



Assessment of potential health risk of major contaminants of groundwater in a densely populated agricultural area

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Abstract As a key part of Bohai New Area development, Haixing County has been undergoing rapid development. In order to estimate potential risks of chemical parameters to human health of local residents, carcinogenic and non-carcinogenic risks via direct ingestion of drinking water were calculated using human health risk assessment (HHRS) based on triangular fuzzy number. The levels of pH, total dissolved solids, total hardness, SO_4^{2-} , Na^+ , Cl^- , SO_4^{2-} , F^- , Fe (total iron), NO_3^- , and NO_2^- were more or less higher than the permissible limits except parameters As and Mn. The analysis results show that risk level for different crowds in the study area demonstrated an obvious variation, generally in the

order of infants > children > adult males > adult females for non-carcinogenic risk values (R^n), while the sequence of the carcinogenic risk values (R^c) are adult males > adult females > children > infants. When the confidence level was 0.8, the non-carcinogenic risk values (R^n) through drinking water intake were higher than 1, and this implied that potential health impacts on human health for local residents. However, the risks of carcinogenic risk values (R^c) were lower than $1.0\text{E}-4$, demonstrating minimal and acceptable health risk. Furthermore, according to the middle values ($\alpha = 1$) of R^n , the total non-carcinogenic risks for local residents were obtained in the following order: GW (Gaowan Town) > XJ (Xinji-Xiangfang County) > ZM (Zhaomaotao County) > HX (Haixing-Suji Town) > ZH (Zhanghuiting County) > XS (Xiaoshan County), and ZM > XJ > GW > HX > XS > ZH for R^c . It was also found that the spatial distribution of fluoride level in drinking water is urgently needed to be identified. In conclusion, the potential health risks to residents should cause enough attention both from society and the academic community.

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Introduction

Contamination of the environment has been accelerated all over the world during the past two decades owing to rapid economic development and industrialization (Solgi et al. 2012; Zang et al. 2017; Wang et al. 2018a). Contaminations can arise from air-borne particles containing pollutions from industries production, agriculture and living activities that indirectly contaminate surface soil by atmospheric deposition or precipitation (Shakoor et al. 2017; Rahman et al. 2018). Land is also directly contaminated by the pollutants from industrial slag, construction waste and household waste-water discharge (Liu et al. 2015; Yang et al. 2016; Muhammad et al. 2018). Meanwhile, natural processes, such as volcanic particles and mineral dissolution, also add the content of chemical parameters in surface soil or groundwater (Ahada and Suthar 2017; Zhang et al. 2017; Chen et al. 2018). Issues may arise when these soil contaminants are bioavailable to plants or enter groundwater through soil percolation (Fig. 1). Contaminations exposure can induce human health risks, including a variety of cancers, intellectual disabilities and neurologic, etc., through a variety of exposure pathways, including inhalation, ingestion, and dermal contact (Nazir and Khan 2006; Liu et al. 2015; Saba et al. 2015; Bao et al. 2017; Wang et al. 2018b).

Therefore, health risk assessment based on consumption of contaminated water is highly significant due to the direct, intense, and continuous human exposure (Siddique et al. 2015; Shakoor et al. 2017; Su et al. 2018; Zhang et al. 2018), and the massive quantities of water pollution incidents have also been reported around the world (Chotpantarat et al. 2014; Ayotte et al. 2015). The most common contaminations in water include heavy metals (Wang et al. 2018a), fluoride (Yousefi et al. 2018), nitrogen (Zhang et al. 2018), and many others (Azizullah et al. 2011; Ali et al. 2018). Meanwhile, some studies identified a strong relation between poverty and disease (Chowdhury et al. 2006; Ahmad et al. 2007). Taking into account the above situation, the need to fully understand the contaminants and the associated health risks through ingestion exposure should be recognized.

In order to assess the relationship between pollutants and human health, human health risk assessment (HHRA) proposed by U.S. Environmental Protection Agency (USEPA 1989), a methodology to accurate

quantitative relationship between “intake and risk” was used in this study. However, there is a need considering about the uncertainties of the prevalence of effects on variables (Arunraj et al. 2013; Yang et al. 2018; Wang et al. 2018b), such as mean values of weight, ingestion quantity, life expectancy, and contaminant level. In this context, fuzzy triangular number (FTN) was employed to determine the risk fluctuation range in different confidence levels to process the uncertainties of the data, so as to avoid overestimate/underestimate of health risk value (Yang et al. 2013; Wang 2017).

As a part of the development of Bohai New Area, northeast China, Haixing County has undergoing rapid development, especially in husbandry is highly developed. The agricultural population in the area was 182,000, which accounts for 84.65% of the total population of Haixing County. Beans, paddy rice and goose are the three mainstay industries in study area, and the residents mainly depended on upper confined water to meet drinking water needs, and the potential contamination of the water in the confined aquifers is a “time bomb.” Meanwhile, groundwater has been overexploited in many areas. It is reported that the quantity of groundwater extraction in 2005 is about $0.042 \times 10^8 \text{ m}^3$, the buried depth of groundwater in dry season increased from 60.48 to 68.76 m during 2000 and 2005, and the water level decreased 8.33 m in the same period. The area of depression cone is 1245.2 km^2 , increasing 693.2 km^2 compared with that of 2000, which was caused by continuous overexploitation of groundwater in the study area. From a literature review, few studies have been found that addressed health problems and consequences of high contaminants exposure to Haixing County residents. Thus, health risk assessment associated with contaminations and identifying the influencing factors are preliminary to environmental protection and human health.

The present study is to develop an integrated risk assessment methodology to assess the health risks of upper confined water as the main drinking water sources, which focus on (1) determining the distribution of major chemical parameters; (2) quantifying the human health risk considering age including infants, children, adult males, and adult females and (3) FTN which was introduced to quantify uncertainty in data. The study results can provide more accurate data and evidence and help decision makers take corresponding

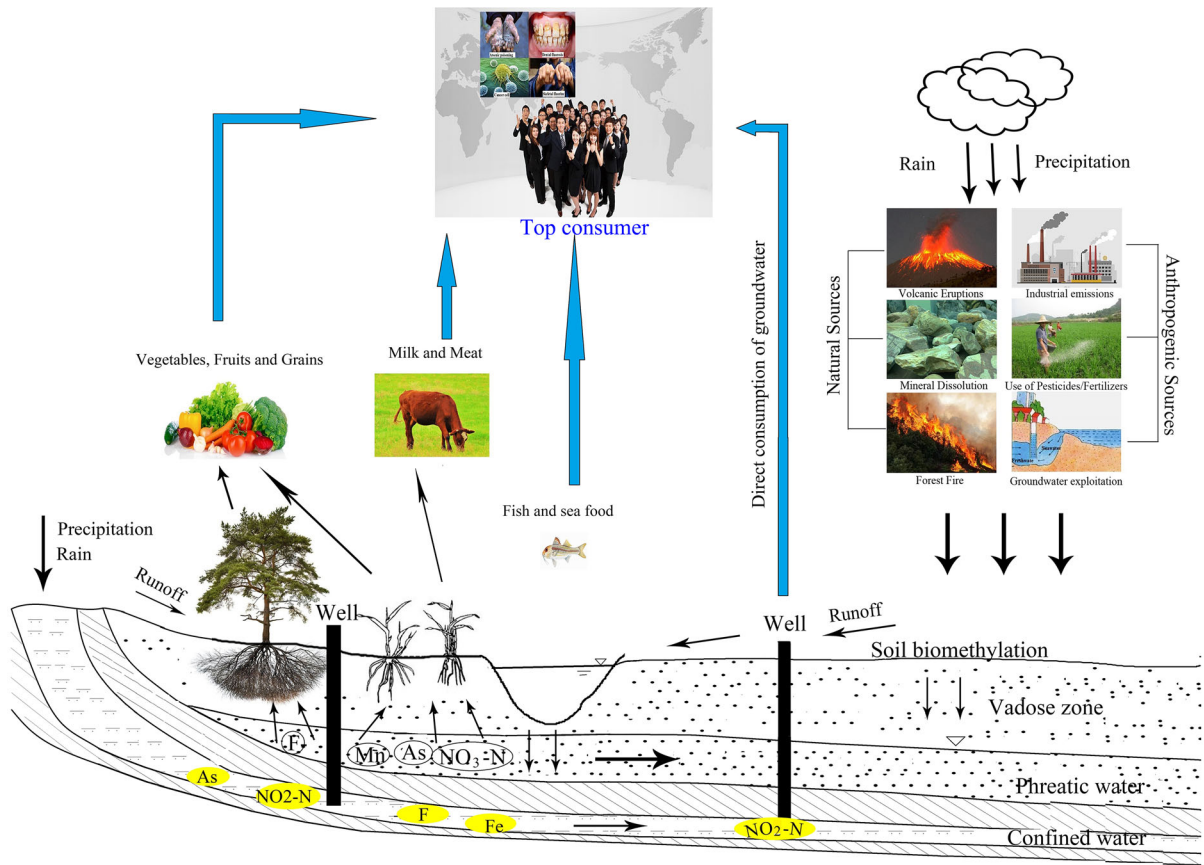


Fig. 1 Different exposure pathways of contaminants to human

measures on groundwater quality protection and management to protect groundwater environment and human health.

Materials and methods

Study area

The study area, Haixing country, is located in eastern part of Hebei Province, Central China, between 37°56'10" to 38°17'31" latitudes N and 117°18'33" to 117°58'09" E (Fig. 2). It covers geographical area of 859.9 km² (East is near Bohai Sea) and 215 thousand populations, where the local people obtain water from groundwater for drinking and domestic consumption. The eastern region of Haixing County is bordered by the Bohai Sea, which results in the 18-km coastline extended from south to the north (Fig. 2). The climate in Haixing County is controlled by warm

temperate humid monsoon, characterized by distinct seasons and significant changes in the temperature difference between seasons. The mean annual temperature was 13.2 °C, rainfall was 600 mm, and evaporation was 1870 mm.

Haixing County is located in coastal alluvial plain. It belongs to the alluvial void saltwater system of Luanhe groundwater system, which is mainly composed of silt fine sand, silty clay, and clay. This aquifer can be divided into phreatic aquifer, upper confined aquifer, and lower confined aquifer from up to down, and these aquifers are separated by less porous silty clay and clay layers that act as semi-confining barriers (Fig. S1). The estimated water-proof floor depths are 120–170 m, 250–350 m, and 350–550 m for phreatic aquifer, upper confined aquifer, and lower confined aquifer. The main recharge sources of confined water are the overflow and lateral runoff of diving water, and the main discharge ways are artificial mining and lateral runoff.

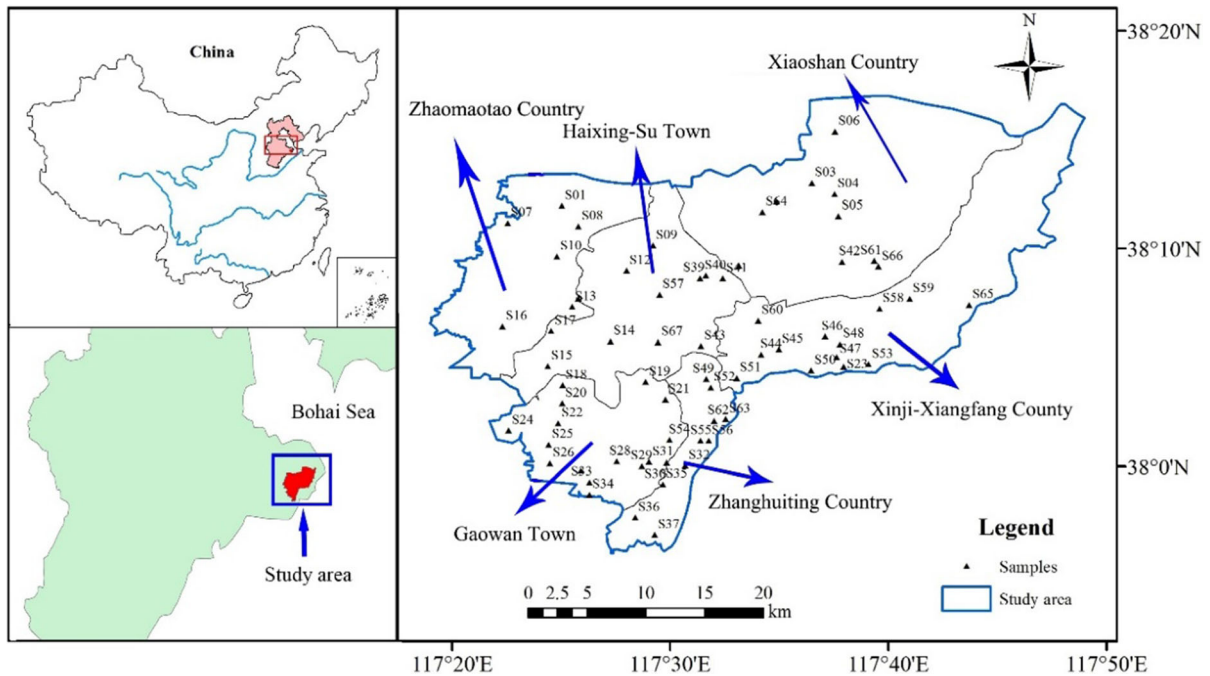


Fig. 2 Location map of study area and sampling points

Sampling and analytical methods

Sample collection and analysis

In Haixing County, 7, 16, 11, 10, 10 and 13 groundwater samples were collected from upper confined aquifer in Zhaomaotao Country (ZM), Gaowan Town (GW), Haixing-Suji Town (HX), Zhanghuiting Country (ZH), Xiaoshan Country (XS) and Xinji-Xiangfang Country (XJ), respectively. These water samples were collected in August 2017 from rural wells (upper confined aquifer), which were mainly used for drinking and domestic consumption, and the positions of selected samples are shown in Fig. 2. The water samples were collected in two plastic bottles, pre-washed with 0.5 N nitric acid (HNO_3) and deionized water, from each sampling point. During sampling, two bottles of water samples were filled with water without bubbles. A suitable amount of dilute nitric acid was added to one bottle to make its PH less than 2 for the determination of metal and nonmetallic elements, while the other bottle was used for other determination without treatment. Finally, seal, label and send it to the laboratory as soon as possible. All sampling procedures, transportation, and conservation followed the

standard methods recommended by the ministry of water resources of People's Republic of China. Accuracy precision of element (Na^+ , K^+ , Ca^{2+} , Mg^{2+} , Cl^- , SO_4^{2-} , HCO_3^- , F^- , NO_2^- , NO_3^- , total dissolved solids (TDS), PH, total hardness (TH), total iron (Fe), Mn, and arsenic (As)) was achieved through the implementation of standard laboratory procedures and quality control techniques.

Methods

Triangular fuzzy numbers method

The concept of set of fuzzy numbers was proposed by Zadeh in 1965, and triangular fuzzy numbers are used to describe problems that contain uncertain or inaccurate information in environments (Tang 2014). In mathematical definition of a fuzzy number, a triangular fuzzy number is used in this paper, which is the most common and simplest realization (Khan and Valeo 2015). A triangular fuzzy number has membership functions defined by only three points: a_1 (the lower value), a_2 (the expected value), and a_3 (the upper value) are the minimum probable, the most probable and the maximum probable values of a certain fuzzy variable x , respectively (Fig. 3). These

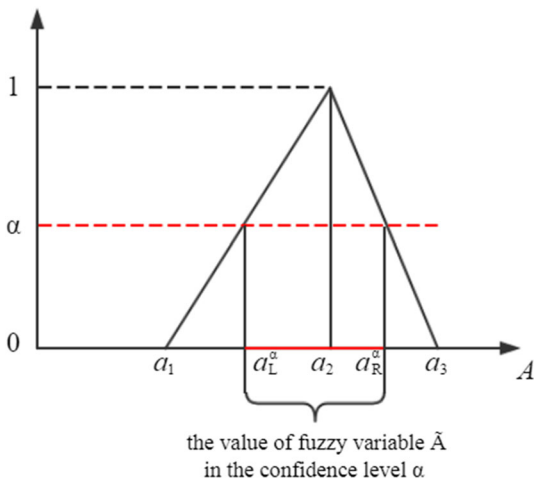


Fig. 3 Sketch map of triangular fuzzy numbers and illustration of α -cut

three numbers are real numbers and $a_1 < a_2 < a_3$, forming triangular fuzzy number $\tilde{A} = (a_1, a_2, a_3)$ (Chen et al. 2018). Being linear functions, its calculation formula is thus as follows:

$$\mu_A = \begin{cases} \frac{x - a_1}{a_2 - a_1} & x \in (a_1, a_2] \\ \frac{a_3 - x}{a_3 - a_2} & x \in (a_2, a_3] \\ 0 & x \in (-\infty, a_1) \cup (a_3, +\infty) \end{cases} \quad (1)$$

Here, a membership level, μ , equal to 0 means that it cannot be regarded as a possible values of \tilde{A} , while a value $\mu = 1$ ($x = a_2$) means that it completely belongs to set \tilde{A} . In triangular fuzzy numbers, the values of a_1 , a_2 and a_3 are generally determined by mathematical statistics. According to the method of mathematical statistics, more than 95% of the data of normal distribution or approximate normal distribution fall into the range of $(\bar{x} - 2\sigma, \bar{x}, \bar{x} + 2\sigma)$ (Gardner and Altman 1986; Chen et al. 2018), among which \bar{x} is the mean and σ is standard deviation of the parameter. Meanwhile, in order to ensure that the interval of index concentration is positive and avoid the influence of abnormal points on the interval, the upper and lower limits of triangular fuzzy numbers is determined based on following criterions: (a) the larger value between minimum and the mean value minus twice standard deviation is set as the lower limit; (b) The smaller value between the maximum and the mean value plus twice standard deviation is set as the upper limit (Yang et al. 2018).

The membership level (μ) of triangular fuzzy numbers represents the relative reliability of each data in the range of minimum and maximum possible values, and different confidence levels α ($0 < \alpha < 1$) represent different data intervals. In calculation, the α -cut technique is used to reduce a fuzzy number into a series of nested intervals (Zhang 2009; Zhang and Achari 2010; Wang et al. 2011; Wang and Huang 2012). The larger α is, the closer it is to the average value, the larger the frequency of occurrence of the data, the smaller the range of the interval, and the data interval where the level of reliability is not lower than α can be expressed as:

$$\tilde{A}^\alpha = \{x | \mu_A(x) \geq \alpha, x \in X\} \quad (2)$$

$$\tilde{A}^\alpha = (a_L^\alpha, a_R^\alpha) = [(a_2 - a_1)\alpha + a_1, (a_2 - a_3)\alpha + a_3] \quad (3)$$

where \tilde{A}^α represents the value of the fuzzy variable \tilde{A} under the condition of confidence level α . Assuming that there are two sets of triangular fuzzy numbers $\tilde{M} = (m_1, m_2, m_3)$ and $\tilde{N} = (n_1, n_2, n_3)$ within the confidence level α , they can be added, subtracted, multiplied and divided (Yang et al. 2018; Chen et al. 2018):

$$\tilde{M}^\alpha \pm \tilde{N}^\alpha = (m_L^\alpha \pm n_L^\alpha, m_R^\alpha \pm n_R^\alpha) \quad (4)$$

$$\tilde{M}^\alpha \cdot \tilde{N}^\alpha = (m_L^\alpha \cdot n_L^\alpha, m_R^\alpha \cdot n_R^\alpha) \quad (5)$$

$$\tilde{M}^\alpha / \tilde{N}^\alpha = (m_L^\alpha / n_L^\alpha, m_R^\alpha / n_R^\alpha) \quad (6)$$

Health risk assessment

Human health risk assessment is to describe the adverse effects of human health caused by environmental pollutions through a variety of exposure pathways. Through health risk assessment, health risks caused by chemical pollutants are obtained intuitively, thus coming up with corresponding suggestion and protection measures for the health of human body and basis treatment of groundwater pollutions in Haixing County (Zhai et al. 2017). For this purpose, a four-step human health risk assessment framework (HHRA), proposed by United States Environmental Protection Agency (USEPA), was used to assessment human health risk in this paper (Fig. 4) (USEPA 2009; Mondal and Polya 2008; Sarabjot and

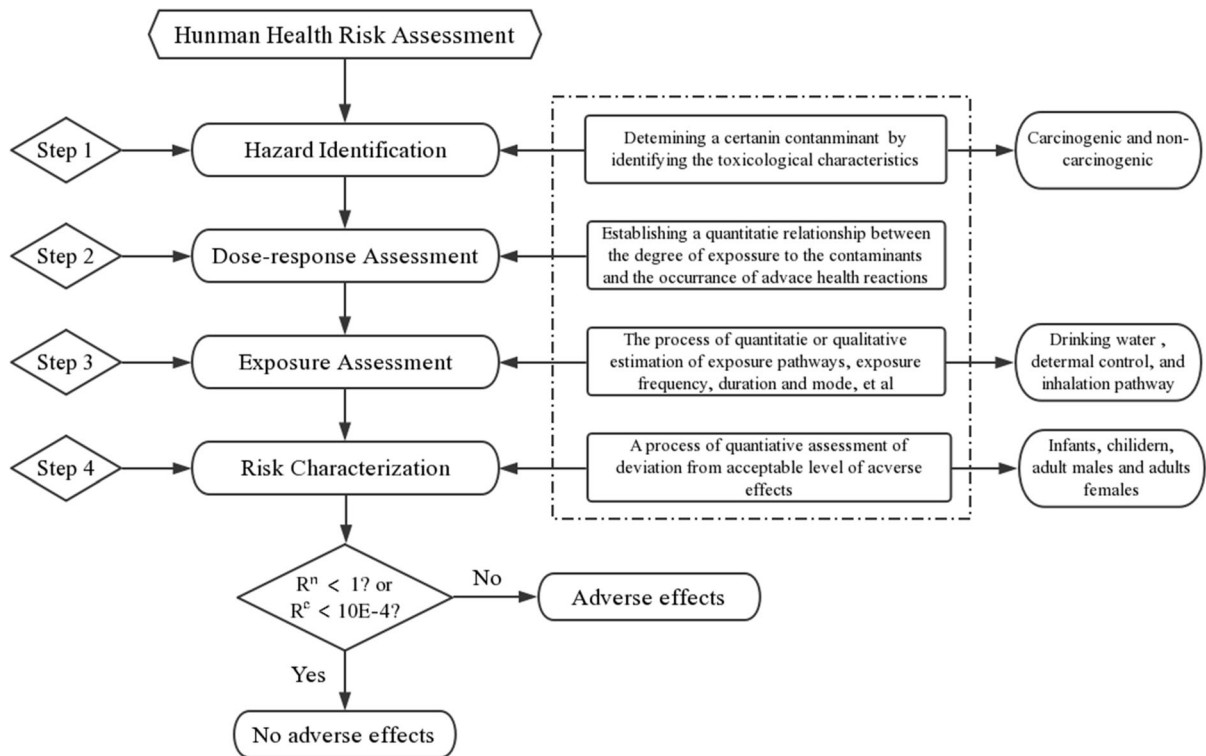


Fig. 4 The framework of human health risk assessment process

Rohit 2019). The method is characterized by the use of risk values to correlate environmental pollutants with human health and quantitatively describe the risk of contamination causing health hazards to humans.

In the third step (Exposure assessment) of HHRA (Fig. 4), potential exposure pathways can be divided into three types: drinking water pathway, dermal control pathway and inhalation pathway. Among them, drinking water containing harmful substances is the main pathway of causing harm to human body by contaminations (Han 2014; Shakoor et al. 2017; Yang et al. 2018). This sentence is changed as: Considering that the massive quantities of upper confined water still used by the residents of the Haixing County to satisfy household demands, which makes health risk assessment through potable water an important step in protecting their health, and the oral exposure of pollutants in drinking water is calculated as follows:

$$D_{i(\text{oral-drinking})} = \frac{C_i \times I_R \times EF \times ED}{B_w \times A_t} \quad (7)$$

where $D_{i(\text{oral-drinking})}$ represents exposure dose of i th pollutant by drinking pathway ($\text{mg}/\text{kg} \text{d}^{-1}$). C_i is the

concentration of i th contaminant in upper confined water ($\mu\text{g}/\text{L}$ or mg/L). I_R represents the daily water quantity (L/day). EF represents the exposure frequency (day/year). ED represents the duration of exposure duration (70 years for carcinogenic pollutants and 30 years for non-carcinogenic pollutants). B_w represents the average body weight (kg), and A_t represents the average exposure time.

Considering the toxicity of substances, risks can be divided into carcinogenic and non-carcinogenic risks. It is generally believed that carcinogens exhibit a non-threshold effect, and even a small amount of exposure has a negative impact on human health to a certain extent. This negative effect usually lasts for a lifetime. However, non-carcinogens exhibit a threshold effect, and non-carcinogenic substances do not have an observable negative impact on the organism below the threshold. Only when the dose threshold is exceeded, it will have an impact on human health (Han 2014). Non-carcinogens are expressed in terms of the effects and severity of human poisoning, rather than the risk of detection or mortality. Then, the

carcinogenic risk and non-carcinogenic risk recommended by EPA are shown as follows (USEPA 1989):

$$R^c = \sum_{i=1}^k R_i^c R_i^c = D_{i(\text{oral-drinking})} \times SF_i \tag{8}$$

$$R^n = \sum_{i=1}^k R_i^n R_i^n = D_{i(\text{oral-drinking})} / RfD_i \tag{9}$$

R_i^c and R_i^n are potential carcinogenic and non-carcinogenic risks by exposure to the related i th pollutant in drinking water, respectively. SF is the cancer slope factor for oral exposure to carcinogenic pollutants (As; kg day/mg), and RfD_i is the reference dosage of non-carcinogenic pollutants (mg/kg day⁻¹). R^c and R^n are the potential total carcinogenic risk and total non-carcinogenic risk posed by the related element in drinking water (Table 1).

Due to differences in respiration, behavior and lifestyle, such as water intake, exposure time, exposure duration, exposure frequency and body weight et al. (Zhai et al. 2017; Yang et al. 2018), the risk values have been estimated for four subgroups, i.e., infants (who are less than 1 years old), children (who are among 2–17 years old), the adult males and the adult females. Meanwhile, to reduce the uncertainties in exposure variation, C_i , B_w , I_R , and A_i are defined as triangular fuzzy numbers, and then the lower, the intermediate and the upper limits for C_i , B_w , I_R and A_i can be gained (Table 2). The risk values of drinking water pathway are given in the following formula:

$$\tilde{R}_i^c = \frac{\tilde{C}_i \times \tilde{I}_R \times SF_i \times EF \times ED}{\tilde{B}_w \times \tilde{A}_i} \tag{10}$$

Table 1 Cancer slope factor of carcinogenic and reference doses of non-carcinogenic

Contaminants	Parameter	
	RfD [mg/(kg day)]	SF [mg/(kg day)]
F ⁻	0.06	–
NO ₂ ⁻	0.1	–
NO ₃ ⁻	1.6	–
Fe	0.3	–
Mn	0.14	–
As	0.0004	1.5

$$\tilde{R}_i^n = \frac{\tilde{C}_i \times \tilde{I}_R}{RfD_i \times \tilde{B}_w} \tag{11}$$

Results and discussion

Water quality variables

It was generally noted that upper confined water was used for drinking and domestic purposes for local people in Haixing County due to lack of awareness of the harmful effects of poor drinking water and having no other alternatives. Some chemical parameters exceeding a threshold are poisonous and may cause severe health problems in humans and other organisms who drink the water. Box and whisker plots for groundwater quality variables were thus used to compare the WHO (2011) and to describe the range of variation of parameters in six regions.

Conventional components

The pH in groundwater indicates the strength of water to react with the acidic or alkaline material present in water (Hem 1991). As shown in Fig. 5, levels of pH found in upper confined water were either comparable to or below permissible upper limit of 8.5 (WHO 2011) except for the only two water samples (S02: 9.16 and S62: 8.9). The data of various region revealed that the mean PH values were 8.22, 8.27, 8.26, 8.15, 8.16 and 8.19 in Zhaomaotao Country (ZM), Gaowan Town (GW), Xiaoshan Country (XS), Xinji-Xiangfang Country (XJ), Zhanghuiting Country (ZH) and Haixing-Suji (HX) Town (Fig. 5), respectively, showing slight alkaline.

The TDS value is a measure of the total amount of dissolved components in water, including all ions, molecules and compounds dissolved in water, but excluding suspended solids and dissolved gases. From the physiochemical in different regions (Fig. 5), it is evident that the concentrations of TDS were be found to vary from 798 to 1926 mg/L, 984 to 2572 mg/L, 1200 to 2218 mg/L, 799 to 1773 mg/L, 103 to 3478 mg/L and 806 to 1817 mg/L in ZM, GW, XS, XJ, ZH, and HX (Fig. 5). The mean TDS content in upper confined water of study area was 1369 mg/L, which may be attributed to salinity intrusion caused by excessive groundwater extraction. In the current study

Table 2 Default factors in HHRA for elements in drinking water

Para	Unit	Meaning	Value			
			Infants	Children	Adult males	Adult females
I_R	L/day	Daily water quantity	(0.45, 0.65, 0.85) ^a	(1.3, 1.5, 1.7) ^a	(1.6, 2.2, 2.8) ^b	(1.4, 2.0, 2.6) ^b
B_W	kg	Body weight	(3, 8, 13) ^a	(20, 30, 40) ^b	(48, 63, 78) ^b	(43.2, 58.2, 73.2) ^b
A_t	day	Exposure time	Carcinogenic pollutants: (23,725, 27,375, 31,025) ^d ; Non-carcinogenic pollutants: ED × EF			
EF	day/a	Exposure frequency	365	365	365	365
ED	a	Exposure duration	0.5 ^a	9 ^a	30 ^c	30 ^c

Para means parameter

^aThese data come from Su et al. (2013)

^bThese data come from Ministry of Environmental Protection, China (2013)

^cThese data come from Ministry of Environment Protection of the People's Republic of China (2013)

^dOffice of the Women and Children's Working Committee of the Hebei Provincial Government (2018)

area, almost all the drinking groundwater source samples (in upper confined aquifer) had significantly higher level of TDS, about 84.1% samples exceeded 1000 mg/L, reflecting the content of groundwater TDS was not suitable for use as a source of drinking water. Researchers reported that continuous use of saline water further contributes toward the increasing risk of skin diseases, diarrhea, indigestion and other related diseases (Tauhid et al. 2017; Javed et al. 2018).

Total hardness (TH) is measured with the concentration of calcium and magnesium and generally is an important reference parameter for drinking water. (Todd 1980). According to WHO international standard, the allowable maximum limit of TH for human drinking purpose is 300 mg/L (WHO 2011). Studies revealed that one and six water samples with were considered as hard water (300–450 mg/L) and super-hard water (700–1000 mg/L), respectively. Short-time intake of a high TH value may not cause direct adverse effect, but ingestion of excess TH for a long time can cause salt deposits in the body (kidneys, gallbladder, and bones). Meanwhile, it can bring many troubles to life, such as scaling on water appliances, reducing washing efficiency of soaps and detergents, etc. In Haixing County total hardness in groundwater samples, with several exceptions, is within the safe limit demonstrated in Fig 5.

Physiological functions of sodium (Na^+) useful in human body and plants are identified; however, excess Na^+ for long times may exert osmotic stress on biota in the water and may cause severe hypertension and

high blood pressure in human (Holden 1970; Kawasaki et al. 1978). The data showed that a large variation of Na^+ throughout the Haixing Country as revealed by different regions. Na^+ concentrations in most water samples were much higher than the WHO standard of 200 mg/L, making it the major pollutant in upper confined water. The observed concentrations of Na^+ were between 379.1 and 558.3 mg/L (the mean value of groundwater samples in every county) from different regions. Salinity intrusion has contributed to sodium enrichment in groundwater, because of the excessive exploitation of groundwater by human beings. Meanwhile, the high concentration of Na^+ may be due to in some cases cation exchange reactions with Ca^{2+} and carbonates precipitation in the process of shallow groundwater seepage (Raza et al. 2007; Farooqi et al. 2007).

Generally, the Cl^- and SO_4^{2-} are considered as the essential elements for human health, but excessive Cl^- and SO_4^{2-} deteriorate the quality of drinking water at certain extent and lead to adverse health (Rao et al. 2012). For example, higher concentration of Cl^- in body may cause some negative effects to health, such as hypertension, risk of stroke, left ventricular hypertrophy, etc. (McCarthy 2004). However, diarrhea, catharsis, dehydration and gastrointestinal irritation may be associated with the ingestion of water containing excessive SO_4^{2-} , which is widely reported by many counties (Garg et al. 2009). For drinking water, WHO set the maximum accepted concentration of 250 mg/L and 250 mg/L for Cl^- and SO_4^{2-} ,

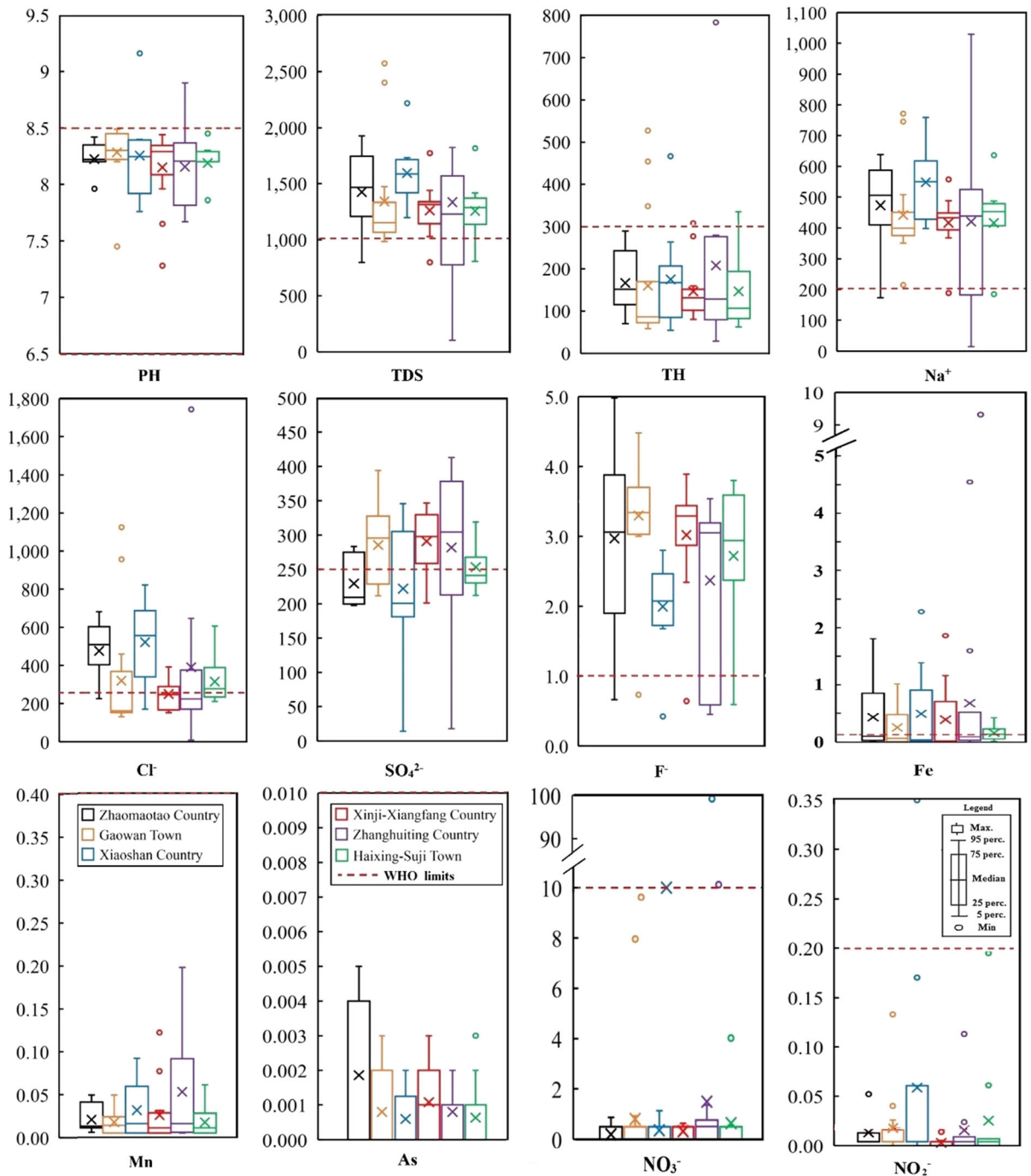


Fig. 5 Physiochemical parameters

respectively (Fig. 5). The groundwater samples of the study area show that the average concentration of Cl^- and SO_4^{2-} in each county ranged from 222.9 to 476.9 mg/L and 203.7 to 289.7 mg/L, respectively. Considering the concentration of chlorine and sulfate,

approximately 50% and 59% of the total groundwater samples were observed to exceed the recommended values for drinking purpose, respectively.

Fluoride (F^-) is known to be an important element in human health, and both its deficiency and

overexposure affect adversely teeth and bones (Disanayake 1991; Ozsvath 2009; Wei et al. 2016; Zhang et al. 2017). In Haixing County, fluoride concentrations of samples, with some exception, were above the WHO safe limits (exceeded 1.0 mg/L in 86.6% of 67 samples), which, after the current study, has stopped being used as a drinking water source. For this situation, various natural processes as well as anthropogenic activities seem to be responsible. There are several reasons that can be used to explain this phenomenon. There are several reasons that can be used to explain this phenomenon, such as the dissolution of fluorine-bearing minerals (biotite, muscovite, apatite and so on) in the stratigraphic structure (Azizullah et al. 2011). As well as the intense groundwater pumping can disturb the original hydrodynamic conditions (Wang et al. 2010; Liu et al. 2015), and further changed the fact that the water pressure of the soil makes the clayey soil with higher fluorine content release water, resulting in higher fluorine content in the aquifer. Meanwhile, fluorine can be introduced to the environment by industrial sewage disposal and using agricultural fertilizers which release F^- into air and then reach the soil with rainfall. Overall, F^- in upper confined aquifer is complex and may cause a challenging health problem deserving proper management and monitoring in the study area.

Toxic metal

Iron (Fe), an important oxygen-carrying substance in heme and myohemoglobin, gives oxygen to tissues, organs and muscles in the body, but its deficiency and overload can cause water pollution and result in severe health problems in animals (Anonymous 2008). In drinking, water the desirable concentration set by WHO is 0.3 mg/L. The measured data show that the percentage of exceeding WHO limit reaches 33.8% for the upper confined water samples. Various counties showed iron concentration ranging from 0.16 to 1.6 mg/L (Fig. 5), and the highest concentration of Fe was reported from Zhanghuiting Country, which may be due to the removal of sewage from surrounding factories. Although iron overload or overexposure is a less common condition, it can lead to several serious health problems like cancer (Beckman 1999; Parkkila et al. 2001), diabetes (Ellervik et al. 2001; Parkkila et al. 2001), liver and heart diseases (Milman

et al. 2001; Rasmussen et al. 2001) and neurodegenerative disorders (Sayre et al. 2000; Berg et al. 2002), compared with its deficiency.

Manganese (Mn) is a naturally occurring mineral in surface and groundwater, but human activities also contribute much to its introduction in water (USEPA 2004). In Haixing County groundwater (as an important part of drinking water), which mainly comes from the dissolution of minerals, the concentration of Mn posed little problem as the concentration of samples is below the WHO standard limit (0.4 mg/L). Meanwhile, manganese is one of the essential trace elements for human beings. However, exposure to excessive doses of Mn affects the major organs and systems of the body such as nervous system (Barbeau 1984; Inoue 1996; Crossgrove and Zheng 2004).

Elevated arsenic (As) in groundwater is recognized as a big threat in both carcinogenic and non-carcinogenic ways to public health all over the world, such as India, Vietnam, Bangladesh, Nepal and Myanmar (Ul-Haque et al. 2007). Meanwhile, exposure to high As level has been confirmed to be associated with various types of acute and chronic health hazards (Edmunds et al. 2015; Joseph et al. 2015; Rahman et al. 2018), and skin disorders and keratosis are the most common external indicators of arsenic poisoning. In this study, the concentrations of As in wells were reported that the variation range of groundwater samples is 0–5 $\mu\text{g/L}$, which was within the WHO standard of 10 $\mu\text{g/L}$.

Three types of nitrogen

The nitrogen pollution ($\text{NH}_4\text{-N}$, $\text{NO}_3\text{-N}$ and $\text{NO}_2\text{-N}$) of groundwater has become a serious and widespread problem all over the world (Chen et al. 2016; Kleinjans et al. 1991; Mehdi et al. 2018). Furthermore, a large number of studies on potable water contamination with nitrogen pollution were reported due to the rapid growing trend of industrialization and urbanization in many parts of China (Zhang 2015; Chen et al. 2017; Su et al. 2018; Zhang et al. 2018). Therefore, three types of nitrogen were analyzed in the present study, but $\text{NO}_3\text{-N}$ and $\text{NO}_2\text{-N}$ were detected in this study and were important from the health point of view.

According to National Water Quality Monitoring Program, nitrate has been considered as the fourth most potential contaminant (PCRWR 2005). Meanwhile, the elevated $\text{NO}_3\text{-N}$ concentration in drinking

water can be expected to have an adverse impact on the health, such as blue baby syndrome that is the most common health impact (Gupta et al. 2000; Weyer et al. 2001; Mehdi et al. 2018; Dippong et al. 2019). In general, in study area, $\text{NO}_3\text{-N}$ concentrations in collected samples were in the range of 0–99.2 mg/L with an average of 2.5 mg/L (Fig. 5). Only two samples showing higher $\text{NO}_3\text{-N}$ concentrations of S64 (99.2 mg/L) and S37 (10.9 mg/L) in Xiaoshan Country and Zhanghuiting Country, respectively, which may due to a large amount of downward leakage from the upper unconfined aquifer with high nitrate concentration (soil fertilization) into the upper confined aquifer, similar to the finding of Liu et al. (2015) that identified the reason for high $\text{NO}_3\text{-N}$ in confined aquifer.

The national standards have set the acceptable limits of nitrite for drinking groundwater as 0.2 mg/L. $\text{NO}_2\text{-N}$ concentrations were in the range of 0–0.35 mg/L, respectively. Only one sample (S42 in Xiaoshan Country) had relatively high concentration of $\text{NO}_2\text{-N}$ (Fig. 5), which was 1.75 times higher than the permissible limit, and may be caused by the mineralization of nitrogenous organic substances from plants and animals (Dippong et al. 2019). Compare with $\text{NO}_3\text{-N}$, $\text{NO}_2\text{-N}$ has more serious adverse impact on human health. Ingestion of more nitrite forms a nitrosamine with a secondary amine in human intestines and stomach, which is a carcinogen that causes a variety of cancers, thus posing serious questions on human physical and mental health (Dippong et al. 2019; Mehdi et al. 2018).

Water environment of health risks

According to the above analysis, the upper confined water as potable water sources in the study area has been polluted to some degree, and main elements that exceed the drinking water standard (WHO) were TDS, Na^+ , Cl^- , SO_4^{2-} , F^- , Fe, NO_3^- and NO_2^- . Based on measurements, human health risk assessment of drinking water was conducted, which was necessary to support groundwater quality assessments and environment management (Ugran et al. 2016). After comparison of various constituents in the samples with the standard and the hazardous effects of primary elements, six elements (including F^- , Fe, Mn, NO_3^- , NO_2^- and As) in drinking water were evaluated (USEPA 1989). The intervals of pollutant

concentration were obtained by triangular fuzzy number method mentioned above. The results of Table 3 indicate that concentration of parameters varies from region to region, which indicated that the health risk assessment of groundwater in the study of zoning has certain practical significance.

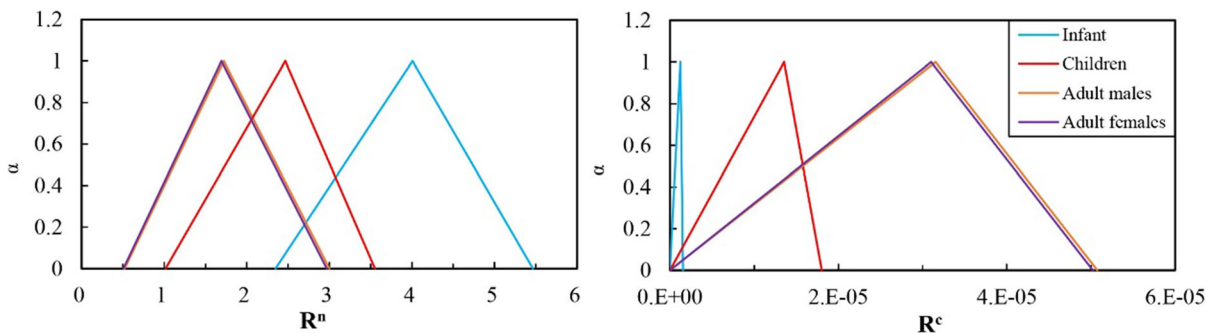
Total risk of Haixing County

On the basis of the reference values proposed by USEPA (1989) and the relevant researches (Siddique et al. 2015; Zhai et al. 2017; Mehdi et al. 2018; Barzegar et al. 2018), the health risk assessment was carried out using the HHRA model based on triangular fuzzy number mentioned above, and the ranges of risk of different parameters for different crowds (including infants, children, adult males, and adult females) were calculated through drinking water pathway. A relatively high level of confidence was adopted to assess public health risks for environment risk management, and the results are summarized in Figs. 6, 7 and 8.

The results indicated that there were differences in health risk of the pollutions among different regions and populations, due to the different impact of parameters on people. For Haixing County as a whole, for infants, the interval values ($\alpha = 0$) of R^n (total non-carcinogenic risk value) and R^c (total carcinogenic risk value) were 2.34–5.46, and 0 to 1.54E–06, and the most probable values ($\alpha = 1$) were 4.0 and 1.2E–06, respectively (Fig. 6). Health risk value of R^n to children was usually 0.5 or 1.3 time lower than to infants because of the different impact of exposure parameters on people, while health risk value of R^c was 11 time higher than infants, due to the toxicity characteristics of different elements. Carcinogens exhibit a non-threshold effect, and even a small amount of exposure has a negative impact on human health to a certain extent, while non-carcinogens exhibit a threshold effect. Only when the dose threshold is exceeded, it will have an impact on human health (Han 2014). Similarly, the values of R^n and R^c to adult males and adult females indicated that compared with infants, the risk of non-carcinogenesis of adults (including adult females and adult males) was higher and the risk of carcinogenesis was lower. For adult males, the related risk intervals ($\alpha = 0$) of R^n and R^c were in the range of 0.52–3 and 0 to 5.07E–05, and the most probable values ($\alpha = 1$) were 1.7 and 3.2E–05, respectively (Fig. 6). Meanwhile, the results

Table 3 Triangular fuzzy numbers of contaminant concentration in drinking water in study area

S	Parameters					
	F ⁻ (mg/L)	Fe (mg/L)	Mn (10 ⁻²) (mg/L)	NO ₃ ⁻ (mg/L)	NO ₂ ⁻ (10 ⁻²) (mg/L)	As (10 ⁻³) (mg/L)
ZM	[0.66, 2.97, 4.98]	[0.01, 0.43, 1.68]	[0.6, 2.07, 4.9]	[0, 0.55, 0.81]	[0, 1.33, 4.58]	[0, 2.43, 5]
GW	[1.71, 3.3, 4.48]	[0.01, 0.26, 0.94]	[0.5, 1.76, 4.9]	[0, 1.04, 4.74]	[0, 1.82, 8.28]	[0, 1.4, 2.26]
XS	[0.71, 2, 2.8]	[0.01, 0.49, 1.96]	[0.5, 3.14, 8.41]	[0, 10.4, 69.6]	[0, 5.9, 27.6]	[0, 1.2, 2]
XJ	[1.46, 3.02, 3.89]	[0, 0.39, 1.51]	[0.81, 2.61, 4.4]	[0, 0.51, 0.59]	[0, 0.48, 1.02]	[0, 1.54, 3]
ZH	[0.45, 2.37, 3.54]	[0.01, 1.61, 7.41]	[0.5, 5.29, 19.8]	[0, 1.64, 7.82]	[0, 1.69, 8.21]	[0, 1.1, 1.7]
HX	[0.62, 2.72, 3.8]	[0.01, 0.16, 0.41]	[0.5, 1.75, 3.36]	[0, 0.83, 2.85]	[0, 2.68, 13.8]	[0, 1.36, 2.65]

**Fig. 6** Comparison of human health risk among different crowds in Haixing County

of adult females were the same to those of adult males except for a little exception considering the discrepancy of water ingestion rate and body weight. These results illustrated that as a whole the sequence of the non-carcinogenic risk value through drinking water intake was infants > children > adult males > adult females, and most of the values were higher than 1 and this implies that the local population of this area can be at risk of non-carcinogenic risk with water being the main source. However, the sequence of the carcinogenic risk was adult males > adult females > children > infants, and the risks of carcinogenic risk value were lower than $1.0E-4$, demonstrating minimal and acceptable health risk.

Health risk assessment based on triangle fuzzy approach

Considering some management policies, different management levels and degree of groundwater pollution from counties to counties, the risk values of each zone was discussed (Fig. 7). Tables 4 and 5 present the calculated results of health risks from pollution for

infants, children, adult males, and adult females through drinking water intake in the study area when $\alpha = 0.8$. As can be seen in Table 4, the total non-carcinogenic risks for infants ranged from 4.85 to 5.18, 4.45 to 4.85, 4.21 to 5.27, 3.64 to 4.34, 3.48 to 4.51, and 3.36 to 4.4 in GW, XJ, ZM, HX, ZH, and XS, respectively. For children, the R^n were in the ranges of 2.83–3.23, 2.62–2.98, 2.53–3.3, 2.18–2.71, 2.09–2.92, and 2.01–2.76 for GW, XJ, ZM, HX, ZH, and XS, respectively. For adult males, the lower and upper risks through drinking water consumption in GW, XJ, ZM, HX, ZH, and XS are 1.91 and 2.36, 1.76 and 2.19, 1.74 and 2.44, 1.50 and 1.98, 1.45 and 2.09, 1.37 and 2.06. The R^n of adult females had the same distribution trend as that of adult males in different regions, but compared with the R^n of adult males, adult females were 0.2 or 0.3 lower than the lower risk values of the total non-carcinogenic risk of adult males, and 0.2–0.4 lower than the upper risk values of the R^n of adult males. Results revealed among the four age groups, infants were at the highest non-carcinogenic risk.

According to the middle values ($\alpha = 1$) of R^n , the total non-carcinogenic risk for local residents was

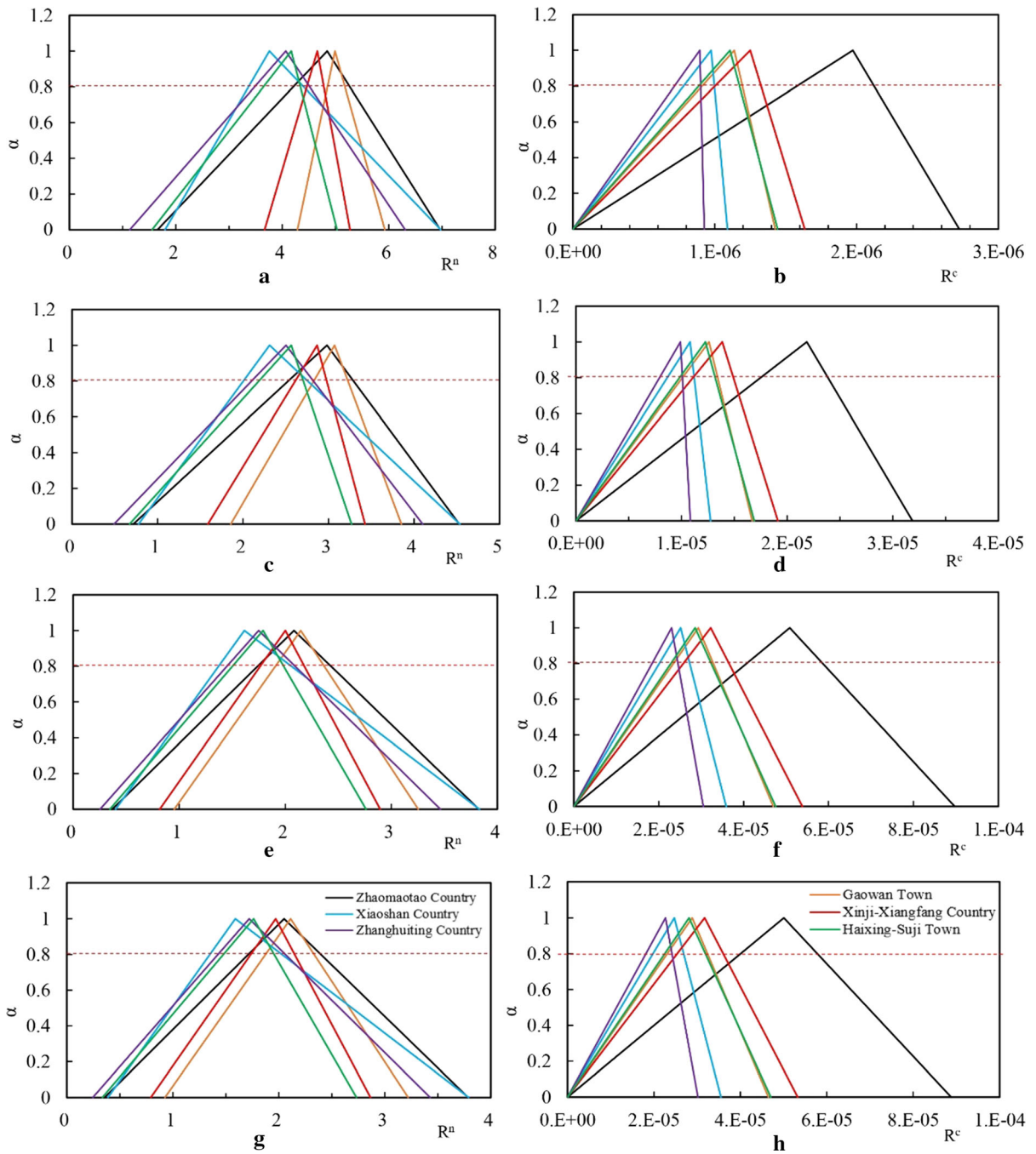


Fig. 7 R^c and R^n indices through consumption of drinking water. **a, b** Non-carcinogenic and carcinogenic risks of infants, respectively; **c, d** non-carcinogenic and carcinogenic risks of

children, respectively; **e, f** non-carcinogenic and carcinogenic risks of adult males, respectively; **g, h** non-carcinogenic and carcinogenic risks of adult females, respectively

obtained in the following order: GW > XJ > ZM > HX > ZH > XS (Fig. 7). The results clearly indicated that total non-carcinogenic risk values are beyond the acceptable limit ($R^n = 1$) recommended

by USEPA of all counties for all the crowds, suggesting the level of pollution may pose considerable adverse health effects to the individuals of all ages and sexes who use it as drinking water. And the

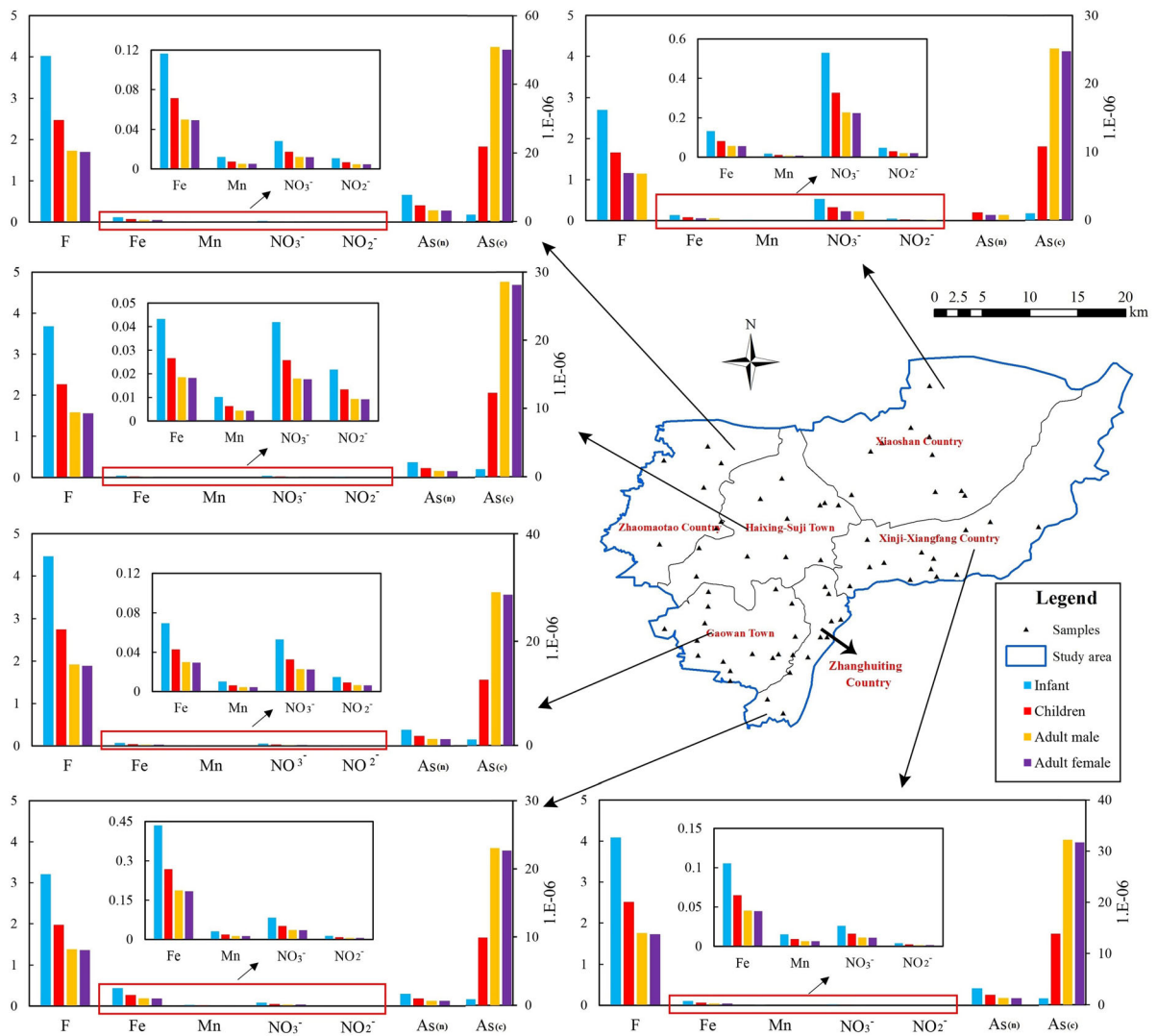


Fig. 8 The maps of risk values of six elements in the six counties

Table 4 Interval values of non-carcinogenic risk of contaminants in drinking water ($\alpha = 0.8$)

Site	Risk values			
	Infants	Children	Adult males	Adult females
GW	[4.85, 5.18]	[2.83, 3.23]	[1.91, 2.36]	[1.88, 2.33]
XJ	[4.45, 4.85]	[2.62, 2.98]	[1.76, 2.19]	[1.73, 2.15]
ZM	[4.21, 5.27]	[2.53, 3.30]	[1.74, 2.44]	[1.71, 2.40]
HX	[3.64, 4.34]	[2.18, 2.71]	[1.50, 1.98]	[1.47, 1.96]
ZH	[3.48, 4.51]	[2.09, 2.92]	[1.45, 2.09]	[1.42, 2.06]
XS	[3.36, 4.40]	[2.01, 2.76]	[1.37, 2.06]	[1.35, 2.03]

highest R^n , namely 5.0 ($\alpha = 1$), of the study area was in Gaowan Town for the infants (Fig. 7a, c, e, g).

The probability interval distribution of total carcinogenic risk values ($\alpha = 0.8$) for the four age groups is shown in Table 5. Huge variation in values of

Table 5 Interval values of carcinogenic risk of contaminants in drinking water ($\alpha = 0.8$)

Site	Risk values			
	Infants (1.0E−6)	Children (1.0E−5)	Adult males (1.0E−5)	Adult females (1.0E−5)
ZM	[1.58, 2.12]	[1.75, 2.39]	[0.41, 0.59]	[0.40, 0.58]
XJ	[1.00, 1.33]	[1.11, 1.49]	[0.26, 0.37]	[0.25, 0.36]
GW	[0.91, 1.20]	[1.01, 1.34]	[0.23, 0.33]	[0.23, 0.32]
HX	[0.89, 1.18]	[0.89, 1.32]	[0.23, 0.32]	[0.22, 0.32]
XS	[0.78, 1.00]	[0.86, 1.12]	[0.20, 0.27]	[0.20, 0.27]
ZH	[0.72, 1.18]	[0.79, 1.76]	[0.18, 0.25]	[0.18, 0.24]

carcinogenic risk was in the ranges of 1.58E−6 to 2.21E−6, 1.0E−6 to 1.33E−6, 0.91E−6 to 1.2E−6, 0.89E−6 to 1.18E−6, 0.78E−6 to 1.0E−6, and 0.72E−6 to 1.18E−6 for infants, and 1.75E−5 to 2.39E−5, 1.11E−5 to 1.49E−5, 1.01E−5 to 1.34E−5, 0.89E−5 to 1.38E−5, 0.86E−5 to 1.12E−5, and 0.79E−5 to 1.76E−5 for children in ZM, XJ, GW, HX, XS, and ZH, respectively. For adult males and adult females in HX and XS, the lower and upper risks through drinking water consumption were the same, 0.23E−5 to 0.32E−5 (HX), 0.2E−5 to 0.27E−5 (XS), respectively, while the upper and lower carcinogenic intervals in adult males were greater than 0.1E−5 in adult females in ZM and XJ. In GW and ZH, the upper and lower carcinogenic ranges were the same for adult males and adult females, while the lower value were less than 0.1E−5 for adult males.

Meanwhile, the mean total carcinogenesis risk values were the maximum at ZM and then decreased in turn in XJ, GW, HX, XS and ZH. Results indicated low probability of pollutant-related diseases in local human population, as in all of drinking well samples, total carcinogenicity $R^c \leq 1.0E-4$. And the highest R^c , namely 2.19E−5 ($\alpha = 1$), of the study area was in Zhaomaotao County for children, while the lowest value (8.9E−7) was observed in Zhanghuiting County for infants (Fig. 7b, d, f, h).

To develop a more detailed understanding of uncertainty, the non-carcinogenic and carcinogenic risk values with different confidence levels in drinking water for infants, children, adult males and adult females from the six counties and towns were calculated. When $\alpha = 0$, the narrow range of risk value reveals the weak uncertainties. Meanwhile, the uncertainties has a significant impact on the total risk values, which means that total risk is more sensitive to variation in data (Chen et al. 2018). The total non-carcinogenic levels for populations (including infants,

Children, Adult males, and Adult females) were generally observed in the order of $XS > ZH > ZM > HX > GW > XJ$, while the orders of R^c were $ZM > XJ > HX > GW > XS > ZH$. As the above discussion attests, the selection of a confidence level for interval has effect on the ranking of risk values in each county.

Spatiotemporal variation of each element

Based on our results, it can be concluded that the total carcinogenic and non-carcinogenic risks have some different degrees of influence on the human health in different counties and towns. However, it is not clear that the main factors affecting the high non-carcinogenic risk value. Therefore, for further assessing the chemical components in drinking water, risk value of each indicator of each county in the study area was calculated. The distributions of mean carcinogenic and non-carcinogenic risk values for 4 groups of people (including infants, children, adult males, and adult females) of the 6 counties are shown in Fig. 8.

The calculated risk value of elements had varying degrees of impact on total carcinogenic and non-carcinogenic risks. According to Fig. 8, potential health risk values of fluoride were significantly large compared with the other parameters for residents in Haixing County, accounting for 71.9–89.4% of the total non-carcinogenic risk. The results indicated that fluoride in groundwater, which was the main factor affecting the non-carcinogenic risk value and above the safe threshold value of 1, can cause high probability of fluoride-related toxicity to local residents of the study area, while the range of F^- was quite higher for infants than other groups, followed by arsenic in the majority of areas. For As^{III} (non-carcinogenic value of arsenic), the mean values (ZM, GW, XS, XJ, ZH, and HX) were in the ranges of 0.3–0.66, 0.18–0.4,

1.28E−1 to 2.82E−1, and 1.26E−1 to 2.78E−1 for infants, children, adult females, and adult males, respectively, and the maximum (mean) risk value occurred in Zhaomaotao County and the minimum appeared in Zhanghuiting County. Compared with fluoride, the risk values of Fe and NO₃[−] were relatively small, ranging from 0.04 to 0.43 and 2.5E−2 to 0.53 (infants), 0.02–0.26 and 1.6E−2 to 0.33 (children), 1.85E−2 to 1.87E−1 and 0.01 to 2.27E−1 (adult females), and 1.82E−2 to 1.84E−1 and 0.01 to 2.24E−1 (adult males). It was noteworthy that the non-carcinogenic risk of nitrate in Xiaoshan County is higher than that in other counties. The reason for this was that the concentration of nitrate in a water sample in Xiaoshan County is 99.2 mg/L, which made the average concentration of nitrate on the high side and increases the risk value of nitrate in the whole county. Among the six regions, the highest iron risk value was found in ZhanghuiTing County, which was caused by the high value of 3 abnormal samples, 9.31, 4.55, and 1.6 mg/L. The non-carcinogens Mn and NO₂[−] were identified as having low risk values (less than 0.03), indicating that no region was threatened by the two chemical parameters.

The total carcinogenic risk value consists of only one arsenic cancer element, and the carcinogenicity analysis of arsenic was the same as previous studies on the total carcinogenic risk, so we would not elaborate on it here.

As the results of the present study show, the levels of fluoride were higher in the study area, which was primarily attributed to geological factors. Nonetheless, as pumping of groundwater and release of pollutants by human can directly or indirectly change the environment of groundwater, it can also affect the fate of chemical parameters (Smith et al. 2000; Radloff et al. 2011; Dragon 2012). Therefore, given that the unacceptable risk value of F[−] coincides with high population density in Haixing County, it would appear that elevated F[−] is an alarming news to the practitioners, researcher, policy makers and people who are using upper confined water as main source of safe drinking water, and relevant measures should be taken to help to control or diminish the problems of deteriorating water quality.

Conclusions

The residents in Haixing County depend heavily on the upper confined water, the characteristics of chemical component were analyzed, and health risks via direct ingestion of drinking water were calculated for infants, children, adult males, and adult females using human health risk assessment (HHRS) based on triangular fuzzy number. The levels of pH, TDS, TH, SO₄^{2−}, Na⁺, Cl[−], SO₄^{2−}, F[−], F, NO₃[−], and NO₂[−] slightly exceeded the permissible limits except As and Mn. Risk level for different crowds in the study area demonstrated an obvious difference, generally in the order infants > children > adult males > adult females for non-carcinogenic risk values (R^n), while the sequence of the carcinogenic risk values (R^c) was adult males > adult females > children > infants. The non-carcinogenic risk values (R^n) through drinking water intake were higher than 1, and this implied potential health impacts on human health for local residents with respect to $\alpha = 0.8$. However, the risks of carcinogenic risk values (R^c) were lower than 1.0E−4, demonstrating minimal and acceptable health risk. Furthermore, considering the uncertainty ($\alpha = 1$) in data, the total non-carcinogenic risks for local residents were obtained in the following order: GW (Gaowan Town) > XJ (Xinji-Xiangfang County) > ZM (Zhaomaotao County) > HX (Haixing-Suji Town) > ZH (Zhanghuiting County) > XS (Xiaoshan County), and ZM > XJ > GW > HX > XS > ZH for R^c . With the help of HHRA, the parameter F[−] in groundwater was identified to be as a significant human health issue of Haixing County, and such exposure (through drinking water) leads to numerous skeletal and physiological disorders in the human especially in infants and children. Finally, as treatment for rural people, drinking clean water that can be obtained by purchasing pure water or abandoning excessive water chemical parameters in drinking water, etc., is essential for disease prevention. In conclusion, the results of this study have advanced the understanding of the chemical parameters fate in the upper confined aquifer system. They can be used as a scientific guidance for formulating more regulations and educating the population on groundwater safety and pumping programs.

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