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ORIGINAL PAPER

Levels, origins and probabilistic health risk appraisal for trace elements in drinking water from Lhasa, Tibet

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Abstract Due to the lack of monitoring systems and water purification facilities, residents in western China may face the risk of drinking water pollution. Therefore, 673 samples were collected from Lhasa's agricultural and pastoral areas to reveal the status quo of drinking water. We used inductively coupled plasma-mass spectrometry to determine trace elements concentrations for water quality appraisal, source apportionment, and health risk assessment. The results indicate that concentrations of V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd, Ba, and Pb are below the guidelines, while As concentrations in a few samples exceed the standard. All samples were classified into "excellent water" for drinking purpose based on Entropy-weighted water quality index. Thereafter by principal component analysis, three potential sources of trace elements were extracted, including natural, anthropogenic, and mining activities. It is worth noting that geotherm and mining exploitation does not threaten drinking water safety. Finally, health risks were assessed using Monte Carlo technique. We found that the 95th percentiles of hazard index are 1.80, 0.80, and 0.79 for children, teenagers, and adults, indicating a non-carcinogenic risk for children, but no risks for the latter two age groups. In contrast, the probabilities of unacceptable cautionary risk are 7.15, 2.95 and 0.69% through exposure to Cr, Ni, As, and Cd for adults, children, and teenagers. Sensitivity analyses reveal As concentration and ingestion rate

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are most influential factors to health risk. Hence, local governments should pay more attention to monitoring and removal of As in the drinking water.

Keywords Drinking water · Trace elements · Source apportionment · Probabilistic health risk · Monte Carlo simulation · Contamination control

Introduction

The drinking water resource is a basis for human survival and development. However, with the population growth and extension of anthropogenic activities, the drinking water is polluted widely (Islam et al., 2015; Li et al., 2011). Among these pollutants in drinking water, trace elements cannot be ignored due to their high toxicity, long persistence, and bioaccumulation potential (Canpolat et al., 2020; Tudi et al., 2019). Although at low concentrations (Xie et al., 2019), trace elements in aquatic ecosystems play an important role for human health. Cu, Zn, Mo, Se are essential elements for human life (Ćurković et al., 2016), but excessive ingestion of them is harmful (Lu et al., 2015), whereas some other trace elements may pose seriously adverse health effects at a low content. For instance, kidney, nervous and hematopoietic system, respiratory tract and skin will be damaged by excessive exposure to Cd, Pb, and Cr (Cao et al., 2019; Panhwar et al., 2016). Ingestion of Ba may cause hypertension (Phan et al., 2013). What is worse, exposure to high levels of As was confirmed to cause different kinds of diseases, including hypertension, cerebrovascular disease, skin lesions, stillbirth, spontaneous abortion, cancers, and so on (Islam et al., 2012; Smith & Steinmaus, 2009; Wu et al., 2012). In particular, the people in developing countries are facing more threats by exposure to trace elements in drinking water, due to the lack of monitoring systems and the proper treatments.

In China, it was reported that more than 200 million people are still using unsafe water (Gao et al., 2019; Qiu, 2009). It is estimated that every year 190 million people in China fall ill and 60,000 people die from diseases caused by water pollution such as liver and gastric cancers (Qiu, 2011). So that it is urgent to conduct studies on drinking water safety nationwide. In recent years, studies on drinking water were carried out in major river basins in China, including the Yangtze River Basin (Gu et al., 2020; Liang et al., 2018), Yellow River Basin (Li et al., 2014), Hai River Basin (Gao et al., 2019), Huai River Basin (Qiu et al., 2021; Wang et al., 2017), Pearl River Basin (Liu et al., 2017), etc. These studies focused on the drinking water quality appraisal, pollutant sources apportionment, and potential health risk assessment. Overall, the water quality in northern China is worse than that in southern China and arsenic is the predominant contaminant (Xiao et al., 2019). These results are the basis for local drinking water management and water resources utilization. However, the previous studies are mainly concerned with high-density population and developed areas in eastern China while those in the western regions are still very scarce.

Lhasa is the most developed city of Tibet; it is relatively isolated from other districts and is usually considered to be less disturbed by humans due to its high elevation and harsh climate conditions (Dai et al., 2019). However, the rapidly growing economy and human development have induced some disturbance on the local environment, especially on the surface water quality (Huang et al., 2010; Li et al., 2013; Mao et al., 2019). In the agricultural and pastoral areas of Lhasa, drinking water is mainly from surface rivers (Ye et al., 2016), trace elements in water may threaten human health for lack of proper treatments. To our best knowledge, there is no comprehensive research on the drinking water of Lhasa. Trace elements in drinking water have an influence not only on local residents but also on more populations downstream at home and abroad. Thus 673 drinking water samples in agricultural and pastoral areas of Lhasa were collected to (1) determine the trace elements concentrations in drinking water of Lhasa, including V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Ba, and Pb; (2) evaluate the suitability of drinking water in Lhasa based on Entropy-weighted water quality index (EWQI); (3) use multivariate statistical methods to analyze the potential sources of trace elements in drinking water; and (4) perform a probabilistic health risk assessment with Monte Carlo simulation, finding out most contributing factors. Results of this study can provide a prior understanding of trace elements in the drinking water, which is essential for contamination monitoring and removal in the future.

Materials and methods

Study area

The study area (Fig. 1) is the agricultural and pastoral areas of Lhasa (excluding Chengguan District), covering an area of 31,662 km^2 (89°44′-92°38’ E, 29°14′-31°3′ N), with the highest population density and industrial activity intensity in the Tibet Autonomous Region. Lhasa is one of the highest cities in the world, with an average elevation of 3650 m. The terrain is high in the north and low in the south. The middle and south regions are the valley plains of the Lhasa River, a main tributary of the Yarlung Zangbo River.

The study area belongs to the semi-arid monsoon climate, with an average annual precipitation of 530 mm, most of which occurs during the monsoon period from June to September (Peng et al., 2015). In the study area, the Lhasa River is the main stream. It originates from the Nyenqintangula Mountains on the Qinghai-Tibet Plateau, extending from northeast to southwest and eventually flowing into the Yarlung Zangbo River. Residents are mainly distributed along the Lhasa River and its five tributaries (Lhachu, Razheng Tsangpo, Xuerong Tsangpo, Meldromarchu, and Tölungchu), where high intensity industrial and agricultural activities occupy, such as manufacturing, service industry, mining, and geothermal exploitation (Zhang et al., 2018). In particular, the Gyama Valley, the Yangbajain and the Yangyi geothermal plants have great potential for exploration (Wang et al., 2020; Ying et al., 2014), which might affect drinking water safety. Moreover, residents mainly drink surface water lacking proper treatments, so the matter of drinking water safety should be focused on.

Sampling and analysis

According to the standard inspection method for drinking water (GB/T 5750.2-2006), we collected 673 drinking water samples from households. Meanwhile,

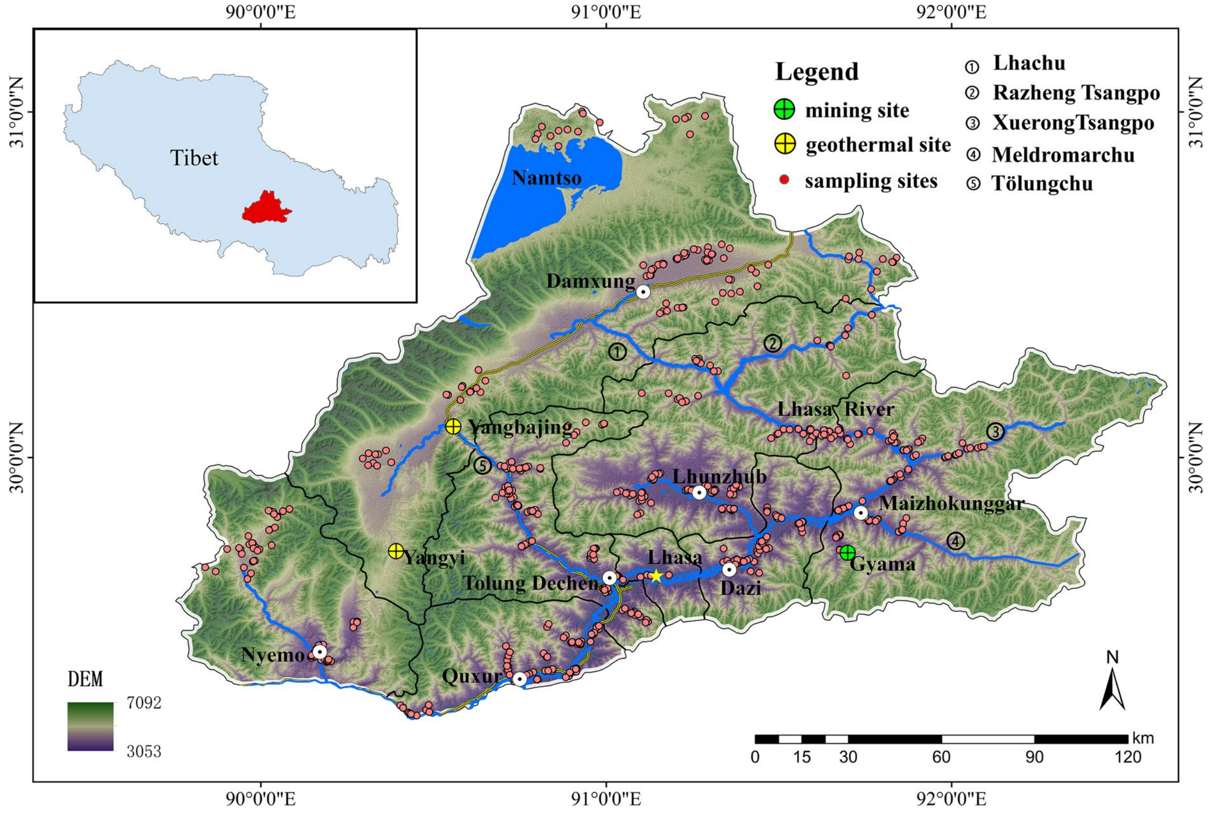


Fig. 1 Location of the study area and sampling sites

a portable GPS was used to record the coordinate information of the sampling sites. After collection, the water samples were filtered through 0.45 µm cellulose acetate membrane (Whatman GmbH, Germany) and stored in 1-L polyethylene bottles, which were pretreated with high-density nitric acid (Pesticide residue grade, Germany MERCK) and ultra-pure water (18.5 MΩ). Afterward, the filtered samples were acidified (nitric acid 65% Suprapure® MERCK, Germany) to pH < 2 in suit and stored at 4 °C, preventing any aging and pollution during transportation and storage.

Subsequently, the samples were sent to the Institute of Tibetan Plateau Research for laboratory analysis. The concentrations of trace elements were determined by inductively coupled plasma-mass spectrometry (ICP-MS; X-7 Thermo Elemental, USA). Each sample was measured twice in parallel, and the relative standard deviations (RSD) of trace elements were lower than 5%.

Entropy-weighted water quality index

Water quality index was proposed by the National Health Foundation of the United States in 2017, currently being applied worldwide (Meng et al., 2016; Sener et al., 2017). It is an indicator for measuring the comprehensive impact of various pollutants on water quality and reflecting the overall status of water quality. The key of the water quality index calculation is to determine the weight of every pollutant. Herein, the principle of information entropy was adopted to determine the weights for eliminating the subjective effects (Wang & Li, 2022; Zhang et al., 2021). EWQI is calculated by four steps:

(1) Construction of eigenvalue matrix X. In Eq. (1), the matrix X contains the information of samples number and elements types:

$$X = \begin{bmatrix} x_{11} & x_{12} & \cdots & x_{1n} \\ x_{21} & x_{22} & \cdots & x_{2n} \\ \vdots & \vdots & \ddots & \vdots \\ x_{m1} & x_{m2} & \cdots & x_{mn} \end{bmatrix} \tag{1}$$

where, m, n equal 673 and 12, respectively.

(2) Calculation of standard-grade matrix. Y is transformed from X by normalization. To eliminate the effect of dimension quantity grade of water quality variables, the normalization can be performed by Eqs. (3) and (4):

$$Y = \begin{bmatrix} y_{11} & y_{12} & \cdots & y_{1n} \\ y_{21} & y_{22} & \cdots & y_{2n} \\ \vdots & \vdots & \ddots & \vdots \\ y_{m1} & y_{m2} & \cdots & y_{mn} \end{bmatrix} \tag{2}$$

$$y_{\mathrm{ij}} = \begin{cases} \frac{x_{\mathrm{ij}} - (x_{\mathrm{ij}})_{\min}}{(x_{\mathrm{ij}})_{\max} - (x_{\mathrm{ij}})_{\min}} \, \textit{Benefit type}(3) \\ \frac{(x_{\mathrm{ij}})_{max} - x_{\mathrm{ij}}}{(x_{\mathrm{ij}})_{\max} - (x_{\mathrm{ij}})_{\min}} \textit{Cost type}\,(4) \end{cases}$$

${x_{\mathrm{ij}}}_{\max}$, ${x_{\mathrm{ij}}}_{\min}$ is the maximum and minimum concentration, respectively.

Equation (4)was selected here.

(3) Calculation of information entropy "e_j" and entropy weight "w_j", according to Eqs. (5), (6), and (7):

$$e_j = -\frac{1}{\ln m} \sum_{i=1}^{m} \left(P_{\mathrm{ij}} \times \ln P_{\mathrm{ij}} \right) \tag{5}$$

$$P_{\mathrm{ij}} = \frac{y_{ij}}{\sum_{i=1}^{m} y_{ij}} \tag{6}$$

$$w_j = \frac{1 - e_j}{\sum_{i=1}^{n} (1 - e_{j)}} \tag{7}$$

where, P_{ij} is the index j value for sample i.

(4) Obtain the EWQI value using Eqs. (8), (9):

$$q_j = \frac{C_j}{S_j} \times 100 \tag{8}$$

$$\mathrm{EWQI} = \sum_{j=1}^{n} (w_j \times q_j) \tag{9}$$

where, C_j is the concentration of ith element; S_i is drinking water guidelines in China.

Statistical analysis

By using principal component analysis (PCA), interrelationships between various indexes are considered, with which multivariate indicators are converted into a few unrelated comprehensive indicators through a linear transformation. Thus we can obtain dimensionality reduction in multivariate data without information loss (Borůvka et al., 2005; Wu et al., 2009). For water quality analysis, this method is mainly used to extract pollution factors and identify major sources of pollutions (Pekey et al., 2004).

PCA was performed in the Statistical Software Package SPSS (Version26.0) for Windows. Primarily, Kaiser–Meyer–Olkin (KMO) and Bartlett's sphericity test were applied to judge the suitability of the dataset for PCA. It is feasible to run PCA when the value of KMO test is greater than 0.5 and the significance of Bartlett's sphericity test is less than 0.05. The appropriate number of principal components is determined by filtrating the eigenvalues and the cumulative contribution of principal components.

Health risk assessment

We adopted the health risk assessment model recommended by the United States Environmental Protection Agency (US EPA). This model is widely used for evaluating current and future health risks of pollutants in drinking water to the exposed population (Jafarzadeh et al., 2022; Zhang et al., 2017). Oral intake, air inhalation, and skin contact are considered as three main pathways through which pollutants pose a health risk to the human body. It was reported that the intake dose of pollutants through the first pathway is higher (Hossain & Patra, 2020; Ijumulana et al., 2020). Thereafter, children, teenagers and adults were separated to assess the health risk through exposure to trace elements in drinking water. According to the risk assessment manual of the US EPA, the exposure dose (ADD) was calculated using Eq. (10):

$$\mathrm{ADD} = \frac{C_w \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \tag{10}$$

Thereafter, the health risk was evaluated based on ADD. Hazard index (HI) and hazard quotient (HQ) are used to characterize the potential non-carcinogenic risk (NCR). Where $\mathrm{HQ_i}$ and HI represent the NCR of the ith element and the total, respectively. The calculation equations are as follows:

$$\mathrm{HQ} = \frac{\mathrm{ADD}}{\mathrm{RfD}} \tag{11}$$

$$\mathrm{HI} = \mathrm{HQ}_1 + \mathrm{HQ}_2 + \cdots + \mathrm{HQ}_i \tag{12}$$

The carcinogenic risk (CR) is the possibility of cancer risk over a lifetime period due to exposure to the trace elements, by using Eq. (13):

$$\mathrm{CR} = \mathrm{ADD} \times \mathrm{SF} \times 10^{-3} \tag{13}$$

All above input parameters were summarized in Table 1 and Table S1.

Probabilistic risk modeling and sensitivity analysis

High uncertainties remain during the health risk assessment when the deterministic method is adopted (Zhang et al., 2017). The input parameters of the health risk assessment model are all single-point values, which often take the upper limit of the possible range, leading to the conservatism and uncertainty of the model (Kaur et al., 2020). To overcome this matter, Monte Carlo simulation technique was introduced, with which the exposure dose was calculated by repeated simulations from randomly chosen values within their range of variability (Glorennec et al., 2007). The simulation was performed using Oracle Crystal Ball (Version 11) loaded in Microsoft Excel with 10,000 times running. Before running the model, we used the "Fit Distribution" tool to obtain the optimal concentration distribution of 12 trace elements. The other parameters were collected from previous studies. Meanwhile, sensitivity analysis was performed to further determine the most contributing variables to health risk assessment. All results were visualized by MATLAB and OriginPro software.

Results and discussion

Statistical characteristics of trace elements

The statistical results of 12 trace elements concentrations measured from 673 samples are shown in Table 1. The mean concentration of 12 trace elements are in the following order: Fe(64.12 μg/L) > Zn(23.38 μg/L) > Ba(14.71 μg/L) > Cu(12.48 μg/L) > As(2.14 μg/L) > Cr(1.67 μg/L) > Mn(1.31 μg/L) > V(0.74 μg/L) > Ni(0.36 μg/L) > Pb(0.15 μg/L) > Co(0.04 μg/L) > Cd(0.02 μg/L). According to the classification principle of Xiao et al. (2014), 12 were divided into three groups: (1) Dominant trace elements: Fe, Zn, Ba and Cu (> 10 μg/L); (2) Moderate trace elements: As, Cr,

Table 1 Concentration of trace elements in the study area and other river basins in comparison to the drinking water limits (μg/L)

River		V	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Cd	Ba	Pb	References
Detection limit		0.031	0.026	0.004	0.704	0.003	0.028	0.079	0.017	0.109	0.000773	0.009945	0.003613	This study
Max		12.25	9.14	300.00	241.20	0.68	3.38	595.10	2172.00	85.28	3.00	252.70	11.56	
Min		0.031	0.193	0.004	11.610	0.004	0.058	0.079	0.018	0.111	0.001	0.046	0.004	
$n = 673$	SD	1.36	0.97	6.26	37.23	0.04	0.28	51.38	122.75	5.22	0.14	26.01	1.08	
CV		1.83	0.58	4.78	0.58	1.18	0.78	4.12	5.25	2.44	9.08	1.77	7.23	
Mean		0.74	1.67	1.31	64.12	0.04	0.36	12.48	23.38	2.14	0.02	14.71	0.15	
Yangtze river basin		–	1.9	41.4	306.1	–	2.5	4.2	21.5	1.4	–	203.9	0.7	Yuan et al. (2020)
Yellow river basin		2.84	6.54	2.34	–	0.17	1.97	2.58	0.77	2.4	0.03	–	0.208	Gao et al. (2019)
Liao river basin		1.78	3.36	0.59	–	0.06	0.75	1.03	2.7	1.23	0.022	–	0.03	Gao et al. (2019)
Hai river basin		4.26	5.46	0.62	–	0.14	1.4	0.84	2.9	2.18	0.031	–	0.045	Gao et al. (2019)
Huai river basin		2.56	6.09	5.82	–	0.12	1.84	3.1	1.81	2.74	0.034	–	0.055	Gao et al. (2019)
Brahmaputra Basin		–	2.7	12.8	–	–	–	1.4	9.8	10.5	1	12	5.6	Qu et al. (2019)
Tarim river basin		1.06	0.43	16.51	61.85	0.1	1.79	1.22	7.11	3.07	0.02	53.03	0.45	Xiao et al. (2014)
Loess plateau China		–	17.38	58.22	67.00	7.28	13.02	19.49	46.76	15.16	0.02	733.0	0.450	Xiao et al. (2019)
Worldwide average		–	0.7	34	–	0.15	0.8	1.48	0.6	0.62	0.08	–	0.079	Gaillardet et al. (2014)
Chinese standard		300	50	300	500	1000	20	1000	1000	10	5	700	10	MHPRC (2006)
WHO standard		300	50	–	–	–	70	2000	–	10	3	700	10	WHO (2011)

Mn, V, Ni and Pb (0.1 μg/L ~ 10 μg/L), and (3) Low trace elements: Co and Cd (< 0.1 μg/L). By comparison, the concentrations of trace elements in our study are lower than the other major rivers in China. The possible reason is less disturbance by anthropogenic activities in the study area than the other major basins, where the studies of water quality often focus on polluted regions such as industrial and agricultural areas. However, the average concentrations of Cr, Cu, Zn, As, and Pb in this study are higher than the worldwide average (Gaillardet et al., 2014). These heavy metals are associated with human production and life, such as industry effluents and domestic sewage (Wang et al., 2017; Xiao et al., 2019), which suggests that the local drinking water has been affected by social and economic activities to a certain extent. Of note, the peak value of As concentration is up to 85.28 μg/L, which significantly exceeds the drinking water standard value (WHO, 2011). Some recent studies indicated that the As concentration ranges from 1.0 to 257.6 μg/L in water of the southern Tibetan Plateau (Huang et al., 2011; Li et al., 2013). Therefore, the local government should pay attention to the monitoring and disposal of arsenic.

The higher the coefficient of variation, the more heterogeneous the spatial distribution of pollutants, which indicates the greater potential disturbance by human activities. Wilding (1985) proposed that the high, moderate and mild variation corresponded to CV > 36%, 36% > CV > 16% and CV < 16%. Thus, the concentrations of 12 TEs in the study area all belong to high variation, of which Cd, Pb, Zn, Mn, and Cu have the highest coefficients of variation. It is likely to be induced by human activities input and spatial heterogeneities of the natural environment.

Water quality appraisal

In the present study, water quality appraisal contains two aspects. Firstly, 12 trace elements concentrations were compared with the standards of drinking water. As shown in Table 1, the mean values of all 12 trace elements concentrations are within the standards. In addition, except for Zn, As and Pb, the peak concentrations of other trace elements are also lower than Chinese standard and WHO standard limits. Of note, the peak concentration of As is 8.5 times higher than the guideline (WHO, 2011), probably causing a threat to human health. It is worth noting that 23 out of 673 samples have excessive concentrations of As, which are located in Nyemo County (8), Damxung County (5), Dazi County (3), Lhunzhub County (3), Maizhokunggar County (2), and Quxur County (2). Previous studies proposed most high As concentrations occur in the central and southern rivers of the Qinghai-Tibet Plateau (Guo et al., 2009; Li et al., 2013), probably for the contributions of arsenic-rich soils and geothermal springs distributed in these regions (Huang et al., 2011; Li et al., 2013). In addition, anthropogenic activities may also contribute to As in water, including mining and smelting activities, the use of arsenical pesticides in agriculture, the discharge of domestic sewage and landfill (Gao et al., 2019; Mao et al., 2019; Qiong et al., 2019), etc. The implementation of environmental protection in Lhasa is backward, and there has been mixing drinking water both for people and animals for a long time, which may cause drinking water pollution.

Secondly, we use EWQI to evaluate the quality status of drinking water comprehensively. Compared with the traditional method, EWQI weakens the comparison error because it has a robust and logical weighting technique (Amiri et al., 2014; Islam et al., 2020). Gorgij et al. (2017) proposed that the physiochemical parameters with larger entropy weight due to the minimal information entropy value have greater impacts on general water quality. Table S2 shows that the contributions of 12 trace elements on EWQI decrease in the order: Ni > Fe > V > Ba > Cr > Cu > As > Zn > Mn > Co > Cd > Pb. The box plot in Fig. 2a shows statistical values of EWQI for 7 counties in Lhasa. According to Meng et al. (2016), the quality of drinking water was classified into five groups based on EWQI, from excellent to undrinkable grade. Hence, the water is suitable for drinking due to the mean values of EWQI being far less than 50. It was reported that the EWQI value of the southwest river basin in China is the smallest (Tong et al., 2021), due to less disturbance by humans. In contrast, mean values in our study are little less than their results. A possible reason is that smaller weights were determined for As, Mn, and Pb as their greater information entropy in our study. Furthermore, IDW (Inverse Distance Weighting) method was applied in ArcGIS10.5 to draw the spatial distribution map of EWQI in agricultural and pastoral areas of Lhasa. Figure 2b shows that high EWQI is mainly distributed in the upper reaches of Laqu, the middle and lower reaches of the

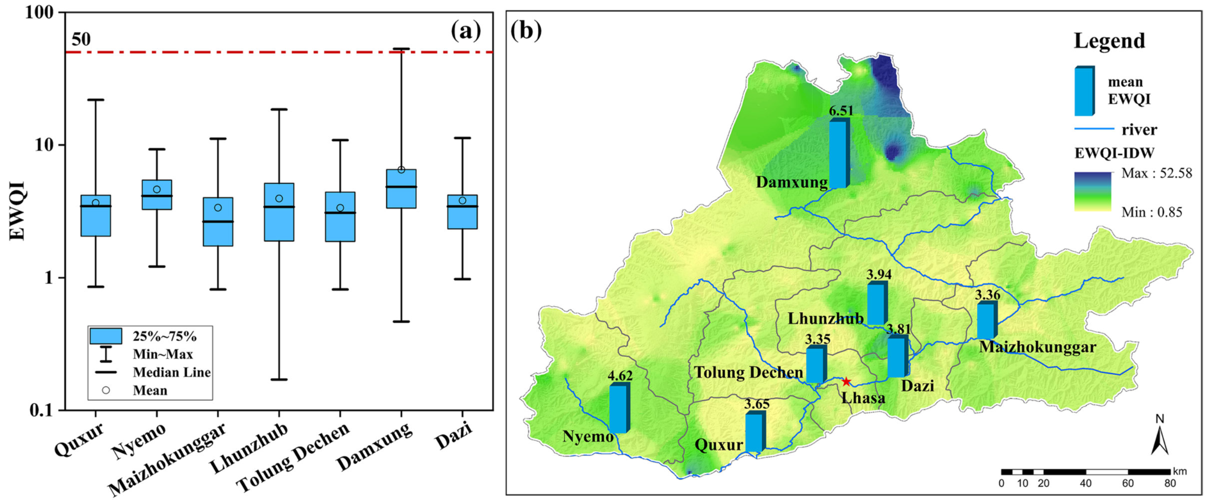


Fig. 2 Box plots of **a** EWQI and **b** mean EWQI value map of drinking water in seven counties of Lhasa

Lhasa River. In particular, the higher EWQI appears in the northeast of Damxung County, so that drinking water in this region should be considered in the future. The exploitation of safe drinking water sources and water quality monitoring equipment is necessary. To sum up, concentrations of trace elements in more than 96% of samples are within safety limits, suggesting that the water in Lhasa's agricultural and pastoral areas is suitable for drinking purposes. However, it is important to note that the effects of As on drinking water safety for residents cannot be ignored.

Source apportionment

Multivariate statistical analysis is often used to identify possible sources of pollutants (Pekey et al., 2004; Wu et al., 2009). Herein, correlation analysis and principal component analysis (PCA) were used to identify the possible sources of trace elements in the drinking water of the study area.

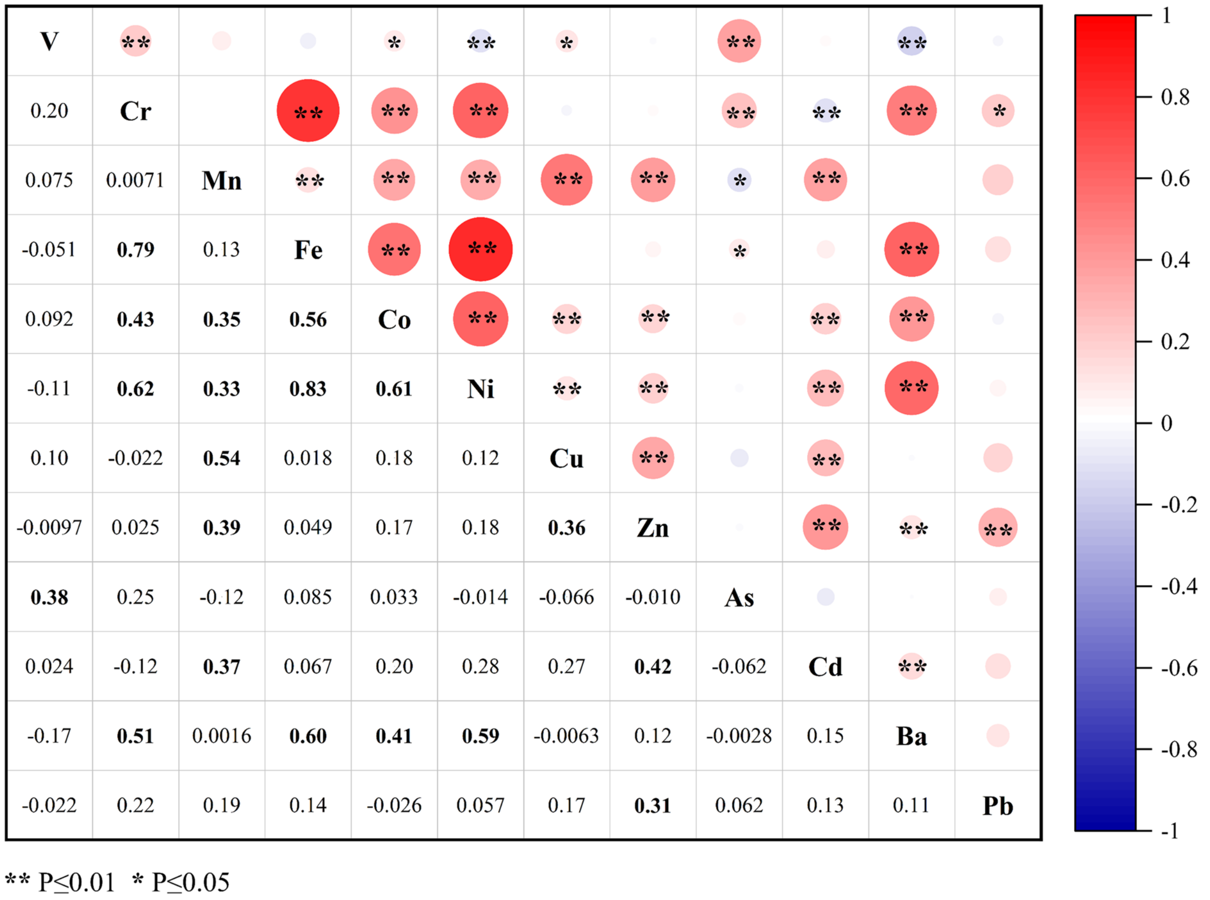
Correlation analysis

A 2-tailed correlation analysis for 12 trace elements was performed. In Fig. 3, except for the pairs V–Ni, V–Ba, and Cr–Cd, the strong correlations ($P \leq 0.01$) between each pair of trace elements, Cr, Mn, Fe, Co, Ni, Cu, Zn, are positive, extending from 0.085 to 0.83. High correlation coefficients ($r \geq 0.30$) are bold in the lower triangle area. For instance, correlation coefficients of the pairs Cr–Fe, Fe–Ni, Fe–Ba, and Co–Ni are 0.79, 0.83, 0.60, and 0.61, respectively. It was confirmed that trace elements with high correlation coefficients might have a common source of origin and mutual dependence during transport (Suresh et al., 2011). So that Cr, Fe, Co, Ni, and Ba probably have common sources, whereas correlations of Zn, As, Cd, Pb with other trace elements are weaker, indicating possibly from multiple sources (Kukrer et al., 2014).

Principal component analysis

Previous studies proposed that the contents of trace elements in the Yarlung Tsangpo River basin varied with the differences in the weathering of rocks, groundwater supply, rainwater, and human inputs (Qu et al., 2017; Tatsi et al., 2015). Thus PCA was carried out to identify the sources of trace elements in detail. To begin with, the raw data were log-transformed to obtain a normal-distribution dataset (Devic et al., 2014). Kaiser–Meyer–Olkin (KMO) value was 0.674 and the significance level of Bartlett's sphericity test was ≤ 0.01, suggesting the measured data were suitable for PCA (Varol, 2011). Three principal components (PCs) (eigenvalue > 1) were extracted from 12 trace elements, which were grouped in rotated space (Fig. 4).

Table S3 shows 64.51% of the total variance is interpreted by three PCs. PC_1 is heavily loaded with



V	**			*	**	*		**		**	
0.20	**Cr**		**	**	**			**	**	**	*
0.075	0.0071	**Mn**	**	**	**	**	**	*	**		
-0.051	**0.79**	0.13	**Fe**	**	**			*		**	
0.092	**0.43**	**0.35**	**0.56**	**Co**	**	**	**		**	**	
-0.11	**0.62**	**0.33**	**0.83**	**0.61**	**Ni**	**	**		**	**	
0.10	-0.022	**0.54**	0.018	0.18	0.12	**Cu**	**		**		
-0.0097	0.025	**0.39**	0.049	0.17	0.18	**0.36**	**Zn**		**	**	**
0.38	0.25	-0.12	0.085	0.033	-0.014	-0.066	-0.010	**As**			
0.024	-0.12	**0.37**	0.067	0.20	0.28	0.27	**0.42**	-0.062	**Cd**	**	
-0.17	**0.51**	0.0016	**0.60**	**0.41**	**0.59**	-0.0063	0.12	-0.0028	0.15	**Ba**	
-0.022	0.22	0.19	0.14	-0.026	0.057	0.17	**0.31**	0.062	0.13	0.11	**Pb**

** P≤0.01 * P≤0.05

Fig. 3 Correlation coefficient heat map of 12 trace elements in drinking water samples

Cr, Fe, Co, Ni, As, and Ba, accounting for 32.86% of the total variance. As mentioned earlier, mean concentrations of all trace elements are far lower than the safety limits, and concentrations of Co and Ni are lower than the worldwide average. Previous studies revealed water chemistry compositions of the Tibetan Plateau are mainly controlled by the bedrock and soil constituents (Huang et al., 2009). It was reported that Fe in the Niyang River mainly comes from the weathering of chlorite and calcite (Wu et al., 2020). Co and Ni are siderophile elements, which are mainly from parent material weathering and the pedogenic process (Xiao et al., 2019). It is confirmed by the high correlation coefficients of the pairs of Fe–Co and Fe–Ni (Fig. 3). In addition, the samples near the Yangbajain and the Yangyi geothermal stations did not show higher As concentrations, indicating As loading in PC_1 is more likely affected by arsenic-rich soils. Hence, PC_1 is interpreted as natural sources, such as bedrock weathering, soil leaching, and atmospheric precipitation. Accounting for 16.56% of the total variance, PC_2 is strongly loaded with Mn and Cu. While Co, Ni, Zn, and Cd are moderate loading factors in PC_2. As is known, these trace metals are affected by human activities to a large extent, especially with Lhasa's accelerated development (Huang et al., 2011). It was reported that Cu, Mn, and Zn are associated with industrial processes such as metallurgy, petrochemical plants, as well as domestic wastewater (Li et al., 2011; Vu et al., 2017). According to previous studies, Zn and Cd may originate from agricultural activities, such as the use of chemical fertilizers and pesticides (Ke et al., 2017; Wang et al., 2015). Hence we speculated that PC_2 represents anthropogenic sources. 15.09% of the total variance is explained by PC_3, in which Zn, Cd, and Pb have strong loading.

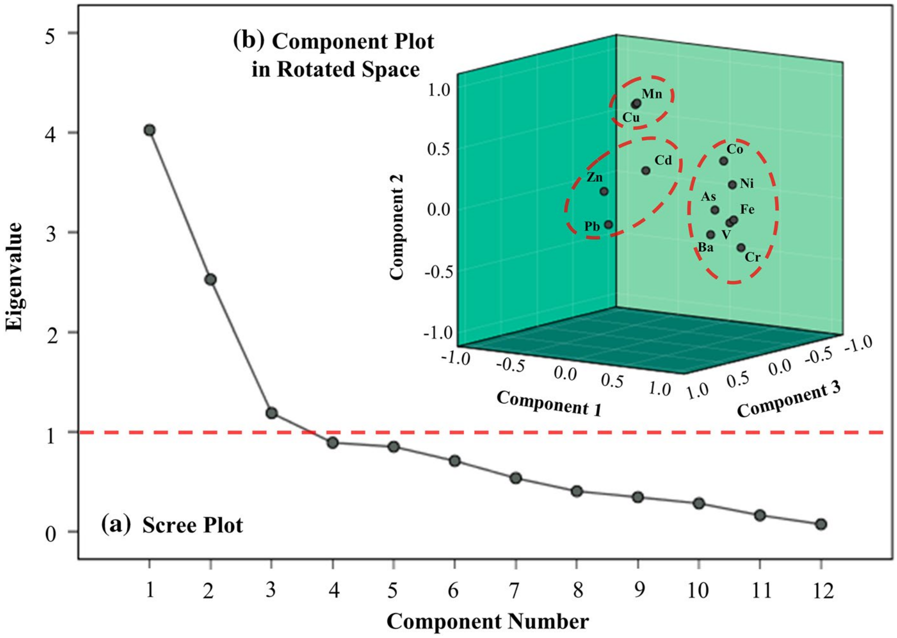


Fig. 4 Principal component analysis of trace elements in drinking water samples: **a** screen plot and **b** component plot in rotated space

According to Huang et al. (2010), the study area is rich in nonferrous mineral resources, such as Cu, Zn, Pb, etc. Mining is an important industry in Lhasa. A recent study suggested that mining production is a possible source of Pb in the Lhasa River (Mao et al., 2019). At the same time, the higher measured values of Zn and Pb appeared close to the Gyama mining area. All of which supports that PC_3 represents a mining source.

Trace elements in the drinking water of the study area are mainly from natural sources, being affected by anthropogenic input and mining production to a lesser extent. The influence on the local environment caused by geothermal exploitation has been a concern for a long time, especially for the Yangbajain geothermal plant. Our results indicate there are no As excess risks in drinking water imposed by geothermal discharge. On the one hand, during the exploitation of the Yangbajain geothermal power plant, much attention has been paid to the control of geothermal effluent recharge (An, 2017), which effectively decreases the discharge of geothermal wastewater. On the other hand, with the dilution and adsorption of riverbed sediment downstream, most geothermal As can be removed from river water at a short distance away from the wastewater discharge sites (Guo et al., 2015). Therefore, local geotherm exploitation has no adverse effects on drinking water downstream.

Probabilistic health risk assessment

Non-carcinogenic risk assessment

By using Monte Carlo simulation technique, we evaluated the probabilistic health risk through exposure to 12 trace elements in drinking water of the study area for different populations (children, teenagers, and adults). Figure 5 shows the cumulative probability distribution of potential NCR. In total, the NCR of 12 trace elements decrease in the order: As > Cr > Mn > Cu > Co > V > Fe > Ba > Zn > Pb > Ni > Cd (Fig. S1). HQ < 1 represents there is no potential NCR to humans (Ravindra & Mor, 2019). In addition, the 95th percentile is usually selected to judge whether the health risk of trace elements exceeds the standard or not, aiming to avoid the effect of extreme values on the evaluation (Saha & Rahman, 2020). Figure S1 shows that the 95th percentile value range of HQ is 2.89E-03 ~ 8.61E-02, 1.30E-03 ~ 3.86E-02, and 1.22E-03 ~ 3.68E-02 for children, teenagers, and adults, respectively, except for As. So that the NCR imposed by the former 11 trace elements are within the safety threshold. The red color padded areas (Fig. S1) represent cautionary risk. For Mn and Cu, the probabilities of excessive NCR are rare, while As poses a high probability that NCR exceeds the safety threshold. The probabilities of excessive NCR

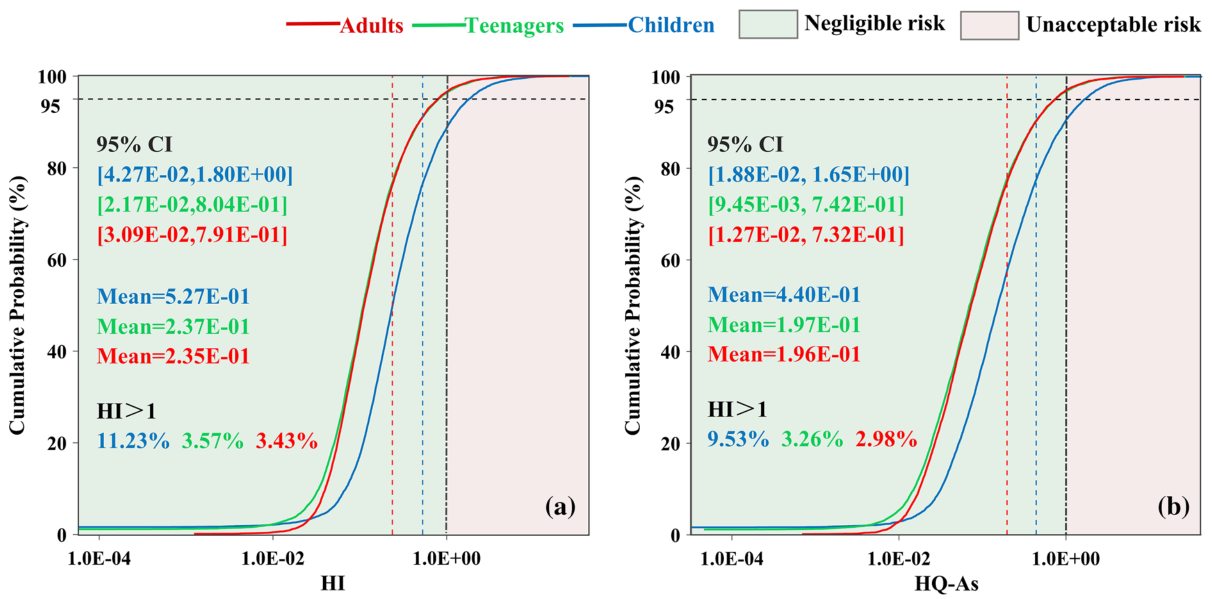


Fig. 5 The cumulative probability distribution of **a** total non-carcinogenic risk, that is HI; and **b** non-carcinogenic risk of As. The blue and red, vertical, dashed line represents the mean value. The cumulative probability reaches 95% at the horizontal dashed lines. The green, vertical, dashed line was omitted as the mean non-carcinogenic risks of teenagers and adults are very close

through exposure to As for children, teenagers, and adults are 9.53, 3.26, and 2.98%, respectively. HI is the sum of HQ of 12 trace elements (Fig. 5a). It can be seen that the 95th percentiles of HI for teenagers and adults are less than 1, whereas the value for children is greater than 1. Thus there is no NCR for teenagers and adults, but excessive NCR for children. The probability of excessive NCR for children is 11.23%, of which As is the main contributor to HI value.

Meanwhile, the results reveal distinctions of NCR exist among different populations. The mean value range of HQ is 8.12E-04 ~ 4.40E-01, 3.69E-04 ~ 1.97E-01, and 3.67E-04 ~ 1.96E-01 for children, teenagers, and adults, respectively. The potential NCR is highest in children, while those of teenagers and adults are comparable, which is in line with previous studies (Jafarzadeh et al., 2022; Kaur et al., 2020; Tong et al., 2021). This might be associated with the lower body weights of children. Overall, there is a need to concentrate on NCR imposed by As.

Carcinogenic risk assessment

According to the NCR results, As was determined to estimate CR. In addition, Cr, Ni, and Cd are also considered to be elements with potential carcinogenic effects (Cao et al., 2014). The cancer slope factors (SF, (kg•d)/mg) of Cr, Ni, As, and Cd are 0.501, 1.7, 1.5, and 0.63, respectively. The CR can be obtained by multiplying SF and ADD. Generally speaking, 1.0E-06 is considered as the threshold of negligible risk (MEPRC, 2019). CR through exposure to these four trace elements are different, the mean values decrease in the order: As > Cr > Ni > Cd. For Cd, the 95th percentile of CR falls into the negligible region (Fig. S2). While for Cr, Ni, and As, there are probabilities of cautionary risk. Fortunately, there are no unacceptable risks for Cr and Ni.

Figure 6b shows that the CR imposed by As is significant. The unacceptable risk probabilities of As are 2.47% for children, 0.64% for teenagers, and 5.61% for adults. Compared with Fig. 6a, As is the main contributor to the total carcinogenic risk, not only for high-value SF of As but also for its excessive concentration (Huang et al., 2008; Tian et al., 2016). If the deterministic method is used for calculation, when the CR of As reaches 1.0E-04, the corresponding concentration of As is 11 μg/L for children, 25 μg/L for teenagers, and 6 μg/L for adults. As above mentioned, the peak concentration of As in the study

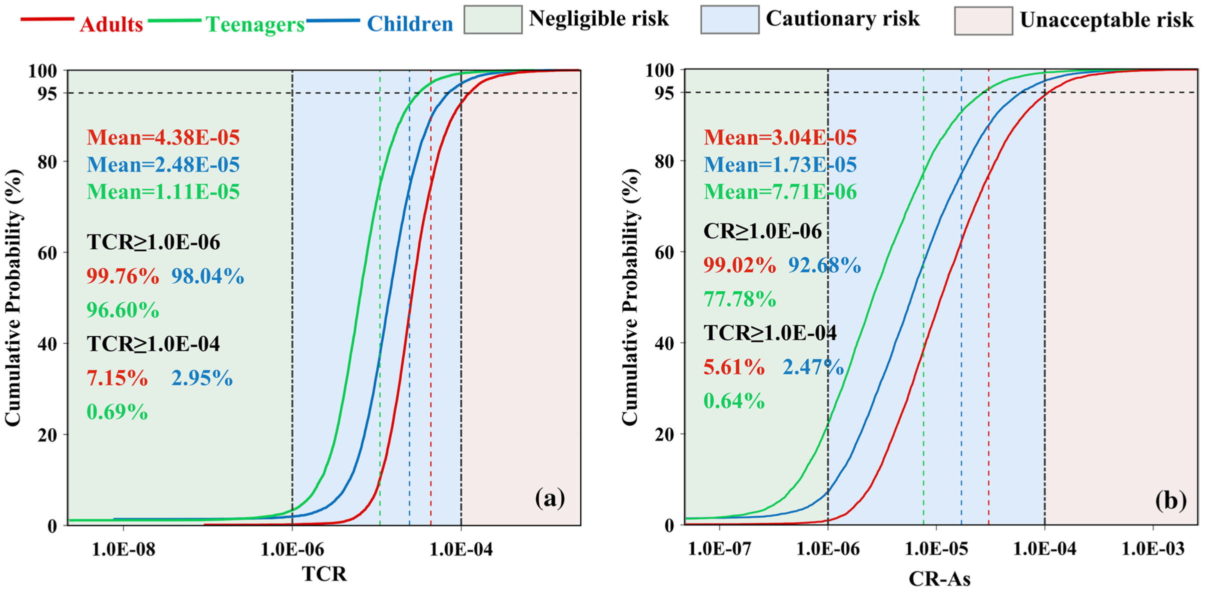


Fig. 6 The cumulative probability distribution: **a** total carcinogenic risk and **b** carcinogenic risk of As. The red, blue and green, vertical, dashed line represents the mean value. And the gray, vertical, dashed lines represent the acceptable/unacceptable thresholds (1.0E-06/1.0E-04), respectively

area reached 85.28 μg/L, resulting in an inevitable probability of unacceptable risk, which may lead to lung, skin, kidney, and liver cancer in human beings (Cogliano et al., 2011). Especially for the susceptible children group, a previous study reported that exposure to As is related to neurobehavioral defects during the early years of life (Calatayud et al., 2019). Further, the CR among the three age groups differs from the NCR results, decreasing in the order of adults, children, and teenagers. The CR imposed by four selected trace elements for adults is rough twice as much as children and four times as much as teenagers. This is possible because adults have a significantly higher exposure duration than the other two populations, while children have lighter body weights than teenagers. Overall, As in drinking water of the study area has potential cancer risks, and adults are the most susceptible group.

Sensitivity analysis

To further determine the contribution of input variables on health risk calculation, sensitivity analyses based on Monte Carlo simulation results were performed (Islam et al., 2020). For NCR, we only considered the sensitivity of As concentration and the rest parameters (Fig. 7a). It indicates Bw has a negative contribution to the NCR for three populations, but the contribution can be negligible, which is in line with the results of previous studies (Gao et al., 2019; Hossain & Patra, 2020). It is worth noting that Bw has a more significant effect on NCR in the children group. By contrast, the other three parameters contribute positively to NCR, the sensitivities decrease in the order: C–As > IR > EF. Among them, As concentration is the most important parameter and its contribution reaches 80.05% for children, 83.35% for teenagers, and 89.84% for adults, respectively. In contrast, as for concentration adults are more sensitive, while for IR children are more sensitive. For TCR, Fig. 7b shows the sensitivity of each parameter decreasing in the order: C–As > IR > C–Cr > EF > C–Ni > Bw > C–Cd. Due to low contents, the contribution of Cd to TCR is negligible. In addition, similar to the results of NCR analysis, Bw is a variable with negative contribution; As concentration and ingestion rate are the primary factors affecting TCR, and the sensitivity of parameter C–As is more than 50% for three populations.

To sum up, concentration and IR are the most influential factors. The variation coefficient of As concentration measured in the study area is significant, which may affect the precision of probabilistic

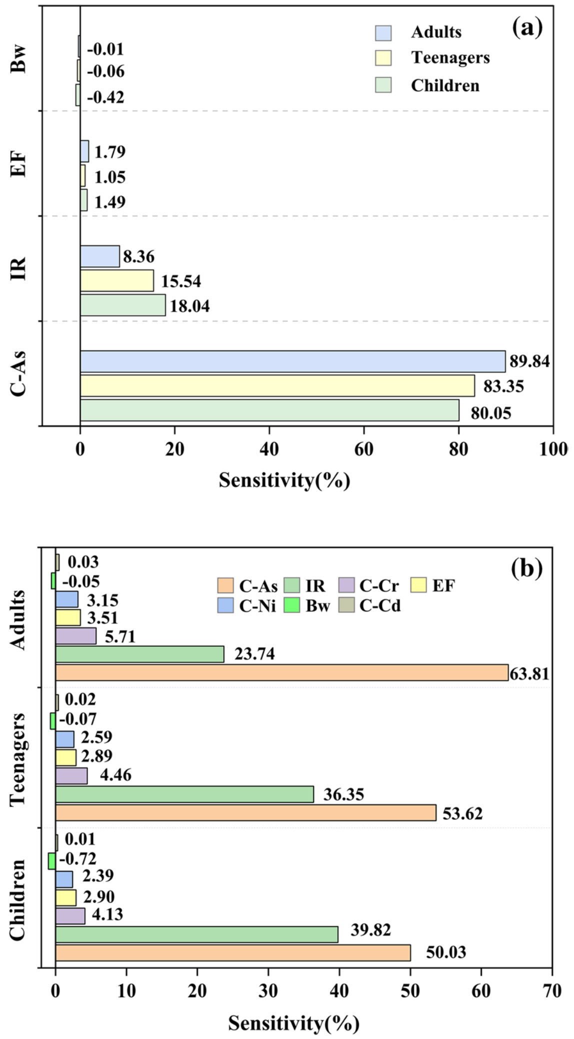


Fig. 7 Sensitivity analysis outcomes to identify the relative contribution of input variables on potential health risk: **a** non-carcinogenic risk caused by As and **b** total carcinogenic risk caused by four trace metals. C-As(Cr/Ni/Cd) = concentration of As(Cr/Ni/Cd); IR = ingestion rate; EF = exposure frequency; Bw = body weight

health risk assessment. Further studies should overcome this problem by increasing the number of samples.

Conclusion

In this study, comprehensive analyses on trace elements in drinking water in the agricultural and pastoral areas of Lhasa were carried out. The main findings are as follows: (1) the water in the study area is suitable for drinking, as the mean concentrations of 12 trace elements are within the guidelines and the values of EWQI are less than 50; (2) by combining correlation analysis with PCA, natural, anthropogenic and mining activities were identified as potential sources of trace elements in drinking water. The contribution of natural processes such as rock weathering and soil leaching is predominant, accounting for 32.86% of trace elements in drinking water; (3) the probabilistic health risk assessment using Monte Carlo technique shows there is no health risk through exposure to trace elements except for As. The probabilities of cautionary risk were attributed to excessive concentrations of As in a few samples, and (4) As concentration and ingestion rate are more sensitive to health risk outcomes for three populations.

Overall, our study provides a basic understanding of trace elements in drinking water in Lhasa's agricultural and pastoral areas for the first time, which is essential for the local drinking water management services. However, we acknowledge there are limitations that we did not obtain the actual exposure parameters of the local residents. Since the probabilistic health risk assessment considerably relies on input parameters, including concentrations of trace elements, body weight, ingestion rate, etc. Thus more precise input parameters of residents are needed to reduce the simulation bias. Meanwhile, temporal variation of trace elements concentrations and drinking water intake can be considered in the future.

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Author contributions All authors contributed to the study conception and design. SP: methodology, formal analysis, data curation, writing—original draft; XX: conceptualization, methodology, supervision, project administration; HZ: validation, formal analysis, writing—review & editing; ZY: methodology, writing—review & editing; UMA: visualization, writing—review & editing; YZ: data curation; HC: methodology; GL: data curation, methodology; XD: editing; GM: conceptualization, supervision; PY: revision. The first draft of the manuscript was written by SP and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Declarations

Conflict of interest The authors have no relevant financial or non-financial interests to disclose.

Ethical approval This study does not require ethics approval.

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