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ORIGINAL PAPER

Occurrence and Distribution of Groundwater Fluoride and Manganese in the Weining Plain (China) and Their Probabilistic Health Risk Quantification

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Abstract

In this paper, the spatial distribution of fluoride (F^-) and manganese (Mn) in groundwater was described and the overall groundwater quality was assessed with the entropy weight water quality index (EWQI) in Weining Plain, northwest China. The probabilistic health risk of F^- and Mn in groundwater was assessed by the Monte Carlo stochastic simulation method. The results show that 50 groundwater samples (a total of 144) belong to poor and very poor quality because of agricultural activities, industrial development, and the local hydrogeological conditions. Groundwater quality in the upper reaches of the Yellow River is better than that in the lower reaches. The average health risks faced by children and adults are 4.59 and 0.62, respectively. If the uncertainty of the model parameters are considered, the mean risks faced by children and adults are 6.84 and 0.92, respectively, indicating that the health risk to residents, especially children, cannot be ignored. Compared with dermal contact, drinking water intake is the main exposure way to harm residents' health. F^- contributed higher to the health risk than Mn, and children face greater risk than adults. Stochastic simulation can reflect health risks more comprehensively than the deterministic methods. The study results are helpful for decision makers to take measures for the safe supply of drinking water in the study area.

Keywords Groundwater pollution · Monte Carlo · Health risk · Uncertainty · Sensitivity analysis

Introduction

Groundwater is of great significance for drinking, irrigation, industrial, aquaculture, and wildlife (Falkenmark 2005; Ghazavi and Ebrahimi 2015; Ren et al. 2021). In many arid and semiarid regions, groundwater has even become the only source of drinking water on account of inadequate rainfall and surface water (Li et al. 2019a; Shukla and Saxena 2021). Meanwhile, water resources are considered as the first priority for sustainable development in some countries (Fekkoul et al. 2013). Owing to the protection of the vadose zone, groundwater is considered to be safer than surface water (Subba Rao 2018; Khan et al 2021).

The quality and quantity of groundwater are equally important (Li 2016). Especially, when groundwater is used as drinking water source, its water quality is directly related to residents' health. However, due to global climate change, the impact of human activities, and unscientific groundwater management, the quantity and quality of the groundwater in various areas have decreased (Soma and Kumar 2015). Deterioration of groundwater quality will seriously hinder the development of society (Li et al. 2017a, 2021a). Contaminated groundwater, once consumed by human beings through drinking, dermal contact, and breathing, can cause harm to human health (Chabukdhara et al. 2017; Wang et al. 2020; Liu et al. 2004). Therefore, conducting groundwater quality studies is essential for the sustainable development of the society (He and Wu 2019). Researchers have founded that the hydrochemical characteristics of groundwater are affected by water–rock interaction and human activities, which hinders the local development and threaten public health (Purushotham et al. 2017; Shukla and Saxena 2020;

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Verma et al. 2021; Subba Rao 2021). Researchers have proposed a number of methods for studying water quality, such as the Technique for Order Preference by Similarity to Ideal Solution (TOPSIS) method (Li et al. 2011a, 2013a, 2013b), water quality index (WQI) (Vasanthavigar et al. 2010), water quality identification index (Zhang et al. 2021), and set pair analysis (Su et al. 2019; Li et al. 2011b; Tian and Wu 2019). These studies have promoted the development of water quality research.

Both fluoride and manganese are trace elements needed by the human body, but excessive intake of them will have adverse effects on health (Hong et al. 2018). Excessive intake of fluoride can cause dental fluorosis and skeletal fluorosis (Adewole et al. 2021; Deepanjan et al. 2021), and excessive manganese intake can harm the liver (Chen et al. 2015). Health risk assessment intends to find and reveal the connection between environmental pollution and human health, and can quantitatively describe the risk of pollution to human health (Li et al. 2021b). The traditional health risk assessment uses a deterministic model, which collects a large number of model parameters and pollutant concentration data, and calculates their mean risk values. However, the parameters of the model are not actually a definite value. They are affected by many uncertain factors, such as amount of drinking water, human body weight, the frequency of bathing, and age. Simultaneously, the concentration of pollutants is also not a definite value, but it varies with time, places, and receptors (Yang et al. 2010). By reason of the randomness of the model parameters, the results obtained by the traditional deterministic health risk assessment model cannot reflect the real situations satisfactorily (Liu et al. 2004). To overcome this drawback, many researchers have proposed methods considering uncertainty, such as grey system theory, fuzzy mathematics theory, and probability statistics theory (Wang 2004). Grey system theory is mainly used for short-term prediction with unclear principles, fuzzy theory is mainly used to deal with things with unclear boundaries, and probability statistics theory is mainly used to study the probability of occurrence of events affected by random factors (Zhou 2017). Therefore, probabilistic statistics theory is the most suitable method to study the probabilistic health risk. Monte Carlo is a simulation method based on the probabilistic statistics theory, and it can yield reliable results (Soleimani et al. 2020; Rajasekhar et al. 2020; Ali et al. 2021).

Weining Plain is a traditional agricultural region in northwest China, and it has undergone rapid industrial development in recent years. However, both industry and agriculture use groundwater for development in addition to river water from the Yellow River, and domestic water is mainly from groundwater (Wang et al. 2017). Recent research has shown that groundwater in this area has been contaminated (Li et al. 2016a,b). In addition, researchers have used the modified DRASTIC model and quantized pollution loading method to quantify the vulnerability and pollution carrying capacity of groundwater in Weining Plain, and found that the areas with high pollution are mainly situated in two industrial parks (Li et al. 2017b). The health risk of an industrial park in the survey region was evaluated by Li et al. (2014a) using deterministic method, and found that the total health risk evoked by drinking contaminated groundwater exceeded the maximum acceptable level. The existing research provides valuable instruction and guidance for groundwater protection in this area. However, the probabilistic health risk caused by groundwater contamination in the entire Weining Plain has not been well discussed.

Therefore, this paper focused on groundwater quality and health risk assessment on the basis of field investigation and sampling. The main purposes of this research are: (1) to learn the groundwater quality in the study area clearly, and (2) to evaluate the human health risks of fluoride (F^-) and manganese (Mn) in groundwater using probabilistic health risk assessment model. The results will be helpful to furnish decision makers with a reasonable basis for the rational utilization and management of local groundwater in the plain.

Study Area

Location and Climate

Weining Plain is located in the central and western part of Ningxia Hui Autonomous region (Fig. 1), with the Xiangshan to the south, Beishan to the north, Tengger Desert to the west and Niushou Mountain to the east. The Yellow River flows through the plain from southwest to northeast. The entire plain covers about 1600 km^2. In terms of the elevation, Weining Plain is high in the west and low in the east, high in the north and south, and low in the middle. Weining Plain is controlled by the continental monsoon climate, with hot and rainy summers, cold and dry winters, warm springs and cool autumns. The annual average temperature is about 10.9℃, and the temperature is usually the highest in July and the lowest in January. The multi-year average precipitation is 179.4 mm, and the multi-year average evaporation is 1390.0 mm (Wang et al. 2017).

Hydrogeology

Weining Plain is a faulted basin formed in the Cenozoic. It is distributed in the east–west structural belt of Weining area. Since the Quaternary, a thick layer of loose sediments has been deposited in the area, and the upper Holocene alluvium is in disconformable contact with the lower Pleistocene (Li et al. 2016a; Lyu 2015). The aquifer is mainly constituted by the gravels and sands, loamy sands and silty clay (Li et al.

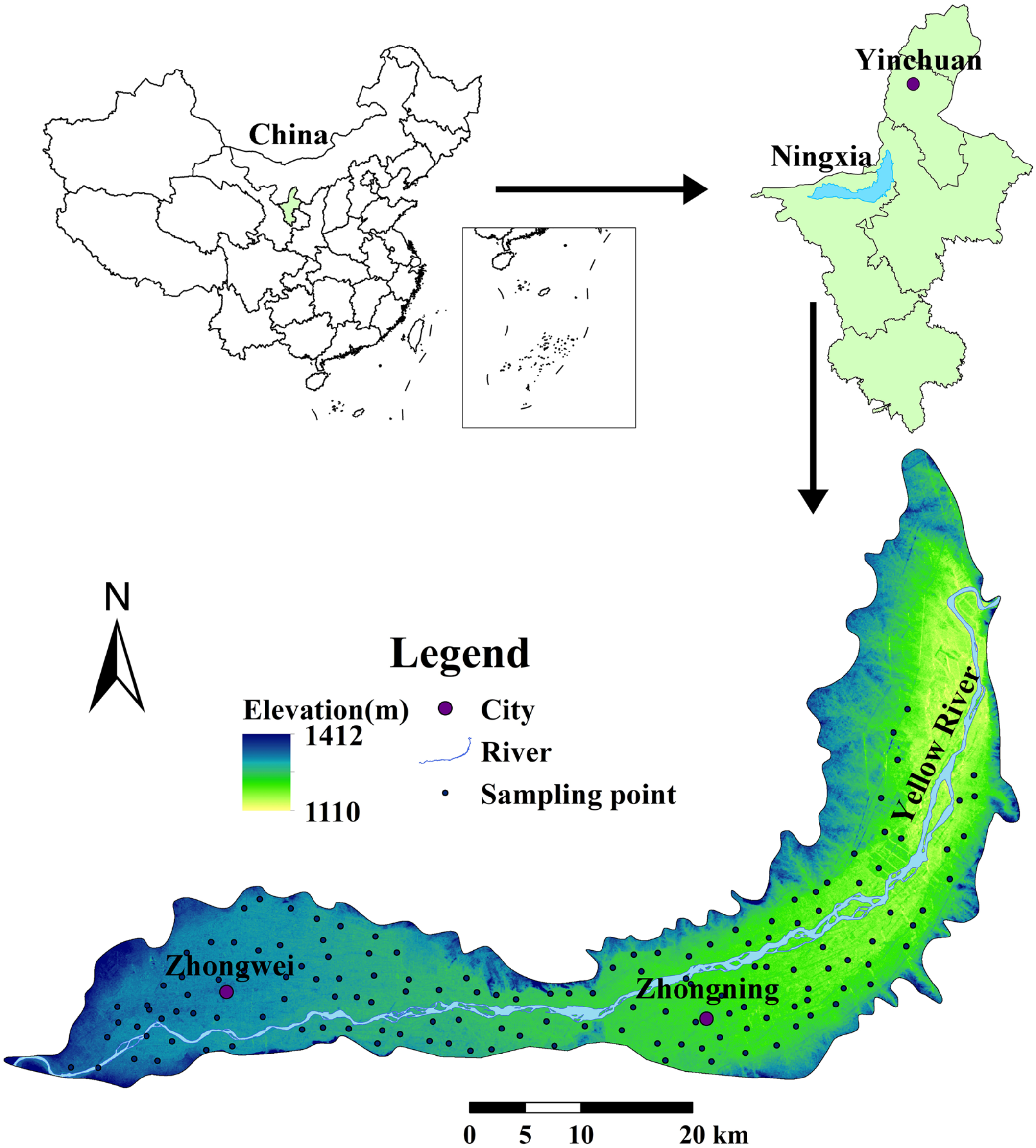


Fig. 1 Location of study area and sampling sites

2016a). Weining Plain is characterized by thick aquifers, loose structure of aquifer media, abundant recharge sources of groundwater and strong water resources bearing capacity (Li et al. 2020). The thickness of the phreatic aquifer ranges from 6 to 48 m, and groundwater level depth for the phreatic aquifer is less than 5 m (Li et al. 2017b).

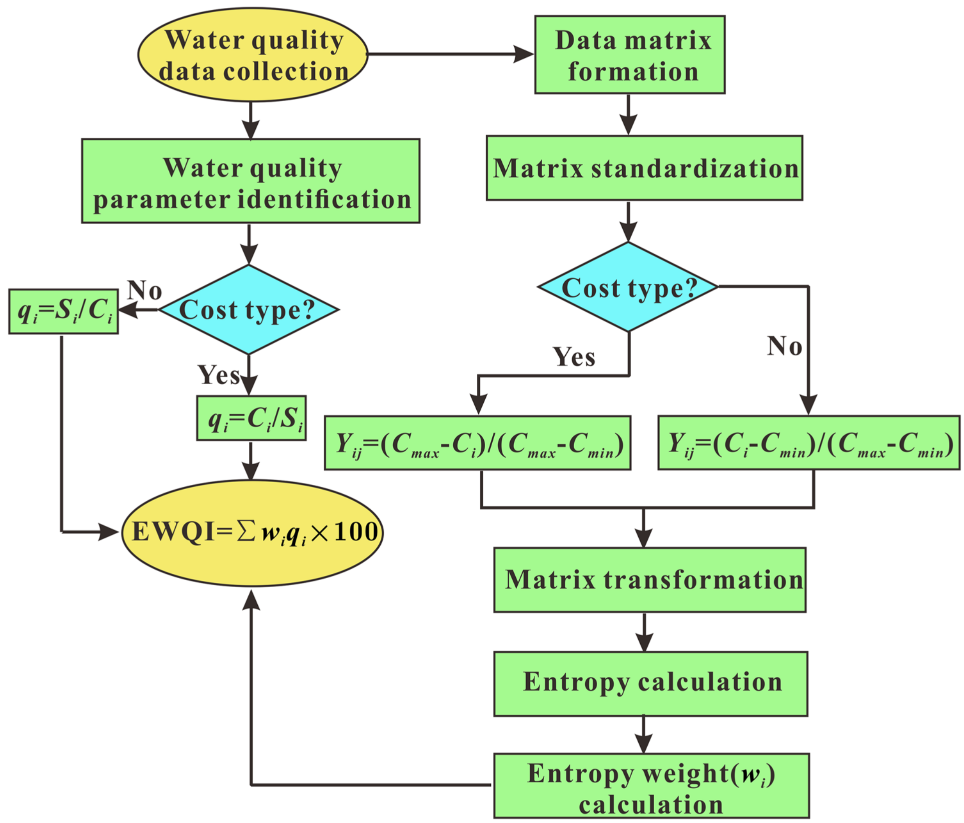
The groundwater generally flows from both sides to the Yellow River. Regionally, it flows from northwest to southeast in the north side of the Yellow River, and the groundwater in the south side flows from southwest to northeast (Wang et al. 2017). Weining Plain is a typical irrigation area by the diversion of the Yellow River, and rainfall is scarce. Thus, infiltration recharge of the Yellow River diversion canal system and irrigation infiltration recharge are the main sources of groundwater recharge (Wang 2017). Groundwater discharge mainly occurs by runoff, evaporation, and artificial exploitation. Groundwater in the survey region is mainly used for daily domestic usage, industrial production and agricultural development.

Materials and Methods

Sample Collection and Analysis

In this study, 144 groundwater samples were collected from the phreatic aquifer from private wells in the Weining Plain (Fig. 1). The sampling, storage, and handling were carried

Fig. 2 Procedures of EWQI



out according to the national standard (Ministry of Environmental Protection of China 2009). The containers were washed three times with the water to be sampled before sampling. After sampling, the samples were sent to the Soil and Water Testing Center of Shaanxi Institute of Engineering Investigation for physicochemical analysis. The physicochemical indices analyzed include pH, total dissolved solid (TDS), total hardness (TH), main cations (Na^+, K^+, Ca^{2+}, and Mg^{2+}), major anions (Cl^-, SO_4^{2-}, and HCO_3^-), nitrate (NO_3-N), nitrite (NO_2-N), ammonia nitrogen (NH_4-N), F^-, and Mn. In the process of sample analysis, standardization, setting blank control and duplicate testing were introduced to ensure the quality of data. In addition, ion charge balance error percentage (%CBE) was used to measure the accuracy of the physicochemical analyses (Li et al. 2018a). The %CBE of each sample is within ± 5%, which manifests that the data is credible. The ion charge balance error percentage was calculated as follows:

$$\%\ \mathrm{CBE} = \frac{\mathrm{TC} - \mathrm{TA}}{\mathrm{TC} + \mathrm{TA}} \times 100 \quad (1)$$

where, TC and TA represents the total concentrations of cations and anions, respectively, meq/L.

Water Quality Analysis

In this study, the overall water quality evaluation is achieved by employing the entropy weight water quality index (EWQI). The method consists of five steps (Fig. 2). First is to establish an initial matrix according to physicochemical data of the water samples. After that, standardization is performed to avoid the unit and order of magnitude differences of each index affecting the calculation results as much as possible. In the next step, the information entropy is calculated and the weight of each index is obtained using it. And the quality rating scale q_j of parameter j should be calculated. The last step is to calculate the value of EWQI. The procedures for calculating EWQI are briefly summarized in Fig. 2 (Li et al. 2010, 2013c, 2014a; Wu et al. 2011).

According to the value of EWQI, groundwater is segmented into five grades, and the water quality classification criteria can be inquired from Table 1 (Li et al. 2018b).

Health Risk Assessment

The effects of pollutants on human health can be characterized by health risk assessment (Li et al. 2014a). The model mainly relied on in this study was published by the Ministry of Environment Protection of the P.R. China (2014). Hazard identification, dose–response assessment, exposure

Table 1 Water quality Classification Standard based on EWQI

EWQI	Grade	Water quality
< 25	I	Excellent quality
25–50	II	Good quality
50–100	III	Medium quality
100–150	IV	Poor quality
> 150	V	Very poor quality

assessment and risk characterization are the four main steps in the process of assessment (Momot and Synzynys 2005).

Carcinogenic risk and hazard quotient (HQ) characterize the harm caused by human exposure to carcinogenic pollutants and non-carcinogenic risk pollutants, respectively (Wei et al. 2021). For the study area, F^- and Mn have non-carcinogenic effects on humans, and local residents can be affected by the contaminants through two exposure pathways: (a) Oral intake of contaminated groundwater. (b) Dermal contact with contaminated groundwater through daily bathing and washing.

Non-carcinogenic risk through oral intake pathway (HQ_o) can be evaluated according to the following formulae (Wang et al. 2021; Li et al. 2021c):

$$Intake_o = \frac{C \times GWCR \times EF \times ED}{W \times T} \tag{2}$$

$$HQ_o = \frac{Intake_o}{RfD_o} \tag{3}$$

where, $Intake_o$ denotes the daily average exposure caused by drinking contaminated groundwater, mg/(kg·day). C represents the concentration of the contaminants considered, mg/L. $GWCR$ indicates the groundwater consumption rate, L/day. EF is the exposure frequency, day/year. ED represents the exposure duration, year. W indicates the body weight, kg. T denotes the average time of non-carcinogenic effect, day. RfD_o refers to the reference dose for non-carcinogenic pollutant through oral intake pathway, RfD_o for F^- and Mn are 0.04 and 0.1 mg/(kg·day), respectively.

The following formulae can be used for calculating the non-carcinogenic risk through dermal intake pathway (HQ_d, Ji et al. 2020; Wu et al. 2019, 2020):

$$Intake_d = \frac{C \times AF \times ET \times EF \times ED \times SA \times CF}{W \times T} \tag{4}$$

$$HQ_d = \frac{Intake_d}{RfD_d} \tag{5}$$

where, $Intake_d$ is the daily average exposure dosage through dermal intake per unit weight, mg/(kg·day). AF is the skin permeability coefficient, cm/hour. ET is the exposure time, hour/day. SA indicates the surface area of skin exposed to pollutants, cm^2. CF is a conversion factor with a value of 0.001 (Wu and Sun 2016). RfD_d is the reference dose for non-carcinogenic pollutant through dermal intake, RfD_d for F^- and Mn are 0.04 and 0.1 mg/(kg·day), respectively.

The total non-carcinogenic risk of F^- and Mn (HQ_{total}) can be obtained by adding up the risks of different exposure pathways caused by the two pollutants (Eq. 6). $HQ_{total} > 1$ indicates an unacceptable risk.

$$HQ_{total} = HQ_o + HQ_d \tag{6}$$

The parameters in the above formulae are mainly derived from the recommended values by the Ministry of Environment Protection of the P.R. China (2014) and related studies that have been carried out in China. The values and descriptions of the parameters are shown in Table 2. The concentrations and statistical distributions of F^- and Mn are summarized in Table 3.

Table 2 Exposure parameters for adults and children

Parameters	Meaning of parameters	Units	Distribution form	Content
$GWCR_a$	Groundwater consumption rate for adults	L/day	Normal	1.67–2.33
$GWCR_c$	Groundwater consumption rate for children	L/day	Normal	0.57–1.53
EF	Exposure frequency	day/year	–	350
ED_a	Exposure duration for adults	year	–	24
ED_c	Exposure duration for children	year	–	6
W_a	Average weight of adults	kg	Normal	50.1–69.9
W_c	Average weight of children	kg	Normal	6.1–25.9
T_a	Average effect time of adults	day	–	10,950
T_c	Average effect time of children	day	–	2190
AF	Skin permeability coefficient	cm/hour	–	0.001
ET	Average exposure time	hour/day	–	0.4
SA_a	Skin surface area of adults	cm^2	Triangular	14,400–17,600
SA_c	Skin surface area of children	cm^2	Triangular	8500–11,500

Monte Carlo Simulation

Monte Carlo method was employing to quantify the indeterminacy factor associated with the health risk model in the Oracle Crystal Ball software, and its basic principle is to define the corresponding probability distribution for various uncertain factors. According to the probability distribution of each factor, a large number of random values can be generated, and simulated values under different confidence degrees can be obtained (Zhao 2013). With the increase of iteration times, the simulation results tend to be stable. The stochastic modeling can make the result more objective. The procedures of Monte Carlo simulation can be generally divided into the following four steps:

The first step is to define the assumptive variables. The random variables with known probability distributions such as the groundwater consumption rate (*GWCR*), average body weight (*W*), surface area of exposed skin (*SA*) and concentrations of different pollutants are defined as the assumptive variables.

The second step is to define the objective to be forecasted. According to the non-carcinogenic risk calculation formulas (Eqs. 2–6), risks to different population groups through different pathways were defined as the objective to be forecasted.

The third step is to determine the running preferences, which include the number of trials to run and the confidence level determined. In this study, the number of trials to run was set to 20,000 and the confidence level was set to 95%.

The last step is to run the simulation. After running the simulation, the statistics tables, frequency charts and the percentile tables can be obtained. The simulation results can also show the influence of different uncertain factors on the results. The absolute value of sensitivity indicates the degree of influence, and the positive or negative of the sensitivity indicates that it has a positive or negative effect on the result.

Results and Discussion

Hydrochemical Characteristics

Water quality parameters were analyzed statistically, and the statistical results and the standards (General Administration of Quality Supervision, Inspection and Quarantine of the People's Republic of China 2017) were shown in Table 4. Table 4 shows that the pH varies between 7.53 and 8.51, and the average value is 7.93, which shows that the groundwater is weakly alkaline. The exceeding standard rates for most groundwater quality parameters in the study area are high. For example, the exceeding standard rates of SO_4^{2-}, Mn and TH are higher than 50%, and the exceeding standard rates of Na^+, K^+, Cl^-, NO_3^-, F^- and TDS are more than 15%. About 55% of the groundwater samples have TDS less than 1 g/L, which belongs to fresh water, and the rest are brackish water with TDS between 1 and 2 g/L. Meanwhile, a few samples with high TDS contain high concentrations of SO_4^{2-} and Na^+, which indicates the strong evaporation and concentration effects and water–rock interactions. Weining Plain has a long history of agricultural development, and NO_3^-, NO_2^-, and NH_4^+ are common pollutants in aquatic environment in agricultural regions (Mahvi et al. 2005; Zhang et al. 2018). Soluble nitrate will transport in the groundwater with groundwater flow (Li et al. 2014a). In the present study, over 20% of the groundwater samples are detected with high-nitrate concentration, indicating the agriculture seriously impacts groundwater quality.

The hydrochemical types of groundwater are delineated by the Piper diagram (Piper 1944; He and Li 2020). Figure 3 shows that the groundwater samples mainly belong to SO_4·Cl-Ca·Mg and HCO_3-Ca·Mg, followed by SO_4·Cl-Na. The aquifer media in the upper reaches of the Yellow River are composed of coarse pebbles and sands, which favors the water circulation. Groundwater in the upstream is mainly recharged by the rainfall and melting ice and snow, as well as recharge from the Yellow River. Towards the downstream, the groundwater level depth becomes smaller than the upstream areas, reducing high evaporation of groundwater. The strong evaporation and concentration effects result in higher concentrations of Na^+ and Cl^- of groundwater in the lower reaches of the Yellow River.

The primary mechanisms affecting groundwater hydrochemical evolution were also analyzed with the Gibbs diagram (Gibbs 1970). As shown in Fig. 4, groundwater geochemistry in the study area is mainly affected by rock weathering, followed by evaporation. It indicates that the formation of chemical types of groundwater in this area is mainly influenced by natural factors, and geological factors have a great influence on it. In the meantime, the groundwater level depth is small, which makes the groundwater evaporation strong. This is consistent with the results of the Piper diagram.

Table 3 Total content of pollutants in groundwater in the study area

Pollutants	Number of samples	Unit	Minimum value	Maximum value	Average value	Distribution form
Fluoride	144	mg/L	0.17	5.10	0.81	Lognormal
Manganese	144	mg/L	0.05	4.20	0.40	Lognormal

Table 4 Statistics of water quality parameters

Parameters	Number of samples	Units	Maximum	Minimum	Average	Standards	Exceeding standard rate
pH	144	–	8.51	7.53	7.93	6.5–8.5	0.69%
TDS	144	mg/L	5240.00	252.00	1192.79	1000	45.83%
TH	144	mg/L	2302.00	145.00	616.39	450	72.92%
Na^+	144	mg/L	1125.00	19.90	167.68	200	25.00%
K^+	144	mg/L	48.30	2.09	11.04	12*	27.08%
Ca^{2+}	144	mg/L	511.00	26.10	119.02	200*	8.33%
Mg^{2+}	144	mg/L	316.00	6.08	78.27	150*	5.56%
Cl^-	144	mg/L	1808.00	17.70	182.03	250	17.36%
SO_4^{2-}	144	mg/L	2113.00	43.20	373.68	250	54.17%
HCO_3^-	144	mg/L	726.00	97.60	396.93	600*	4.86%
NO_3-N	144	mg/L	21.00	0.56	14.56	20.0	22.22%
NO_2-N	144	mg/L	1.89	9.13×10^{-4}	0.06	1.00	1.39%
NH_4-N	144	mg/L	8.711	0.012	0.105	0.50	2.08%
F^-	144	mg/L	5.10	0.17	0.81	1.0	21.53%
Mn	144	mg/L	4.20	0.05	0.40	0.10	50.00%

* World Health Organization (WHO) standards (WHO 2017)

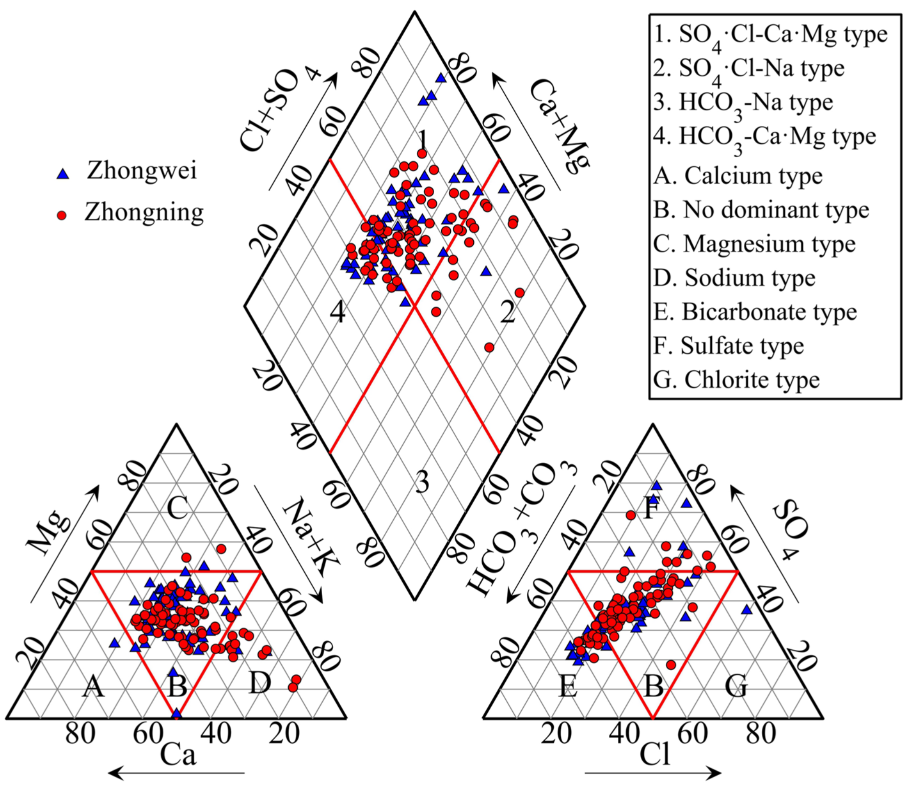


Fig. 3 Piper diagram of groundwater samples

Fluoride and Manganese

F^- is a trace element that is essential and significant to human. Too high or too low F^- content in the environment may cause the imbalance of F^- content in human body, which is harmful to health (Hong et al. 2018). Especially, excessive F^- intake can accumulate a large amount of F^- in hard tissues such as bones and teeth, leading to skeletal fluorosis and dental fluorosis. Drinking water with F^- content higher than 1.5 mg/L for a long time can easily lead to dental fluorosis (Adewole et al. 2021). If the content of F^- in water is higher than 4 mg/L, it can lead to bone spur,

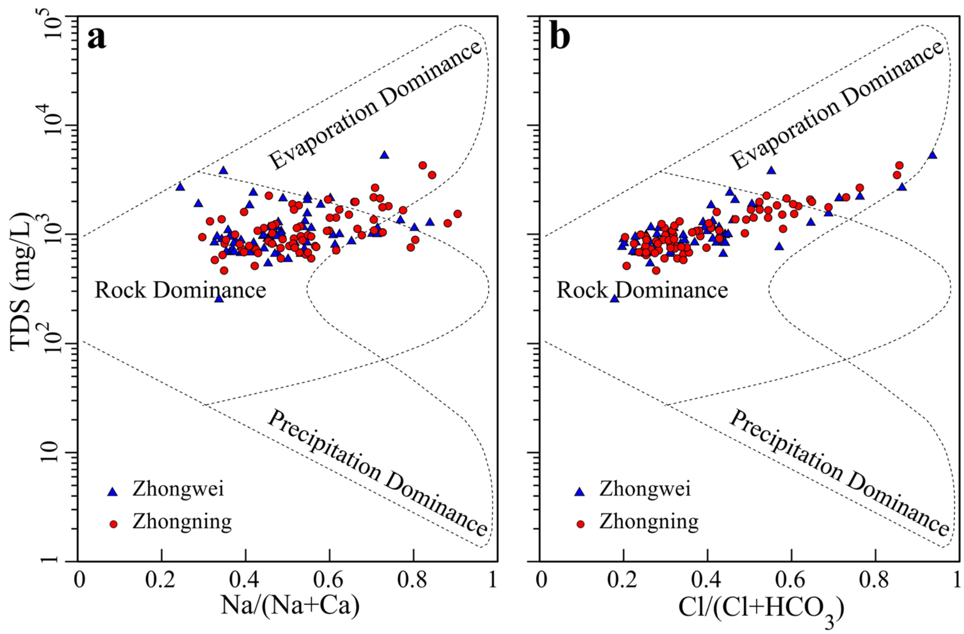


Fig. 4 Gibbs diagram of groundwater samples

bone sclerosis, osteoporosis and skeletal fluorosis (Deepanjan et al. 2021). Excessive F^- intake can also have negative effects on human intellectual development, reproductive hormone levels, digestion and cardiovascular system in varying degrees (He et al. 2020a,b). Groundwater with a high concentration of F^- is widely distributed in the world, particularly in semi-arid and arid areas (Wu et al. 2015). More than 70 million people around the world suffer from fluorosis (Jadhav et al. 2015). The impact of high F^- groundwater on human health in China is very serious (He et al. 2020b, 2021). The high F^- groundwater in the Loess Plateau can cause non-carcinogenic effects on human, and in these areas, the provision of low F^- drinking water is beneficial to people's health (Wu and Sun 2016; Li 2016).

Mn is a trace element needed by human body for normal metabolism, but excessive intake of Mn can affect children's intellectual function and impair their intellectual development (Bouchard et al. 2011). When people drink groundwater directly, heavy metal such as Mn will accumulate in the human body, causing serious damage to human tissue and mechanism, especially to liver, lungs and other organs (Chen et al. 2015). Further, Mn is not biodegradable, so it is very difficult to eliminate the Mn pollution in the environment.

Among the 144 samples in the survey region, F^- in 31 samples and manganese concentration in 72 samples exceed the standard limit (Table 4). This statistical result indicates that contaminant of F^- and Mn is serious, and exposure to the contaminated groundwater may be risky to local people. The distributions of F^- and Mn in the study area were investigated (Fig. 5). As shown in Fig. 5, the contents of F^- and Mn in the lower reaches of the Yellow River are in general higher than that in the upper reaches of the Yellow River. Researchers have shown that the sharp decline in the groundwater level around the water source areas caused by groundwater exploitation can aggravate the dissolution of Mn and fluoride-bearing minerals (Guo et al. 2018). The increase of F^- and Mn concentration from the upstream to the downstream may be related to the development and utilization of local groundwater resources. The specific hydrogeological settings of the study area such as the fine lithology, flat terrain, and small hydraulic gradient can also make it easier to enrich Mn in groundwater in the downstream areas (Khozyem et al. 2019). In the lower reaches of the Yellow River, the hydraulic gradient is small and groundwater flow is slow, which is easy to form a reducing environment and is conducive to the dissolution of Mn. The reduction environment can also promote the decomposition of organic matter, produces CO_2 and H_2O, and increases the content of HCO_3^-, which promotes the gathering of F^- (Li et al. 2014b).

To further understand the correlation of F^- and Mn with other physicochemical parameters, Pearson correlation was used (Table 5). The Pearson correlation matrix reflects that there is a positive correlation between F^- and Na^+ ($r=0.272$) and pH ($r=0.295$), and a negative correlation between F^- and Ca^{2+} ($r=-0.037$) (Table 5). Although they are not significant, it can be inferred that F^- will enrich in alkaline groundwater, this has been confirmed by Li et al. (2019b). Under alkaline conditions, the exchangeable F^- from fluoride-bearing minerals can be replaced by OH^-, thereby increasing the concentrations of F^- (Li et al. 2014b). Studies have revealed that the accumulation of F^- is positiely related to Na^+ and HCO_3^- in groundwater, while negatively related with Ca^{2+} (Wu et al. 2015; Singh et al. 2013). Ca^{2+} is significantly and positively correlated with SO_4^{2-} ($r=0.739$),

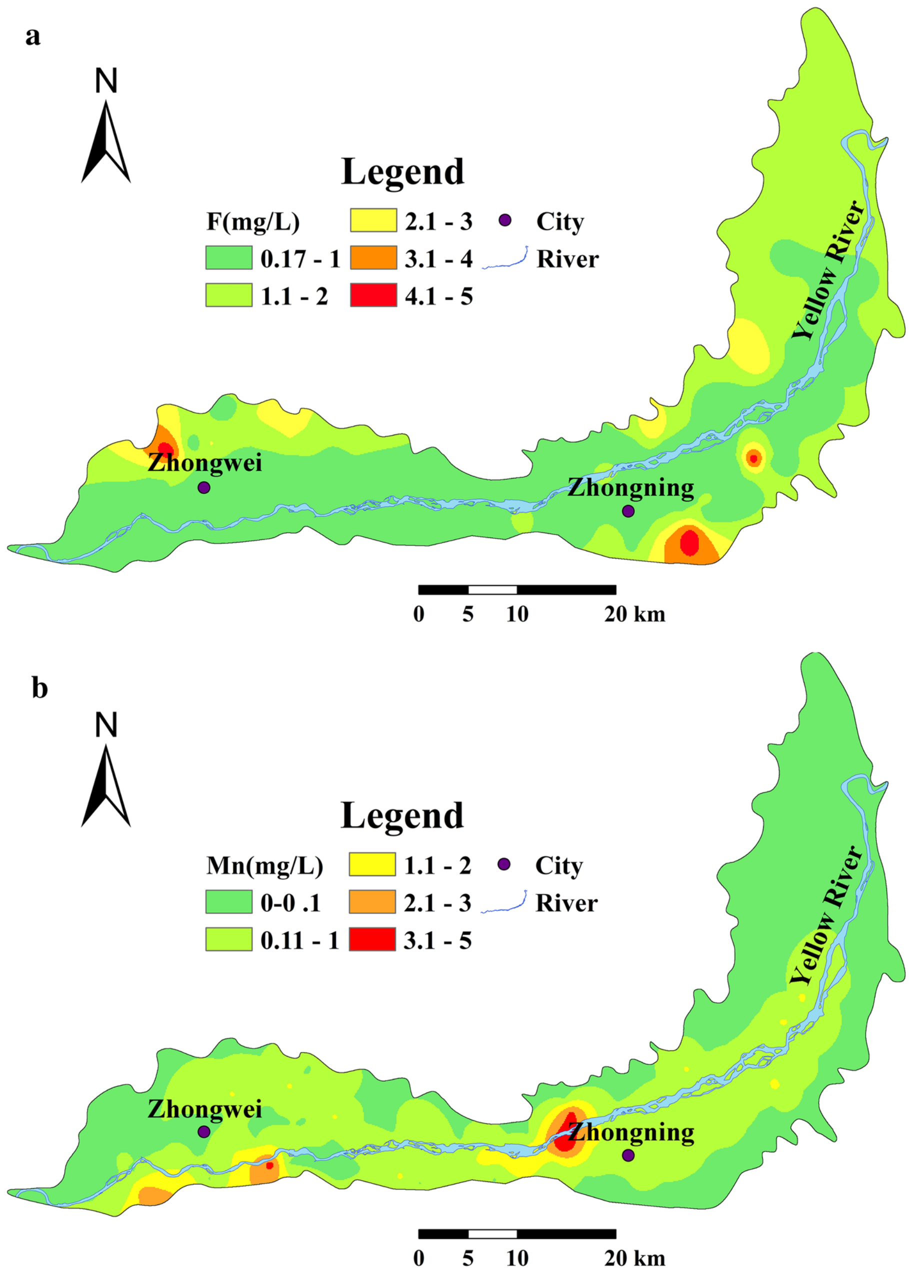


Fig. 5 Spatial distribution of **a** F^- and **b** Mn

and TH shows a significant positive correlation with Ca^{2+} ($r=0.898$) and SO_4^{2-} ($r=0.785$) (Table 5). This manifests that the dissolution of gypsum is a significant geochemical process in groundwater system (Li et al. 2019b). Due to the precipitation of fluorite (CaF_2), the concentration of F^- may decrease with the increase of Ca^{2+}. This is confirmed by the negative correltion between TH and F^- in Table 5.

It has been revealed that the concentration of trace metals in the groundwater is mainly affected by the hydrogeological structure of the aquifer and the chemical environment (Carretero and Kruse 2015; Amiri et al. 2021; Snousy et al. 2021). The main source of Mn in groundwater may be manganese-bearing minerals in the stratum (Sharma et al. 2021). The formation of the study area contains manganese ore, which is the reason for the development of the local manganese industry. The manganese ore in the formation will dissolve under the influence of rock weathering. At the same time, the concentration of manganese in the groundwater can be affected by other factors. As demonstrated in Table 5, Mn and pH are negatively correlated ($r=-0.277$). Environment with low pH value can promote the dissolution of Mn (Khozyem et al. 2019). It can be inferred that the accumulation of Mn in groundwater is related to pH. In addition, as mentioned previously, the decomposition of organic matter in the reducing environment produces CO_2 and H_2O, promotes the dissolution of calcite, dolomite and

Table 5 Pearson correlation matrix of physicochemical parameters

	K^+	F^-	HCO_3^-	Na^+	pH	Ca^{2+}	Mg^{2+}	NH_4^+	Cl^-	SO_4^{2-}	NO_3^-	NO_2^-	Mn	TDS	TH
K^+	1.000	0.148	**0.347**	**0.286**	−0.102	**0.307**	**0.402**	**0.356**	**0.325**	**0.250**	0.194	0.045	0.196	**0.373**	**0.391**
F^-		1.000	−0.053	**0.272**	**0.295**	−0.037	0.042	−0.041	0.121	0.190	0.003	−0.027	−0.144	0.173	−0.007
HCO_3^-			1.000	−0.171	**−0.342**	−0.092	0.193	**0.257**	−0.149	**−0.269**	0.092	0.103	**0.315**	−0.107	0.073
Na^+				1.000	0.102	**0.405**	**0.547**	0.080	**0.932**	**0.778**	0.078	-0.004	−0.014	**0.885**	**0.470**
pH					1.000	**−0.379**	**−0.231**	−0.113	0.001	−0.082	−0.143	-0.162	**−0.277**	−0.107	**−0.345**
Ca^{2+}						1.000	**0.617**	0.151	**0.539**	**0.739**	**0.309**	0.019	0.180	**0.730**	**0.898**
Mg^{2+}							1.000	0.039	**0.604**	**0.748**	**0.391**	0.055	0.191	**0.809**	**0.876**
NH_4^+								1.000	0.187	-0.010	−0.074	−0.023	0.156	0.095	0.106
Cl^-									1.000	**0.731**	0.092	0.014	0.003	**0.887**	**0.590**
SO_4^{2-}										1.000	0.133	-0.027	0.129	**0.930**	**0.785**
NO_3^-											1.000	0.154	-0.113	**0.284**	**0.393**
NO_2^-												1.000	0.049	0.016	0.045
Mn													1.000	0.109	0.213
TDS														1.000	**0.813**
TH															1.000

Bold numbers indicate that the correlation is significant at the 0.01 level (2-tailed)

Underlined numbers indicate that correlation is significant at the 0.05 level (2-tailed)

Mn minerals in the formation, and produces HCO_3^-. This is confirmed by the positive correlation of Mn with Ca^{2+} ($r=0.180$), Mg^{2+} ($r=0.191$) and HCO_3^- ($r=0.315$). The reducing environment of the groundwater reduces the Mn^{4+} in the groundwater to Mn^{2+} (Weng et al. 2007). Therefore, the reduction and dissolution of manganese ore increase the concentration of manganese in the groundwater. It can be considered that the enrichment of manganese is affected by local natural factors, such as formation lithology and hydrochemistry environment.

Water Quality Analysis and Assessment

In this procedure, pH, Na^+, Cl^-, SO_4^{2-}, NO_3^-, NO_2^-, F^-, Mn, TDS, and TH were selected for calculating the value of EWQI. The results show that EWQI of groundwater ranges from 31.66 to 316.30. For the 144 groundwater samples, 75 samples are classified as medium quality water (Class III) which can be used for domestic purpose with caution and pretreatment, accounting for about 52.0% of the total groundwater samples. Poor quality water samples (Class IV) account for 25.7% of the total sample, and these water samples are not suitable for drinking and may be used for recreational purpose. The good quality (Class II) and very poor quality (Class V) groundwater accounts for 13.2% and 9.0%, respectively. Unfortunately, there is no excellent quality groundwater found in this study. Overall, medium and poor quality groundwater is prevalent in the entire study area.

To aid the management of groundwater quality, the groundwater quality distribution map was generated on the basis of the EWQI results (Fig. 6). Figure 6 shows that groundwater quality of the survey region is generally poor, but the groundwater quality in the upstream area is better than that in the downstream area, and the water quality in the middle part of the upstream area is better than that in the boundary areas of the upstream. Statistically, around 40% of the water samples in the downstream belong to Class IV and Class V, while in the upstream the percentage is 29.0%. Groundwater quality in Zhongning is worse than that in Zhongwei, because there are more industrial plants in and around Zhongning. Groundwater quality in the rural areas and wetlands is better than that in the urban areas and agricultural areas.

As mentioned previously, Weining Plain is a traditional agricultural region, and it has undergone rapid industrial development in recent years. The infiltration of agricultural irrigation water and the use of agricultural fertilizers will affect the quality of groundwater (Wu et al. 2015). Mineral deposition and strong evaporation in the lower reaches of the region will also deteriorate the water quality. Therefore, to protect the groundwater quality it is suggested that residents and related enterprises should make rational use of groundwater to avoid groundwater overexploitation, and reduce the excessive use of agricultural chemical fertilizer. In addition, groundwater level increase during the irrigation seasons should be controlled to reduce the groundwater evaporation and concentration effects. More importantly, the supervision of environmental protection in the area must be strengthened to eliminate illegal activities such as sewage discharge and industrial waste disposal.

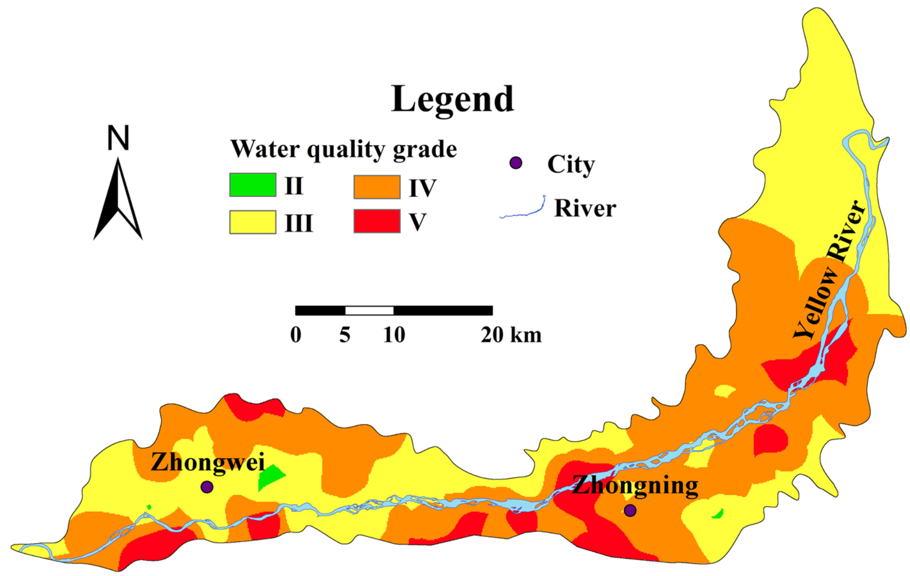


Fig. 6 Zoning of groundwater quality

Table 6 Deterministic health risks

Statistics	Adults					Children				
	HQ_o		HQ_d		HQ_{total}	HQ_o		HQ_d		HQ_{total}
	F^-	Mn	F^-	Mn		F^-	Mn	F^-	Mn	
Min	0.11	0.01	3.48×10^{-4}	2.05×10^{-5}	0.14	0.92	0.01	1.02×10^{-3}	5.99×10^{-5}	1.15
Max	3.26	1.07	1.04×10^{-2}	3.44×10^{-3}	3.30	27.51	2.27	3.06×10^{-2}	1.01×10^{-2}	27.61
Mean	0.52	0.10	1.66×10^{-3}	3.21×10^{-4}	0.62	4.38	0.21	4.86×10^{-3}	9.41×10^{-4}	4.59

Deterministic Health Risk Assessment

Deterministic health risk assessment was conducted, and F^- and Mn in groundwater were considered in this assessment. HQ values range from 0.14 to 3.30 (0.62 on average) and from 1.15 to 27.61 (4.59 on average) for adults and children, respectively (Table 6). There are 17 samples with HQ value exceeding 1 for adults and 144 samples with the HQ value higher than 1 for children. This suggests that both adults and children face no-carcinogenic risk, but the risk faced by children is higher than that faced by adults, because children generally have smaller weight than adults (Li et al. 2016b). In addition, the main exposure pathway to harm people's health is the drinking water intake exposure pathway, and the risk to human health caused by dermal contact is very small and sometimes may be ignored. Similar conclusions have appeared in the research of Wu and Sun (2016) and Li et al. (2016b).

Different pollutants have different effects on human health. The HQ of F^- ranges from 0.11 to 3.26 (0.52 on average) for adults, and that of Mn ranges from 0.01 to 1.07 with a mean of 0.10, indicting higher risk of F^- than Mn for adults. This also applies to children. In addition, the average HQ of F^- exceed 1, manifesting that the health risk of F^- cannot be ignored. To summarize, F^- intake through drinking water is the main source of health risk for both adults and children. Therefore, much attention should be paid to the health risk induced by F^- in drinking water and the risk of excessive F^- in drinking water should be controlled.

Probabilistic Health Risk

The Monte Carlo stochastic method was used to simulate the groundwater health risk in the study area. With respect to the total risk at the 95% confidence level, it ranges from 0.23 to 3.09 (0.92 on average) for adults, and ranges from 1.54 to 22.67 (6.84 on average) for children (Fig. 7). Figure 7 shows that the health risk probabilities for both population groups conform to a lognormal distribution, but children face greater health risks than adults. This conclusion is in agreement with that achieved from the deterministic health risk assessment. Simulation results show that the probability

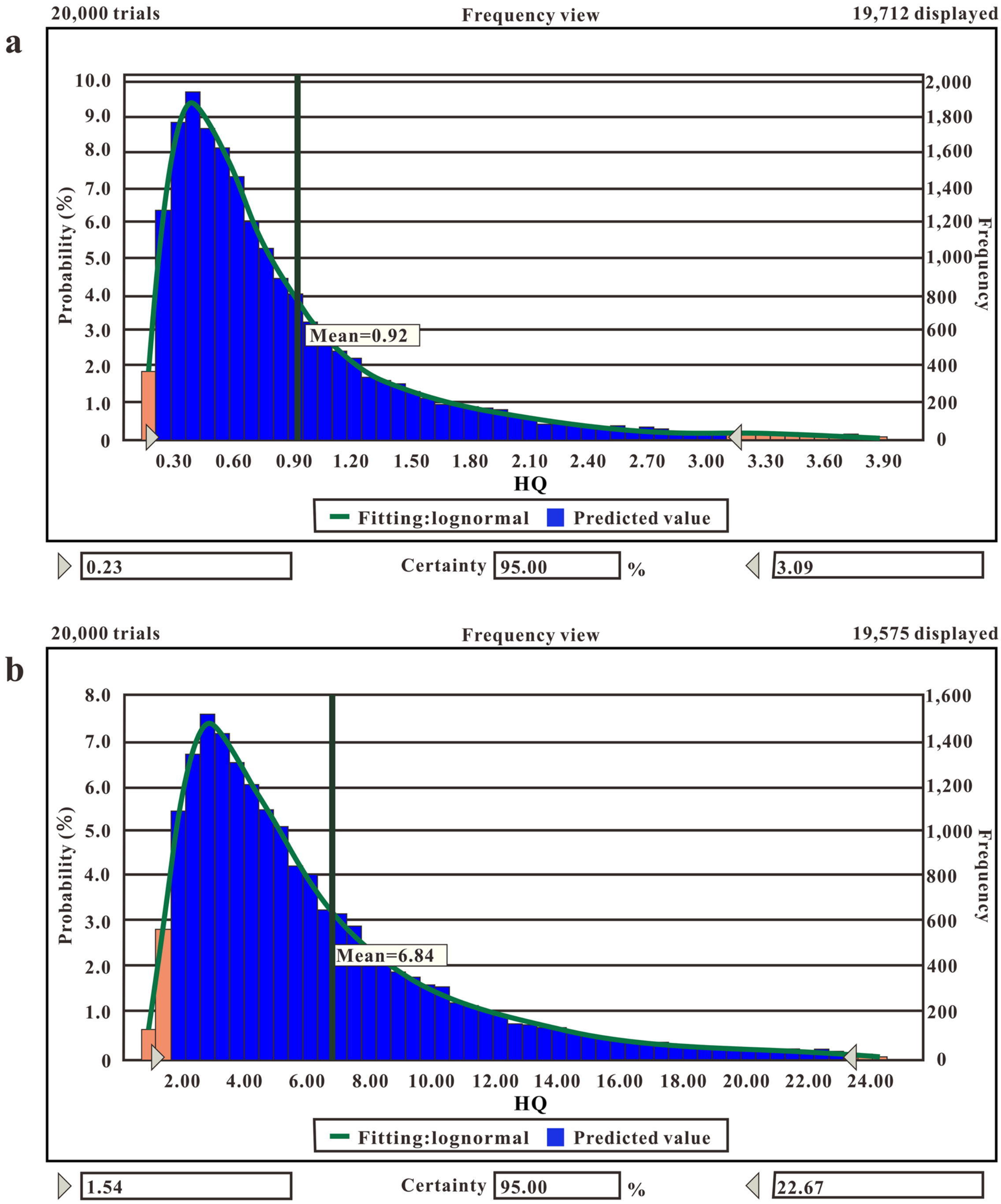


Fig. 7 Simulation results of total hazard quotient for **a** adults and **b** children

of HQ exceeding the threshold for children and adults are 95.0% and 30.0%, respectively. The probability of HQ exceeding the threshold can be obtained by summing the probability values corresponding to HQ exceeding 1.

The health risk simulation results of F^- and Mn for different population groups via different exposure pathways are shown in Fig. 8. Figure 8a, b show that the health risks caused by F^- and Mn for adults through oral intake averaged 0.63 and 0.29, respectively. F^- is the major pollutant causing health risk to the adults. As shown in Figs. 8a-d, compared with dermal contact exposure pathway, oral intake is the main exposure pathway for F^- and Mn to impose adverse effect on the health of adults. The health risks caused to adult residents are less than 1, indicating the health risk is acceptable to adults. Through the contrast of Figs. 8e-h, it can be found that he health risks caused by the ingestion of

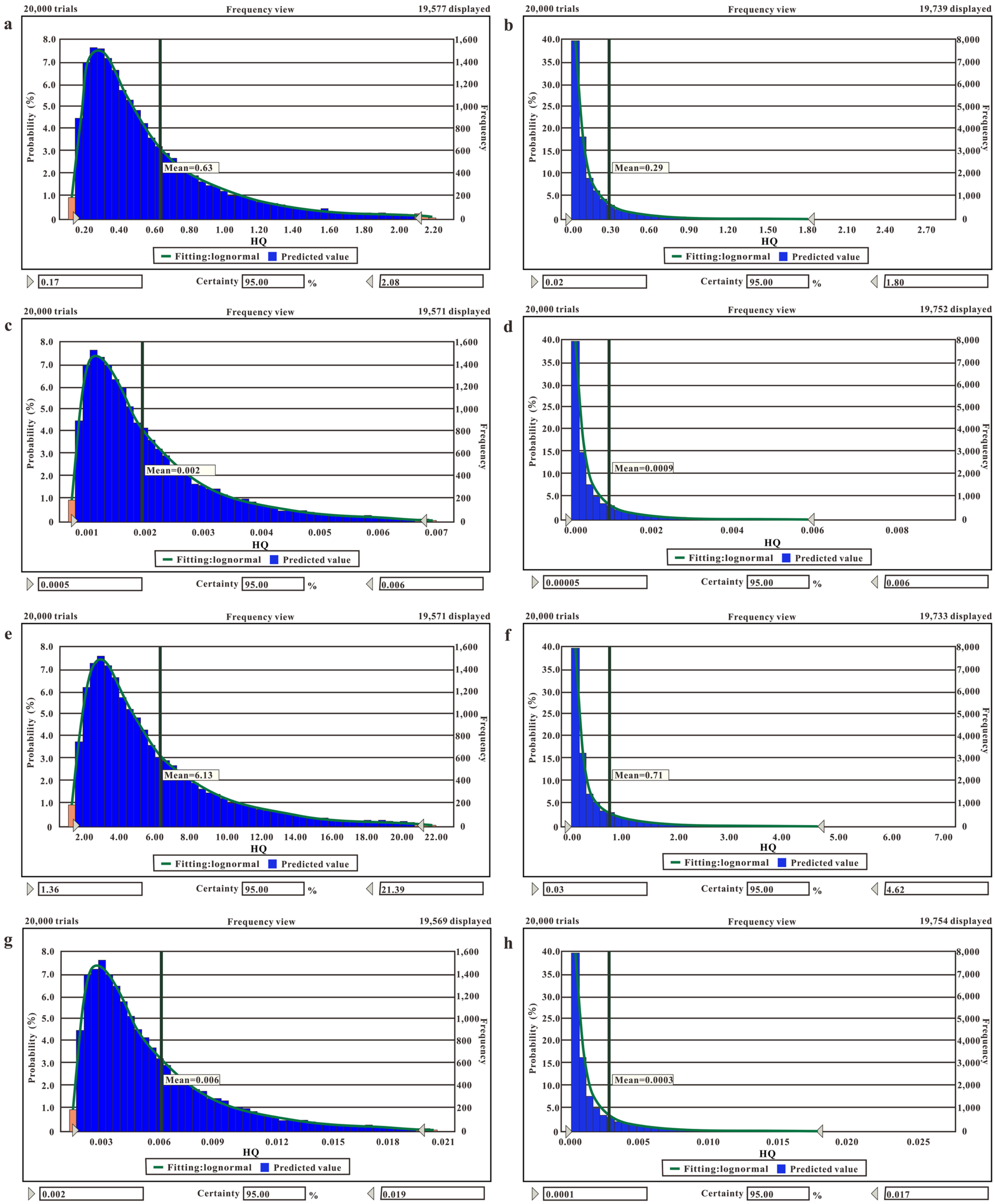


Fig. 8 Health risk simulation results under different conditions. HQ_o of **a** F^- and **b** Mn for adults, HQ_d of **c** F^- and **d** Mn for adults, HQ_o of **e** F^- and **f** Mn for children, HQ_d of **g** F^- and **h** Mn for children

Table 7 Results of parameter sensitivity analysis

Random variable	Sensitivity degree (%)
Concentration of fluoride in groundwater	79.8
Drinking water rate	14.2
Concentration of manganese in groundwater	5.5
Body weight	−0.6

F^- and Mn through the drinking water pathway is 6.13 and 0.71, respectively, to children, and the health hazards that children are exposed to F^- and Mn by skin are 0.006 and 0.0003, respectively. Therefore, the health risk through dermal contact is acceptable to children. However, children face greater health risks than adults, and this validates the results of Fig. 7. Therefore, for the control of non-carcinogenic risk, the concentrations of contaminants especially the F^- concentration should be reduced. At the same time, the spatial distribution of F^- must be well understood to control the high F^- groundwater in the region.

As shown in Table 6, Figs.7 and 8, similar results can be found in deterministic and probabilistic health risks. F^- is more harmful to health than Mn, oral intake is the major exposure pathway, and children are generally facing greater health risk than adults. The advantage of the probabilistic simulation is that it can visually show the probabilities of different simulated values and can use limited data to get more comprehensive results.

Sensitivity analysis can be used to analyze the sensitivity of HQ to various pollutants and exposure parameters. Table 7 shows that the sensitivity of non-carcinogenic risk to uncertain factors is in the following order: concentration of F^- > drinking water rate > concentration of Mn > body weight. Among them, HQ is positively correlated with the concentration of F^-, concentration of Mn and daily drinking water rate, and is negatively correlated with human body weight. The higher the absolute value of sensitivity indicates greater influence on the total hazard quotient. This shows that drinking more water will yield greater health risk, and people with smaller body weight will associate higher risk. Therefore, it is crucial to focus on preventing the health risks caused by pollutants to children.

Conclusions

This paper reported the occurrence and distribution of F^- and Mn in groundwater and the overall groundwater quality in the Weining Plain. The non-carcinogenic health risks induced by F^- and Mn via drinking and dermal contact exposure pathways were assessed using the deterministic and probabilistic models. The following conclusions can be summarized:

1. The groundwater samples mainly belong to the types of SO_4·Cl-Ca·Mg and HCO_3-Ca·Mg. The enrichment of F^- and Mn is related to the groundwater geochemical which is affected by both natural and human factors, including rock weathering, evaporation, agricultural and industrial activities.
2. The results of EWQI reveal that most of the samples are of medium and poor quality. Upstream areas have better groundwater quality than the downstream areas. Taking relevant measures will benefit for achieving better groundwater quality.
3. Both deterministic and probabilistic health risk results show that compared with Mn, F^- is the main pollutant causing health risk. Drinking water intake causes greater health risk than dermal contact. Children face greater health risk than adults. The results of stochastic simulation can reflect the health risks more comprehensively.
4. The results of sensitivity analysis reveal that particular attention should be paid to reducing the concentrations of F^- and Mn in drinking water, and the health of children needs more attention.

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Author Contribution Leining Liu developed the suitable methodology, and was involved with the writing of the manuscript. Jianhua Wu conducted the investigation and data collection, provided supervision in the presented work, and involved in the writing and editing of the manuscript. Song He and Lei Wang helped in methodology selection and visualization, and participated in editing of the earlier versions of the manuscript.

Data Availability All the data used for the study appear in the article.

Declarations

Conflict of interest The authors declared no conflict of interest.

Consent to Participate and Publish All authors agreed to participate and publish.

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