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RESEARCH ARTICLE

Spatial analysis and health risk assessment of heavy metals concentration in drinking water resources

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Abstract The heavy metals available in drinking water can be considered as a threat to human health. Oncogenic risk of such metals is proven in several studies. Present study aimed to investigate concentration of the heavy metals including As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, and Zn in 39 water supply wells and 5 water reservoirs within the cities Ardakan, Meibod, Abarkouh, Bafgh, and Bahabad. The spatial distribution of the concentration was carried out by the software ArcGIS. Such simulations as non-carcinogenic hazard and lifetime cancer risk were conducted for lead and nickel using Monte Carlo technique. The sensitivity analysis was carried out to find the most important and effective parameters on risk assessment. The results indicated that concentration of all metals in 39 wells (except iron in 3 cases) reached the levels mentioned in EPA, World Health Organization, and Pollution Control Department standards. Based on the spatial distribution results at all studied regions, the highest concentrations of metals were derived, respectively, for iron and zinc. Calculated HQ values for non-carcinogenic hazard indicated a reasonable risk. Average lifetime cancer risks for the lead in Ardakan and nickel in Meibod and Bahabad were shown to be 1.09×10^{-3}, 1.67×10^{-1}, and 2×10^{-1}, respectively, demonstrating high carcinogenic risk compared to similar standards and studies. The sensitivity analysis suggests high impact of concentration and BW in carcinogenic risk.

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Keywords Groundwater · Metals · Health risk assessment · Monte Carlo simulation · Sensitivity analysis · Geographic information systems

Introduction

Water is a compound with specific chemical properties which can dissolve diverse compounds or keep them suspended (WHO 2007). In doing so, a serious environmental concern is currently the pollution of groundwater resources (Vodela et al. 1997). Among the pollutants which can affect water resources, the heavy metals are more paid into attention due to their high toxicity in low concentration (Marcovecchio et al. 2007). Heavy metals in water may exist as colloidal, particulate, or dissolved phase modes (Adepoju-Bello and Alabi 2005). Heavy metals such as iron, copper, cobalt, molybdenon, manganese, and zinc are useful for body but in low doses and can serve as catalyst for enzyme activities. However, they are toxic in high doses (Adepoju-Bello et al. 2009).

In case of uncontrolled heavy metals in the environment, they can lead to anti-health effects such as poor growth and development, cancer, nervous system damage, and death. Touching some heavy metals including lead and mercury may result in occurrence of autoimmunity diseases where one's immune system acts against the body's cells and damages them (Barakat 2011). Maximum contaminant level (MCL) standard is presented in Table 1. This standard is published by the United States Environmental Protection Agency (USEPA) for the heavy metals (in particular those used in present study) along with their effects on human health (Babel and Kurniawan 2003).

With increasing expansion of industries and uncontrolled waste disposal in the environment, pollution of drinking water

Table 1 The MCL standards for the most hazardous heavy metals (Babel and Kurniawan 2003)

Heavy metal	Toxicities	MCL (mg/L)
Arsenic	Skin manifestations, visceral cancers, vascular disease	0.050
Cadmium	Kidney damage, renal disorder, human carcinogen	0.01
Chromium	Headache, diarrhea, nausea, vomiting, carcinogenic	0.05
Copper	Liver damage, Wilson disease, insomnia	0.25
Nickel	Dermatitis, nausea, chronic asthma, coughing, human carcinogen	0.20
Zinc	Depression, lethargy, neurological signs, and increased thirst	0.80
Lead	Damage the fetal brain; diseases of the kidneys, circulatory system, and nervous system	0.006
Mercury	Rheumatoid arthritis and diseases of the kidneys, circulatory system, and nervous system	0.00003

by heavy metals may result in adverse effects on human health (Geng et al. 2016; Alves et al. 2014). The knowledge on pollution quality in the groundwater polluted by heavy metal is so useful to employ such waters as a resource for diverse applications (Karbassi et al. 2007). Given the studies conducted on disadvantages of heavy metal pollution on human health, the International Agency for Research on Cancer (IARC) developed a classified system for assessment of carcinogenic properties of chemical pesticides, and for this purpose, a health hazard model was generated by USEPA for carcinogens and non-carcinogens (USEPA 2005; Wiltse and Dellarco 1996). Most recently, quantitative health risk assessment has frequently been used as a reasonable method to calculate metal pollutants' risk potential (Ferre-Huguet et al. 2009).

Geostatistics is widely used to determine changes and spatial distribution of pollutants (Liu et al. 2004). ArcGIS is used as a powerful software for environmental modeling and

Table 2 Some studies on health risk assessment of heavy metals among different environments

Authors	Heavy metals studied	Study environment	Results
M. Javed et al. (2016)	Mn, Fe, Co, Ni, Cu, and Zn	Freshwater fish, *Mastacembelus armatus*	Non-carcinogenic risk for Co and Ni only; hazard index (HI) was high; Carcinogenic risk TR was 3.43×10^{-3} and 3.91×10^{-3} for male and female, respectively for Ni.
N. Maghakyan et al. (2016)	Cd, As, Pb, Cr, Ni, Co, Zn, Cu, Ag, Hg, and Mo	Yerevan's tree leaves	Non-carcinogenic risk (HI > 1) for children and adults at 13 and one sampling sites, respectively. Carcinogenic risk (>1:1,000,000) for Cr and As was observed in 14 and 25 samples, respectively.
D. M. Cocârță et al. (2016)	Be, Cd, Cr, Ni, and Pb	Soil	Potential risk of human health was higher than the acceptable one after World Health Organization.
Lingming Lei et al. (2015)	Hg, As, Cd, Cr, Pb, Cu, Zn, and Ni	Wheat flour	HMs did not cause non-carcinogenic risks in the study area (HI < 1); Cd generated the greatest carcinogenic risk
V. Demir et al. (2015)	Sb, As, Cd, Cr, Cu, CN, Pb, Hg, Ni, and Se	Drinking water	In the samples collected from Mazgirt area HQ was >1 for As; HQ values were <1 in the other samples.
J. Nawab et al. (2016)	Ni, Mn, Cr, Pb, and Cd	Drinking water	For both adults and children, the HRI values of HMs were <1.
P. Wongsasuluk et al. (2014)	As, Cd, Cr, Cu, Hg, Ni, and Zn	Shallow groundwater wells in an agricultural area	The highest HQs for non-carcinogenic risk was for Cu and Pb, with a range of 0.053–54.818 and 0.007–26.80, respectively; the HI values exceeded acceptable limits in 58% of the wells.
P. Chanpiwat et al. (2014)	As and Ba	Groundwater	Only As was contributing to non-carcinogenic health risks in all studied areas; As groundwater concentration, average daily dose of As, exposure duration, and subject body weight were the important factors in the non-carcinogenic and carcinogenic risks

Table 3 Population and geographic information of cities and wells

City	Population	City location	Study area (wells) location	Number of wells	Average flow (lit/s)
Ardakan	77,875	31° 36′ N–55° 24′ E	31° 28′ N–31° 40′ N, 55° 17′ E–55° 41′ E	9	32.22 ± 2.63
Meybod	82,840	32° 15′ N–54° 0′ E	32° 12′ N–32° 13′ N, 53° 57′ E–54° 00′ E	8	28.12 ± 2.58
Abarkouh	46,662	31° 7′ N–53° 17′ E	30° 52′ N–31° 12′ N, 52° 50′ E–53° 10′ E	9	22.22 ± 2.63
Bafgh	53,161	31° 36′ N–55° 24′ E	31° 28′ N–31° 40′ N, 55° 17′ E–55° 41′ E	8	30.62 ± 3.20
Behabad	27,800	31° 52′ N–56° 1′ E	31° 28′ N–31° 40′ N, 55° 17′ E–55° 41′ E	5	24.00 ± 4.18

geostatistics studies (Fedra 1994). So far, geographic information system (GIS) has been used to model and evaluate diverse pollutants including those of water (Merchant 1994; Khosravi et al. 2017), soil (Cattle et al. 2002), and air (Yan et al. 2015; Briggs et al. 1997) in several studies. GIS is a suitable tool to study and simulate quality of groundwaters in diverse regions, and it is used to prepare risk assessment map to estimate the corresponding human health impacts (Mimi and Assi 2009; Ghodeif et al. 2013; Arnous et al. 2011).

The health risk assessment of heavy metals is conducted in different studies for various foodstuffs (Javed and Usmani 2016; Lei et al. 2015), soil (Cocârță et al. 2016), dust (Maghakyan et al. 2016), and water (Demir et al. 2015; Nawab et al. 2016; Wongsasuluk et al. 2014; Chanpiwat et al. 2014; Lu et al. 2015). Table 2 shows a summary of these studies.

Due to improper management of industrial wastewater and solid waste in the studied areas, presence of high concentrations of heavy metals and the corresponding risk for environment and inhabitants are assumed. Therefore, investigations on concentration of heavy metals in groundwater resources are necessary and helpful. The present study aimed to estimate concentration of heavy metals in groundwaters of five cities including Abarkouh, Ardakan, Meibod, Bafgh, and Bahabad located in Yazd Province at geographical center of Iran. For this purpose, not only metals were estimated in groundwater of mentioned cities by the software ArcGIS but also quantitative spatial distribution of such metals were conducted in water. Then, the non-carcinogenic hazard and lifetime cancer risk (LTCR) associated with the metals studied here were investigated to estimate the corresponding human health impacts.

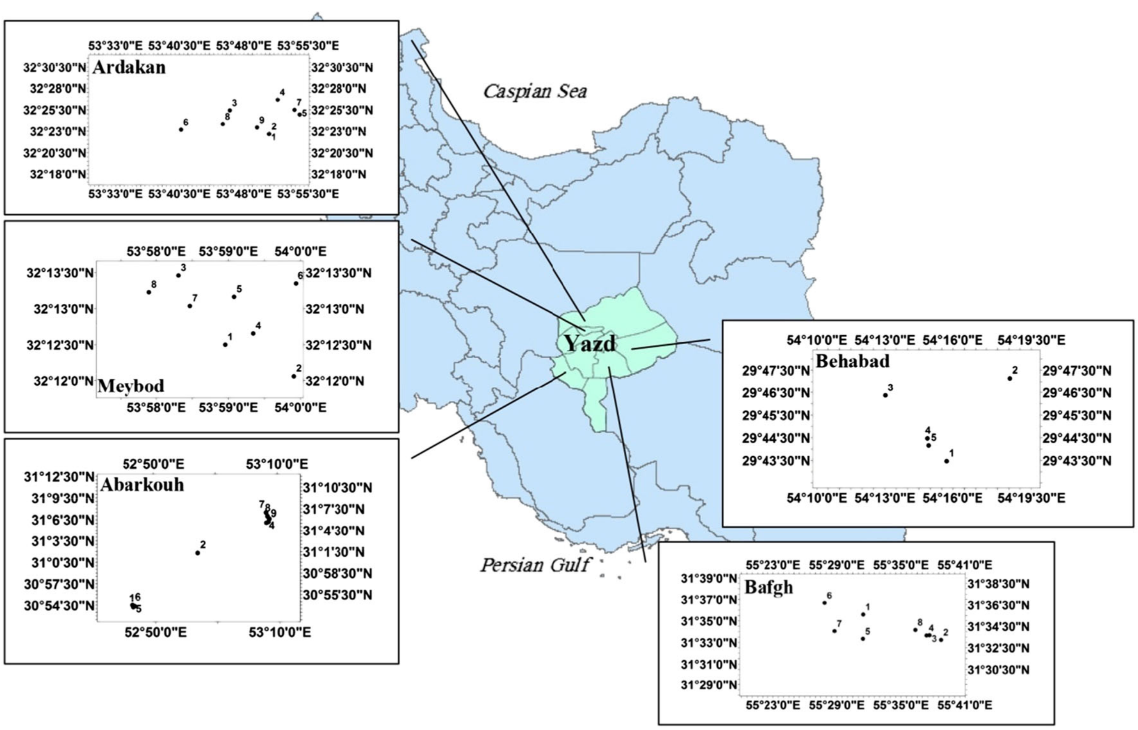


Fig. 1 Geographic position of wells in studied cities

Table 4 Concentration of heavy metals in water of the wells supplying drinking water among the studied cities

City	Well no.	Cr (ppb)	Cu (ppb)	Fe (ppb)	Mn (ppb)	Ni (ppb)	Pb (ppb)	Zn (ppb)
Abarkouh	1	4.05 ± 0.02	12.10 ± 0.73	40.21 ± 4.21	1.12 ± 0.91	2.93 ± 0.89	ND	40.49 ± 5.21
	2	4.13 ± 0.23	9.76 ± 1.25	39.56 ± 6.12	1.03 ± 0.48	2.55 ± 0.05	ND	18.31 ± 3.31
	3	3.21 ± 1.01	11.03 ± 3.18	105.09 ± 9.31	3.52 ± 1.06	ND	ND	26.46 ± 5.12
	4	3.09 ± 0.92	13.42 ± 2.63	23.16 ± 5.02	1.14 ± 0.77	ND	ND	26.10 ± 2.43
	5	3.64 ± 0.61	14.37 ± 3.05	21.60 ± 3.66	1.94 ± 1.07	1.21 ± 0.12	ND	22.91 ± 4.09
	6	3.00 ± 0.91	11.70 ± 2.11	25.03 ± 2.75	1.20 ± 0.49	1.06 ± 0.69	ND	20.12 ± 3.71
	7	2.12 ± 1.04	9.64 ± 0.99	27.14 ± 5.10	1.00 ± 0.61	ND	ND	134.42 ± 9.15
	8	2.03 ± 0.43	10.21 ± 2.15	45.30 ± 7.72	1.62 ± 0.87	ND	ND	16.78 ± 3.12
	9	2.29 ± 0.31	13.82 ± 0.32	16.10 ± 3.19	1.49 ± 1.12	1.87 ± 0.43	ND	12.30 ± 4.01
Ardakan	1	18.46 ± 3.15	15.36 ± 2.81	353.21 ± 10.95	12.10 ± 2.03	ND	4.20 ± 1.10	16.61 ± 2.17
	2	11.14 ± 2.60	13.21 ± 3.16	345.72 ± 16.23	17.32 ± 4.19	ND	ND	14.91 ± 3.98
	3	5.14 ± 1.14	8.07 ± 1.06	436.20 ± 11.94	4.82 ± 1.19	ND	5.91 ± 2.14	44.03 ± 6.09
	4	5.03 ± 0.91	14.19 ± 3/21	847.31 ± 13.83	27.40 ± 3.34	ND	ND	16.21 ± 2.15
	5	3.85 ± 1.84	9.43 ± 1.19	225.62 ± 5.59	13.90 ± 2.19	ND	ND	100.44 ± 8.95
	6	3.31 ± 0.74	12.81 ± 1.73	53.03 ± 4.11	1.49 ± 0.88	ND	2.43	36.10 ± 4.15
	7	3.04 ± 0.96	9.54 ± 2.11	303.10 ± 8.91	5.32 ± 1.68	ND	ND	18.64 ± 2.98
	8	3.11 ± 0.12	13.40 ± 2.67	1459.25 ± 21.48	20.14 ± 4.01	ND	ND	14.40 ± 3.66
	9	2.41 ± 1.21	10.63 ± 3.13	701.51 ± 8.12	22.31 ± 2.95	ND	ND	26.12 ± 2.10
Meybod	1	12.60 ± 1.08	9.28 ± 0.52	66.92 ± 3.09	1.06 ± 0.99	4.04 ± 1.16	ND	12.31 ± 4.01
	2	6.30 ± 2.90	13.21 ± 1.77	81.98 ± 2.97	1.54 ± 1.04	4.51 ± 1.75	ND	14.69 ± 2.69
	3	5.01 ± 1.11	11.26 ± 3.01	28.30 ± 5.62	1.10 ± 0.65	3.59 ± 0.99	ND	20.01 ± 5.11
	4	3.09 ± 2.65	15.67 ± .44	212.61 ± 10.42	4.76 ± 1.01	3.18 ± 1.74	ND	22.94 ± 2.33
	5	3.51 ± 0.41	13.12 ± 3.10	166.95 ± 8.39	8.40 ± 2.59	3.00 ± 1.57	ND	18.12 ± 3.70
	6	3.26 ± 0.19	12.06 ± 1.69	185.14 ± 10.14	4.16 ± 1.04	2.64 ± 1.06	ND	16.41 ± 1.96
	7	1.04 ± .09	14.11 ± 2.01	42.09 ± 4.17	1.32 ± 0.77	3.92 ± 0.57	ND	16.20 ± 3.52
	8	1.00 ± 0.53	11.67 ± 1.93	48.49 ± 6.03	1.00 ± 0.04	3.02 ± 1.31	ND	10.85 ± 1.19
Bafgh	1	5.05 ± 1.14	11.21 ± 3.71	910.83 ± 16.39	33.41 ± 3.39	2.50 ± 0.66	ND	172.90 ± 15.93
	2	3.14 ± 1.94	15.04 ± 2.18	3.12 ± 1.10	47.19 ± 3.95	1.31 ± 1.02	ND	70.40 ± 6.75
	3	3.08 ± 0.33	21.43 ± 4.01	22.05 ± 3.94	1.16 ± 0.91	ND	ND	28.98 ± 4.14
	4	3.26 ± 0.82	8.90 ± 3.10	29.41 ± 0.71	1.05 ± 1.01	1.01 ± 0.49	ND	26.12 ± 3.67
	5	3.00 ± 1.51	11.05 ± 0.51	1308.91 ± 21.09	29.41 ± 2.20	2.14 ± 0.94	ND	10.51 ± 1.19
	6	2.87 ± 1.04	12.43 ± 2.70	1436.06 ± 14.96	24.85 ± 3.96	ND	ND	428.61 ± 13.44
	7	2.46 ± 0.64	9.06 ± 1.41	799.31 ± 18.01	28.73 ± 5.01	1.72 ± 0.04	ND	184.11 ± 8.56
	8	2.09 ± 1.01	12.11 ± 2.97	149.92 ± 4.71	4.68 ± 1.33	ND	ND	92.04 ± 5.15
Behabad	1	5.43 ± 1.30	11.67 ± 1.29	28.11 ± 3.31	1.30 ± 0.58	3.44 ± 1.19	ND	20.21 ± 3.95
	2	4.14 ± 2.01	14.15 ± 4.01	71.93 ± 5.18	1.75 ± 0.30	2.61 ± 0.93	ND	12.09 ± 1.13
	3	3.70 ± 0.93	13.90 ± 2.92	103.60 ± 2.97	1.09 ± 0.86	1.39 ± 0.56	ND	18.12 ± 2.90
	4	3.12 ± 0.22	11.01 ± 2.14	132.38 ± 4.19	2.15 ± 1.49	3.10 ± 1.12	ND	14.61 ± 3.67
	5	2.43 ± 0.96	11.93 ± 3.21	113.26 ± 2.98	8.41 ± 2.93	7.71 ± 2.05	2.40 ± 0.95	36.19 ± 5.06
Drinking Groundwater standard (μg/L)	EPA (2012)	100	1300	NM	NM	NM	15	800
	WHO (2011)	50	2000	NM	400	70	10	NM
	PCD (2000)	50	1500	1000	500	20	10	5000

Ar, Cd, and Hg were not detected

ND not detected, *NM* not mentioned

Table 5 Concentration of heavy metals in the water reservoirs separated by the city

City	Cr (ppb)	Cu (ppb)	Fe (ppb)	Mn (ppb)	Ni (ppb)	Pb (ppb)	Zn (ppb)
Abarkouh	2.90 ± 0.93	11.45 ± 2.64	36.65 ± 3.62	1.20 ± 0.55	1.22 ± 0.92	ND	33.75 ± 4.18
Ardakan	5.82 ± 1.25	11.39 ± 1.79	504.44 ± 12.67	13.13 ± 2.61	ND	2.71 ± 0.85	32.27 ± 2.93
Meybod	4.22 ± 1.92	12.26 ± 0.63	103.86 ± 9.37	2.66 ± 1.19	3.13 ± 1.05	ND	15.95 ± 3.10
Bafgh	2.85 ± 0.68	12.04 ± 1.09	586.57 ± 10.12	21.02 ± 4.18	1.24 ± 0.43	ND	127.38 ± 12.09
Behabad	3.41 ± 1.73	11.91 ± 2.12	90.95 ± 6.94	2.41 ± 0.43	3.08 ± 1.19	2.22 ± 0.56	19.41 ± 3.15

Ar, Cd, and Hg were not detected

ND not detected

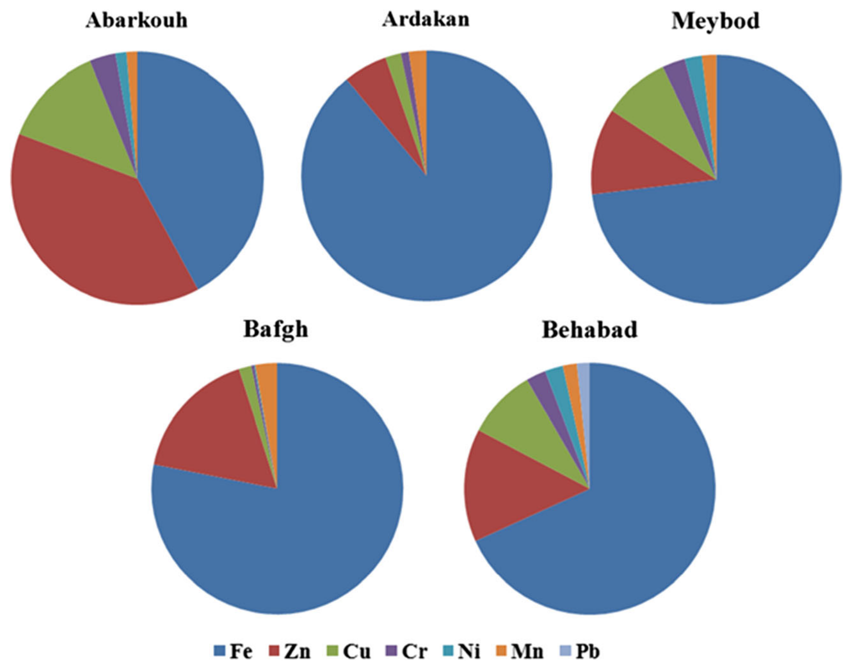


Fig. 2 The water type in diverse cities in terms of concentration of heavy metals relative to each other

Ultimately, sensitivity analysis was carried out to determine the most important and effective factors for LTCR calculation.

Materials and methods

Studied area

Our studied area consisted of five cities including Ardakan, Meibod, Abarkouh, Bafgh, and Bahabad located in Yazd Province at geographical center of Iran. Yazd Province with a hot and dry desert climate is located in piedmont of Yazd-Ardakan plain at coordinates of eastern 54° 22′ 3″ and northern 31° 53′ 50″ (Fallahzadeh et al. 2016). The drinking water in mentioned cities is supplied through groundwater resources. Demographic and geographic details of these cities and region are presented in Table 3. Figure 1 shows geographic position of wells in the studied cities.

Data collection and analysis

A sample of 500 mL was provided from each of 39 drinking water wells and 5 storage tanks in Ardakan, Abarkouh, Meibod, Bafgh, and Bahabad in 2016 at four times (once a season). Totally, 176 analyses were implemented to determine concentration of heavy metals. The samplings were conducted in polyethylene dishes, while by increasing 2 mL of concentrated acid nitric, pH of the samples reached to lower than 2 and the samples were transferred to the laboratory, keeping a temperature of 4 °C. The heavy metal measurement method for the samples was based on what mentioned in the book "Standard Method" with the number A3120, 21st edition (2015), and was analyzed by inductively coupled plasma-mass spectroscopy (ICP-MS; PerkinElmer, model ELAN DRCe) (Federation and Association 2005). In this study, the metals arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), mercury (Hg), manganese (Mn), nickel (Ni), lead (Pb), and zinc (Zn) were quantified.

The limits of detection (LODs) for As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, and Zn were 11.6, 7.6, 92, 51, 71, 2.0, 4.17, 3.63, 1.24, and 11.8 ng L^{-1}, respectively; the limits of quantity (LOQs) for these metals were 165.62, 75.96, 310, 170, 240, 6.0, 41.68, 36.29, 12.36, and 117.53 ng L^{-1}, respectively.

In order to assure accuracy of data, standard reference materials (Merck, Germany) were included in every batch of sample and analysis as a part of the quality control protocol. Each sample was analyzed three times and two standards were used as test references after every 10 samples. The calibration curves were linear within the concentration range, with the regression coefficients (R^2) > 0.999. Relative standard deviations of repeated measurements were <10%. These results showed that the elemental analysis method was both reliable and precise. Recovery

Table 6 Order of heavy metal concentration in the studied cities

City	HM concentration
Abarkouh	Fe > Zn > Cu > Cr > Ni > Mn > Hg
Ardakan	Fe > Zn > Mn > Cu > Cr > Pb > Ni > Hg
Meybod	Fe > Zn > Cu > Cr > Ni > Mn > Hg
Bafgh	Fe > Zn > Mn > Cu > Cr > Ni > Hg
Behabad	Fe > Zn > Cu > Cr > Ni > Mn > Pb > Hg

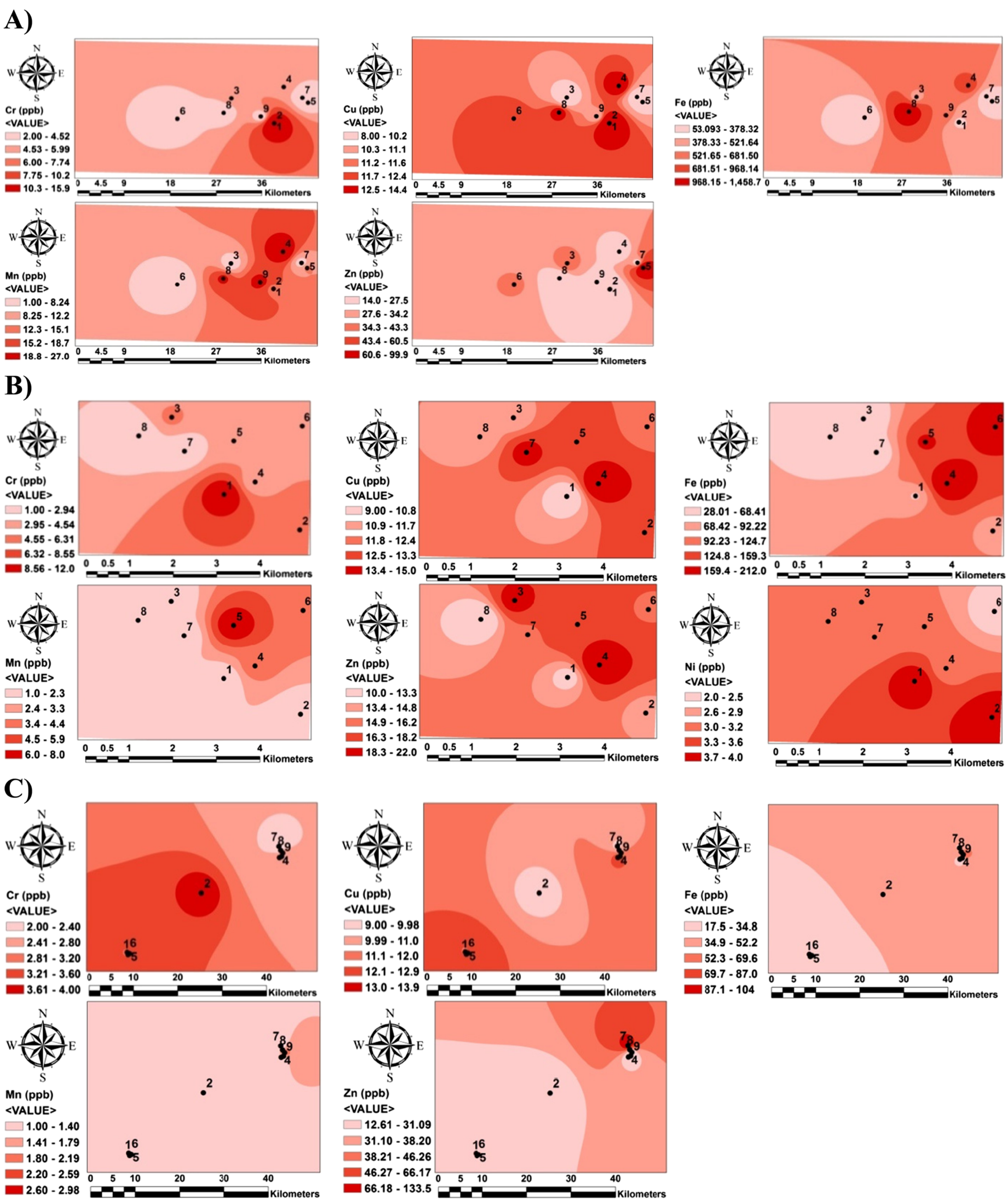


Fig. 3 Spatial distribution of heavy metal concentration for **a** Ardakan, **b** Meibod, **c** Abarkouh, **d** Bafgh, and **e** Bahabad

studies of metal determination were conducted to demonstrate the efficiency of the methods. Only the elements with given certified values and mean recoveries ranging from 90 to 110% were included in the data analysis.

Spatial distributions

The software ArcGIS version 10.0 created by ESRI Co. was used for zoning/spatial distribution of heavy metals in drinking

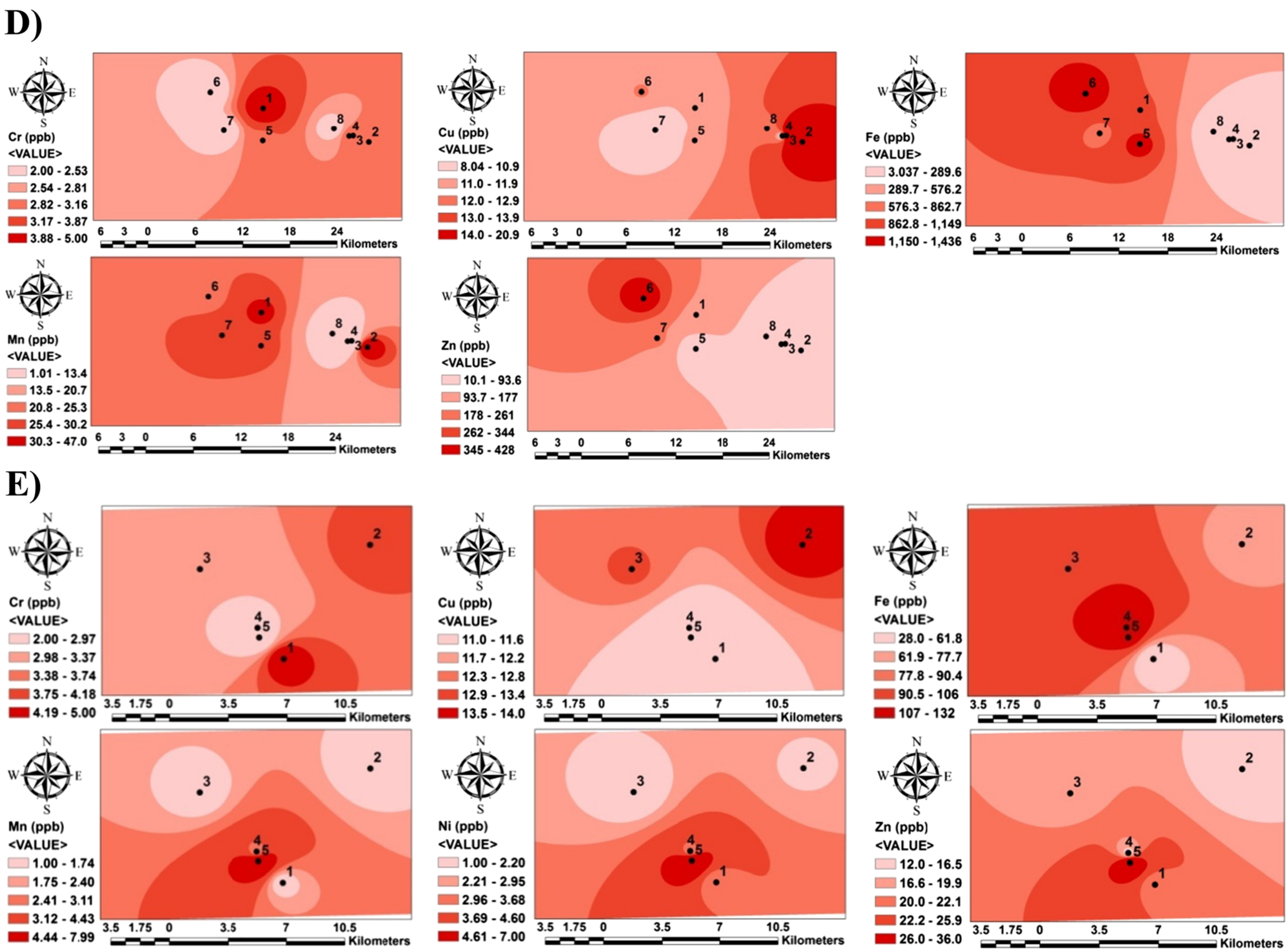


Fig. 3 (continued)

water. Interpolation technique or inverse distance weighting (IDW) was employed for spatial distribution of each heavy metal and development of the independent raster layer associated with concentration of contaminant in different points of this area. IDW has been used in several studies for spatial distribution of contaminants including assessment of heavy metals in groundwater of West Bokaro and its spatial distribution (Tiwari et al. 2016) as well as investigation of groundwater quality of Eğirdir Lake basin and risk assessment using GIS (Şener et al. 2016). IDW is a non-statistical method which is employed for environmental studies to predict concentration of contaminants at unmeasured locations through the optimal spatial prediction technique (Miri et al. 2016). IDW assumes that prediction values have a linear function with existing data. IDW is calculated as follows (Xie et al. 2011):

$$Wi = \frac{Di{-}\propto}{\sum_{i=1}^{n} Di{-}\propto,} \tag{1}$$

where W is the weight of the station i, D_i is the distance between the point i and the place with unspecified values, $\propto$ is the weighting power, and n is the total number of specified points used in a spatial distribution.

Health risk assessment

In this section, the non-carcinogenic hazard and the LTCR associated with the studied metals in this work were studied to evaluate the corresponding human health impacts. The non-carcinogenic hazards associated with the studied metals were calculated by the following equation as hazard quotient (HQ):

$$\mathrm{HQ} = \frac{\mathrm{LEC}}{\mathrm{RfC}} \tag{2}$$

In the present study, LEC and RfC are annual average of daily received concentration (mg L^{-1}) and non-carcinogenic reference concentration of the pollutant, respectively. LTCR is calculated through multiplication of chronic daily intake (CDI) by cancer slope factor (CSF), which is determined by Integrated Risk Information System (IRIS) as follows (Miri et al. 2016):

Table 7 Non-carcinogen risk of elements in different city of Yazd Province in terms of hazard quotient (HQ)

City	Element	RfC (μg/kg/day)	Concentration (μg/L)		Hazard quotient (HQ)
			Mean	SD	
Abarkouh	Cr	3	2.88	0.73	0.03
	Cu	40	11.33	1.69	0.01
	Fe	300	37.88	25.45	0.003
	Mn	20	1.22	0.62	0.001
	Ni	20	1.22	0.42	0.001
	Zn	300	34.88	35.34	0.003
Ardakan	Cr	3	5.88	4.97	0.06
	Cu	40	11.44	2.66	0.008
	Fe	300	524.66	399.85	0.052
	Mn	20	13.44	8.66	0.02
	Pb	1.4	3.86	1.08	0.027
	Zn	300	31.55	26.19	0.003
Meybod	Cr	3	5.33	3.19	0.04
	Cu	40	12.25	1.78	0.009
	Fe	300	103.5	67.81	0. 01
	Mn	20	2.62	3.39	0.003
	Ni	20	3.12	0.59	0.004
	Zn	300	16	3.74	0.001
Bafgh	Cr	3	2.87	0.92	0.028
	Cu	40	12.37	3.8	0.009
	Fe	300	582	564	0.058
	Mn	20	20.87	15.93	0.031
	Ni	20	2	0	0.0007
	Zn	300	126.25	129.43	0.021
Behabad	Cr	3	3.4	1.01	0.034
	Cu	40	12	1.26	0.009
	Fe	300	89.4	36.51	0.008
	Mn	20	2.6	2.72	0.003
	Ni	20	3.75	1.92	0.004
	Pb	1.4	2.4	0	0.01
	Zn	300	20	8.48	0.002

$$LTCR = \mathrm{CDI} \times \mathrm{CSF} \quad (3)$$

CDI is found as follows:

$$\mathrm{CDI} = \frac{\mathrm{C} \times \mathrm{IR} \times \mathrm{ED} \times \mathrm{EF}}{\mathrm{BW} \times \mathrm{AT}} \quad (4)$$

where C is the concentration of the pollutant (μg L^{-1}), IR is the polluted water's daily ingestion rate (L day^{-1}), ED is the annual exposure duration (year), EF is the annual exposure frequency (days $year^{-1}$), BW is the average body weight (kg), and AT is the average lifetime (days).

Given the literature review and the values presented by EPA, the parameters involved in calculation of CDI, LTCR, and HQ were used. The values of ingestion rate (IR), exposure duration (ED), average body weight (BW), average time (AT), and exposure frequency (EF) used in previous studies are 2.2 L day^{-1}, 70 years, 70 kg, 25,550 days, and 360 days $year^{-1}$, respectively (Wu et al. 2009; Liang et al. 2011). The CSF values for the lead (Pb) and nickel are 0.009 and 1.7 μg g^{-1} day^{-1}, respectively (Koki et al. 2015). CSF values have not been reported for additional metals measured in present study. Therefore, the risk of carcinogenicity was calculated only for lead and nickel.

In our study, sensitivity analysis technique was used to determine how different values of input variables can affect risk estimation in a given set of assumptions. In addition, risk assessment was carried out by Monte Carlo simulation technique using the software Crystal Ball version 11.1.2.3 created by Oracle Co. (Oracle® Crystal Ball software version 11.1.2.3) (Dan et al. 2016; Ren et al. 2016; Greenland 2001).

Results and discussion

Heavy metal concentration

In this study, concentration of the heavy metals including As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, and Zn was quantified in ppb and compared to guidelines of USEPA (2012), World Health Organization (WHO) (2011), and Pollution Control Department (PCD) (2000). Among the metals studied in water of 39 wells of the 5 cities, the metals As, Cd, and Hg were recognized in none of the samples. Ni was not recognized in wells of Ardakan while the Pb was not seen in groundwater of Abarkouh, Meibod, and Bafgh. Totally, As, Cd, and Hg were found in none of the samples; Cr, Cu, Fe, Mn, and Zn were seen in 100% of the samples; and Ni and Pb were found in 58.92 and 10.25% of the samples, respectively. Concentration of the metals measured in the drinking water is presented in Table 4. The results indicated that concentration of all elements (except Fe) reveals the level mentioned at the guidelines of EPA, WHO, and PCD in some wells of Bafgh and Ardakan. High concentration of iron in underground water resources of the cities Bafgh and Ardakan is due to industrial activities for iron extraction and its industrial processing. There are iron ore mines in Bafgh, as well as iron processing industries such as pelletizing and rolling. The sludge and effluent flowing from sewage treatment plants of these industries will be released into the environment. There are also Chadermullo iron ore and iron reduction and pelletizing industries in the Ardakan that flow from sewage treatment plants are released to environment somewhere upstream of some drinking water wells which can penetrate to aquifer. The drinking water studied here had a concentration lower than that of previous studies (Tiwari et al. 2016; Zhao et al. 2016).

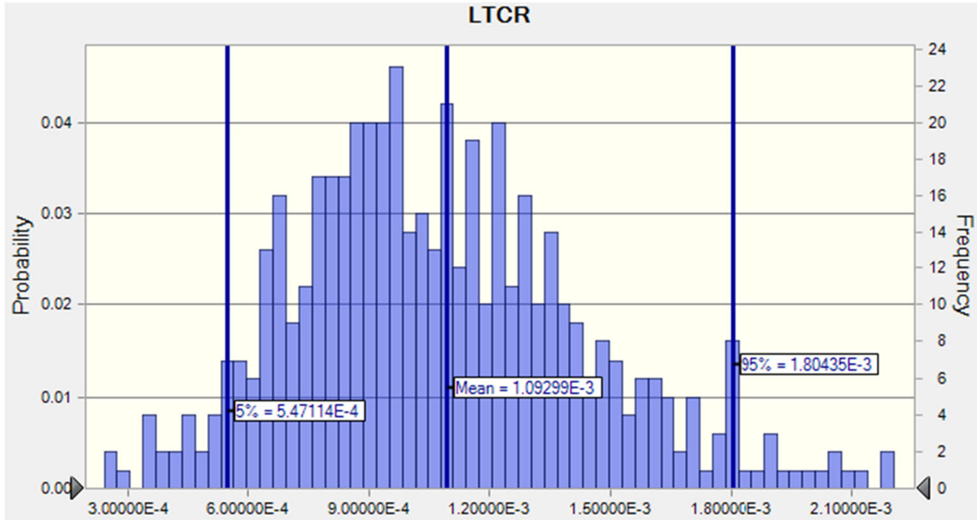


Fig. 4 Predicted probability of lifetime cancer risk (LTCR) for Pb in Ardakan

Upon exiting the wells, the underground water at catchment area of the studied cities is transferred to the storage tanks from which underground resources are transferred to the distribution network. Water accumulation in a tank can affect the concentration of heavy metals and result in its dilution. Table 5 shows concentration of heavy metals in the storage tanks separated by the city. As it can be seen in Table 5, concentrations of the studied metals are in the level mentioned by EPA, WHO, and PCD standards and that of Fe in Ardakan and Bafgh was measured by dilution, which is in standard range.

Figure 2 shows quantity of heavy metals relative to each other in different cities. The highest concentrations of metals were obtained by Fe and Zn, respectively. Order of heavy metal concentrations in the five cities are presented in Table 6.

Spatial distribution

Using the software ArcGIS version 10.0 and IDW technique, spatial distribution map for heavy metal concentration was provided for each studied city. Figure 3 shows spatial distribution of heavy metal concentration separated by metal type and the city.

Health risk assessment

Table 7 presents the non-carcinogenic hazard associated with the elements studied in each city. As it can be seen, the highest risk in the cities Abarkouh, Ardakan, Meibod, and Bahabad is associated with Cr, while it is associated with Fe in Bafgh. If HQ exceeds 1, the risk will be unacceptable, and in case of HQ < 1, the risk will be acceptable (Wongsasuluk et al. 2014). Given the values obtained for HQ of heavy metals, the non-carcinogenic hazard is concluded to be acceptable for all metals. In some of previous studies, this risk is greater than 1 for arsenic, which is recognized to be due to anthropogenic activities such as the use of herbicides (Kavcar et al. 2009; Alves et al. 2014).

Using Monte Carlo method, the carcinogenic hazard associated with the lead in Ardakan and nickel in Meibod and

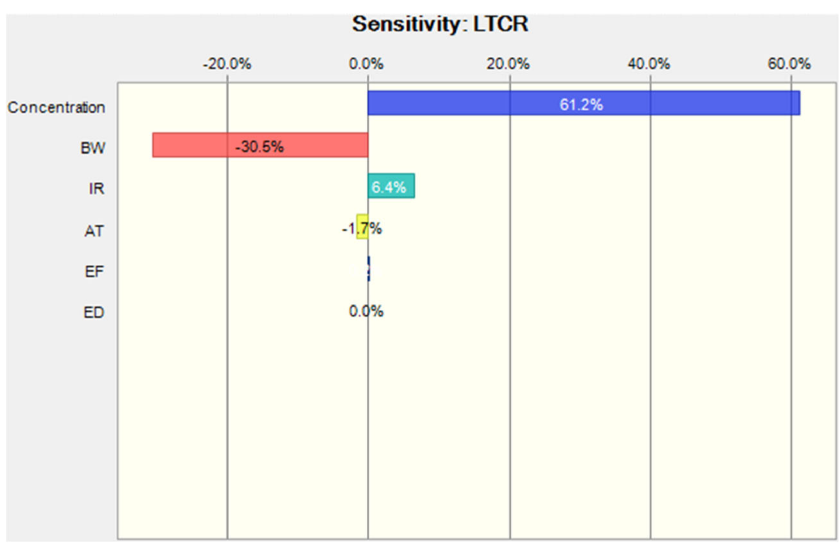


Fig. 5 Sensitivity analysis of LTCR model for Pb in Ardakan

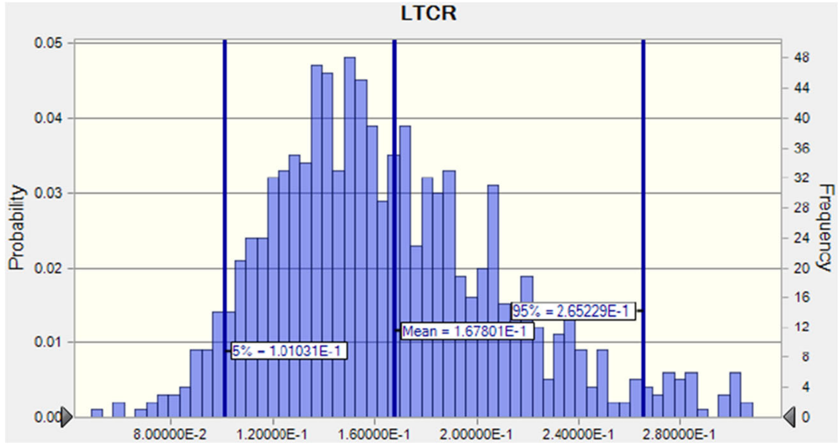


Fig. 6 Predicted probability of lifetime cancer risk (LTCR) for Ni in Meibod

Bahabad was studied. Figure 4 shows the prediction for probability of lifetime cancer risk for the lead in Ardakan. The mean LTCR for this metal is 1.09×10^{-3}. The risks of 5 and 95% were obtained as many as 5.47×10^{-4} and 1.8×10^{-3} for this metal. This risk is very high compared to the maximum reported acceptable risk (1×10^{-4}) (Lu et al. 2015). The risk value for the lead was calculated 4.4×10^{-5} in the similar study conducted in Shenzhen, China, indicating a value lower than what calculated in present work (Lu et al. 2015).

Sensitivity analysis was conducted to determine importance of the variables involved in LTCR calculation (Ma et al. 2000). Such an analysis for the lead in Ardakan showed that the two factors including heavy metal concentration and BW are the most important factors on LTCR values. The concentration of 61.2% and BW of −30.5% had, respectively, the highest positive and negative impacts on LTCR calculation, compared to additional parameters (Fig. 5).

The mean probability of lifetime cancer risk for nickel in Meibod was 1.67×10^{-1}, while the risks of 5 and 95% were equivalent to 1.01×10^{-1} and 2.65×10^{-1} (Fig. 6). The maximum determined acceptable risk by WHO is 1×10^{-2} for nickel (Turdi and Yang 2016), which is lower than the values reported in present study. Sensitivity analysis for LTCR calculation regarding nickel showed that the two variables including metal concentration of 44.4% and BW of −36.8%, respectively, had the highest positive and negative impacts on carcinogenic hazard calculation. In a study conducted by Geng regarding risk assessment of drinking water in two regions of China, the risk value in the studied areas was lower than maximum acceptable risk. Sensitivity analysis also indicated that heavy metal concentration has the highest positive impact on risk calculation (Geng et al. 2016) (Fig. 7).

LTCR calculation for nickel in Bahabad indicated that the mean carcinogenic risk for this metal is 2×10^{-1} while the risks of 5 and 95% are 3.6×10^{-2} and 3.88×10^{-1}, respectively, which are lower than the determined acceptable risk by WHO (Turdi and Yang 2016). Also, sensitivity analysis indicated that the concentration and BW of 87.5 and −8.2%,

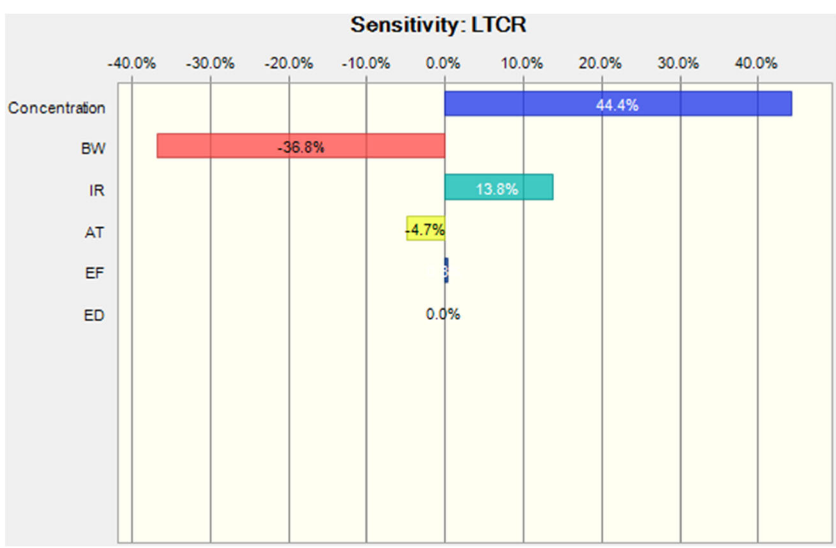
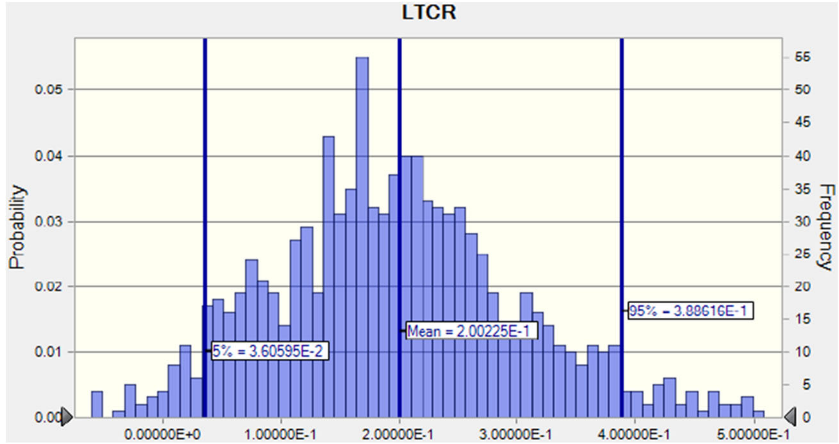


Fig. 7 Sensitivity analysis of LTCR model for Ni in Meybod

Fig. 8 Predicted probability of lifetime cancer risk (LTCR) for Ni in Behabad



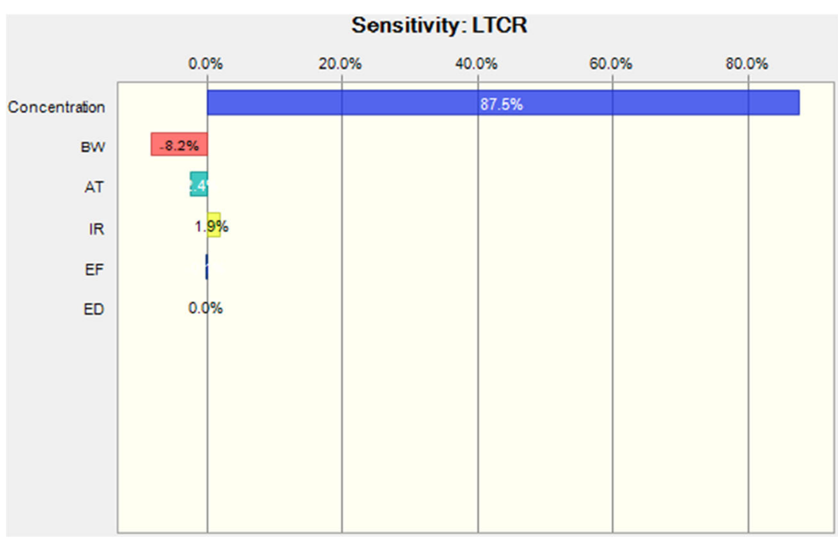
respectively, have the highest positive and negative impacts on LTCR calculation (Figs. 8 and 9).

Ardakan and Meybod are the two cities with the highest industrial settlements in Yazd Province. Industrial wastewater from the industrial settlements of these two cities is collected without pre-treatment and eventually enters into the central wastewater treatment plant. In the treatment plant, the coagulation and flocculation processes are done in order to remove suspended solids. Soluble contaminants such as heavy metals cannot be eliminated completely with this process and released into the environment by the effluent of wastewater treatment plant which increases the potential for groundwater contamination and health risks.

Conclusion

Present work studied concentration of heavy metals such as Cd, Cr, Cu, Fr, Hg, Mn, Ni, Pb, and Zn in 39 water supplying wells and 5 water storage tanks among the cities Ardakan, Meibod, Abarkouh, Bafgh, and Bahabad. The results of present study indicated that among the 10 studied metals, concentration of iron is higher than standard level in water of 3 wells of Ardakan and Bafgh. However, combination of these wells with other wells resulted in a heavy metal concentration lower than maximum valid limit which is not considered as a threat for public health. Iron and zinc, respectively, had the highest concentration among the metals studied here. Non-carcinogenic risk analysis showed that the risk is acceptable for all metals of the wells but carcinogenic risk analysis for lead in Ardakan and nickel in Meibod and Bahabad estimated the risk relative to the maximum determined risk in the standard, and thereby, it indicates a high risk. The sensitivity analysis was conducted to determine the impact of the variables involved in LTCR calculation, and the results showed that the two factors including heavy metal concentration and BW, respectively, are the most important factors with positive and negative impacts on LTCR values among the studied metals.

Fig. 9 Sensitivity analysis of LTCR model for Ni in Behabad



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