Shallow groundwater quality and health risk assessment of fluoride and arsenic in Northwestern Jiangsu Province, China

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Abstract

Assessing groundwater quality is critical to regional water resource conservation and human health safety, especially in areas with co-existence of toxic constituents fluoride (F^-) and arsenic (As). In this study, fourteen groundwater samples were collected in Feng County, Northwestern Jiangsu Province to identify dominant contaminants and their spatial distribution and health risk. The composition and variation characteristics of major ions (K^+, Na^+, Ca^{2+}, Mg^{2+}, Cl^-, SO_4^{2-}, HCO_3^-, and NO_3^-) and trace elements (F^-, As, and Mn) were analyzed. The hydrochemical results revealed that high F^- groundwater was mainly distributed in the northern areas whereas As-riched groundwater was primarily distributed in southern areas. Notably, over 85.7% and 21.4% of the shallow groundwater samples exceeded the drinking water quality standard of 1.5 mg/L for F^- and 10 μg/L for As, respectively. Based on the water quality index (WQI) appraisal result, 71.4% of the groundwater in the study area is classified as "poor", and thus unsuitable for drinking directly. We assessed the human non-carcinogenic health risk of F^- ($HQ_{Fluoride}$) and As ($HQ_{Arsenic}$) and the carcinogenic health risk of As ($CR_{Arsenic}$). The calculated hazard quotient (HQ) for F^- indicated nearly all groundwater samples have an unacceptable risk (HQ > 1) for each age group. However, $HQ_{Arsenic}$ values revealed that 28.6%, 21.4%, 21.4%, and 21.4% of groundwater samples posed potential non-carcinogenic health risks for infants, children, females, and males, respectively. The calculated results of $CR_{Arsenic}$ showed that 0%, 21.4%, 28.6%, and 28.6% of groundwater samples posed unacceptable health risks (CR > 1.0×10^{-4}) to infants, children, females, and males, respectively. The groundwater irrigation suitability assessment results showed that 21.4% of samples were doubtful to unsuitable for irrigation, and 85.7% owed magnesium hazards. The findings of this study will assist policymakers in formulating proper remedial policies and mitigation strategies to ensure the safety of drinking and irrigation water.

Keywords Groundwater quality · Drinking and irrigation · Water quality index · Fluoride · Arsenic · Health risks

Introduction

Worldwide, over 2.5 billion people rely on groundwater as a reliable freshwater source for drinking (Liu et al. 2019a, b; Mahmud et al. 2020). Simultaneously, about 20% of agricultural irrigation and 15% of industrial water also depend on groundwater (Adimalla et al. 2018). In 2015, the United Nations made a universal call to ensure that all people have "clean drinking water and sanitation facilities" till 2030 in Sustainable Development Goal 6 (SDG-6) (Ruidas et al. 2024). However, an increasing number of countries and regions are experiencing serious water security problems due to natural environmental changes and intensive anthropogenic activities (industrial production, agricultural activities, wastewater discharge, etc.) (Xu et al. 2018; Liu et al. 2020b; Tanwer et al. 2023). In recent decades, human health risks triggered by groundwater contamination have received

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much attention in many countries, particularly in developing countries (Pal et al. 2022; Jaydhar et al. 2023; Ruidas et al. 2024). India and China are the worldwide hotspots in terms of fluoride (F^-), nitrate (NO_3^-), and arsenic (As) contamination in groundwater aquifers (Liu et al. 2015a, b; Ruidas et al. 2022, 2023a; Jaydhar et al. 2023). According to Cao et al. (2023), 260 million people are reportedly exposed to endemic fluorosis in 25 countries worldwide, with China being one of the most vulnerable countries (> 60 million people). It is reported that the accumulation of F^- in groundwater primarily resulted from the weathering of fluorine-hosting minerals like fluorite, topaz, and apatite (Bhattacharya et al. 2017; Enalou et al. 2018; Elumalai et al. 2019; Xu et al. 2023). Long-term consumption of high-fluoride groundwater (> 1.5 mg/L) may cause dental fluorosis, osteosclerosis, and mental retardation in children, despite fluoride's role as an essential trace element (Jia et al. 2018; Hossain and Patra 2020; Jiang et al. 2022). Notably, the prolonged consumption of arsenic-containing groundwater (> 10 μg/L) can result in several health problems such as arsenic poisoning, cancer, and increased all-cause mortality (Pincetti-Zúniga et al. 2020; Podgorski and Berg 2022).

It is reported that natural processes or anthropogenic input of F^- and As have a significant impact on human health and crop growth (Bhattacharya et al. 2017; Ruidas et al. 2023b, 2024). Irrigating crops and vegetables with F^- and As contaminated groundwater is a key transfer route that F^- and As entry into the human food chain via the water-soil-plant-food system (Huang et al. 2006; Bhattacharya et al. 2010). Therefore, the combined effect of F^- and As in groundwater will enhance the probability of related diseases including chronic arsenic poisoning and endemic fluorosis (Bhattacharya et al. 2020; Ruidas et al. 2023a, 2024). A detailed scientific assessment of groundwater quality for various uses is imperative to guide water resource management. For example, Ruidas et al. (2024) and Pal et al. (2022) have been working on groundwater F^- and As contamination along with associated health risks individually and combinedly in Bangladesh and India by utilizing various machine learning (ML), Random forest (RF), Multi-collinearity analysis, and GIS-based techniques.

Feng County, located in northwestern Jiangsu Province, is highly contaminated by F^- since the Yellow River flood sediment contains a huge amount of F^- to be released into the groundwater aquifer (Wang et al. 2023a; Chen et al. 2023). Until now, rural area still relies on groundwater for industrial, agricultural, and domestic use although 5% of the residents have suffered from skeletal fluorosis and 70% from dental fluorosis (Zhu 2007). Wang et al. (2023a) mainly focus on the groundwater cycle and hydrochemical evolution processes in Feng County using stable isotope and geochemical modeling. Chen et al. (2023) integrated self-organizing maps with the Monte Carlo method to evaluate groundwater fluoride exposure and related health risks in the Fengpei plain. Further, Zhou et al. (2023) reported that the mean F^- concentration reached 2.81 mg/L in the shallow groundwater of Feng County. Previous studies mainly reported natural processes and anthropogenic factors affecting the hydrochemical characteristics of high-fluoride aquifers. However, little literature has been reported in this region regarding the co-existence of toxic constituents F^- and As and their associated health risks. Additionally, groundwater suitability for drinking and irrigation purposes was also not studied.

Based on the above context, a systematic and comprehensive evaluation of groundwater quality in the study area is urgently needed to ensure the local water resources safety. The main objectives of this study are to (1) evaluate the groundwater quality and its spatial variation based on GIS; (2) assess the groundwater suitability for drinking and irrigation purposes; and (3) determine the potential human health risks of F^- and As for different age groups. This study can provide an important foundation for formulating sustainable groundwater resource protection schemes in the study area and similar areas experiencing serious F^- and As contamination.

Study area

Feng County (116° 21′ 15″–116° 52′ 03″ E, 34° 24′ 25″–34° 56′ 27″ N) is located in northwestern Jiangsu Province with an area about 1450 km² (Fig. 1). This area is characterized by a semi-humid monsoon climate, with an average annual temperature of 15.3 °C, a maximum temperature of 38.3 °C, and a minimum temperature of − 9.9 °C. The average annual precipitation and evaporation from 1956 to 2020 were 733.82 mm and 1472.32 mm, respectively (Wang et al. 2023a). Evaporation is much greater than precipitation. The main surface water bodies in the study area include the Fuxin River and the Dasha River.

The study region belongs to the North China stratigraphic area geologically. The Quaternary strata are extensively developed and are approximately 270 m thick (Wang et al. 2023a). The thickness of the Quaternary Holocene strata is 11.0–15.0 m, mainly chalky sand and loamy soil. The thickness of Middle and Upper Pleistocene strata is about 130 m, and the thickness of Lower Pleistocene and Tertiary Pliocene strata is about 180 m. The bedrock is Tertiary, and the lithology is mainly red sandstone and conglomerate interbedded with colored shale, sandstone, and mudstone. There are three aquifers in the formation: the first aquifer is located above 50 m, with medium water-richness, mainly used for rural life and agricultural irrigation. The second aquifer (50–120 m) has low water-bearing and high mineralization. The third aquifer (100–200 m) has a strong water abundance
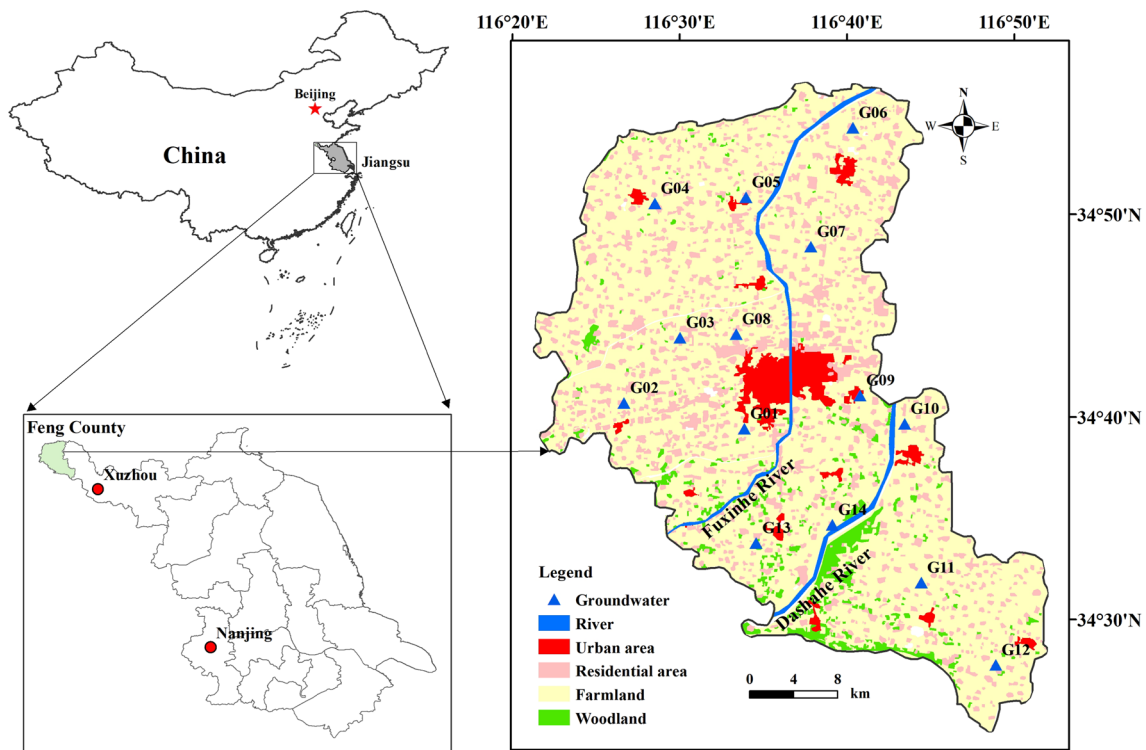
Fig. 1 Location of sampling sites and land use types in the study area

and good water quality, with a single well with a production rate of 70 m³/h, which is mainly used as urban industrial and domestic water.

Based on the land use data of the raster image in 2018 (30m × 30m), the land use types in the study area mainly included agricultural, construction, and other land uses (Fig. 1). The agricultural land was the highest accounting for 77.4% of the total area, followed by construction and other lands use with the proportion of 18.2% and 4.4%, respectively. As reported by Liu et al. (2020a), land use patterns may influence the distribution and transport of pollutants and have a major impact on groundwater quality.

Material and methods

Sample collection and testing

Groundwater samples from 14 shallow wells were collected to investigate the concentrations of major ions and trace elements in November 2021 (Fig. 1). Most groundwater samples were taken from wells for domestic and irrigation supplies with groundwater levels ranging from 33 to 40 m. A calibrated portable multi-parameter digital meter (HQ30d, HACH) was used to measure the pH and electrical conductivity (EC) of groundwater samples during the field investigation. The parameter values were recorded when the data on

the dashboard remained stable. After pumping for approximately 5 min, the bottles were rinsed 2–3 times with sample water before being placed in polyethylene sampling bottles. The samples were filtered using 0.45 μm microporous filters and stored at 4 °C before being analyzed. 50 ml groundwater samples were acidified to pH < 2 with ultrapure HNO_3^- to measure cations (K^+, Na^+, Ca^{2+}, Mg^{2+}). However, the other was kept without acidification for testing Cl^-, SO_4^{2-}, NO_3^-, F^-, As, and Mn.

The major cations (K^+, Na^+, Ca^{2+}, and Mg^{2+}) were measured by using an inductively coupled plasma optical emission spectrometer (Perkin-Elmer Optima 5300DV, USA). whereas major anions (Cl^-, SO_4^{2-}, NO_3^-, and F^-) were determined by ion chromatography (Thermo Fisher ICS600, USA). Alkalinity (HCO_3^-) was measured by using a titration method within 24 h of sampling. The As and Mn content of the groundwater samples was analyzed by inductively coupled plasma mass spectrometer ICP-MS (NexION 2000, Perkin Elmer, USA). The original dataset in this study is sourced from Wang et al. (2023a), except Mn and As.

To ensure the reliability of the results, the charge balance errors (CBE) of cation and anion were checked by Eq. (1). The acceptable ion balance errors in this study were within ± 10%.

$$CBE(\%) = \frac{\sum N_c - \sum N_b}{\sum N_c + \sum N_b} \times 100\% \tag{1}$$

where N_c and N_b represent the concentrations of anions and cations in meq/L, respectively.

Water quality index

The water quality index (WQI) approach assessed groundwater suitability for drinking purposes by integrating data sets (Verma et al. 2020; Gao et al. 2021; Tanwer et al. 2023). In this study, 14 key parameters (pH, EC, TDS, K^+, Na^+, Ca^{2+}, Mg^{2+}, Cl^-, SO_4^{2-}, HCO_3^-, NO_3^-, F^-, As, and Mn) were incorporated into the WQI to assess the quality of groundwater. All these chemical parameters have a direct and indirect influence on groundwater quality.

Based on its relative importance in human health assessment, each physicochemical parameter was assigned a specific weighting (2 to 5) (Hossain and Patra 2020; Piyathilake et al. 2022). In this study, the weights of SO_4^{2-}, NO_3^-, F^-, As, and Mn were assigned 5 due to their importance in water quality assessment (Table 1). The WQI calculation consists of the following three steps:

$$W_i = \frac{w_i}{\sum_{i=1}^{n} w_i} \tag{2}$$

$$Q_i = \frac{c_i}{s_i} \times 100 \tag{3}$$

$$WQI = \sum_{i=1}^{n} W_i \times Q_i \tag{4}$$

where W_i represents the relative weight of each water quality parameter; w_i is the weight of each water quality parameter; C_i is the concentration of each chemical parameter "i" in each sample (mg/L); S_i is the World Health Organization standard (WHO 2011) for each chemical parameter; Q_i is the quality rating scale of the chemical parameter "i", and "n" is the total number of parameters.

According to the WQI value, groundwater quality can be classified into the following five categories: "Excellent" (WQI < 50); "Good" ($50 \leq$ WQI < 100); "Poor" ($100 \leq$ WQI < 200); "Very poor "($200 \leq$ WQI < 300); "Unsuitable" (WQI $\geq$ 300).

Irrigation water quality evaluation

Excessive dissolved ions in irrigation water will have physical and chemical effects on plants and agricultural soil, thus reducing productivity. Therefore, it is critical to evaluate the suitability of groundwater for irrigation. In this study, irrigation assessment parameters such as sodium adsorption ratio (SAR), sodium percentage (%Na), residual sodium carbonate (RSC), permeability index (PI), and magnesium hazard (MH) were used to evaluate the suitability of irrigation water (Bian et al. 2018; Hou et al. 2023; Hasan et al. 2023; Taloor et al. 2024). These indexes were calculated by Eqs. (5)–(9), respectively, where all ion concentrations are expressed in meq/L:

Table 1 Statistical value of the physical and chemical indicators of groundwater samples in the study area

Chemical parameters	Groundwater				Parameters for WQI calculation		
	Min	Max	Mean	SD	WHO standards	Weight (w_i)	Relative weight (W_i)
pH	7.3	7.8	7.5	0.2	6.5–8.5	4	0.0755
EC (µS/cm)	729.0	3030.0	1667.4	695.3	1500	4	0.0755
TDS (mg/L)	391.7	2132.3	1041.2	561.0	1000	4	0.0755
K^+ (mg/L)	2.8	32.4	16.9	7.7	12	2	0.0377
Na^+ (mg/L)	51.1	547.8	263.6	171.6	200	3	0.0566
Ca^{2+} (mg/L)	29.8	95.5	56.9	22.9	200	2	0.0377
Mg^{2+} (mg/L)	24.7	103.8	59.2	24.7	150	2	0.0377
Cl^- (mg/L)	54.5	506.4	173.8	119.2	250	4	0.0755
SO_4^{2-} (mg/L)	8.1	945.7	262.8	283.4	250	5	0.0943
HCO_3^- (mg/L)	195.2	585.6	398.9	93.5	500	3	0.0566
NO_3^- (mg/L)	0.9	88.5	24.9	31.9	50	5	0.0943
F^- (mg/L)	1.3	5.5	3.4	1.4	1.5	5	0.0943
Mn (µg/L)	0.3	1536.4	250.1	437.1	100	5	0.0943
As (µg/L)	0.1	72.4	8.6	19.2	10	5	0.0943

$$SAR = \frac{Na^+}{\sqrt{\frac{(Ca^{2+}+Mg^{2+})}{2}}} \qquad (5)$$

$$Na\% = \frac{(Na^+ + K^+) * 100}{(Ca^{2+} + Mg^{2+} + Na^+ + K^+)} \qquad (6)$$

$$RSC = (HCO_3^- + CO_3^{2-}) - (Ca^{2+} + Mg^{2+}) \qquad (7)$$

$$PI = \frac{\left(Na^+ + \sqrt{HCO_3^-}\right) \times 100}{\left(Ca^{2+} + Mg^{2+} + Na^+\right)} \qquad (8)$$

$$MH = \frac{Mg^{2+}}{Ca^{2+} + Mg^{2+}} \times 100 \qquad (9)$$

Health risk assessment

Non-carcinogenic risk

Non-carcinogenic risk (NCR) was characterized by the hazard quotient (HQ) and HQ > 1 indicates the presence of non-carcinogenic risk (Nawale et al. 2021; Wang et al. 2023b). In this study, $HQ_{Fluoride}$ and $HQ_{Arsenic}$ represent the non-carcinogenic health risks from F^- and As contamination, respectively. In general, ingestion and dermal are the primary pathways of human exposure to contaminants (Li et al. 2021b; Liu et al. 2022). Given the potential hazards from oral intake are greatly higher than those from dermal contact (Liu et al. 2022), only the oral ingestion exposure pathway is considered to assess the potential health risks in this study. Fluoride exposure in different age groups was calculated using Eq. (10) (Jaydhar et al. 2022). HQ can be calculated by dividing the estimated daily intake (CDI) by the safe dose (RfD), as expressed in Eq. (11).

$$CDI = \frac{C_w \times IR \times ED \times EF}{BW \times AT} \qquad (10)$$

$$HQ = \frac{CDI}{RfD} \qquad (11)$$

where CDI indicates the daily intake dose from ingestion in drinking water (mg/kg/day); C_w is the contaminant (F^- or As) level in groundwater (mg/L); IR is the intake rate (L/day; 0.65 L/day for infants, 1.5 L/day for children, and 2.66 L/day for adults) (Xiao et al. 2022a; Pasupuleti et al. 2022); ED is the exposure duration (years; 0.5, 6, 30, and 30 years

for infants, children, females, and males, respectively) (Xiao et al. 2022b; Zeng et al. 2023); EF is exposure frequency (days/year; EF = 365 days/year); BW indicates average body weight (kg; 6.94 kg, 25.9 kg, 52 kg and 73 kg for infants, children, female, and male, respectively) (Liu et al. 2022; Hu et al. 2022; Zeng et al. 2023); AT is the average exposure time (AT = 365 × ED); RfD denotes the reference dose (mg/kg/d), F^- is 0.06 mg/kg/d (Zeng et al. 2023; Chen et al. 2023) and As is 0.0003 mg/kg/d (Zhang et al. 2019; Qaiser et al. 2023).

Carcinogenic risk

Arsenic (As) is a typical carcinogen in groundwater that affects the public health of inhabitants through oral exposure pathway (Rapant and Krčmová 2007; Bhattacharya et al. 2020). Chronic exposure to As-rich groundwater can lead to carcinogenic risks (CR), such as skin cancer and kidney cancer (Zhu et al. 2023). Therefore, As was selected to assess the carcinogenic risk of groundwater in the study. The carcinogenic risk value of As was calculated using the following formula:

$$CR_{Arsenic} = CDI_{oral} \times SF_{oral} \qquad (12)$$

where SF indicates the slope factor for carcinogenic contaminants $(mg/kg/day)^{-1}$ (Zhu et al. 2023). The SF value for As was set at 1.5 $(mg/kg/day)^{-1}$ in this study (Li et al. 2021a; Rashid et al. 2023). For carcinogenic effects, AT = 70 × 365 (25550 days) because 70 years represent the adverse human health effects of As on a person's lifetime (Zhang et al. 2019; Yuan et al. 2023; Rashid et al. 2023). The carcinogenic risk is an acceptable level if CR values of As via ingestion pathway between 1×10^{-6} and 1×10^{-4} (USEPA 2004); while an unacceptable range when the CR values are larger than 1×10^{-4} (Kumar et al. 2017; Rahman et al. 2022; Goswami et al. 2022).

Geostatistical analysis

The Geographic Information System (GIS) techniques have been widely adopted to identify the spatiotemporal variation of groundwater quality (Ruidas et al. 2021, 2023a; Jaydhar et al. 2022; Biswas et al. 2023). In this study, the spatial distribution map of hydrochemical constituents and water quality index was generated by the inverse distance weighted (IDW) interpolation method in ArcGIS 10.2 software (Rahman et al. 2022). The IDW method estimates the values of an attribute at unsampled points using the distance-weighted average of all data of the neighboring sampled sites (Atikul Islam et al. 2017; Biswas et al. 2023).

Results and discussion

Groundwater quality assessment

Hydrochemical characteristics and spatial distribution

The summary statistics of the physical and chemical parameters in the groundwater samples are shown in Table 1. The pH varied from 7.3 to 7.8 with an average value of 7.5, indicating groundwater was slightly alkaline. As specified by WHO (2011), the pH of all samples is below the limit (6.5–8.5). Total dissolved solids (TDS) varied from 391.7 to 2132.3 mg/L, with a mean value of 1041.2 mg/L. Approximately 42.9% of the water samples exceeded the permissible limit of 1000 mg/L. As shown in Fig. 2a, the TDS exceedance points were mainly distributed in the northern of the study area, which is an important industrial zone, implying that the discharge of industrial pollutants will inevitably lead to groundwater contamination.

The trace elements can be effective in determining the potential contamination status of groundwater. It is noteworthy that F^- concentrations in groundwater in the study area ranged from 1.3 to 5.5 mg/L, with a mean value of

3.4 mg/L. Notably, more than 85.7% of the groundwater samples had fluoride concentrations higher than the WHO guideline-recommended standard of 1.5 mg/L. As shown in Fig. 2b, the F^- concentration of the shallow groundwater rose from south to north in the study area. According to previous studies, high levels of F^- concentration in groundwater are primarily due to fluoride-rich minerals existing in groundwater aquifers, including clay particles, muscovite, biotite, and fluorite (Wang et al. 2023a; Chen et al. 2023). Arsenic concentrations in groundwater ranged from 0.1 to 72.4 µg/L, with a mean value of 8.6 µg/L (Table 1). The geological origins of high-As groundwater are mainly associated with the sedimentary environment of strata, such as the reductive dissolution of Fe (hydr) oxides and the oxidation of sulfide minerals containing As (Zhang et al. 2019; Goswami et al. 2022). Nearly 21.4% of groundwater samples showed arsenic concentrations above the allowable limit of 10 µg/L set by the WHO (2011). Figure 2c shows that the As contamination in groundwater was mainly concentrated in the southeastern part of the study area, such as Liangzhai and Dashahe. Particularly, the As concentration in Huashan town reached 72.4 µg/L, indicating severe As contamination. Notably, co-occurrence of high F^- and As in 21.4% of groundwater samples increased the health risk relative to one toxicant,
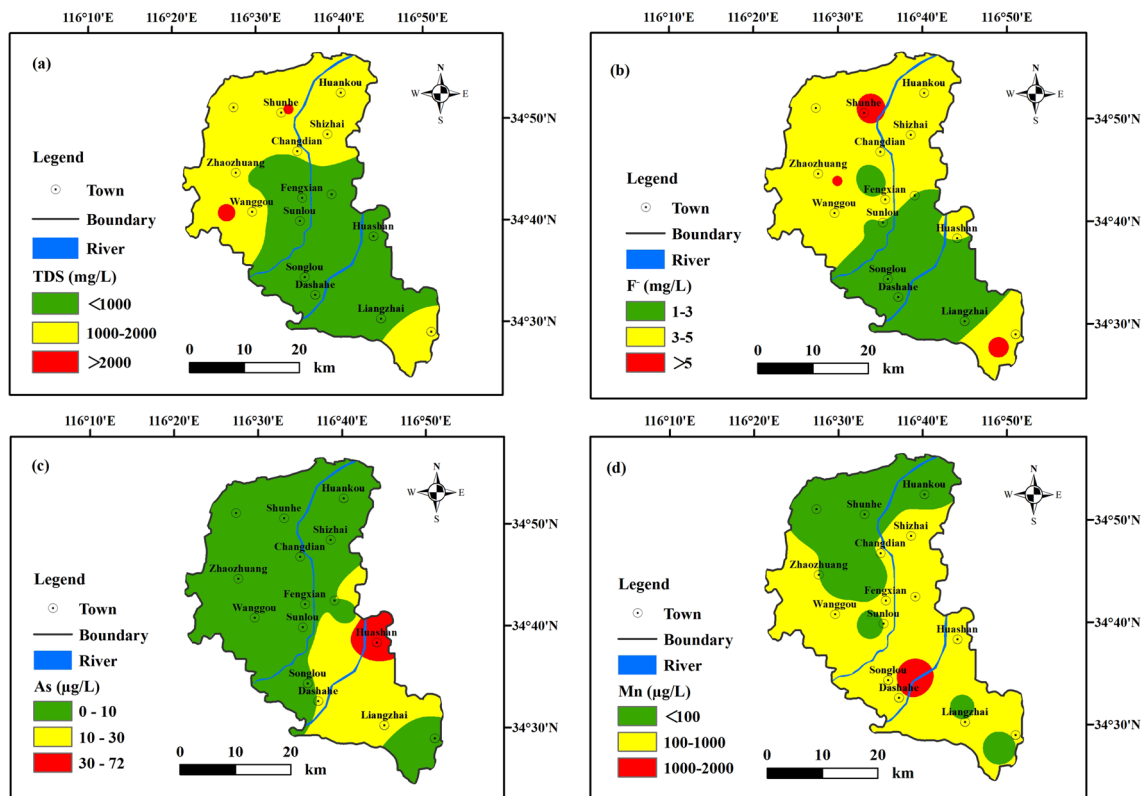

Fig. 2 Spatial variation of TDS, F^-, As, and Mn concentration in shallow groundwater

which was also observed in areas of Dharmanagar region (Bhattacharya et al. 2020) and Huaihe River Basin (Xu et al. 2023). The concentration of Mn in the current study varied between 0.3 and 1536.4 μg/L with a mean value of 250.1 μg/L, and 42.9% of water samples were above WHO guideline for drinking water (100 μg/L) (Fig. 2d). It was notable that the high Mn concentrations (> 100 μg/L) of groundwater were mainly found in the south of the study area (Fig. 2d).

In the present study, K^+ and Na^+ concentrations in groundwater ranged from 2.8 to 32.4 mg/L (mean 16.9 mg/L) and 51.1 to 547.8 mg/L (mean 263.6 mg/L), respectively, indicating that Na^+ concentrations in groundwater were significantly higher than K^+ concentrations. More than 78.6% of sampling sites exceeded the WHO-recommended K^+ permissible limit (12 mg/L), whereas 57.1% of samples had Na^+ exceeding the threshold value of 200 mg/L (WHO 2011). As shown in Fig. 3a and b, the areas with excessive K^+ and Na^+ contents are mainly distributed in the southeast and northwest agricultural areas. According to Wang et al. (2023a) and Chen et al. (2023), the dissolved Na^+ in groundwater may not only be related to the widespread distribution of feldspar and halite in the aquifer but also be probably attributed to cation exchange processes between groundwater and soil. It is noted that the widespread use of potassium fertilizers in agriculture

can result in elevated K^+ levels in groundwater, which can harm the nervous and digestive systems of humans through oral intake (Sharma et al. 2017). On the other hand, Ca^{2+} and Mg^{2+} concentrations ranged from 29.8 to 95.5 mg/L and 24.7 to 103.8 mg/L with mean levels of 56.9 and 59.2 mg/L, respectively. It can be seen that Ca^{2+} and Mg^{2+} levels are all below the acceptable limits of 200 mg/L and 150 mg/L (WHO 2011), respectively (Fig. 3). Related studies have shown that Ca^{2+} and Mg^{2+} in groundwater are mainly derived from the dissolution of carbonate and gypsum by weathering (Liu et al. 2020a; Guo et al. 2023).

The statistical values and spatial variation of major anions (Cl^-, SO_4^{2-}, HCO_3^-, and NO_3^-) are depicted in Table 1 and Fig. 4, respectively. According to WHO guidelines, the maximum permissible limit for Cl^- is 250 mg/L (Liu et al. 2022). In this study, Cl^- concentration in the shallow groundwater ranged from 54.5 to 506.4 mg/L, with about 21.4% of the samples exceeding the recommended permissible limit. Groundwater Cl^- levels were high in Zhaozhuang, Wanggou, and Huankou, although mean values did not exceed the limit (Fig. 4a). High Cl^- concentration in groundwater can induce hypertension, osteoporosis, and asthma in humans. Besides halite dissolution, industrial wastewater, compost, and waste leachate may also contribute to high Cl^- concentrations in groundwater (Chen et al. 2023). The concentrations of SO_4^{2-} in
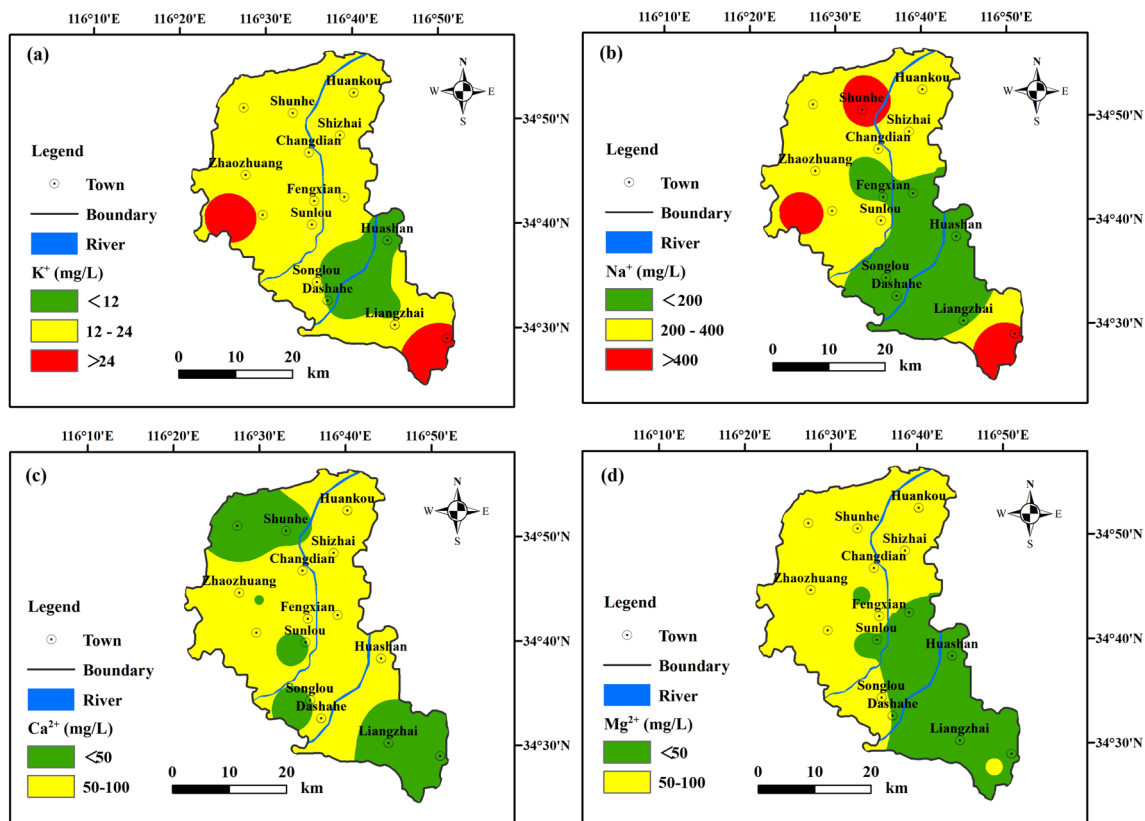

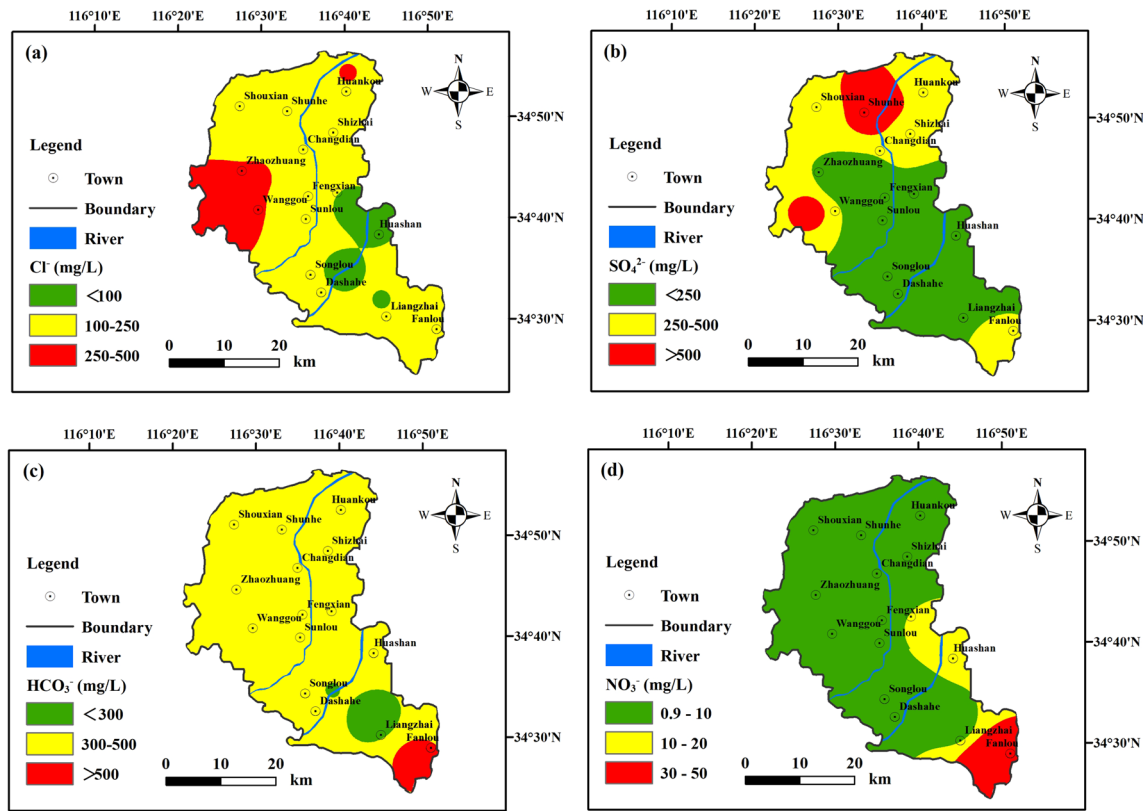**Fig. 3** Spatial variation of major cation concentration in shallow groundwater

Fig. 4 Spatial variation of major anionconcentration in shallow groundwater

groundwater in the study area ranged from 8.1 to 945.7 mg /L (mean 262.8 mg/L). About 42.9% of sampled groundwaters exceeded the safety limit of 250 mg/L (WHO 2011). Groundwater samples with high SO_4^{2-} content mainly occurred in Shouxian, Changdian, Huankou, Shizhai, Shunhe, and Fanlou (Fig. 4b). Although SO_4^{2-} has no toxicity it does change the level of methemoglobin and sulphaemoglobin in the human body when consumed in large amounts through drinking water (WHO 2011; Sharma and Kumar 2020). Gypsum dissolution and other anthropogenic sources (e.g., mining, fertilizer, etc.) may contribute significantly to high SO_4^{2-} concentrations in groundwater (Kumar et al. 2017; Goswami et al. 2022). The concentration of HCO_3^- in groundwater ranged from 195.2 to 585.6 mg/L with a mean value of 398.9 mg/L. About 7.1% of the samples exceeded the limit of 500 mg/L allowed by WHO (2011). As reported by Wang et al. (2023a), silicate and carbonate weathering in the study area may have been an important factor in increasing HCO_3^- concentration of groundwater. Furthermore, the alkaline environment promotes the enrichment of F^- content (Chen et al. 2023). According to the spatial distribution map of HCO_3^- in the study area (Fig. 4c), all samples were within the permissible range, except for the Fanlou. Shallow groundwater NO_3^- concentrations in the study area ranged from 0.9 to 88.5 mg/L, with a mean of 24.9 mg/L. Naturally, precipitation or soil organic nitrogen are the main sources of

groundwater NO_3^-, which is present in the environment at low amounts (below 10 mg/L) (Mao et al. 2023). However, more than 28.6% of samples exceeded the natural environment concentrations and the contaminated areas were mainly located in the southeast (Fig. 4d). Therefore, NO_3^- contamination in groundwater in the study area cannot be ignored. Chronic ingestion of NO_3^--contaminated groundwater can cause methemoglobinemia in humans (Wang et al. 2023b). According to the field investigation, farmers in the study area use various nitrogenous fertilizers to increase crop yields, which inevitably elevated NO_3^- levels in groundwater (Wang et al. 2023a).

In summary, the dominant orders of major cations and anions concentration in the shallow groundwater of the study area were $Na^+ > Ca^{2+} > Mg^{2+} > K^+$ and $HCO_3^- > SO_4^{2-} > Cl^- > NO_3^- > F^-$. The comparative analysis revealed that the groundwater suffered serious contamination from TDS, K^+, Na^+, Cl^-, SO_4^{2-}, NO_3^-, F^-, Mn, and As, with exceedance rates of 42.9%, 78.6%, 57.1%, 21.4%, 42.9%, 28.6%, 85.7%, 42.9%, and 21.4%, respectively.

Drinking water suitability

WQI is a worthwhile approach for assessing groundwater quality and its suitability for drinking purposes (Hossain and Patra 2020; Liu et al. 2021). Several researchers (such

as Nizam et al. 2022 and Taloor et al. 2024) have employed this method to assess groundwater quality in their studies. The WQI values of the groundwater samples ranged from 61.6 to 206.7, with a mean value of 125.7. According to the WQI classification, 28.6% of the groundwater samples were "good" quality, 64.3% were "poor" quality, and 7.1% were "very poor" quality. As shown in Fig. 5, the areas with "good" water quality were mainly concentrated in Sunlou and Fengcheng in the central part of the study area and Liangzhai in the southeast. However, a majority of the shallow groundwater samples were not suitable for drinking due to WQI values between 100 and 200. Human health is expected to be jeopardized if contaminated groundwater is consumed for an extended period. Consequently, seeking alternative clean water sources for drinking is urgent for this area.

Irrigation water quality evaluation

Shallow groundwater is one of the main water sources for agricultural irrigation in the study area. Therefore, groundwater irrigation suitability has a direct impact on the sustainable development of local agriculture. High Na^+ concentration in irrigation water reduces the permeability of the soil, resulting in alkaline soils unsuitable for cultivation

(Li et al. 2018). High salinity inhibits water from reaching the branches and leaves of plants, which is detrimental to crop growth (Sellamuthu et al. 2022). To check the suitability of the groundwater for irrigation uses, a set of determinants for irrigation water suitability was studied and outlined in Table 3 (Atikul Islam et al. 2017; Mohamed et al. 2022). Typically, SAR and %Na are used to evaluate alkali damage of irrigation water (Wang et al. 2022; Mukherjee et al. 2022). SAR were classified as excellent (< 10), good (10–18), doubtful (18–26), and unsuitable (> 26) (Table 3) (Liu et al. 2022). Results showed that SAR values ranged from 1.21 to 12.82, with a mean of 5.61, indicating that all groundwater was suitable for irrigation (Table 2). Furthermore, the %Na in irrigation water should not exceed

Table 2 Summary statistics of groundwater quality indicators for irrigation

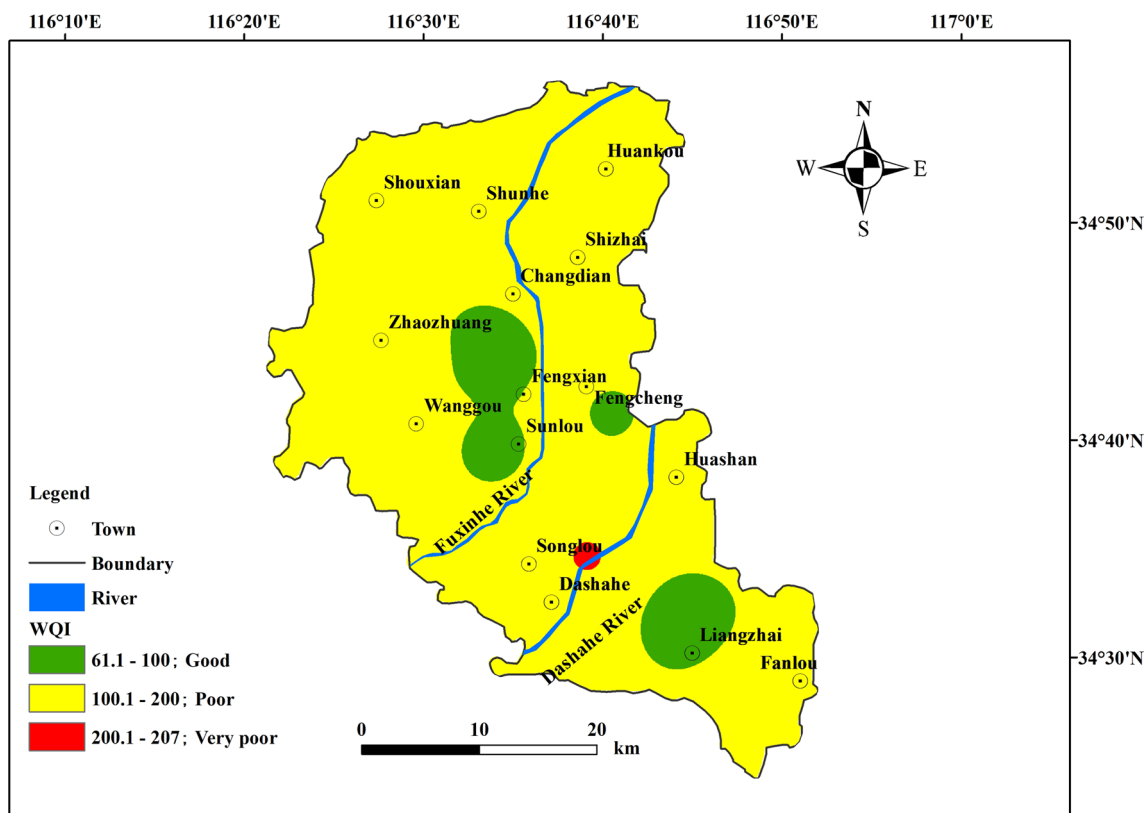| Parameters	Min	Max	Mean	SD
SAR	1.21	12.82	5.61	3.56
%Na	25.73	80.77	55.51	15.51
RSC	− 5.48	3.88	− 1.24	2.34
PI	48.81	90.94	69.87	12.04
MH	31.11	76.12	62.31	14.72

Fig. 5 Spatial variation of WQI of shallow groundwater in the study area

Table 3 Groundwater quality classification for irrigation purposes in the study area

Parameters	Range	Category	Samples Number	Samples % of samples
Sodium hazard (SAR)	< 10	Excellent	12	85.7%
	10–18	Good	2	14.3%
	18–26	Doubtful	–	–
	> 26	Unsuitable	–	–
Residual sodium carbonate (RSC)	< 1.25	Suitable	13	92.9%
	1.25–2.5	Doubtful	–	–
	> 2.5	Unsuitable	1	7.1%
Sodium percentage (%Na)	< 20	Excellent	–	–
	20–40	Good	2	14.3%
	40–60	Permissible	7	50.0%
	60–80	Doubtful	4	28.6%
	> 80	Unsuitable	1	7.1%
Permeability index (PI)	> 75	Good	4	28.6%
	25–75	Permissible	10	71.4%
	< 25	Unsuitable	–	–
Magnesium hazard (MH)	< 50%	Suitable	2	14.3%
	> 50%	Unsuitable	12	85.7%
Salinity hazard (SH)	< 250	Excellent	–	–
	250–750	Good	1	7.1%
	750–2250	Doubtful	10	71.4%
	> 2250	Unsuitable	3	21.4%

60%, and when it does, groundwater becomes unsuitable for agricultural irrigation (Zhou et al. 2021). In Table 2, the variation of %Na ranged from 25.73 to 80.77%, with a mean value of 55.51%. In terms of %Na, about 7.1% of the water samples in the study area were unsuitable for irrigation (Table 3). EC can be used to assess the groundwater salinity suitability for irrigation purposes. Based on the EC values, salinity hazards can be classified as excellent (EC < 250 µS/cm), good (250 < EC < 750 µS/cm), doubtful (750 < EC < 2250 µS/cm), and unsuitable (EC > 2250 µS/cm) (Sharma et al. 2017). The EC of groundwater in this study area ranged from 729 to 3030 µS/cm with a mean value of 1667.4 µS/cm and 21.4% of the groundwater was unsuitable for irrigation regarding salinity hazards (Table 3).

According to the RSC classification (Table 3), groundwater with an RSC value < 1.25 is considered suitable for irrigation purposes. In contrast, if the RSC value varies between 1.25 and 2.5, the water quality is doubtful; water with an SAR value above 2.5, signifies that the water quality is unsuitable for irrigation (Panneerselvam et al. 2021). The results show that the RSC ranged between −5.48 to 3.88 with a mean value of −1.24 (Table 2), and 92.9% of the samples were suitable for irrigation (Table 3). Only one sample (G12) had a SAR value of 3.88, indicating a higher risk of sodium toxicity and a negative impact on crop growth. The permeability index (PI) is an index to assess the permeability of the soil. Long-term groundwater irrigation with rich minerals such as Na^+, Ca^{2+}, Mg^{2+}, and HCO_3^- will

lower the soil permeability and reduce the crop yield. According to the PI classification (Table 3), PI > 75% was considered suitable for irrigation; 25% < PI < 75% was permissible for irrigation; and when PI < 25%, it was not suitable for irrigation (Liu et al. 2022). Table 2 shows that the PI values in the study area ranged from 48.81 to 90.94 with a mean of 69.87, indicating that groundwater is suitable for irrigation. Overall, 28.6% of the samples were "good", and the remaining 71.4% of samples were in the "permissible" category (Table 3). Therefore, the groundwater samples show no evidence of a soil infiltration problem. The MH in groundwater samples ranged from 31.11 to 76.12 (average 62.31) (Table 2), with 85.7% of the water samples exceeding the permissible limit (> 50%) (Table 3). This meant that the majority of the groundwater in the study area posed a significant magnesium hazard.

Besides the above parameters, USLL and Wilcox plots correlating alkalinity and salinity have been widely used to evaluate groundwater suitability for irrigation (Mukherjee et al. 2022; Sellamuthu et al. 2022). The United States Salinity Laboratory plot (Fig. 6a) showed that 71.4% of the water samples are plotted in zone C3S1, indicating high salinity and low sodium hazards. One sample (7.1%) is scattered in zone C3S2, illustrating high salinity and medium sodium hazards. However, 21.4% of the samples are plotted in zone C4S2, showing very high salinity and medium sodium hazards. Wilcox plots (Fig. 6b) showed that 7.1%, 14.3%, and 42.9% of the water samples were plotted in the categories of "unsuitable", "doubtful to unsuitable", and
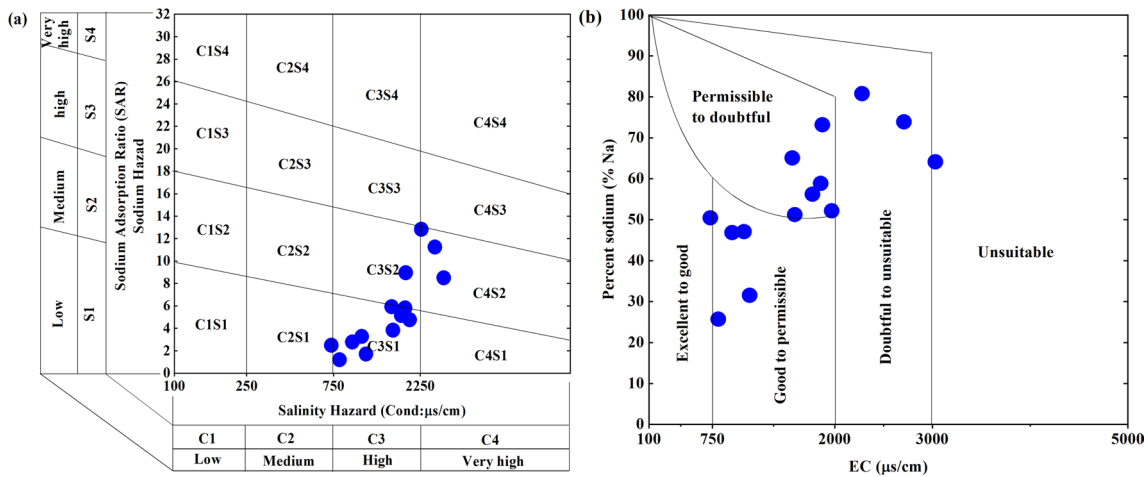
Fig. 6 Irrigation water quality assessment in the study area: **a** USSL plot; **b** Wilcox plot

"permissible to doubtful", respectively. Therefore, alternate irrigation with fresh water is necessary to prevent the accumulation of sodium in the soil (Li et al. 2016).

To sum up, the most important factors affecting the irrigation water quality are the high salinity and the magnesium hazard. High Mg^{2+} in agricultural water will reduce soil fertility and thus affect crop yields (Shaikh et al. 2020). As a result, groundwater that has not been treated for salinity reduction and Mg^{2+} control is not suitable for irrigation directly. Furthermore, sodium hazards in irrigation water should not be ignored.

Health risk assessment for F^- and As in terms of drinking water

Non-carcinogenic risk of F^- and As

Given the study area is facing severe groundwater F^- and As contamination, an assessment of the human health risks is needed. According to Table 4, $HQ_{Fluoride}$ ranged from 2.03 to 8.62 (average 5.35), 1.25 to 5.33 (average 3.31), 1.08 to 4.60 (average 2.85), and 0.77 to 3.28 (average 2.03) for infants, children, females, and males, respectively. Therefore, it can be presumed that all age groups have an alarming non-carcinogenic if prolonged ingestion of high F^- groundwater in the study area.

Table 4 Health risk assessment result for F^- and As in terms of drinking water

Samples ID	HQ$_{Fluoride}$				HQ$_{Arsenic}$				CR$_{Arsenic}$			
	Infants	Children	Females	Males	Infants	Children	Females	Males	Infants	Children	Females	Males
G01	**4.64**	**2.87**	**2.48**	**1.77**	**1.45**	0.90	0.78	0.55	4.7E−06	3.5E−05	**1.5E−04**	**1.1E−04**
G02	**6.43**	**3.98**	**3.43**	**2.45**	0.45	0.28	0.24	0.17	1.5E−06	1.1E−05	4.7E−05	3.3E−05
G03	**8.02**	**4.96**	**4.28**	**3.05**	0.16	0.10	0.09	0.06	5.1E−07	3.8E−06	1.6E−05	1.2E−05
G04	**6.57**	**4.06**	**3.51**	**2.50**	0.28	0.17	0.15	0.11	9.1E−07	6.7E−06	2.9E−05	2.1E−05
G05	**8.62**	**5.33**	**4.60**	**3.28**	0.14	0.09	0.08	0.05	4.6E−07	3.4E−06	1.5E−05	1.1E−05
G06	**4.88**	**3.02**	**2.61**	**1.86**	0.13	0.08	0.07	0.05	4.1E−07	3.1E−06	1.3E−05	9.4E−06
G07	**6.95**	**4.30**	**3.71**	**2.64**	0.08	0.05	0.04	0.03	2.7E−07	2.0E−06	8.5E−06	6.0 E−06
G08	**3.52**	**2.18**	**1.88**	**1.34**	0.03	0.02	0.02	0.01	1.0E−07	7.4E−07	3.2E−06	2.3E−06
G09	**4.32**	**2.67**	**2.31**	**1.64**	0.76	0.47	0.40	0.29	2.4E−06	1.8E−05	7.8E−05	5.5E−05
G10	**4.97**	**3.08**	**2.66**	**1.89**	22.61	13.98	12.07	8.60	7.3E−05	**5.4E−04**	**2.3E−03**	**1.7E−03**
G11	**2.05**	**1.26**	**1.09**	0.78	**4.81**	**2.97**	**2.57**	**1.83**	1.5E−05	**1.1E−04**	**4.9E−04**	**3.5E−04**
G12	**8.12**	**5.02**	**4.33**	**3.09**	0.54	0.33	0.29	0.21	1.7E−06	1.3E−05	5.6E−05	4.0E−05
G13	**3.71**	**2.30**	**1.98**	**1.41**	0.89	0.55	0.48	0.34	2.9E−06	2.1E−05	9.2E−05	6.5E−05
G14	**2.03**	**1.25**	**1.08**	0.77	**5.29**	**3.27**	**2.83**	**2.01**	1.7E−05	**1.3E−04**	**5.5E−04**	**3.9E−04**

Bold indicates the values exceed the health risk safe limit

Also, the values of $HQ_{Arsenic}$ were 0.03 to 22.61 (average 2.69) for infants, 0.02 to 13.98 (average 1.66) for children, 0.02 to 12.07 (average 1.43) for females, and 0.01 to 8.60 (average 1.02) for males. Notably, 28.6%, 21.4%, 21.4%, and 21.4% of the groundwater samples have high arsenic non-carcinogenic risks for infants, children, females, and males, respectively.

In summary, it can be seen from average that the order of non-carcinogenic health risks caused by F^- and As is infants > children > females > males, which is consistent with the findings of Jiang et al. (2022) and Chen et al. (2023).

Carcinogenic risk of As

The results of the carcinogenic risk assessment showed that the potential $CR_{Arsenic}$ values for infants, children, females, and males ranged from 1.0E−07 to 7.3E−05 (mean 8.6E−06), 7.4E−07 to 5.4E−04 (mean 6.4E−05), 3.2E−06 to 2.3E−03 (mean 2.8E−04), and 2.3E-06 to 1.7E-03 (mean 2.0E−04), respectively. Accordingly, 0%, 21.4%, 28.6%, and 28.6% of groundwater samples surpassed the carcinogenic tolerable risk values of 10^{-4} recommended by USEPA (2004). This indicated that As might pose a potential carcinogenic risk to the residents who used groundwater for their daily purposes (Rahman et al. 2022). Therefore, continuous monitoring of groundwater arsenic levels is recommended to identify spatiotemporal variations of potential As risks,

particularly in the southeast of the study area where samples (G01, G10, G11, and G14) exceeded the limit values.

Groundwater sustainable management strategies

Based on the above analysis, an integrated conceptual model of groundwater quality and associated health risk assessment was developed (Fig. 7). As stated in Sect. "Health risk assessment for F^- and As in terms of drinking water", groundwater F^- and As has a significant impact on groundwater quality and associated health hazards in Feng County, particularly in the infants group. Hence, constant monitoring of groundwater F^- and As levels is needed to gain better insight into the contamination dynamics and variation of health hazards (Goswami et al. 2022). Additionally, the Water Affairs Bureau should establish special funding for machine learning projects to predict groundwater quality and enhance sustainable groundwater management (Haggerty et al. 2023).

Monitoring and evaluating environmental pollutants are merely the first steps in improving groundwater quality. Promoting low-cost technologies to remove F^- and As could be a viable alternative for this region. Particularly, rural households should be provided with low-cost water purifiers, such as deionization and de-fluoridation filters. Water administration authorities should prioritize initiating water quality improvement projects to reduce the human health risk of F^- and As contamination in groundwater. More importantly,
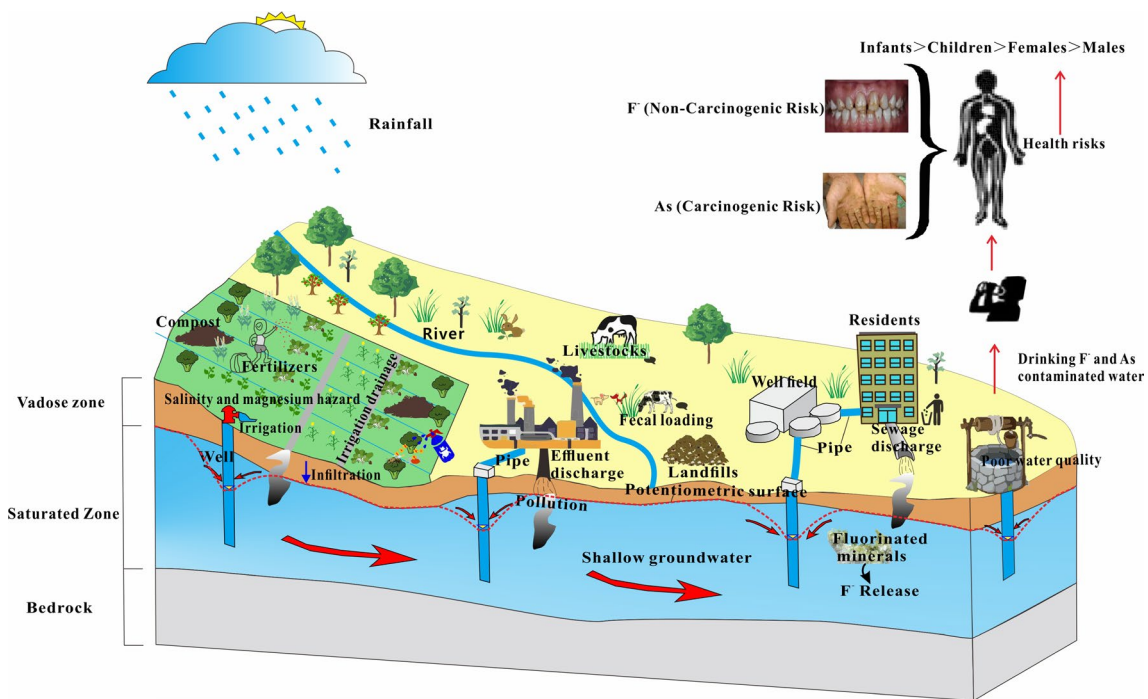

Fig. 7 The conceptual diagram demonstrating groundwater quality and associated health risks

this region urgently needs to use the unpolluted deep groundwater and surface water resources for drinking and irrigation purposes. Irrigation problems caused by high salinity and magnesium content in groundwater can be treated by diluting with different freshwater (Mukherjee et al. 2022).

Conclusions

In the present study, shallow groundwater quality and its suitability for drinking and irrigation, as well as F^- and As health risks were evaluated in Feng County. Results indicated that about 85.7% of shallow groundwater samples exceeded the WHO drinking criterion for F^-, followed by K^+ (78.6%), Na^+ (57.1%), TDS (42.9%), SO_4^{2-} (42.9%), Mn (42.9%), Cl^- (21.4%), and As (21.4%). Based on WQI assessment results, 28.6% of the groundwater was good quality, while 71.4% was unsafe for drinking. According to non-carcinogenic risk appraisal results, almost all age groups hold $HQ_{Fluoride}$ values > 1 and followed the order of infants > children > females > males. Nevertheless, only 28.6%, 21.4%, 21.4%, and 21.4% of samples had $HQ_{Arsenic}$ values > 1 for infants, children, females, and males, respectively. For carcinogenic risk, 0%, 21.4%, 28.6%, and 28.6% of groundwater samples have unacceptable $CR_{Arsenic}$ values ($> 1 \times 10^{-4}$) for infants, children, females, and males, respectively. According to the salinity and alkalinity indicators (SAR, RSC, %Na, PI, MH, and SH) and USSL and Wilcox plot, about 21.4 and 85.7% of groundwater samples were unsuitable for irrigation due to they contained high salinity and magnesium hazards, respectively. The findings of this study could help government agencies or decision-makers adopt appropriate remedial measures to ensure residents' health and help in achieving SDG-6 i.e., supplying clean and hygiene drinking water. Future in-depth studies could integrate aquifer geology and machine learning for groundwater quality forecasting and safeguard public and ecological health.

Declarations

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