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RESEARCH ARTICLE

Study and health risk assessment of the occurrence of iron and manganese in groundwater at the terminal of the Xiangjiang River

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Abstract The pollution of the surface water in the Xiangjiang watershed in China has received much attention, whereas the groundwater conditions in the area have long been ignored. This study investigates the occurrence of Fe and Mn in the groundwater of Chengxi Town located at the terminal of the Xiangjiang River. The study area was monitored for an entire year. Thereafter, the associated health risks were identified. Results showed that approximately 81 and 73 % of the measured samples exceeded the limits for Fe and Mn in Chinese drinking water, respectively. pH level was found to be negatively correlated with the concentrations of Fe and Mn in the groundwater in the study area. The occurrence of Fe in the groundwater showed significant seasonal fluctuations and was possibly affected by the change in environment conditions within the aquifer. By contrast, Mn remained relatively stable in most of the area during the whole year. Overall, no health threats for adults and children in the study area were determined according to the low health index values. Nevertheless, research attention and the implementation of relevant measures are needed for certain villages with exceptionally high Mn concentrations in the groundwater.

Keywords Groundwater · Xiangjiang River · Iron and manganese · Lakeshore · Health risk assessment

Introduction

Groundwater serves as the most important source of drinking and irrigation water for many rural areas in China where available treated tap water is scarce (Qiu 2010). As a result of the unequal distribution of water resources and different climate conditions, the southern area of China has more abundant water storage compared with the northern area. For decades, great attention has been paid to the rapid depletion of groundwater in the arid North China Plain (Feng et al. 2013; Li et al. 2012; Zhang et al. 2012). However, in certain places in the south, serious pollution has also led to severe condition in groundwater. A recent survey (Qiu 2011) showed that 90 % of the shallow groundwater in China is polluted and that an alarming 37 % of this water source is so foul that it cannot be treated for use as drinking water. This situation presents a potential health risk for those who heavily depended on the groundwater in the area.

Iron and manganese are important indicators of groundwater quality, but studies on these metals are fewer than those on other trace metals (Chai et al. 2010). Fe and Mn share many similarities including similar ionic radius, valence charge in physiological conditions, absorptive mechanisms for individuals, and being usually enriched in groundwater environment with close correlation (Elsner and Spangler 2005). Although

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these trace elements are indispensable to human survival, excessive exposure to them can damage human health. The excessive intake of Fe can lead to chronic intoxication while the excessive intake of Mn can result in lung embolisms, bronchitis, impotency, and nerve damage, even to the point of parkinsonism (Zoni et al. 2007). Wasserman et al. (2006) showed that the high Mn concentrations in the drinking water in Bangladesh significantly influenced the intellectual functions of children aged 10 years. However, removal of Fe and Mn from groundwater is difficult and expensive. The World Health Organization (WHO) now recommends 0.4 mg/l for Mn as drinking water guideline (WHO 2011). In China, the national standard qualities for groundwater are 0.3 mg/l and 0.1 mg/l for Fe and Mn, respectively (Ministry of Environmental Protection 1994).

Under natural conditions, Fe and Mn in groundwater originate from the dissolution of rock minerals, and their occurrence is controlled by several environmental factors, such as redox status, ions (type and concentration) in water, and microbiological activity (Zaporozec 1981). Human-generated Fe and Mn, similar to many other metals, can enter aquifers through the direct emission of wastewater, leaching of solid wastes, or polluted surface water intrusion. The groundwater–surface water interaction process plays an important role in influencing Fe and Mn concentrations in aquifers of alluvial areas where groundwater is widely utilized for drinking and irrigation. On the one hand, the mutual supply between rivers and aquifers could provoke the appearance of redox-sensitive elements such as Fe and Mn (Bourg and Bertin 1994; Kim et al. 2008). On the other hand, these elements could migrate to aquifers from polluted rivers through interaction (Kayabalı et al. 1999; Von Gunten and Kull 1986). Generally, occurrence and migration of Fe and Mn in aquifers under the groundwater–surface water interaction are a complicated process. Health risk assessments are usually carried out to estimate the probability of adverse health effects in humans who may ingest heavy metals from groundwater during their daily life. However, limited studies have considered the seasonal fluctuations of Fe and Mn concentrations in the aquifers of alluvial areas.

The Xiangjiang River is the second largest tributary of the Yangtze River and is a vital water source for people living, industry, and agriculture production in Hunan Province. A remarkable array of ore deposits has been found in the upper river areas, and with a long-time mining and smelting activities of non-ferrous metals, wastewater typically discharged to the surrounding environment and thus increased the heavy metal levels in the Xiangjiang River (Zhang et al. 2009). To date, numerous studies have investigated the contamination levels in the surface water (Zhang et al. 2010), sediments (Zeng et al. 2006; Li et al. 2013), and surrounding paddy soils (Huang et al. 2014, 2015) of the river; however, little is known about the extent and severity of the problem in the groundwater, especially in the rural areas that use this water source daily (Peng et al. 2011). In this research, we selected a town located at the terminal of the Xiangjiang River as the study area. The shallow groundwater serves as the main source of drinking water for the local residents, whereas the surface water is used for irrigation. A previous study (Jiang et al. 2013) indicated that Fe and Mn concentrations in the local surface water exceed the national standard. Moreover, the groundwater interacts frequently with surface water because of special geologic conditions and seasonal hydrological changes. This interaction further increases the risk of excessive human intake of Mn and Fe.

The objectives of this study are to (1) investigate the contamination levels of Fe and Mn in the groundwater in the terminal of the Xiangjiang River, (2) examine a number of factors that influence the Fe and Mn concentrations in the groundwater, and (3) assess the health risks of the exposure of the local residents to Fe and Mn. Through this study, we intend to provide residents in the locality or in other similar areas with a reference for safe groundwater utilization. We also offer relevant recommendations that the government could consider in establishing a set of cost-effective measures for groundwater remedy and management.

Study area

The town of Chengxi (28° 62′ to 28° 76′ N, 112° 71′ to 112° 87′ E, Fig. 1) is located at the terminal of the Xiangjiang River, which is 60 km away from Changsha City (Hunan Province, China). The region includes 38 villages with a population of over 72,000. The town has a total area of 122 km^2, of which 55.33 km^2 serves as arable land and 14.13 km^2 is utilized for pond culture. Chengxi is completely surrounded by the Xiangjiang River, which flows from south to north and finally flows into the nearby Dongting Lake. Another river (the Zi River) joins the Xiangjiang River in the northwest. As a result of the influence of the subtropical monsoon climate, the annual mean temperature in the area is 17.1 °C and the mean precipitation is 1392 mm, of which 54.4 % occurs during the rainy season (March to July) (Zhang et al. 2008). No industrial production is found in this area. The ditch water is used for irrigation, whereas the groundwater supplies drinking water to a large portion of the town. The groundwater recharge and discharge are affected by the water levels of the Xiangjiang River, precipitation infiltration, and artificial exploitation. Banks reaching 3 to 7 m above land surround the whole town to protect it from river flooding. During the rainy season, floodwater usually causes intrusion phenomenon inside the embankment. Thus, several pump stations near the riverside have been installed to drain intrusive water.

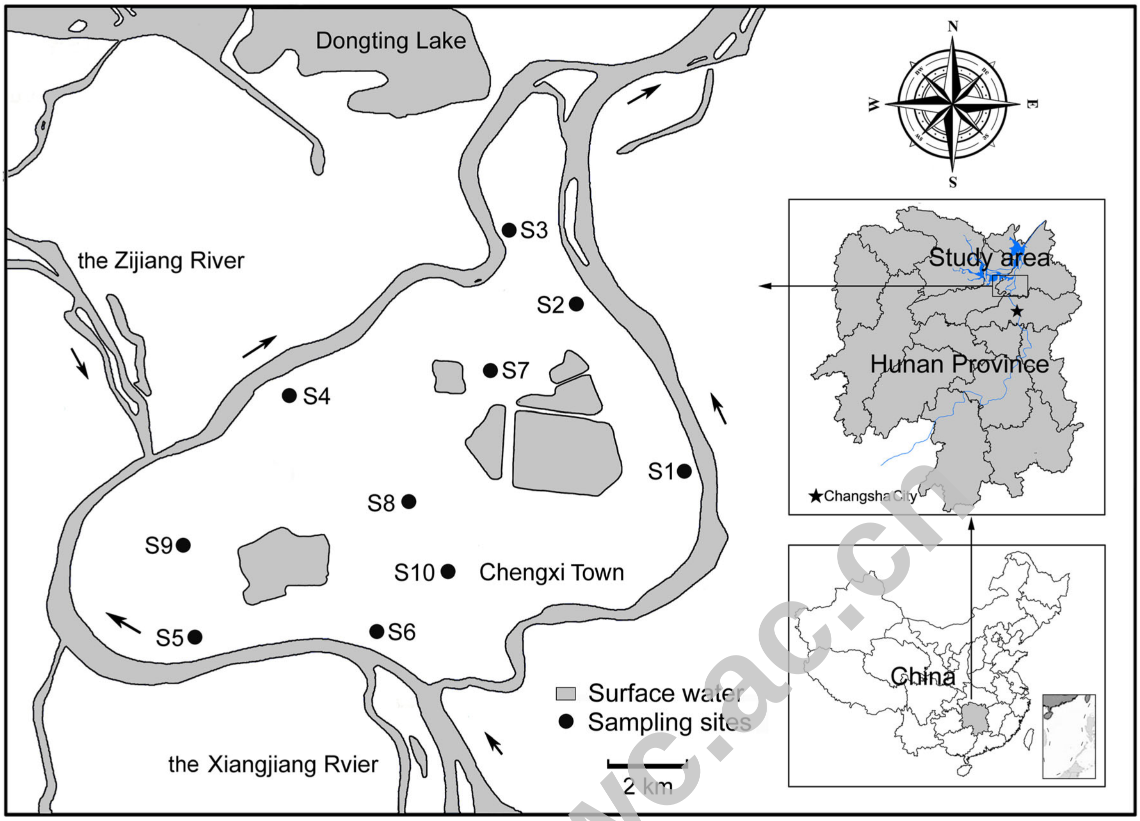

Fig. 1 Location of the study area (Chenxi Town) and distribution of sampling points

Materials and methods

Sample collection

With 2-month intervals between sampling, 30 groundwater samples were collected periodically (in the middle of February, April, June, August, October, and December) from 10 densely populated villages (Fig. 1). The selected villages were Chengxi (S1), Yumin (S2), Xianglin (S3), Gutang (S4), Baoshi (S5), Haohe (S6), Zhonghe (S7), Shunfeng (S8), Hejia (S9), and Jinxing (S10). These villages are geographically distributed evenly and can thus represent the whole study area. Surface water samples from the nearby river (S1, S2, S3, S4, S5, and S6) were also collected. The groundwater samples were mainly obtained from family-owned groundwater tube wells after flushing each tube for several minutes. The water samples were collected and stored in two polythene bottles (500 mL) pre-washed with 5 % (*v*/*v*) HNO_3 solution and deionized water for each sampling point. A few drops of HNO_3 were added into one of the samples used for heavy metal ion analysis; the second sample (not acidified) was used for anion analysis. All the water samples collected in situ were then transported to the laboratory within a few hours and were kept in a refrigerator at 4 °C. Several indicators, including pH, Pb, and SO_4^{2-}, which are closely related to the occurrence of Fe and Mn in groundwater, were tested using different analysis methods.

Sample analysis

All the samples for laboratory analysis were filtered with 0.45-μm cellulose membranes. Reagent blanks and replicate samples were performed as quality control system during all chemical analyses. The pH levels were measured with a HI 3221 pH meter (Hanna Instruments Inc., USA). The concentrations of Fe and Mn, as well as those of other metals (Cu, Zn, and Pb), in the water samples were determined using a TAS-990 flame atom adsorption spectrophotometer (Persee Inc., China). Total N (TN), total P (TP), SO_4^{2-}, and NH_3-N were measured via spectrophotometry (UV-2550, Shimadzu Corp., Japan) (Ministry of Environmental Protection 1990a, b; 2007; 2009).

Health risk assessment model

Fe and Mn are both non-carcinogens according to the division of toxicants, as suggested by the United States Environmental Protection Agency (USEPA). In this work, only the oral intake path was considered because all the other paths were nearly negligible. The human health risk associated with ingesting Fe or Mn was assessed according to a hazard index (HI) as follows (USEPA 2008):

$$HI = \frac{CDI}{RfD} \qquad (1)$$

where *RfD* (mg/(kg·day) is the reference dose of Fe or Mn as suggested by USEPA (0.3 and 0.14 mg/(kg·day) for Fe and Mn, respectively) (USEPA 2002). *CDI* is the chronic daily intake of Fe and Mn and is calculated with the following dose equation:

$$CDI = \frac{C \bullet IR \bullet EF \bullet ED}{BW \bullet AT} \qquad (2)$$

where *C* is the Fe or Mn concentration in environmental media (mg/l), *IR* is the human ingestion rate (l/day), *EF* is the exposure frequency (days/year), and *ED* is the average exposure duration (year). *BW* is the average body weight (kg), and *AT* is the averaging time ($AT = 365 \times ED(d)$). Potential ingestion risks were calculated for two population subgroups, namely, adult and children. In this study, *EF* was supposed to be 365 days/year, and *ED* was considered to be 58 and 10 years for the adults and children, respectively (Ministry of Health 2007). The average body weight values of the adults and children were considered to be 58.6 and 22.3 kg, respectively (Ministry of Health 2007), and their ingestion rates (daily water intake) were 2.2 and 1.8 l/person (Chen et al. 2008), respectively. The values of the parameters were determined according to the statistical mean values in this province in recent years. These data could reasonably reflect the local population characteristics.

Statistical analysis

Statistical analyses including correlation and regression analyses among parameters were conducted using SPSS 10.0 software.

Result and discussion

Characteristics of the measured groundwater quality indicators

Table 1 lists the statistical results for Fe and Mn and the other measured groundwater quality indicators during the whole year, along with their background values in the Dongting Lake basin (Lian and Xiao 2010). Some indicators (especially for Mn, Cu, Zn, Pb, and TN) in the groundwater of the study area showed significantly higher concentrations compared with the background values. Almost all the indicators showed relatively large ranges with max values that were several or even a hundred times larger than the minimum values. For Fe and Mn, their maximum values were 16.9 and 124.4 times larger than their minimum values, respectively. The mean value of Fe was 0.580 mg/l, which was nearly two times that of Fe limit (0.3 mg/l) in Chinese drinking water. The mean value of Mn was 3.79 times larger than the limit of 0.1 mg/l but remained slightly below the WHO health guideline of 0.4 mg/l.

Figure 2 shows the cumulative distribution of the measured Fe and Mn concentrations. Approximately 81 and 73 % of the measured samples exceeded the limit for Fe and Mn in drinking water in China, respectively, and 24 % of the samples exceeded the WHO acceptable limit for Mn. No obvious pollution was observed among the other indicators except for Pb. The maximum and mean values of Pb were respectively15.46 and 2.4 times larger than the drinking water limit of 0.05 mg/l in China. The results suggested that most of the local groundwater was not suitable for drinking. The normal distribution test showed that the Fe concentrations in the groundwater were normally distributed (P=0.557), whereas the Mn concentrations were not. Abnormally high Mn concentrations (Fig. 2b, n=12) were observed in a village (S1) located in the southeast of the town throughout the whole year. The reason for the phenomenon was not determined, but it could be the result of the exogenous manganese pollution existing in the area. Chai et al. (2010) found that the variation ranges of

Table 1 Descriptive statistics of several groundwater quality indicators

Parameters	Units	Min	Max	Mean	Median	CV	Background
Fe	mg/l	0.096	1.625	0.580	0.547	0.310	0.776
Mn	mg/l	0.022	2.737	0.379	0.243	0.579	0.197
pH	–	6.240	7.260	6.876	6.940	0.250	6.730
Cu	mg/l	0.030	0.140	0.081	0.081	0.048	0.001
Zn	mg/l	0.001	0.460	0.068	0.044	0.918	0.014
Pb	mg/l	0.040	0.120	0.773	0.081	0.025	0.0005
TN	mg/l	5.260	56.970	25.210	20.260	0.826	3.646
TP	mg/l	0.010	0.550	0.086	0.048	0.107	0.080
NH_3-N	mg/l	0.003	0.960	0.211	0.167	0.174	0.229
$SO4^{2-}$	mg/l	0.100	29.610	10.484	9.247	8.056	9.880

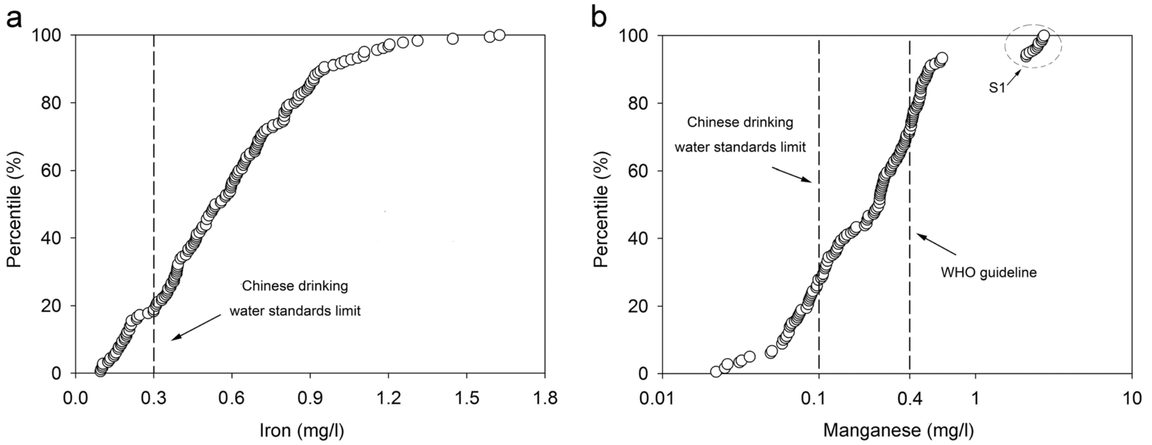

Fig. 2 Cumulative frequency plot for iron (n=180) (**a**) and manganese (n=180) (**b**) in groundwater

Fe and Mn concentrations in the groundwater in Changsha City were 0.1 to 0.48 and 0.16 to 0.34 mg/l, respectively, from 2002 to 2008. This finding indicates that the Fe and Mn concentrations in the groundwater were higher than those in the upper stream. Previous research has confirmed that particular characteristics of alluvial aquifers could provoke the appearance of Mn and Fe, which are sensitive to redox conditions (Kim et al. 2008; Bourg and Bertin 1994). Fe leaching can take place easily under existing anoxic conditions because the local area is primarily covered with laterites, which may be attributed to high concentrations of total Fe in the groundwater.

Relationship among Fe, Mn, and other indicators in groundwater

Correlation analysis was performed to describe the relationship between the pairs of groundwater quality indicators (Table 2). The exceptionally high concentrations of Mn (n=12) in S1 were abnormal data and were thus excluded from the analysis. A good correlation was observed between the pH values and the other five indicators (Zn and TP, $P<0.05$; Fe, Mn, and Pb, $P<0.01$). This correlation implied that such physicochemical parameters in the groundwater were sensitive to changes in environment pH. Especially for Fe and Mn, the Pearson correlation coefficients (−0.946 and −0.643, respectively) were obviously higher than those of other indicators. A close relationship was also observed between Fe and Mn (Pearson correlation coefficient=0.602, $P<0.01$). Regression analysis was performed to further examine the relationship among pH, Fe, and Mn (Figs. 3 and 4). The calculated R^2 indicated that the relationship between pH and Fe could be described well by a liner equation (R^2=0.8947, $P<0.001$). In this case, the pH value could be used to directly estimate the Fe concentrations in the local aquifer. Comparatively, the liner relationships between Mn and pH (R^2=0.4128, $P<0.001$) and Mn and Fe (R^2=0.3622, $P<0.001$) were poor, and predicting the Mn concentrations in the groundwater by using Fe concentrations or pH seemed infeasible. The significant negative correlations of Fe and Mn with pH can be explained by the fact that they are released easily from host rocks and metal–organic compounds under more acidic conditions (Subba Rao 2006). Homoncik et al. (2010) also indicated that Mn exists mainly in the form of reduced soluble Mn^{2+} at lower pH and

Table 2 Correlation matrix of groundwater quality *indicators*

	Fe	Mn	pH	Cu	Zn	Pb	TN	TP	NH_3-N	SO_4^{2-}
Fe	1.000									
Mn	0.602**	1.000								
pH	−0.946**	−0.643**	1.000							
Cu	−0.112	−0.031	0.113	1.000						
Zn	0.443**	−0.031	−0.391*	0.228	1.000					
Pb	0.325	−0.333*	−0344**	−0.093	0.429*	1.000				
TN	0.137	−0.072	−0.249	0.387	−0.159	0.264	1.000			
TP	−0.333**	0.048	0.339*	0.230	0.092	0.215	−0.226	1.000		
NH_3-N	−0.138	−0.008	0.145	−0.05	−0.006	0.007	−0.288	0.082	1.000	
SO_4^{2-}	0.245	−0.215	−0.272	0.631**	0.266	0.741**	0.159	0.270	0.057	1.000

*Correlation is significant at the 0.05 level; **correlation is significant at the 0.01 level

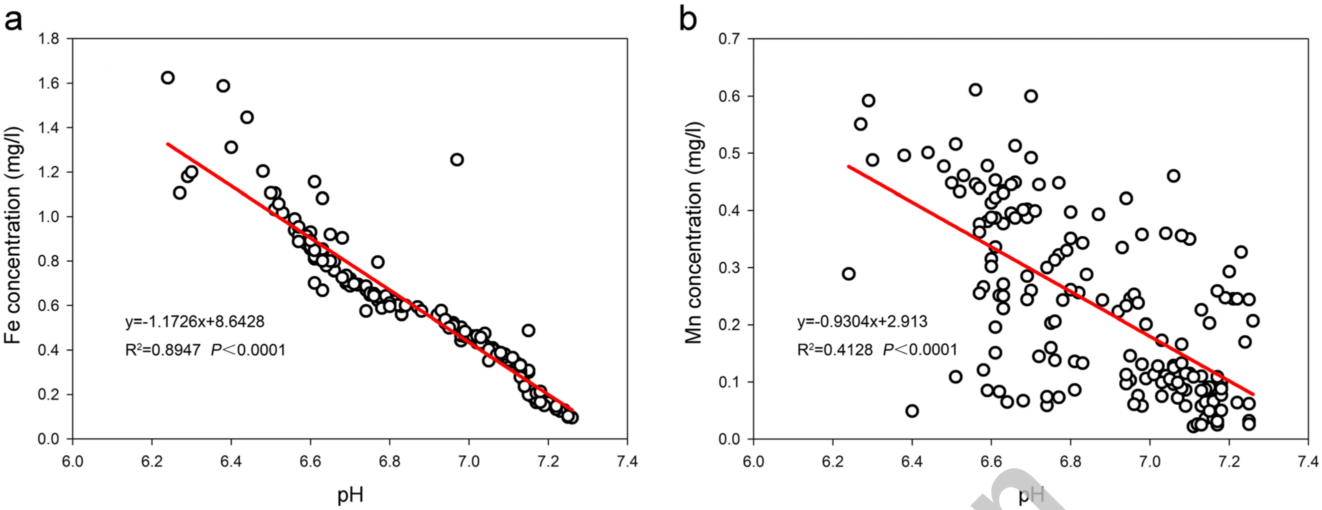

Fig. 3 Relationship between pH and Fe (*n*=180) (**a**), and Mn (*n*=168) (**b**) concentrations

Eh but that it can be oxidized to form precipitates under oxygen and higher pH conditions. For example, the oxidation of organic matter from the rivers leads to the dissolution of Mn oxides from aquifer solids: $CH_2O+2MnO_2+4H^+=CO_2+2Mn^{2+}+3H_2O$ (Bourg and Bertin 1994), and more hydrogen ions in groundwater can accelerate the release of Mn^{2+} into solutions. Mn and Fe are often closely related in natural waters (Homoncik et al. 2010; Collins and Buol 1970; Zhou et al. 2015). Ferric oxides have high affinity for Mn^{2+}. The precipitation of Fe oxides at low Eh may lead to the removal of Mn by occlusion and sorption to precipitated Fe. However, the calculated R^2 between Fe and Mn in the present study was low (R^2=0.3622), which indicated that the occurrence of the two elements in the groundwater was controlled by numerous factors in the groundwater environment, such as Eh, DO, and pH.

Seasonal variation of Fe and Mn in groundwater

Figure 5 shows the temporal variations of the Fe and Mn concentrations in the groundwater and nearby river at the sampling sites S1 (Fig. 5a), S2 (Fig. 5b), S3 (Fig. 5c), S4 (Fig. 5d), S5 (Fig. 5e), and S6 (Fig. 5f) during the whole year. An apparent seasonal fluctuation was observed only in the Fe concentrations. For most of the sampling sites, the highest Fe values appeared in February or April and the lowest appeared in August. The seasonal fluctuation trend of Mn was far less obvious than that of Fe; high values mainly appeared in February. The Fe concentrations in the river showed fluctuation characteristics that were similar to those in the groundwater. Unlike the variation trend in the groundwater, the Mn concentrations in the river exhibited an obvious fluctuation. In most cases, the Fe concentrations in the river were higher than those in the groundwater, and opposite results only appeared during the second half of the year for S3, S4, and S5. For Mn, the concentrations in the river were lower than those in the groundwater for S1, S3, and S4. For S2 and S5, the concentrations in groundwater were higher. The correlation analysis indicated positive correlations between the Fe concentrations in groundwater and those in the river for four villages (S1, S2, S3, and S4), whereas Mn concentrations in only one village (S4) appeared to be positively correlated (Table 3). The occurrence situation of Fe and Mn in alluvial aquifers is determined by several factors, such as the diffusion of pollutants from surface soil and water, degree of rock weathering, oxidation–reduction condition, microbiological activity, and the resulting degradation of organic matter controlled by temperature (Bourg and Bertin 1994; Laluraj and Gopinath 2006; Zhou et al. 2014). As mentioned previously, there is no industrial production and other pollution source in the study area. Therefore, the variation of the two elements probably contributed to the changing of the groundwater environment condition and the influence of the surface water (Xiangjiang River). During the rainy season, the high groundwater table caused by the rising of the river stage made the groundwater environment more reducible, and the suitable temperature could

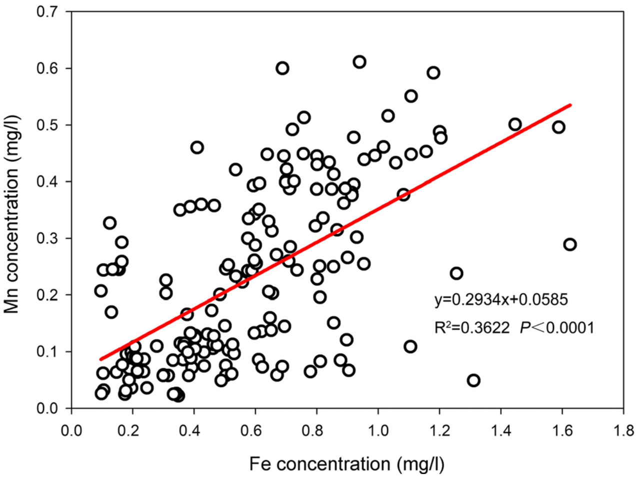

Fig. 4 Relationship between Fe and Mn (*n*=168) concentrations

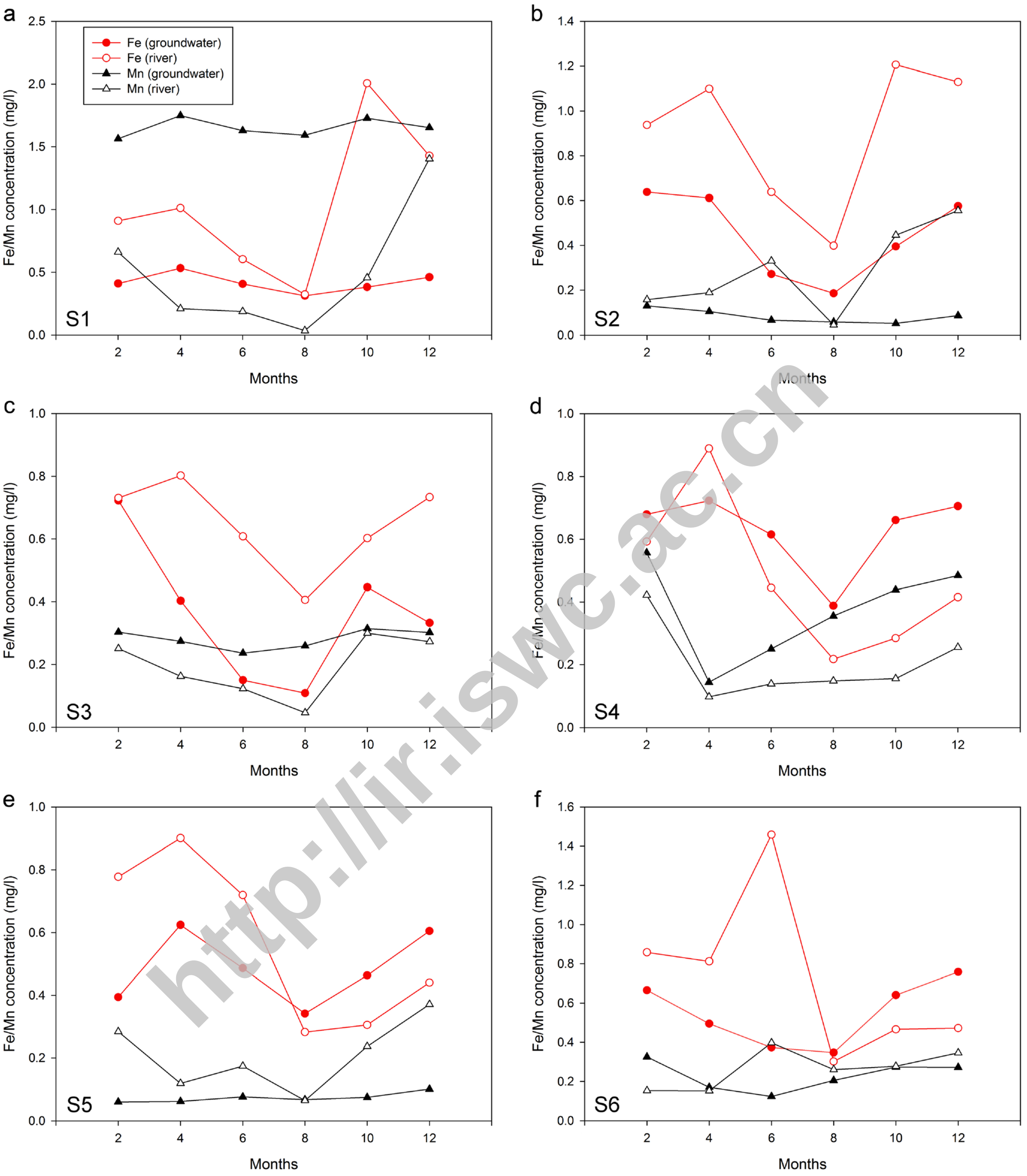

Fig. 5 Temporal variations of Fe and Mn concentrations in the groundwater and nearby river at the sampling sites S1 (**a**), S2 (**b**), S3 (**c**), S4 (**d**), S5 (**e**), and S6 (**f**)

promote microorganism activity which caused the release of the elements in solid phase (Bourg and Bertin 1994; Rajmohan and Elango 2005). The correlation analysis indicated that Mn in the aquifer was possibly not influenced by the river water and that this element did not show any season variation, which may be due to its relatively low concentration (except for S1) in the soil and groundwater environment. Although a good correlation was found between the Fe concentrations in the river and groundwater, determining whether the former influenced the latter was difficult because the

Table 3 The result of correlation analysis between Fe/Mn concentrations in groundwater and nearby river at S1, S2, S3, S4, S5, and S6

Sampling sites	Fe	Mn
S1	0.498 *	0.148
S2	0.613**	−0.189
S3	0.675**	0.416
S4	0.492*	0.745**
S5	0.135	0.341
S6	0.199	−0.031

*Correlation is significant at the 0.05 level; **correlation is significant at the 0.01 level

background value of the area was high (0.776 mg/l). Considering the close relationship between pH and Fe in the groundwater, we speculate that the Fe concentration was more affected by the changes in the environment conditions within the aquifer. Further research is needed to identify whether Fe in the river can infiltrate the aquifer of the area.

Assessment of human health risks

According to our survey, the lack of a safe water supply in the study area forces most local residents to depend on the groundwater as their primary or only source of drinking and household water. As discussed above, the excessive elements including Fe and Mn concentrations in the groundwater pose potential health hazards, particularly when such groundwater is used as drinking water. Therefore, a quantitative health risk assessment is necessary to be carried out in this area. The risk values for both adults and children, who are highly exposed individuals, were determined.

The statistic results of the risk assessment are presented in Fig. 6. A high level of HI corresponds to a high human health threat. Risk values (HI) >1 indicate the potential adverse effects and the need to improve groundwater quality. Generally, no health threat to the adults and children in the study area was determined according to the relatively low mean values (less than 0.25). The maximum HI value of Fe (0.734) was less than 1, which indicated the absence of any related health threat, although Fe in 81 % of the measured samples exceeded the limit for drinking water. As for Mn, the mean values for adults and children are higher than those for Fe, and the maximum value for children ($HI_{children}$) is 1.57. The high $HI_{children}$ values could be attributed to the exceptionally high levels of Mn in Chengxi Village (S1), and they were indicative of high health risks, particularly for the children. The source of foreign Mn in the groundwater of S1 should be identified. Certain measures, such as controlling pollution and lowering Mn levels in groundwater must be employed to reduce health risks. As demonstrated in equation and selected parameters, adverse health effects (HI) increase as the concentration of pollutants increases. Children are more susceptible to the effect of pollution compared with adults. Moreover, health risks during the dry season are higher because Fe and Mn concentrations could reach their maximum values during this period. Nevertheless, the calculated result in this study revealed the absence of any risk of ingesting Fe and Mn from the groundwater in the area.

Ingestion was considered as the sole exposure pathway in this study. Other possible exposure routes include inhalation, daily cleaning, and food chains. In addition, to Fe and Mn, other metals and organic contaminants in the groundwater could pose a threat to human health. These factors were not considered in this study. As a result, the actual health risk was certainly underestimated. Furthermore, the uncertainties during the assessment process (such as the uncertainty with regard to the parameters and the model itself) (Zeng et al. 2009; Wang et al. 2011) made the accurate calculation of the actual health risks relatively difficult. Hence, analyzing the actual health risk of hazardous elements in the local groundwater requires an effective model that considers other factors, as well as uncertainly analysis methods.

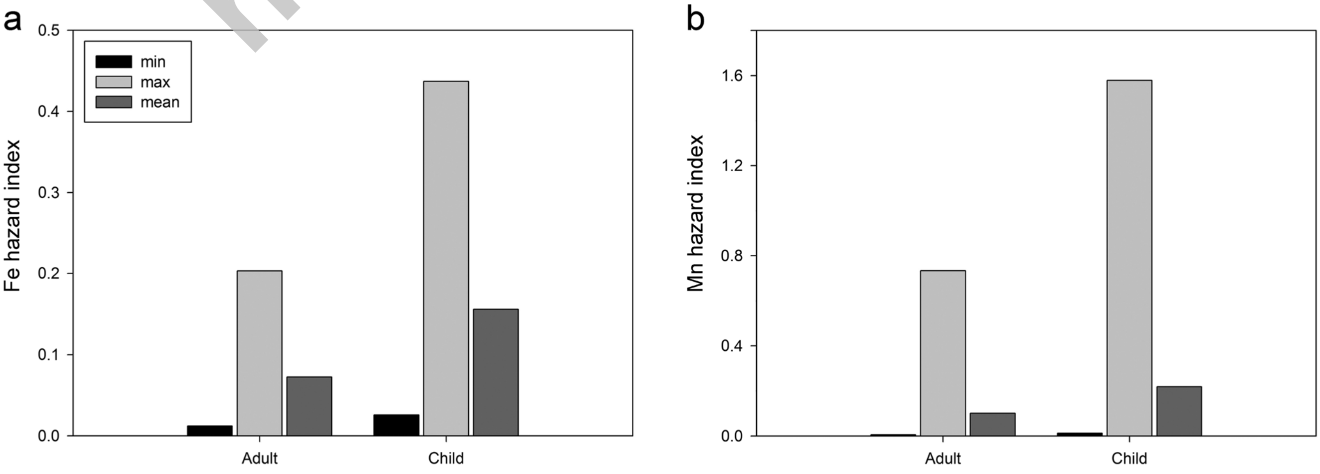

Fig. 6 Hazard indexes of Fe (**a**) and Mn (**b**) for different individuals

Conclusion

This study investigated the occurrence of Fe and Mn in the groundwater of Chengxi Town, which is located at the terminal of the Xiangjiang River. Approximately 81 and 73 % of the measured samples exceeded the limits for Fe and Mn in Chinese drinking water, respectively. The pH value was an important factor that influenced the Fe and Mn concentrations in the groundwater and demonstrated significant negative correlations with Fe in the study area. The occurrence of Fe showed obvious seasonal fluctuations, whereas the Mn concentrations remained relatively stable in the groundwater. Neither of the two elements seemed to be affected by the surface water. In particular, the changes in the environment conditions within aquifer only influenced the occurrence of Fe. The result of the health risk assessment revealed the absence of any health threats to both the adults and children in the study area, as determined from the relatively low HI values. Hence, at present, remediation actions for Fe and Mn pollution in the groundwater are not necessary, except for one village where the Mn concentration was found to be exceptionally high. We recommended long periods of monitoring for the study area, as well as extensive investigation into the whole watershed, to find potential groundwater environmental problems caused by the pollution conditions in the area and the lack of relevant information.

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Compliance with ethical standards The manuscript has not been submitted to more than one journal for simultaneous consideration.

The manuscript has not been published previously (partly or in full).

A single study is not split up into several parts to increase the quantity of submissions and submitted to various journals or to one journal over time.

No data have been fabricated or manipulated (including images) to support the conclusions.

No data, text, or theories by others are presented as if they were our own. Proper acknowledgments have been given, quotation marks are used for verbatim copying of material, and permissions are secured for material that is copyrighted.

Consent to submit has been received explicitly from all co-authors.

Authors whose names appear on the submission have contributed sufficiently to the scientific work and therefore share collective responsibility and accountability for the results.

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

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