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Hydrochemical characteristics and a health risk assessment of the use of river water and groundwater as drinking sources in a rural area in Jiangjin District, China

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Abstract

To determine the hydrochemical characteristics and health risks associated with their use as drinking water, 54 river water and 44 groundwater samples were collected and analyzed in a rural area of Jiangjin District, Chongqing City. The concentration of solutes in river water and groundwater showed significant spatial variability in the region. A number of dissolved chemical constituents including nitrate (NO_3^-), nitrite (NO_2^-), aluminum (Al), arsenic (As), boron (B), barium (Ba), iron (Fe), manganese (Mn), nickel (Ni), lead (Pb) and selenium (Se) exceeded their respective recommended drinking water limits at some locations. Ternary plots of cations, anions and silica indicated that carbonate weathering was the primary source of major ions in water, followed by silicate weathering. Elevated concentrations of some chemical constituents including NO_3^- and chloride (Cl^-) were found in water samples in areas with the most intensive agricultural land use activities. A health risk assessment indicated that Cl^- and NO_3^- were the most important chemical constituents that were a non-carcinogenic health concern. A carcinogenic health risk assessment indicated that chromium (Cr) and As were the chemical constituents of most concern in water from the area that might be used as a drinking water source. The average annual carcinogenic risks for Cr in drinking water were determined to be in the range of 3.14E−05 and 7.90E−05 for adults and children, respectively. Similarly, the average annual carcinogenic risks for As in drinking water for adults and children were calculated to be in the range of 4.43E−07 and 1.11E−06, respectively. These values are within the risk values of 10^{-6} to 10^{-4}, which generally indicate that there are health concerns that need to be addressed in more detail. The highest values of carcinogenic risk for drinking water were mainly located in the northern part of the study area where there are industrial activities that are potential sources of arsenic and chromium in surface water and groundwater.

Keywords Hydrochemical characteristics · Health risk assessment · Carcinogenic risk · Rural area of Jiangjin · Anthropogenic influences

Introduction

Adequate and safe water is essential for our survival. However, water security problems are increasing due to high rate of population growth, rapid economic development and community expectations of an increasing quality of life. China is the largest developing country that had approximately 560 million people still living in rural areas at the end of 2018. Therefore, it is extremely important to ensure the safety of drinking water in Chinese rural areas.

The characteristics of drinking water in rural areas of China is complicated due to widespread presence of pollution sources, small water volumes and dispersion (especially in mountainous areas) and poor wastewater disposal facilities and practices, making it difficult to ensure the sanitary

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status of safe drinking water in rural areas (Ou et al. 2012; Wang et al. 2012).

Low concentrations of many chemical constituents in drinking water are derived from rock weathering, industrial wastewater discharges and agricultural runoff. Although the low dosage of these chemical constituents may not present an acute health hazard, their accumulation over time can produce chronic health impacts such as the effects of fluorosis from the long-term exposure of populations to high concentrations of fluoride in drinking water (Luo et al. 2012; Mandinic et al. 2010), chronic arsenism via drinking water (Villaescusa and Bollinger 2008; Wen et al. 2013) and many 'cancer villages' in China (Dong et al. 2014). Therefore, it is important to identify chemical constituents of health concern in water from rural area to undertake effective health risk assessments (HRAs).

The Three Gorges Dam (TGD) is the largest water conservancy project in the world. The water quality of Three Gorges Reservoir Region (TGRR) directly impacts the water security of cities and rural areas in the upper, middle and lower reaches of the Yangtze River. Therefore, water resources and water quality in TGRR is of great importance and seeks attention (Gao et al. 2017; Huang et al. 2016; Liu et al. 2012; Wang et al. 2018a, b).

Due to the rise of water levels in reservoirs and flow reduction, most of the previous researches mainly focused on water environmental problems in the hinterland of the TGRR such as the effects of eutrophication (Huang et al. 2016; Liu et al. 2012), algal bloom (Ji et al. 2017), excessive nitrogen and phosphorus loads (Li et al. 2009b; Liu et al. 2012) and water temperature stratification in surface water bodies (Yang et al. 2012). Systematic studies have shown that high concentrations of some of the trace elements are present in water in rural areas of the TGRR; however, there are limited studies that reported health risk assessment for the use of river water and groundwater as drinking sources in a rural area of the upstream of the TGRR. Jiangjin District, where the rural population constitutes over 50% of the total population, is located at the upstream of the TGRR and is an important industrial and agricultural region of Chongqing. The district contains industrial parks including power plants, chemical and textile industries which are potential sources of water pollution.

Based on the above considerations, the characteristics of both surface water and groundwater quality in the Jiangjin rural area of the upstream area of Three Gorges Reservoir is studied and evaluated. Consequently, the objectives of this study are as follows: (1) to identify the hydrochemical characteristics and characterize the spatial distribution of chemical constituents of health concern in water in Jiangjin District, China; (2) to identify the effective processes that control the water chemistry; and (3) to assess the health risk associated with exposure to dissolved chemical constituents of health concern in water in rural areas of the region, especially trace elements via oral ingestion.

Materials and methods

Regional setting

Jiangjin is located in the southwestern part of Chongqing and the upstream section of the Yangtze River (Fig. 1). The study area has a subtropical wet monsoon climate with an average temperature of 18.4 °C and total annual rainfall of 1031 mm. Jiangjin district overlies two landforms consisting of the parallel ridge–valley landform of east Sichuan and a mountainous landform that occurs in the southeastern margin of Sichuan basin. Due to the influence of the geomorphology and the geological structure of the region, the regional landscape slopes downwards from the north to the south towards the Yangtze River valley.

On the basis of geological and topographical characteristics, three distinct geomorphological landforms can be distinguished in the study area. These are: valley terraces, hills and mountains (Fig. 1a) (JJPG 2015; Zhao 2015). The valley terrace area occupies 3.1% of the total area, mainly distributed along Yangtze River; the hills occupy 65.1% of the total area and are mainly distributed in syncline valley. Low-elevation mountains are distributed in northern part of the study area, and the medium- to low-elevation mountains are mainly located in the southern part of the study area (JJPG 2015; Zhao 2015). Based on its geomorphological characteristics and variations in levels of socio-economic development, Jiangjin district is divided into three sub regions: the northern region (the area with low-elevation mountains); the central region (the hilly region); and the southern region (the area with medium- to low-elevation mountains) (JJPG 2015).

Jiangjin district, in the southern part of the east Sichuan fold belt, is located at the transitional area between the east Sichuan fold belt and the Sichuan–Guizhou SN tectonic belt. Sediments in the core of anticline are of Triassic age, while they are of Jurassic age in the core of the syncline (Fig. 1b). From west to east, a number of anticlinal and synclinal structures are arranged sub-parallel to each other. These are successively: the Libixia Anticline (A1); the Bishan Syncline (S1); the Wentangxia Anticline (A2); the Beibei Syncline(S2); the Shilongxia Anticline (A3); the Zhongfeng Temple Syncline (S3); and the Guanyinxia Anticline (A4) (Fig. 1b).

About 78.7% of the study area is underlain by sediments of Jurassic age which is the largest, 13.7% of the area is underlain by Cretaceous sediments, 5.3% by Triassic sediments, and 2.3% by sediments of Quaternary age. Clastic rocks and carbonate rocks of Triassic age are mainly
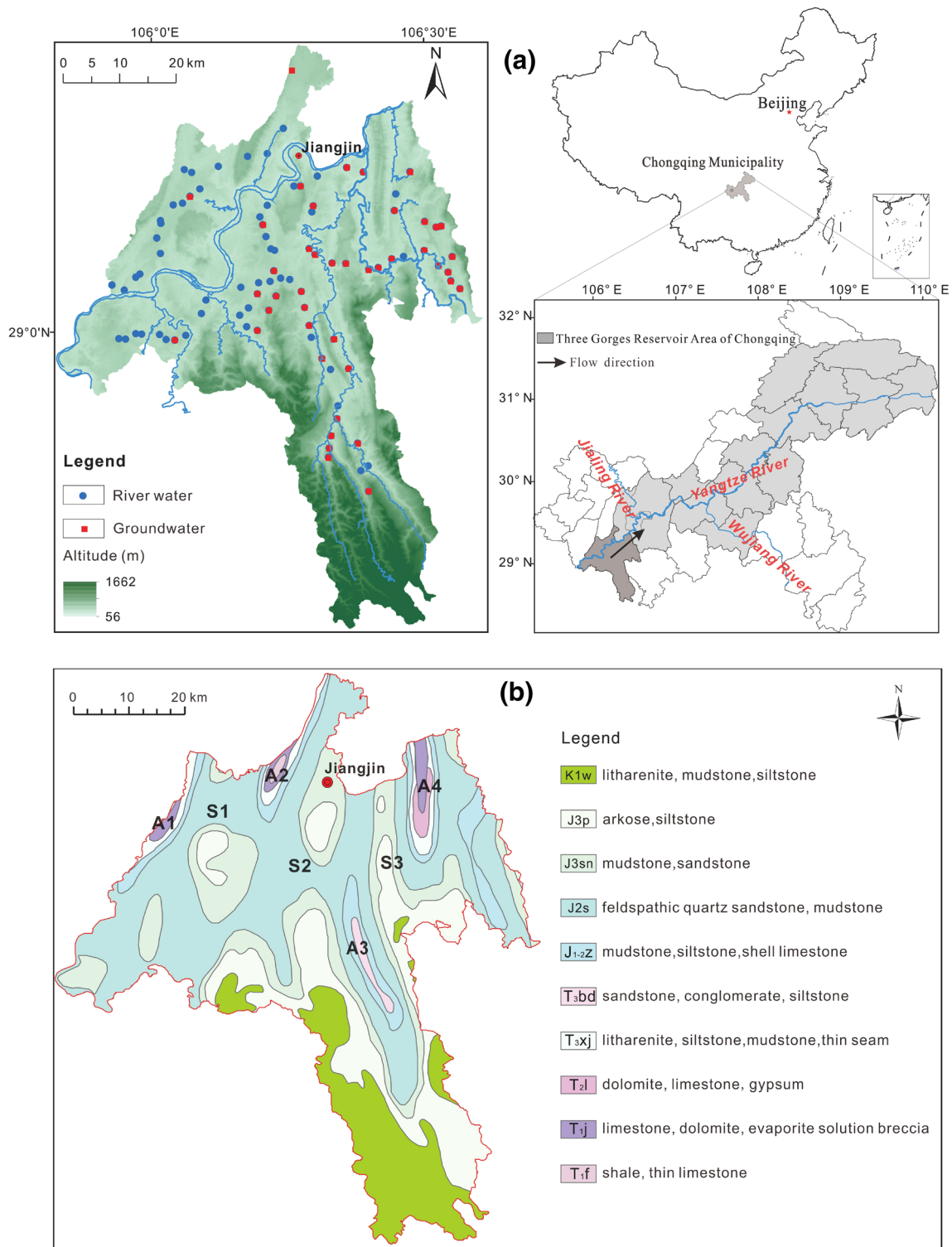
Fig. 1 Map showing the Jiangjin District, Chongqing City of China. Note: **a** the DEM and water sampling point of Jiangjin, **b** the geological map of Jiangjin; A1: Libixia Anticline; A2: Wentangxia Anticline; A3: Shilongxia Anticline; A4: Guanyinxia Anticline; S1: Bishan Syncline; S2: Beibei Syncline; S3: Zhongfeng Temple Syncline

distributed in the northern part of the study area (Fig. 1b), whereas sandstones and mudstones of Cretaceous age are distributed in the southern part of the study area (Fig. 1b). Jurassic strata are mainly exposed in hills in the central part of the study area (Fig. 1b).

The weathered nature of bedrock gives high porosity and permeability that favor the storage and flow of water. The shallow weathering fissure water (including well and spring) is the drinking water source for the dispersed inhabitants of Jiangjin rural area (Liao et al. 2018; Zhou 2007). Aquifer lithology of shallow weathering fissure water is Middle–Upper Jurassic and Upper Cretaceous clastic rock (Zhou 2007). Due to the climate, landform, and geological structure, the recharge sources of shallow weathering fissure water are mainly rainfall and surface water, while the discharge zones are distributed in low terrains suitable for human habitation (Zhou 2007).

Sampling and analytical methods

The drinking water sources in the rural areas of Jiangjin District include river water, groundwater from wells, and spring water in mountainous areas. Fifty-four river water and forty-four groundwater samples were collected in June of 2016 in Jiangjin, Chongqing City (Fig. 1). Water samples at each site were collected in colorless polythene plastic barrels with a capacity of 1 l. The pH and total dissolved solids (TDS) content of each sample were measured in the field using a Mettler Toledo SevenGo pH meter and a Mettler Toledo SevenGo conductivity meter which were previously calibrated. The remaining hydrochemical parameters, including major elements and trace elements, were analyzed at the laboratory of the Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences (CAS). The concentrations of the major cations (Ca^{2+}, Mg^{2+}, Na^+, K^+, SiO_2, B) and sulfate (SO_4^{2-}) in natural water were determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES, Perkin Elmer, Optima 5300 DV). Nitrite (NO_2^-) and chloride (Cl^-) concentrations were measured using ion chromatography (IC, Dionex 600 IC). Bicarbonate (HCO_3^-) was determined using the acid–alkali neutralization titrimetric method. Nitrate (NO_3^-) concentrations in water were determined through UV spectrophotometry (MEPPRC 2002). The fluoride (F^-) concentrations in water were determined by the fluoride ion-selective electrode (ISE) method. Blank samples were determined in parallel to the sample treatment using identical procedures. The precision of analyses was tested throughout by running duplicate analyses of selected samples. The detection limits for major cations (Ca^{2+}, Mg^{2+}, Na^+, K^+, SiO_2) and sulfate (SO_4^{2-}) were 0.01 mg/L using ICP-AES. The detection limit was 0.02 mg/L for Cl^-, 0.08 mg/L for NO_3^-, and 0.03 mg/L for NO_2^- using IC. The concentrations of trace elements

(Ag, Al, Ba, Cd, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Zn) were determined by inductively coupled plasma mass spectrometry (ICP-MS, Perkin Elmer, Elan DRC-e). The detection limits for trace elements (Ag, Al, Ba, Cd, Cr, Cu, Fe, Mn, Mo, Ni, Pb, and Zn) were 0.01 μg/L using ICP-MS. Selenium (Se) and arsenic (As) concentrations in water were measured by hydride generation atomic fluorescence spectrometry (HG-AFS) (AFS-9780, Beijing Kechuang Haiguang Instrument Co., LTD, Beijing, China). The detection limits for Se and As were 0.2 μg/L and 0.1 μg/L, respectively.

The precision of analyses was tested throughout the operation by running duplicate analyses of selected samples. The error percentage in the samples ranged from ± 1 to ± 10%, suggesting a high reliability of the water quality data. Total hardness (TH) was calculated by the concentration of Ca^{2+} and Mg^{2+}.

Risk assessment on human health

A rigorous health risk assessment (HRAs) is an effective approach to determine health risk levels which can be attributed to various contaminants (Giri and Singh 2015; USEPA 2009). This method had been applied to evaluate the potential adverse health effects as a result of being exposed to contaminated water (Giri and Singh 2015; Manli et al. 2014; Zhang et al. 2014).

Human beings can be exposed to hazardous substances via direct ingestion, inhalation and dermal absorption. However, ingestion is generally the most significant pathway for drinking water (Keller 1978; USEPA 2009). The estimated dose intake through this approach was determined using Eqs. (1) and (2):

$$\text{ADD}_{\text{adult}} = \frac{1.542 \times C_W}{58.1} \tag{1}$$

$$ADD_{\text{child}} = \frac{1.0 \times C_w}{15} \tag{2}$$

where ADD is average daily dose, mg/(kg day); C_w is concentration of a chemical constituent of health concern in water, mg/L; 1.542 L/day and 1.0 L/day are average daily water intake for adults and children in Chongqing, L/day (Duan 2013), for adults weighing 58.1 kg and children weighing 15 kg, respectively (Duan 2013).

Risk characterization was quantified by potential non-carcinogenic risks, reflected by the hazard index (HI) using Eq. (3):

$$\text{HI} = \sum \text{HQ}_i = \sum \frac{\text{ADD}_i}{\text{RfD}_i \times 70} \times 10^{-6} \tag{3}$$

where HQ_i is the hazard quotient (HQ)of the ith chemical constituent; RfD_i is the reference dose of the ith chemical constituent, mg/(kg day) (USEPA 2009; Wu et al. 2009b).

Carcinogenic risks (R) were evaluated by Eq. (4):

$$R_i = \frac{ADD_i \times SF_i}{70} \tag{4}$$

where SF_i is the carcinogenic potency index [(kg day)/mg] (USEPA 2009; Wu et al. 2009b); 70 is the average life span (year).

The Mann–Whitney U test and the Kruskal–Wallis test were the non-parametric statistical methods that were used for testing differences. Effects were considered statistically significant with $p < 0.05$ based on two-tailed tests. SPSS 18.0 and Excel 2010 were used for the statistical analysis.

Results and discussion

Hydrochemical characteristics of water from rural areas

The parameters measured in river water and groundwater samples from rural area of Jiangjin District are listed in Table 1. The mean values of pH in river water and groundwater samples were 7.83 and 7.43, respectively, which indicated that river water and groundwater in rural area of Jiangjin were neutral to weakly alkaline. The variation range of TDS values was significant with higher average value in groundwater (11–811 mg/L) than in river water (58–533 mg/L) (Table 1). But the TDS values in river water and groundwater did not exceed the recommended limits for drinking water (MHPRC 2006; WHO 2011). The average

Table 1 Statistical summary of the parameters determined in river water and groundwater samples from rural area of Jiangjin District, Chongqing City

Parameters	River water				Groundwater				MHPRC (MHPRC 2006)	WHO (WHO 2011)	Num[a]
	Min	Max	Mean	SD	Min	Max	Mean	SD			
pH	7.1	8.9	7.8	0.4	5.6	8.4	7.4	0.6	6.5–8.5	6.5–9.5	4
TDS (mg/L)	58	533	183	99.2	11	811	303	154.7	1000	1000	0
TH (mg/L)	7	276	108	58.6	3	546	178	106.2	450	500	1
Ca^{2+} (mg/L)	2	75	31	15.1	1	178	51	32.1	–	75	8
Mg^{2+} (mg/L)	0	24	7	5.4	0	30	12	7.6	–	30	9
Na^+ (mg/L)	1	142	12	19.1	2	83	25	19.2	200	200	0
K^+ (mg/L)	0	6	2	1.4	0	15	2	3.2	–	–	0
HCO_3^- (mg/L)	13	367	107	66.0	9	434	203	122.0	–	120	55
Cl^- (mg/L)	0	31	12	7.3	0	176	18	27.4	250	250	0
SO_4^{2-} (mg/L)	2	173	42	32.7	0	339	52	54.6	250	500	1
NO_3^- (mg/L)	0	72	13	12.6	0	164	16	27.8	50	50	5
NO_2^- (mg/L)	0	1	0	0.2	0	5	0	0.9	1.0	3.0	3
F^- (mg/L)	0	1	0	0.1	0	0	0	0.1	1.0	1.5	0
SiO_2 (mg/L)	1	50	11	8.7	1	76	25	12.4	–	–	0
Ag (μg/L)	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	50	100	0
Al (μg/L)	0.6	76.7	17.2	16.5	0.5	1702.9	138.0	299.3	200	200	8
As (μg/L)	0.0	2.2	0.3	0.5	0.0	37.0	1.4	5.7	10	10	1
B (μg/L)	6.8	377.5	60.5	63.0	5.7	606.6	71.1	97.4	500	500	1
Ba (μg/L)	2.1	334.9	61.4	57.6	4.1	770.4	203.9	173.4	700	700	1
Cd (μg/L)	0.0	0.1	0.0	0.0	0.0	0.6	0.0	0.1	5	3	0
Cr (μg/L)	0.4	7.1	2.2	1.6	0.3	4.3	1.9	1.0	50	50	0
Cu (μg/L)	0.4	70.9	3.2	9.8	0.1	130.6	4.2	19.5	1000	2000	0
Fe (μg/L)	8.7	316.0	131.7	59.1	0.9	1243.4	306.1	253.1	300	300	19
Mn (μg/L)	0.1	54.0	2.0	7.4	0.2	659.6	41.4	113.7	100	400	6
Mo (μg/L)	0.0	4.8	0.8	0.8	0.0	4.8	0.6	0.9	70	70	0
Ni (μg/L)	0.5	47.7	2.5	6.3	0.4	16.9	2.5	2.6	20	20	1
Pb (μg/L)	0.0	9.7	0.2	1.3	0.0	21.5	0.7	3.2	10	10	1
Se (μg/L)	1.0	2.3	1.8	0.3	0.6	11.1	2.3	1.6	10	10	1
Zn (μg/L)	0.0	159.5	15.8	31.2	0.0	575.3	21.5	87.0	1000	3000	0

Note: a: the number exceeding the recommended values

total hardness (TH) value for groundwater (178 mg/L) was higher than that of river water (108 mg/L) ($p = 0.000 < 0.01$; Mann–Whitney U test).

Mesozoic terrestrial clastic rock is widely distributed in Jiangjin District (Fig. 1). Due to the effect of geological structure, lithology and climate, the groundwater type in Jiangjin district is mainly weathering fissure water with shallow depth and convenient exploitation (Liu 2006; Zhou 2007). And the shallow weather fissure water with simply treatment could be sometimes used for drinking water in rural area of Jiangjin (Liao et al. 2018; Zhou 2007). The spatial variability of Ca^{2+}, Na^+, HCO_3^-, SO_4^{2-} and NO_3^- in river water and groundwater from the rural area of Jiangjin was relatively large (SD > 10), suggesting that factors, such as hydrological conditions, topography and human activities have influenced the concentrations of these ions. The pH, TH and SO_4^{2-} in 4.1%, 1.0% and 1.0% of water samples in rural area of Jiangjin exceeded the recommended limits for drinking water (MHPRC 2006). Nitrate (NO_3^-) and nitrite (NO_2^-) in 5.1% and 3.1% of natural water samples exceeded the recommended drinking water limits of 50.0 and 1.0 mg/L (MHPRC 2006).

The concentration of F^- varied from 0.1 to 1.0 mg/L in river water, with an average of 0.2 mg/L. Additionally, the concentration of F^- in groundwater varied from 0.1 to 0.4 mg/L, with a mean of 0.15 mg/L (Table 1). Consequently, the concentration of F^- in river water and groundwater from the rural area of Jiangjin does not exceed the recommended drinking water limit of 1.0 mg/L (MHPRC 2006). The concentration of SiO_2 in river water and groundwater was 1.0–50 mg/L and 1–76 mg/L, respectively (Table 1).

According to their average concentration, trace elements were classified into having "high concentrations"

(> 10.0 μg/L; Al, B, Ba, Fe, Mn, Zn), "moderate concentrations" (0.1–10.0 μg/L; As, Cr, Cu, Mo, Ni, Pb, Se) and "low concentrations" (< 0.1 μg/L; Ag, Cd). The standard deviation (SD) was used to quantify the amount of variation or dispersion of a set of data values. A low standard deviation indicated that the data points tended to be close to the mean of the set, while a high standard deviation indicates that the data points were spread out over a wider range of values (Bland and Altman 1996). So, the standard deviation can reflect the spatial variability of trace element concentrations in water indirectly (Giri and Singh 2015). The spatial variability of "low concentration" trace elements was low (SD < 0.1), whereas concentrations of Al, B, Ba, Cu, Fe, Mn and Zn had a large spatial variability (SD > 10.0) (Table 1). The spatial variation of trace elements in river water and groundwater from rural area of Jiangjin may be related to geology, hydrogeology and human activities, which needs further study.

Table 1 also showed that concentrations of Al, As, B, Ba, Fe, Mn, Ni, Pb and Se in 8.2%, 1.0%, 1.0%, 1.0%, 19.4%, 6.1%, 1.0%, 1.0% and 1.0% of natural water samples, respectively, exceeded the recommended drinking water limits. Concentration of Ag, Cd, Cr, Cu, Mo and Zn in all water samples were within their permissible Chinese drinking water limits.

Relationships between pH, TDS, TH and ions were examined for all sampling sites (Table 2). HCO_3^- had a strong positive correlation ($p < 0.01$) with Cl^-, NO_3^-, Ca^{2+}, Mg^{2+} and Na^+. Similarly, Cl^- was correlated with SO_4^{2-}, Ca^{2+} and Mg^{2+} in a significantly positive way. Additionally, SO_4^{2-} had a significant positive correlation with all cations. There were positive correlations between SiO_2 and Ca^{2+}, Mg^{2+}, Na^+, HCO_3^-, SO_4^{2-}. Both TH and

Table 2 Correlation matrix of the dissolved ions of natural water in Jiangjin

	pH	TDS	TH	HCO_3^-	Cl^-	SO_4^{2-}	NO_3^-	NO_2^-	Ca^{2+}	Mg^{2+}	Na^+	K^+	SiO_2
pH	1.000												
TDS	0.189	1.000											
TH	0.266**	0.904**	1.000										
HCO_3^-	0.328**	0.819**	0.766**	1.000									
Cl^-	0.019	0.455**	0.353**	0.284**	1.000								
SO_4^{2-}	-0.030	0.630**	0.556**	0.133	0.229*	1.000							
NO_3^-	0.030	0.427**	0.332**	0.252*	0.133	0.160	1.000						
NO_2^-	0.004	0.186	0.147	0.168	-0.005	0.141	0.025	1.000					
Ca^{2+}	0.281**	0.882**	0.982**	0.752**	0.354**	0.542**	0.295**	0.159	1.000				
Mg^{2+}	0.195	0.834**	0.906**	0.694**	0.303**	0.514**	0.370**	0.099	0.810**	1.000			
Na^+	0.159	0.719**	0.463**	0.728**	0.154	0.339**	0.178	0.232*	0.462**	0.403**	1.000		
K^+	-0.003	0.102	0.139	-0.033	-0.068	0.241*	-0.039	-0.079	0.091	0.231*	-0.017	1.000	
SiO_2	-0.197	0.600**	0.496**	0.468**	0.180	0.343**	0.172	0.086	0.438**	0.559**	0.430**	0.123	1.000

Note: *Correlation is significant at the 0.05 level. **Correlation is significant at the 0.01 level

TDS had a strong positive correlation ($p < 0.01$) with all cations and anions except for NO_2^- and K^+.

Spatial pattern of major ions and minor ions

Significant spatial variability of major ion concentrations could reflect the influence of different lithology and anthropogenic activities (Chen et al. 2002; Liu et al. 2014). Based on the geomorphological classification

of Jiangjin district into the three subregions previously referred to, the average TDS concentration in river water and groundwater can be classified as follows: the TDS in the central region ($TDS_{mean} = 250$ mg/L) > northern region ($TDS_{mean} = 23$ mg/L) > southern region ($TDS_{mean} = 145$ mg/L) ($p = 0.033 < 0.05$; Kruskal–Wallis test) (Fig. 2a; Table 3). Additionally, NO_3^- concentrations were in the order of central region ($NO_3^-{}_{mean} = 16$ mg/L) > northern region
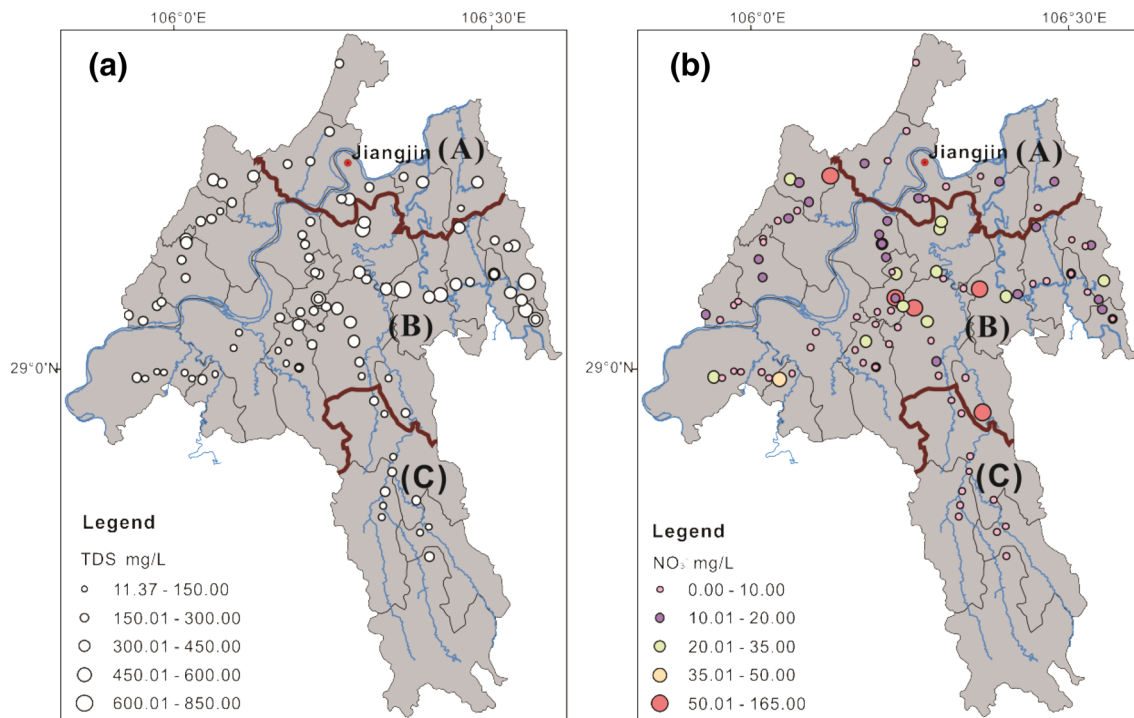

Fig. 2 The distribution of TDS (**a**) and NO_3^- (**b**) in natural water from Jiangjin. Note: **a** Northern region (low mountain area), **b** Central region (hilly region), **c** Southern region (middle–low mountain area)

Table 3 Major ion concentrations in the present study and with other natural water from the literature (mg/L)

	TDS	Ca^{2+}	Mg^{2+}	Na^+	K^+	HCO_3^-	SO_4^{2-}	Cl^-	NO_3^-	NO_2^-	SiO_2	References
Jiangjin	237	40	10	18	2	150	46	15	14	0	17	This study
Northern	234	37	10	20	3	154	44	13	8	0	22	This study
Central	251	42	10	19	2	156	50	17	16	0	17	This study
Southern	145	30	5	8	2	105	23	5	6	0	16	This study
Upper Han River	170	38	8	3	1	148	32	6	7	–	–	(Li et al. 2009a, b)
Tibet	312	55	19	35	3	170	55	48	–	–	13	(Tian et al. 2015)
Yangtze River Basin	206	34	8	8	0	134	12	3	–	–	3	(Chen et al. 2002)
Huai River Basin	509	45	22	87	7	143	107	81	10	–	8	(Liang Zhang et al. 2011)
Upper Wujiang Basin	263	72	12	4	1	199	58	4	12	–	–	(Huang et al. 2017)
Xiangxi Bay	205	46	12	10	2	110	43	10	24	0	4	(Ran et al. 2010)
Daning Bay	222	48	8	16	1	110	35	21	33	0	5	(Ran et al. 2010)
Liangtan River Basin	281	52	36	21	6	73	92	23	13	–	–	(X. Liu et al. 2010)
Global median	65	8	2	5	0	31	5	4	1	–	10	(Meybeck and Helmer 1989)

($NO_3^-{}_{mean} = 8$ mg/L) > southern region ($NO_3^-{}_{mean} = 5$ mg/L) ($p = 0.039 < 0.05$; Kruskal–Wallis test) (Fig. 2b; Table 3). The southern sub-region, where clastic rocks are very common, is likely to have limited land use impacts due to the low population in this area. In contrast, the higher concentrations of major ions and nitrate in the northern and central areas can be attributed to the lower topographic gradients (Fig. 1) and the presence of intensive industrial–agricultural activities. This is especially the case in the central region which is the major farming and animal husbandry district in Jiangjin (JJPG 2015).
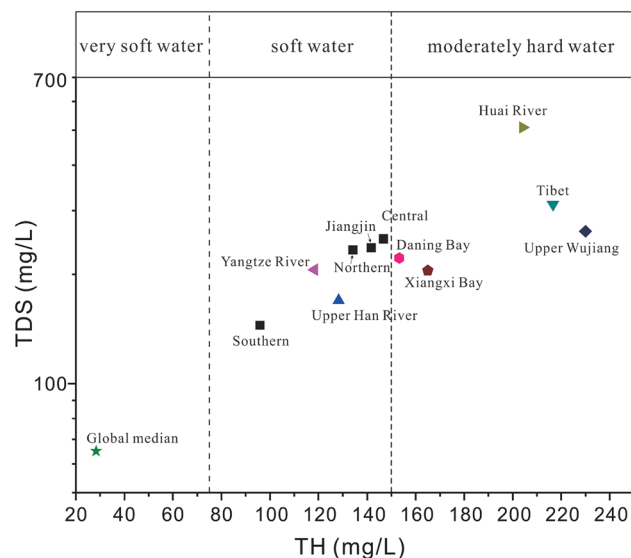
Comparison with studies from other regions

The hydrochemical characteristics of river water and groundwater from rural areas of Jiangjin district were compared with the chemical properties of natural waters from different regions in China (Table 3, Fig. 3). Table 3 and Fig. 3 showed that the hydrochemical characteristics differ in different watersheds. This difference is mainly caused by hydrological conditions, geographic and geomorphic conditions, geologic setting, and human activities in different areas.

The average arsenic concentrations of river water and groundwater samples in the rural area of Jiangjin was in the order: the northern sub-district (4 μg/L) > central sub-district (0.5 μg/L) > southern sub-district (0.10 μg/L) ($p = 0.048 < 0.05$; Kruskal–Wallis test) (Table 4). The highest As concentration measured in the study area (37 μg/L) was from a groundwater sample in the northern sub-district of Jiangjin. The concentration of As in this sample was more

Fig. 3 The graph of TDS vs TH (total hardness) in water

Table 4 Minor ion concentrations in the present study and with other natural water from the literature (μg/L)

Parameters	Jiangjin	Northern	Central	Southern	TGR	Tianjin	Hetao Plain	Sub-arnarekha River Basin, India	Mid-level area, Hong Kong
Ag	0.0	0.0	0.0	0.0	–	–	–	–	1.9
Al	71.4	23.5	74.1	100.8	–	4.0	–	–	1.0
As	0.8	3.7	0.5	0.1	1.7	1.0	43.1	0.2	2.7
B	65.3	108.8	60.1	57.5	–	–	–	–	146.1
Ba	125.4	93.4	136.5	80.5	–	–	143.0	46.4	57.2
Cd	0.0	0.0	0.0	0.0	0.3	0.3	–	0.0	0.1
Cr	2.0	2.9	2.0	1.0	2.3	2.0	–	0.8	1.6
Cu	3.7	1.1	4.5	0.7	3.4	50.0	–	2.0	1.3
Fe	210.0	262.4	205.3	190.4	–	61.0	1706.8	221.2	4.9
Mn	19.7	23.7	21.8	0.9	–	30.0	315.7	183.9	89.9
Mo	0.7	0.8	0.7	0.6	–	–	5.4	2.5	1.6
Ni	2.5	2.5	2.7	1.0	0.8	–	–	4.3	–
Pb	0.5	0.0	0.6	0.2	4.3	1.3	–	–	1.0
Se	2.1	1.7	2.1	1.8	0.4	0.2	–	0.6	8.6
Zn	18.4	4.4	19.9	21.7	6.4	30.0	25.8	60.5	33.7
References	This study	This study	This study	This study	(Lv et al. 2007)	(Fu et al. 2015)	(Yilong Zhang et al. 2014)	(Giri and Singh 2015)	(Leung and Jiao 2006)

TGR: Three Gorges Reservoir

than three times the recommended drinking water limit of 10 μg/L (MHPRC 2006; WHO 2011). Additionally, this groundwater sample collected was close to the Hetao Plain (where the average As content of water is 43 μg/L; Table 4) (Zhang et al. 2014) where arsenic poisoning associated with drinking water has been reported (Wen et al. 2013).

When present at low concentrations in drinking water, selenium (Se) can have a beneficial effect (Fordyce 2007), but, unlike many other trace elements, selenium has a very narrow range between beneficial and harmful health impacts. The average selenium concentration (2.0 μg/L) of river water and groundwater in the rural area of Jiangjin was higher than that of Three Gorges Reservoir (0.43 μg/L) (Lv et al. 2007); Tianjin (0.20 μg/L) (Fu et al. 2015); and the Yangtze River Basin (0.27 μg/L) (Yao et al. 2007). However, the average selenium concentration in water in the study area was lower than that of the Mid-level area in Hong Kong (8.60 μg/L) (Leung and Jiao 2006) (Table 4). The average concentration of Pb (0.5 μg/L) in river water and groundwater in the study area was lower than that of the Three Gorges Reservoir (4.34 μg/L) (Lv et al. 2007); Tianjin (1.30 μg/L) (Fu et al. 2015); and the Mid-level area, Hong Kong (0.95 μg/L) (Leung and Jiao 2006) (Table 4). However, the highest concentration of Pb measured in the study area (22.0 μg/L) was from a groundwater sample in the northern sub-district of Jiangjin which was about twice the recommended drinking water limit for this element (10 μg/L) (MHPRC 2006; WHO 2011). The presence of locally elevated concentrations As, Se, and Pb in potential drinking water sources in the study area means that an additional health risk assessment should be undertaken to determine the potential health impacts of this source of contamination.

Mechanisms controlling the major ion chemistry of water in the study area

Dissolved chemical constituents in river water and groundwater can be derived from a number of sources including: sea salts carried in the atmosphere and deposited on land and in surface water bodies inland (cyclic salts); the weathering of silicate, carbonate and sulfide minerals; the dissolution of evaporate minerals; and anthropogenic inputs (Berner and Berner 2012).

Cyclic salts

The dissolved matters in water could be from sea salts carried in the atmosphere and deposited in precipitation or dry deposition (Berner and Berner 2012). However, the average detention time of sea salts in the atmosphere is only about 3 days (Junge 1972). As a result, the contribution of cyclic salts to riverine-dissolved salt loads tended to decrease with increasing distance inland from marine

source areas of atmospheric precipitation (Stallard and Edmond 1981). Na^+ and Cl^- had less adsorbance to soils than metallic elements do (Stallard and Edmond 1983). The mean value of mNa/mCl (on meq/L basis) from sea salts in natural water was about 0.86 (Stallard and Edmond 1983). However, the value of mNa/mCl in river water and groundwater from the rural area of Jiangjin varied from 0.11 to 46.32, with an average of 4.04. Consequently, it is considered to be unlikely that sea salts contribute a significant proportion of Na^+ and Cl^- in river water and groundwater in the study area.

Weathering

Gibbs (Gibbs 1970) proposed that atmospheric precipitation, rock weathering, and evaporation crystallization were the three major mechanisms controlling the chemistry of groundwater and surface water bodies. The weathering of different host rocks can yield different combinations of dissolved cations and anions (Gibbs 1970; Stallard and Edmond 1981, 1983; Zhang et al. 2011). For instance, Ca and Mg ions are mainly from the weathering of carbonates, silicate rocks and evaporites; Na and K ions are derived mainly from the weathering of silicate minerals and evaporates; and silica is supplied by the weathering of silicate minerals.

Plots of TDS versus the weight ratio of Na/(Na + Ca) and Cl/(Cl + HCO_3) were undertaken for samples from the study area to determine the likely sources of these ions (Fig. 4). These plots suggest that major ions in river water and groundwater from the rural area of Jiangjin District are derived from rock weathering (Fig. 4).

To explain the relative importance of different weathering regimes, ternary plots of cations (Fig. 5a) and of anions and silica (Fig. 5b) were established. Most sites fall in the cluster towards the Ca^{2+} apex and HCO_3^- apex, suggesting that major ion chemistry of river water and groundwater in Jiangjin is dominated by the weathering of carbonate minerals. However, a few sites fall towards the $Na^+ + K^+$ apex and $Cl^- + SO_4^{2-}$ apex, suggesting that they were influenced by the weathering of evaporite minerals. Additionally, SiO_2 comprised about 20% of the total anions and silica in some water samples, suggesting that silicate weathering also plays an important role in determining the major ion composition of natural waters in the study area.

Although the most common rock types distributed in Jiangjin District are clastic rocks including mudstones, siltstones and sandstones (Fig. 1), the primary and secondary cements these rocks are carbonates and mud (Zhang 2009), respectively. Consequently, the preferential weathering of the carbonate cements in these rocks is likely to be the dominant source of dissolved ions in waters, augmented with minor weathering of silicate minerals in these rocks.
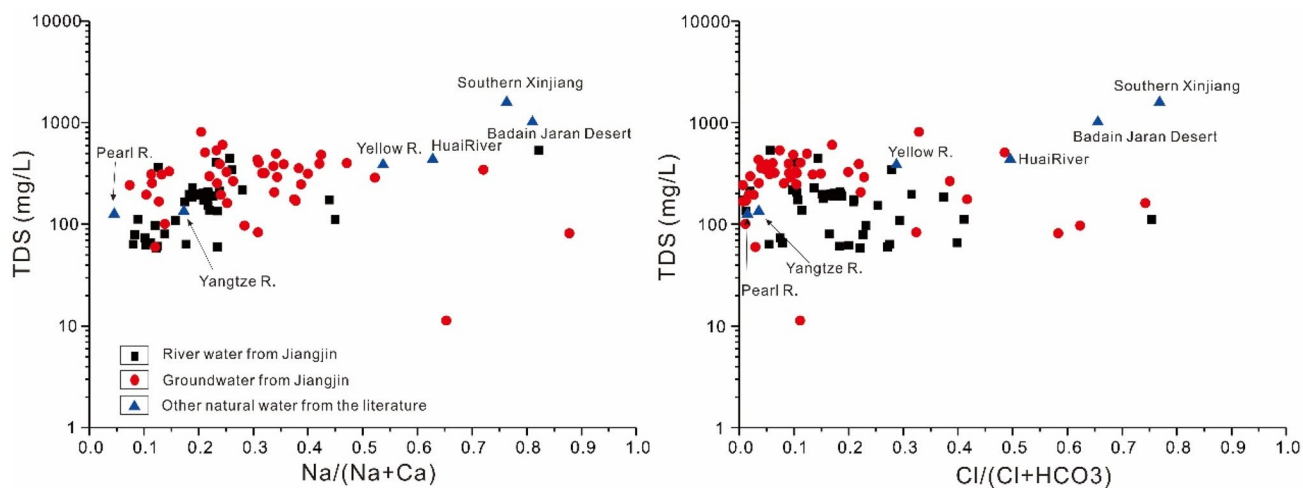
Fig. 4 The Gibbs graph of natural water from Jiangjin. References: Southern Xinjiang (Pang et al. 2010; Zhang et al. 1995); Badain Jaran Desert (Yang and Williams 2003); Yellow river basin (Chen et al. 2005); Huai river basin (Zhang et al. 2011); Pearl River Basin (Chen and He 1999); Yangtze River Basin (Chen et al. 2002)

Fig. 5 Ternary plots showing the relative abundances of cations, anions and silica
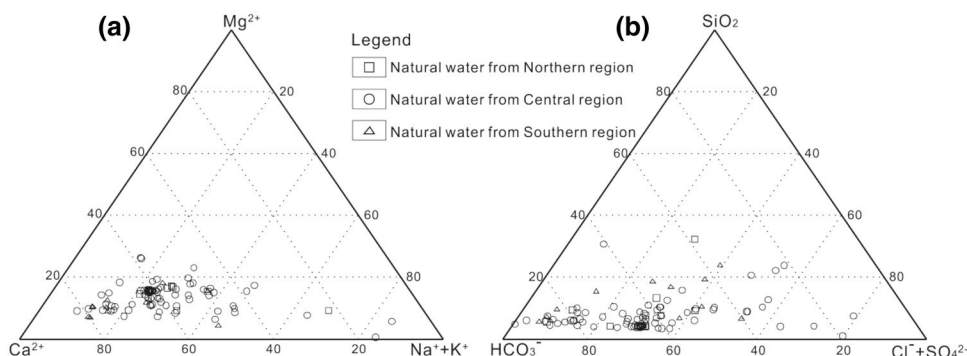
Anthropogenic sources of chemical constituents

Agricultural activities can adversely affect water quality in many watersheds (Liu et al. 2014; Meybeck 2003; Meybeck and Helmer 1989; WHO 2011). In particular, the use of nitrogenous fertilizers use has caused extremely high nitrate concentrations in natural water of many areas (Li et al. 2009a; Liu et al. 2010; Niu et al. 2017; Zhang et al. 2011). In the study area, nitrate and nitrite show a different spatial variability (Table 1). Compared to the mean concentration of NO_3^- in river water (13 mg/L) and groundwater (16 mg/L) (Table 1), ten groundwater samples collected from central sub-district in Jiangjin show higher concentrations of nitrate (32–164 mg/L) than elsewhere in the study area. This suggests that this part of the study area, which is the main center of agricultural, livestock and poultry production in the region, had been greatly influenced by the local use of agricultural fertilizer and by the discharge of animal wastes to ground.

Health risk assessment

In this study, the HQ values of all hydrochemical solutes using their average concentrations was less than 1×10^{-6} (Table 5), indicating that these hydrochemical solutes are not a significant health hazard in drinking water (Giri and Singh 2015; Wu et al. 2009a). The order of HQ values was determined to be $Cl^- > NO_3^-$ $> F^- > As > NO_2^- > Fe > Cr > Ba > Se > Pb > B > Mn > Mo$ $> Ni > Cu > Zn > Cd > Ag$ for adults and children (Table 5).

The range of HQ values for Cl^- was 1.49E−09–6.69E−07 for adults and 3.74E−09–1.68E-06 for children, suggesting that elevated Cl^- concentrations in drinking water have the potential to cause adverse health effects at some locations and is a potential non-carcinogenic health concern for children. And the maximum HQ of other hydrochemical elements was lower than 1×10^{-6}, suggesting that only Cl^- was of a potential non-carcinogenic heath concern.

Table 5 Hazard quotient (HQ) and hazard index (HI) for harmful hydrochemical elements in natural water in Jiangjin

Elements	Jiangjin		Northern		Central		Southern	
	HQ_{adult}	HQ_{child}	HQ_{adult}	HQ_{child}	HQ_{adult}	HQ_{child}	HQ_{adult}	HQ_{child}
Cl^-	5.64E−08	1.42E−07	5.02E−08	1.26E−07	6.29E−08	1.58E−07	1.75E−08	4.40E−08
NO_3^-	3.36E−09	8.44E−09	1.88E−09	4.72E−09	3.87E−09	9.72E−09	1.31E−09	3.28E−09
NO_2^-	9.29E−10	2.33E−09	2.05E−10	5.15E−10	1.09E−09	2.75E−09	5.16E−10	1.30E−09
F^-	9.99E−10	2.51E−09	1.05E−09	2.64E−09	1.07E−09	2.68E−09	4.87E−10	1.22E−09
Ag	4.05E−13	1.02E−12	3.65E−13	9.18E−13	4.21E−13	1.06E−12	3.31E−13	8.31E−13
As	9.84E−10	2.47E−09	4.69E−09	1.18E−08	5.71E−10	1.43E−09	1.32E−10	3.32E−10
B	1.24E−10	3.11E−10	2.06E−10	5.18E−10	1.14E−10	2.86E−10	1.09E−10	2.74E−10
Ba	2.38E−10	5.97E−10	1.77E−10	4.45E−10	2.59E−10	6.50E−10	1.53E−10	3.83E−10
Cd	1.98E−11	4.99E−11	1.20E−11	3.01E−11	2.24E−11	5.63E−11	1.01E−11	2.53E−11
Cr	2.56E−10	6.42E−10	3.72E−10	9.35E−10	2.57E−10	6.45E−10	1.30E−10	3.26E−10
Cu	3.47E−11	8.71E−11	9.94E−12	2.50E−11	4.24E−11	1.06E−10	6.24E−12	1.57E−11
Fe	2.65E−10	6.67E−10	3.32E−10	8.33E−10	2.59E−10	6.52E−10	2.41E−10	6.04E−10
Mn	5.32E−11	1.34E−10	6.43E−11	1.61E−10	5.90E−11	1.48E−10	2.50E−12	6.27E−12
Mo	5.08E−11	1.28E−10	6.19E−11	1.55E−10	5.06E−11	1.27E−10	4.16E−11	1.05E−10
Ni	4.68E−11	1.18E−10	4.71E−11	1.18E−10	5.07E−11	1.27E−10	1.96E−11	4.93E−11
Pb	1.26E−10	3.16E−10	1.16E−11	2.92E−11	1.55E−10	3.89E−10	3.96E−11	9.94E−11
Se	1.55E−10	3.90E−10	1.27E−10	3.18E−10	1.62E−10	4.08E−10	1.36E−10	3.42E−10
Zn	2.32E−11	5.83E−11	5.56E−12	1.40E−11	2.52E−11	6.32E−11	2.74E−11	6.88E−11
HI	6.41E−08	1.61E−07	5.94E−08	1.49E−07	7.10E−08	1.78E−07	2.09E−08	5.24E−08

Northern: Northern area of Jiangjin; Central: Central area of Jiangjin; Southern: Southern area of Jiangjin

The HI values of potentially harmful hydrochemical elements in river water and groundwater were ranked in the order: the central sub-district (between 7.10E−08 and 1.78E−07) > northern sub-district (between 5.94E−08 and 1.49E−07) > southern sub-district (between 2.09E−08 and 5.24E−08) ($p = 0.001 < 0.05$; Kruskal–Wallis test). This difference is likely to be due to the intensity of agricultural activities in the central sub-district by comparison with other parts of the study area. The use of potassium (mainly potassium chloride) and nitrogenous fertilizers and excrement of livestock are likely to increase the content of Cl^-, NO_3^- and NO_2^- in river water and groundwater in the central sub-district (Li et al. 2009a; Zhao et al. 2011).

The average annual carcinogenic risk of As, Cd and Cr consumed in river water and groundwater from the study area was 4.43E−07 and 1.11E−06, 6.05E−08, respectively, for adults and 1.52E−07, 3.14E−05 and 7.90E−05, respectively, for children. Carcinogenic risk

values between 10^{-6} and 10^{-4} suggest that there is a potential health risk that requires further assessment (Wu et al. 2009a). Moreover, the order of carcinogenic risk was: Cr (between 3.14E−05 and 7.90E−05) > As (between 4.43E−07 and 1.11E−06) > Cd (between 6.05E−08 and 1.52E−07) for adults and children (Table 6).

Therefore, the results reported in this study suggested that the Cr and As in river water and groundwater in rural area of Jiangjin pose potential health risk to rural residents who use this water as a drinking source, especially for children. The magnitude of the carcinogenic risk of Cr and As for adults and children was in the order of the northern sub-district (between 4.58E−05 and 1.15E−04 for Cr and between 2.11E−06 and 5.30E−06 for As) > central sub-district (between 3.16E−05 and 7.94E−05 from Cr and between 2.57E−07 and 6.45E−07 for As) > southern sub-district (between 1.59E−05 and 4.00E−05 for Cr and

Table 6 Cancer risk for As, Cd and Cr in water in rural area of Jiangjin

Elements	Jiangjin		Northern		Central		Southern	
	R_{adult}	R_{child}	R_{adult}	R_{child}	R_{adult}	R_{child}	R_{adult}	R_{child}
As	4.43E−07	1.11E−06	2.11E−06	5.30E−06	2.57E−07	6.45E−07	5.94E−08	1.49E−07
Cd	6.05E−08	1.52E−07	3.66E−08	9.19E−08	6.83E−08	1.72E−07	3.07E−08	7.71E−08
Cr	3.14E−05	7.90E−05	4.58E−05	1.15E−04	3.16E−05	7.94E−05	1.59E−05	4.00E−05
Total	3.19E−05	8.02E−05	4.79E−05	1.20E−04	3.19E−05	8.02E−05	1.60E−05	4.03E−05

Northern: Northern area of Jiangjin; Central: Central area of Jiangjin; Southern: Southern area of Jiangjin

between 5.94E−08 and 1.49E−07 for As) ($p = 0.004 < 0.05$ and $p = 0.04 < 0.05$; Kruskal–Wallis test).

There is sufficient evidence to demonstrate the carcinogenicity in humans of chromium and arsenic (Johnson et al. 2006; Villaescusa and Bollinger 2008; Wen et al. 2013). Chromium sources can often be ascribed to anthropogenic activities (Johnson et al. 2006) and weathering of Cr-bearing mineral (Liu et al. 1984; Fallahzadeh et al. 2018). Sandstone, shale, and carbonate rocks in Jiangjin District have a low Cr content (Zhao 2015; Wang et al. 2018a, b). Therefore, elevated Cr in water is not largely derived from geological weathering. Chromium is an important metal used in stainless and alloy steels, automobile, leather tanning, chrome plating, and chemicals (Alimonti et al. 2000; Giri and Singh 2015; Johnson et al. 2006). Additionally, the northern subdistrict in Jiangjin contains industrial areas that are potential sources of this metal for groundwater and surface water in the area.

Traditionally, high-As groundwater is controlled by lithology, geologic structure, landform, hydrochemical environment, and climate condition and so on (Smedley and Kinniburgh 2002; Tang et al. 2013; Villaescusa and Bollinger 2008). High concentrations of arsenic in natural waters are often derived from rocks that contain high concentrations of this element. In the vicinity of the study area, these include arseniferous coal-bearing strata in the Hetao Plain (As: 2–10 mg/kg) (Zhang and Wen 2010). High-As groundwater is mainly distributed in Mesozoic–Cenozoic fault basin (Tang et al. 2013; Zhang and Wen 2010), Cenozoic coastal plain (Liao et al. 2011), and diluvial–alluvial plains (Zhou et al. 2017). High As concentrations in groundwater are also controlled by hydrogeochemical environment (Tang et al. 2013; Wen et al. 2013) and are all found in reducing or oxidizing conditions, which helped to release As from sorption sites in ferruginous minerals (Liao et al. 2011; Park et al. 2006; Wen et al. 2013; Zhang and Wen 2010; Zhou et al. 2017). Moreover, groundwater flow system in closed basin is favorable for As enrichment (Liao et al. 2011; Park et al. 2006; Tang et al. 2013; Zhou et al. 2017). Examples of such groundwater flow systems include those on the Hetao Plain, the Datong Basin, and the Tarim Basin (Smedley and Kinniburgh 2002; Wen et al. 2013). In addition to these, high-As groundwater in China is mainly located in arid and semi-arid regions, such as Kuitun of Xinjiang province (Zhou et al. 2017), Hetao Plain of Inner Mongolia (Zhang and Wen 2010) and so on.

The northern part of Jiangjin with total annual rainfall of 1031 mm is situated at the southern margins of a regional parallel ridge–valley landform. The land dips to the south which increases drainage from the area (Li 2013). The outcropped lithology in northern Jiangjin mainly consists of clastic rocks and carbonate rocks that generally have a low As content (Fig. 1) (Liu et al. 1984; Taylor and McLennan 1985). Consequently, the northern part of Jiangjin is not considered to have suitable geographic, geological, climatic and hydrogeochemical conditions to produce naturally elevated concentrations of arsenic in groundwater. Consequently, it is considered to be more likely that the measured elevated arsenic concentrations in groundwater in this area were derived from human activities, industrial land uses and the use of coal-fired power generation.

The average annual carcinogenic risk was 10^2–10^3 times higher than the average annual non-carcinogenic risk (Tables 5, 6), indicating that the health risk associated with using river water and groundwater as a drinking source in the rural area of Jiangjin mainly came from the carcinogenic heavy metals, especially Cr and As. Drinking water contaminated by As and Cr in the long time may potentially cause carcinogenic effects or other diseases (Phan et al. 2013; Villaescusa and Bollinger 2008; Wen et al. 2013). Therefore, the potential effects of elevated As and Cr concentrations for local rural residents should be carefully managed in the area, particularly the potential impacts on children. In future, Cr, As and other high mental elements should become priority-controlled pollutants by local government authorities in the region.

In the study, although the mean concentration of metal elements such as Cr and As in river water and groundwater was far below the recommended limits for drinking water, the carcinogenic risk of these exceeded the recommended value set by the International Commission on Radiological Protection (ICRP). Therefore, it is recommended that the water quality guidelines should be jointly used with health risk assessment to ensure that susceptible populations are fully protected from potential health problems associated with the use of natural waters as drinking sources in the region.

There might be some uncertainties in this study, which included parameters obtained from USEPA and WHO might not be specific to Chinese conditions (Wu et al. 2009a). Additionally, the manner in which concentrations of solutes varied with time in water in the study area could not be determined in this study. Similarly, concentrations of many chemical constituents in water in the region are likely to be more variable that indicated in this study. The limited number of samples collected in this study led to the underestimation of the spatial distribution of some dissolved solutes, especially for As and Cr. Therefore, detailed sampling and risk characterization would be required to fully evaluate the health risk that the use of local water sources for drinking poses to rural communities in the region.

Conclusion

There was significantly spatial variability in the concentration of major ions and other dissolved chemical constituents in river water and groundwater in rural area of Jiangjin

District in China. The mean concentrations of TDS and NO_3^- were highest in the central sub-district of Jiangjin (251 mg/L and 16 mg/L for TSD and nitrate, respectively), followed by average levels in the northern sub-district (234 mg/L and 8 mg/L for TDS and nitrate, respectively), and by low levels in the southern sub-districts (145 mg/L and 6 mg/L for TDS and nitrate, respectively) ($p = 0.033$ and $p = 0.039$; Kruskal–Wallis test). At some locations, the concentrations of NO_3^-, NO_2^-, Al, As, B, Ba, Fe, Mn, Ni, Pb and Se exceeded their respective recommended Chinese and WHO drinking water limits.

At most sampling sites, Ca^{2+} and HCO_3^- were the dominant ions in river water and groundwater, suggesting that the chemical composition of natural water in Jiangjin was mainly dominated by the weathering of carbonate minerals. SiO_2 occupied above 20% of the total anions and silica in some river water and groundwater samples, suggesting that silicate weathering also played an important role in determining chemical composition of water in the region. Generally, the weathering of carbonate and silicate minerals was the primary and secondary contributors for the major ions, respectively. Concentrations of NO_3^- were generally high in areas where intensive agricultural activities were conducted, predominantly in the northern and central rural areas of Jiangjin.

A human risk assessment using an exposure risk assessment model indicated that Cl^- and NO_3^- were the most important chemical constituents for the assessment of non-carcinogenic health risks for drinking water. The highest non-carcinogenic health risks associated with the use of drinking water were distributed in the central rural area of Jiangjin where most of the cultivated agriculture and livestock rearing takes place in the region.

The average annual carcinogenic risks for Cr in drinking water were determined to be in the range of 3.14E−05 and 7.90E−05 for adults and children, respectively. Similarly, the average annual carcinogenic risks for As in drinking water for adults and children were calculated to be in the range of 4.43E−07 and 1.11E−06, respectively. These values are within the risk values of 10^{-6} to 10^{-4}, values that generally indicate that there are health concerns that need to be addressed in more detail. The highest values of carcinogenic risk for drinking water were mainly located in the northern part of the study area where there are industrial activities that are potential sources of arsenic and chromium in surface water and groundwater.

This study has indicated that water quality (especially in groundwater) in rural parts of Jiangjin has been strongly affected by anthropogenic activities. The average annual carcinogenic risk exceeded the average annual non-carcinogenic risk, indicating that the health risk for the use of river water and groundwater in the study area is mainly associated with elevated concentrations of metals and metalloids, especially for Cr and As. To reduce the estimated risk, the local authorities should make great efforts to control the concentration of Cr and As in river water and groundwater in the area.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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