, the HQ values for the 5th and 95th percentile of adults for Mn and Ba were  $1 \times 10^{-5}$  to  $6.7 \times 10^{-4}$  and  $4.1 \times 10^{-4}$  to  $5.3 \times 10^{-3}$ , respectively. In the case of children, the 5th and 95th percentile of the HQ values for Mn and Ba were  $1.3 \times 10^{-5}$  to  $1.08 \times 10^{-3}$  and  $5 \times 10^{-4}$  to  $8.6 \times 10^{-3}$ , respectively. Based on the results shown in Fig. 2, all the HQ

Table 3Non-carcinogenic riskvalues of selected heavy metals in<br/>drinking water samples

	Mn		Ba		Hazard Index	
	Adult	Children	Adult	Children	Adult	Children
Dry season Wet season	$1.78 \times 10^{-4}$ $2.02 \times 10^{-4}$	$2.7 \times 10^{-4}$ $3.08 \text{E} \times 10^{-4}$	$2.09 \times 10^{-3}$ $1.88 \times 10^{-3}$	$3.18 \times 10^{-3}$ $2.86 \times 10^{-3}$	$2.27 \times 10^{-3}$ $2.08 \times 10^{-3}$	$3.54 \times 10^{-3}$ $3.17 \times 10^{-3}$
Average	$1.9 \times 10^{-4}$	$2.89 \times 10^{-4}$	$1.98 \times 10^{-3}$	$3 \times 10^{-3}$	$2.17 \times 10^{-3}$	$3.29 \times 10^{-3}$

values for Mn and Ba were less than 1. Also, the HO values in Ba were higher than those in Mn, however, all the values were in the acceptable ranges. Therefore, the human health risk assessment of Mn and Ba exhibited that the HO values had an acceptable level of non-carcinogenic adverse health risk. Also, the results exhibited that the estimated average values of the HQ index obtained from the deterministic approach were nearly identical to the corresponding mean values from the Monte Carlo simulation model (Table 4). Qualitative sensitivity analyses were used to classify the parameters that have the greatest effect on the output values of the non-carcinogenic risk model (Fig. 3). The outcomes of the sensitivity analyses for adults showed that two variables including heavy metal concentration (Mn = 93.9%, Ba = 82.9%) and IR (Mn = 5.8%, Ba = 15.6%)had the highest positive effects on the non-carcinogenic risk assessment, compared to other input parameters. In the case of children, the results showed that two parameters including metal concentration (Mn = 89.1%, Ba = 72.8%) and IR (Mn = 10.4%, Ba = 25.4%) had the maximum positive effects on the non-carcinogenic risk calculation. It should be noted that, in both children and adults, BW had a negative impact on the output of the health risk assessment model.

Probabilistic health risk assessment (Monte-Carlo simulation technique) for heavy metals in drinking water samples of Singhbhum, India, was studied by Giri et al. Based on the findings of sensitivity analyses, two variables including heavy metal levels and exposure duration had the maximum impact on the output of the risk assessment. Also, their results showed that the 95th percentile of the HI values obtained by the Monte-Carlo technique for children, females, and males were 4.57, 2.54, and 2.87 for the dry season, and 3.75, 2.02, and 2.28 for wet season, respectively. Also, based on the results obtained from this study, the values of non-carcinogenic risks in children were higher than those in women and men [34].

 Table 4
 The estimated mean values of the HQ index from two assessment methods in the studied area

Assessment method	Adults		Children		
	Mn	Ba	Mn	Ba	
Deterministic method Monte-Carlo simulation					

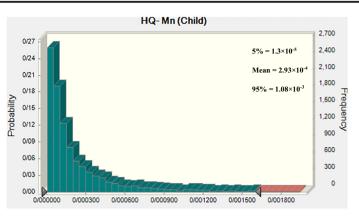
The non-carcinogenic human health risk of  $NO_3$  in Haryana water sources (India) was assessed using probabilistic (Monte-Carlo technique) and deterministic health risk assessment methods by Kaur et al. Their results displayed that the mean values of HQ index in both deterministic and probabilistic methods were almost the same, which correspond with the results of our study. Furthermore, based on their observations,  $NO_3$  concentration had an extreme impact on the output of the non-carcinogenic risk assessment model [14].

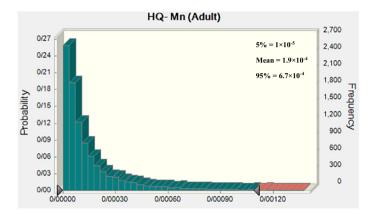
Probabilistic health risk assessment of Pb and Ni in drinking water sources of the city of Yazd was investigated by Fallahzadeh et al. According to their study, the lifetime cancer risks for the 5th and 95th percentile for Pb and Ni were  $5.47 \times 10^{-4}$  to  $1.8 \times 10^{-3}$  and  $1.01 \times 10^{-1}$  to  $2.65 \times 10^{-1}$  with mean values of  $1.09 \times 10^{-3}$  and  $1.67 \times 10^{-1}$ , respectively. The results of their sensitivity analysis illustrated that the heavy metal concentrations (Pb = 61.2%, Ni = 44.4%) and BW (Pb = -30.5%, nickel = -36.8%) were the most important parameters on the output values of lifetime cancer risk [6].

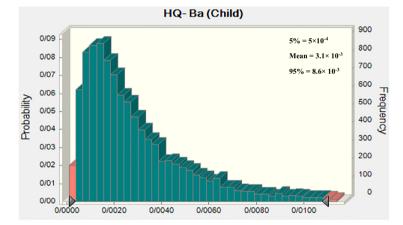
# Conclusion

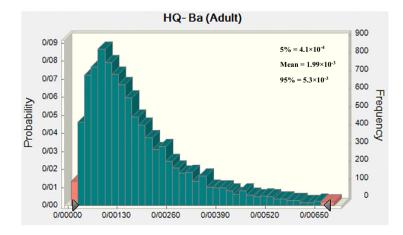
In the current study, the levels of heavy metals (Ba, Mn, Pb, and Cd) in 328 drinking water sources of the villages of Hamadan province, west of Iran, was assessed, and their non-carcinogenic health risks were analyzed based on the recommendations of the U.S EPA. Also, sensitivity analysis and uncertainty of the effective parameters were achieved using the Monte Carlo simulation. The findings indicated that the concentrations of Mn and Ba in all sampling sites were in accordance with the standards of the WHO for drinking water. Also, the levels of Pb and Cd in all the samples were lower than the limits of detection (LOD) of the ICP-OES apparatus. According to the results, in dry season, the HI values for children and adults were  $3.54 \times 10^{-3}$  and  $2.27 \times$  $10^{-3}$ , respectively. Also, in wet season the HI values for children and adults were  $3.17 \times 10^{-3}$  and  $2.08 \times 10^{-3}$ , respectively. The HI values <1 indicate that there is no significant non-carcinogenic risk for the examined heavy metals (Mn and Ba) in the studied area. In the case of Ba and Mn, the results revealed that the estimated average values of the HQ index obtained from the deterministic approach were nearly identical to the corresponding mean values from the Monte Carlo simulation model. Moreover, the outcomes of the sensitivity analyses showed that the heavy metals

**Fig. 2** Probabilistic risk assessment of HQ values for both age groups (adults and children)









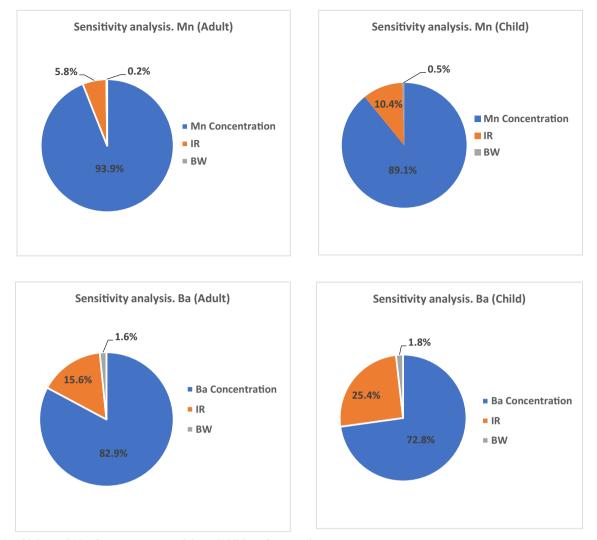


Fig. 3 Sensitivity analysis of two age groups (adults and children) for Ba and Mn

concentration for adults (Mn = 93.9%, Ba = 82.9%) and children (Mn = 89.1%, Ba = 72.8%) had the highest positive effects on the output of non-carcinogenic risk assessment, compared to other variables. It was also found that there was no significant non-carcinogenic risk for the examined heavy metals due to the consumption of drinking water for the local residents living in the studied area. Although, the drinking water sources of the villages of Hamadan province are not contaminated with heavy metals, due to the possibility of unpredictable pollution through the industrialization of communities, agricultural activities, and leakage of pollutants from various sources, regular monitoring of pollutants in drinking water sources is essential.

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### Declaration

Conflict of interest The authors declare that they have no conflict of interest.

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