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Elements in potable groundwater in Rugao longevity area, China: Hydrogeochemical characteristics, enrichment patterns and health assessments

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ABSTRACT

Rugao city is a typical longevity area taking shallow groundwater as the primary drinking water source. To determine the relationship between longevity and groundwater conditions, the hydrogeochemical characteristics and related causes of potable groundwater were investigated. On this basis, the water quality index (WQI) and hazard index (HI) of groundwater were evaluated. Meanwhile, the nutrient indicators beneficial to human health, like Ca and Mg concentrations, were also considered to explore the relationship. The results were as following: (1) 91.3% of water samples fell under the Ca/Mg–HCO_3 water type, which resulted from the dissolution of silicate rock. Na, Cl^-, Br, B in groundwater emanated from seawater intrusion. The abnormal concentrations of NO_3^- and As also indicated that anthropogenic activities had exerted significant influences on groundwater quality. (2) The average WQI value was 30.19, which meant that the overall groundwater quality in Rugao city was pretty good. However, 8 water samples were found to have HI values above 1, which might be attributed to the high concentration of As (maximum value 0.0407 mg/L; mean value 0.0076 mg/L). In general, low WQI and HI values corresponded to towns with a high longevity population; what's more, WQI and HI values of Rugao city were lower than those of non-longevity areas. (3) Comparing with adjacent non-longevity areas, the potable groundwater in Rugao city had the characteristics of high Ca (mean value 123.57 mg/L), high Mg (mean value 50.33 mg/L) and high SO_4^{2-} (mean value 525.19 mg/L). The daily intake of Ca and Mg from drinking water could meet 12.4% and 22.4% of daily Ca and Mg requirements, respectively. Also, the areas where the Sr and B concentrations were higher usually had higher life expectancy. The high concentrations of Ca, Mg, SO_4^{2-}, Sr and B in drinking water, as well as low WQI and HI values, probably contribute to physical health and longevity. This research helps provide an insight into the relationship between groundwater quality and health and can serve as a reference for drinking water quality management.

1. Introduction

Recently, the relationship between groundwater environment and physical health is a hot issue in the field of medical geology (Selinus et al., 2016). Several studies have looked into As, F, and heavy metals in poor-quality groundwater and their potential risks to human health (Podgorski and Berg, 2020; Sarvestani and Aghasi, 2019; Srivastava and Flora, 2020). In contrast, the groundwater quality in longevity areas receives relatively less attention. Groundwater is the primary source of drinking water in many longevity areas in the world. It is widely known that the groundwater environment is the most basic, active, and influential factor in sustaining human life (Lv et al., 2011). In addition, potable groundwater quality is critical because the beneficial and harmful substances in drinking water are more easily assimilated than those in food (Yousefi et al., 2018). As a result, studying the groundwater environment in longevity areas may aid in the search for the possible causes of longevity and provide valuable insights into the influences of drinking water quality on physical health.

Numerous studies have investigated the relationship between groundwater quality and longevity. These studies proposed that the

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chemical composition of groundwater in longevity areas varied conspicuously compared to non-longevity areas. For instance, the research conducted by Liu et al. (2014) analyzed drinking water characteristics in Tianshan Mountain, Xinjiang, China, and found that the total hardness (TH) of drinking water in longevity region was obviously higher than that in adjacent non-longevity regions. Furthermore, Deng et al. (2018a) investigated and compared the drinking water quality in longevity and non-longevity counties in Hechi city, Guangxi province, China, and they proposed that the contents of trace elements and mineral levels, such as H_2SiO_3, Ca, Na, Fe, Mg, and Li, in drinking water in longevity counties were higher than those in non-longevity counties. The aforementioned studies come up with conclusions by simply comparing the concentration levels of groundwater components in longevity and non-longevity areas. However, the impact of groundwater quality on physical health results from the synergistic effect of various groundwater components. The level of a single indicator can not sufficiently reflect the groundwater quality. Thus, it is not reasonable to reveal the effects of drinking water quality on longevity by directly comparing the single indicator level. For example, another research conducted in Hechi city reported that the concentrations of Mg, Na, Fe, Li and Mn in drinking water in longevity counties were significantly lower than those in non-longevity counties (Cai et al., 2019). This contradicts the findings of Deng et al. (2018a). The comprehensive evaluation of groundwater quality requires the use of multiple indicators which could reveal the groundwater characteristics. Long and Luo (2020) applied the WQI method to evaluate the groundwater quality in the North China Plain. Based on the national drinking water standards, different drinking water components will be assigned weights according to their importance during WQI evaluation; we can then assess the water quality comprehensively. Besides, HI is commonly applied for drinking water quality evaluation (Kubicz et al., 2021). HI can assess the non-carcinogenic risk of human exposure to nitrates, heavy metals, etc., in groundwater from the oral and dermal intake. These evaluation methods provide insights about the water quality in terms of groundwater safety, but they fail to elucidate the influence of Ca, Mg, etc., in groundwater on human health in terms of mineral nutrition (World Health Organization, 2005). As a result, according to WQI and HI assessments and analysis of some nutrient indicators (like Ca and Mg contents), a more comprehensive evaluation of groundwater quality in the longevity area can be obtained. This provides a new approach to reveal the relationship between groundwater quality and longevity.

Rugao city is one of the famous longevity areas in China. There were 134 centenarians per 1 million people, which far exceeded the longevity standard, 75 centenarians per 1 million people, prescribed by the United Nations. Besides, the longevity people in this city have low mobility and present spatial aggregation characteristics (Yang et al., 2005). Moreover, shallow groundwater is the long-term drinking water source for these elderly people. These two characteristics make Rugao city an ideal place to study the relationship between shallow groundwater environment and longevity. In this study, we investigated the distribution of longevity population and collocated shallow groundwater samples to obtain the groundwater composition. Subsequently, WQI and HI were conducted to evaluate the groundwater quality based on these groundwater components; meanwhile, a comparative analysis of nutrient indicators such as Ca and Mg levels in longevity and non-longevity areas was carried out. The objectives of the study are: (1) to analyze the hydrogeochemical characteristics of shallow groundwater in Rugao city and determine the related genesis; (2) to reveal the influence of groundwater quality on longevity in the study area by comparing the spatial distribution of longevity indices and groundwater quality indicators. This research will help us understand the effects of drinking water quality on physical health and human longevity. At the same time, this study could provide a valuable reference to establish drinking water health standards.

2. Materials and methods

2.1. Study area description

Rugao city is within latitude 32°00′ N to 32°30′ N and longitude 120°20′ E to 120°50′ E and has a subtropical humid monsoon climate. The average annual temperature and precipitation are 14.6 °C and 1059.7 mm, respectively. The study area is located in the alluvial plain of the Yangtze River Delta, which has a well-developed canal network. The terrain of the whole city is relatively flat, with no significant variation in climatic conditions. However, there exist evident regional differences in geological background, soil properties, and so on.

The study area is located in the easternmost section of the Yangtze quasi-platform, and its substrate mainly consists of light metamorphic rock series. From Sinian to Paleogene period, huge clastic rocks and carbonate rocks were deposited alternately; the crustal movement was dominated by lifting movement, and several sea transgressions and retreats occurred in the meantime. The Indosinian movement in the late Triassic resulted in folds and fractures in the stratus; thus, northeast-oriented uplifts and sags were gradually formed, which eventually led to the formation of a terrestrial environment. In addition, the Yanshan movement in this area caused many faults in the strata, creating northeast-oriented barrier-type fault zones. In general, the faults were mainly northeast-oriented longitudinal faults, accompanied by northwest-oriented transverse faults and east-west-oriented faults. This geological structure makes the terrain, geomorphology, aquifer structure, etc., vastly different in this area; thus, even in the contiguous regions, the groundwater hydrogeochemistry characteristics present noticeable differences (Zhao et al., 2017).

The groundwater in Rugao city is mainly stored in the Quaternary (Q4) unconsolidated sedimentary sand aquifers and Neogene (N2) sedimentary sand aquifers. The main drinking water in Rugao is extracted from shallow pore aquifers, which mainly consist of Quaternary Holocene alluvial facies having grayish-yellow and grey silty sands and silty fine sands, as well as localized sandy loam soil. The thickness of the aquifer is about 20 m. The buried depth of the water table is from 1 to 3 m on average, and that in some local areas is less than 1 m. The unconfined aquifers and the underlying confined aquifers communicate with each other (see Fig. 1). Hilly mountainous areas in the west are the recharge area of the regional groundwater aquifers. This results in the high groundwater level in the west and low in the northeast and southeast; as a consequence, groundwater flows into the sea in the direction from the west towards the northeast and southeast. Groundwater also discharges into the Yangtze River in the areas close to it. Nevertheless, groundwater flow in Rugao city is limited resulting from the flat topography. Due to the Holocene transgression, the groundwater was salinized; then, it was gradually desalinated by the Yangtze River freshwater and atmospheric precipitation (Zhao et al., 2017). Moreover, Rugao is a highly-developed industrial and agricultural area; thus, the groundwater pollution problems caused by human activities and groundwater over-extraction cannot be ignored. Therefore, water quality is quite complex in Rugao city.

2.2. Demographics

The demographic data was based on the 2018 census data provided by the Rugao city's Civil Affairs Bureau. The data included the total population in different affiliated towns and the total number of people in different age groups. Thus far, there are no unified longevity indices (Deng et al., 2018b). Centenarian prevalence (OC) has always been regarded as one of the most important indicators to measure the level of longevity, which refers to the number of centenarians in every 100,000 people. It can directly reflect the percentage of the population with extreme longevity in an area. Other longevity indices include longevity index (LI) (the proportion of the population over 90 years old to the population above 65 years old, 90+/65+), longevity level (the ratio of

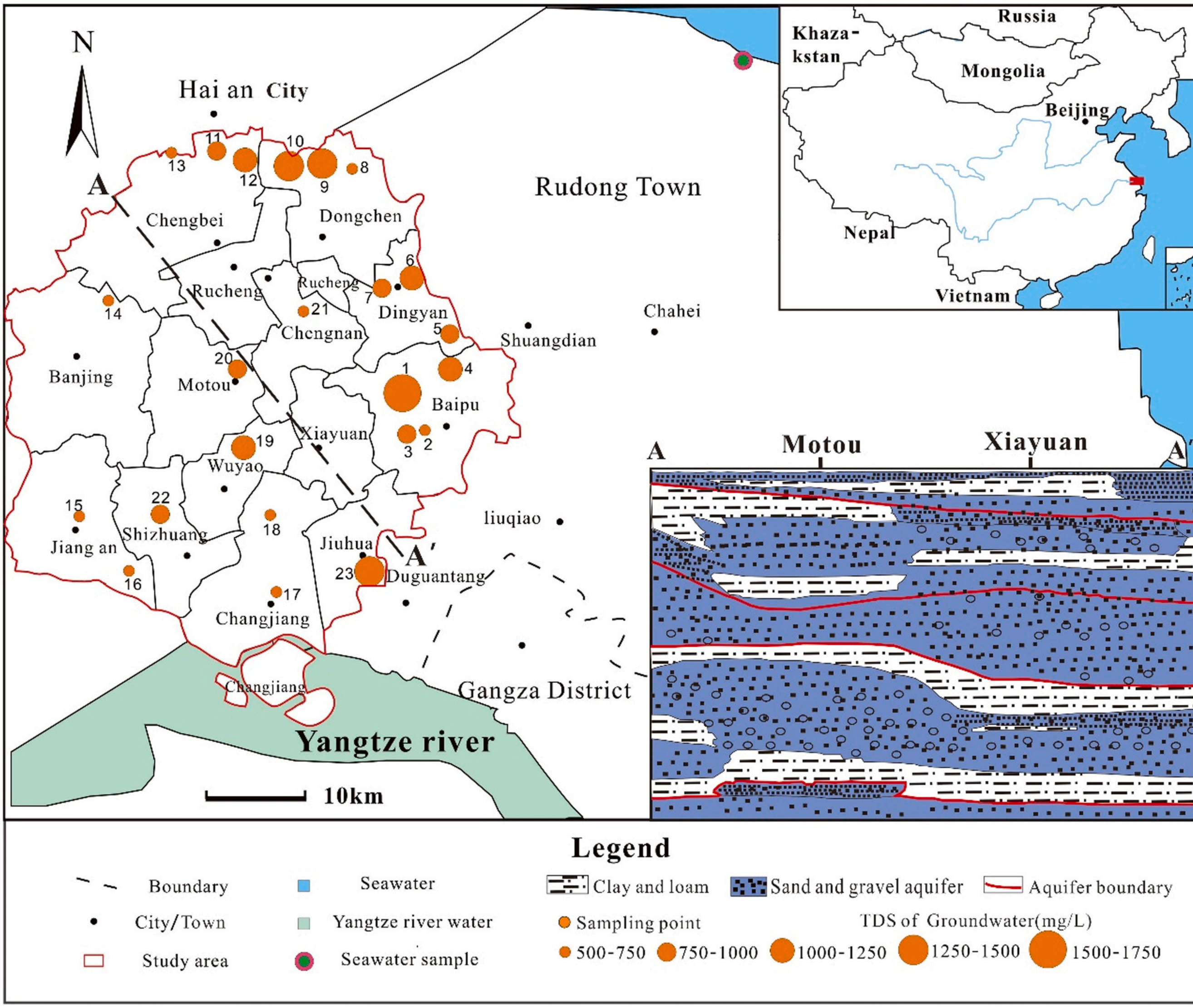

Fig. 1. Distribution of sampling points in the study area and typical hydrogeological section (A–A′) across the study area.

the population over 80 years old to the population above 60 years old, 80+/60+), aging tendency (the proportion of aging population to the total population, 60+/total population), UC index (the number of centenarians per 10,000 people over 65 years old), CI index (the ratio of centenarians to the population over 90 years old), and the proportion of the elderly population at different ages to the total population (80+(%), 85+(%), 90+(%), 95+(%), 100+(%)) (Deng et al., 2018a). Above mentioned indicators are widely used to measure regional life expectancy. Because a single indicator cannot fully reflect the region's longevity level, it is necessary to select several indicators to conduct the studies. In this research, OC, 80+(%), 90+(%), and 100+/80+(%) were used to comprehensively reflect the longevity of Rugao city. Among them, OC and 90+(%) can reflect the proportion of people having an extreme life expectancy; 80+(%) can reflect the potential of future longevity population, and 100+/80+(%) can reduce the effect of age structure on the evaluation of extreme longevity population.

2.3. Sample processing and measurement

In August 2019, 23 groundwater samples were collected from domestic wells located in shallow groundwater aquifers in the study area. The wells were cleaned by pumping water for 5–10 min before sampling. Aside from this, one seawater sample was collected from the coast near the study area (Fig. 1). All groundwater samples were filtered using 0.45 μm filter membranes (Sartorius Minisart) and then collected by two 50 mL HDPE bottles and one 200 mL HDPE bottle. These HDPE bottles had been soaked in acid, then rinsed with deionized water to remove impurities before use. Prior to collection, these bottles were rinsed multiple times with the filtered groundwater sample. For the cation analysis, one water sample in 50 mL HDPE bottle was acidified (pH < 2) using high-grade concentrated nitric acid; the water sample in another 50 mL HDPE bottle was used for anion analysis; the water sample in 200 mL HDPE was used for carbonate titration. All the water samples were kept at 4 °C during transportation to our laboratory. All chemical reagents used in this study were guaranteed reagent grade.

Because some groundwater quality indicators are sensitive to environmental changes, parameters such as TDS, pH, DO, and Eh were measured on-site using a portable multi-meter (Hach, HQ40D). For anion measurement, F^-, Cl^-, NO_3^-, Br^-, and SO_4^{2-} were measured by ion chromatography (IC, 761COMPACTIC, Metrohm AG). CO_3^{2-} and HCO_3^- were determined by standard hydrochloric acid titration method with methyl orange and phenolphthalein as indicators. For cation measurement (Ca, Mg, Na, K, As, Cd, Cr, Pb, Fe, Mn, Cu, Zn, Ag, Al, Sr, Se, Ba, Be, B, Li, Mo, Hg, Co, Sb), the inductively coupled plasma emission mass spectrometer (ICP-MS, Agilent 7700, Agilent) was used. In order to maintain the accuracy of measurement, besides the initial calibration, a new calibration curve was established after measuring every 10 samples. A standard solution (8500–6940, Agilent) was also applied to check ICP-MS measurement accuracy. The recovery rates of standard samples were in the range of 90–110%. In addition, the relative standard deviations of the samples were all below 10%. In data analysis, the concentration of elements that were not detected or below the detection limit were replaced by 0.55 times the detection limit (Gan et al., 2018). The concentrations of major elements and trace elements were rounded off to 2 decimal places and 4 decimal places, respectively.

SPSS software (IBM, edition 23) was applied for statistical analysis of data. In the correlation analysis of groundwater components in the study area, firstly, the single sample K-S test and Shapiro-Wilk test were used to determine whether the samples conformed to the normal distribution. The Spearman model was then applied because Al, Ba, Fe, As, Zn, and Mn severely disobeyed the normal distribution. ArcGIS softwater (edition 10.5) was used to draw the spatial distribution diagrams of the Water Quality Index (WQI), Hazard Index (HI), As and Mn in the research area based on the Kriging interpolation method.

2.4. Groundwater quality assessment and health risk assessment

The groundwater quality of Rugao city was assessed by the WQI method which is widely applied to evaluate the overall quality of surface water and groundwater.

The water quality index for drinking purpose was shown in Eqs. (1) and (2) (Abbasnia et al., 2019)

$$W_i = \frac{w_i}{\sum w_i} \tag{1}$$

Where W_i is the relative weight of element i; w_i is the weight indicator of element i ranging from 1 to 5 (1 means the least effect of the element on water quality; 5 represents the most serious effect of the element on water quality, as shown in Table S1); Σw_i is the sum of the weightings of all elements.

$$WQI = \sum [W_i \times (\frac{C_i}{S_i}) \times 100] \tag{2}$$

Where c_i is the concentration of the element i in the water sample, s_i represents the corresponding upper limit of the element i in *China National Standard for Drinking Water Quality* (GB5749-2006). In general, the groundwater quality can be classified into 5 categories based on different WQI values: excellent quality water (WQI < 50), good quality water (50 ≤ WQI < 100), fair quality water (100 ≤ WQI < 200), poor quality water (200 ≤ WQI < 300), and water unsuitable for drinking (WQI ≥ 300) (Long and Luo, 2020).

In this study, the health risk assessment is evaluated from two main exposure pathways: dermal contact and oral ingestion. Hazard index (HI) is calculated to evaluate the health risk by following equations (Qasemi et al., 2019, 2020).

$$HQ_{ingestion}(individual) = \frac{MC_i \times DR \times EF_i \times ED_i}{BW \times AT \times RfD} \tag{3}$$

$$HQ_{dermal}(individual) = K_p \times MC_i \times \frac{ET \times ED_d \times EF_d \times SA \times 10^{-3}}{BW \times AT} \times \frac{1}{RfD \times GIABS} \tag{4}$$

$$HI = \sum HQ_{ingestion}(individual) + \sum HQ_{dermal}(individual) \tag{5}$$

Where $HQ_{ingestion}$(individual) and HQ_{dermal}(individual) are the health risk caused by oral ingestion and dermal contact of element i, respectively; MC_i is the measured concentration of element i in groundwater sample (mg/L); DR is the water consumption rate (L/day); EF_i is the ingestion exposure frequency (days/year); ED_i is the ingestion exposure duration (years); BW is the body weight (kg); AT is the average time for non-carcinogens (days); RfD_i is the reference dose of element i via oral exposure pathway (mg/kg/day). K_p is the dermal permeability coefficient of pollutant (cm/h); ET is the exposure time (h/day); ED_d is the dermal contact exposure duration (years); EF_d is the dermal contact exposure frequency (days/year); $GIABS$ is the fraction of chemical absorbed in the gastrointestinal tract (its value is 1); SA is the exposed skin area (cm^2); 10^{-3} is the volume conversion factor (L/cm^3). All parameter values are shown in Table S2. Generally, 1 is the threshold value of HI. When HI < 1, the adverse effects on physical health caused by element i could be ignored, and vice versa (Dehghani et al., 2019).

3. Results

3.1. Spatial distributions of the longevity population in Rugao city

As shown in Table S3, there were 440 centenarians among 1.42 million citizens in the study area. Thus, there were 30.9 centenarians pre 100,000 people, which is far more than the average value (2.7 centenarians/100,000 people) in China. The highest ratio of centenarians was found in Baipu town with the OC value of 44.68, followed by Dongcheng town and Jiuhua town; while the lowest OC (15.12) was found in Dingyan town, followed by Wuyao town, Shizhuang town, Jiang'an town and Chengnan street. There were some differences among the proportions of 80+%, 90+% and OC, as seen in Fig. 2. The highest 80+ (%) was found in Baipu town, Dongcheng town, and the lowest was in Rucheng town and Jiang'an town. As to the 90+(%), Baipu town, Dongcheng town, and Jiuhua town had relatively higher values, while Rucheng street and Motou town had the lowest value. The maximum values of 100+/80+(%) were obtained in Baipu town, Jiuhua town, and Dongchen town; whereas the minimum ratios were found in Dingyan town, Wuyao town, and Chengnan street. Although there are some differences in the distribution of these indicators, in general, the longevity population is mainly located in Dongchen town in the northeast of the city, Baipu town in the east of the city as well as Jiuhua town in the southeast of the city, respectively. Generally, the tendency of longevity distribution is declining from east to west and from north to south. The longevity distribution presents a bow shape (see Fig. 2).

3.2. Hydrogeochemical characteristics of groundwater in Rugao city

The main components in groundwater samples were shown in Table S4. After statistics and analysis, the characteristics of these components were listed in Table 1. The pH values of groundwater samples were from 6.90 to 7.50, with an average of 7.15, which meant most of the groundwater samples were weakly alkaline. The TDS ranged from 521.0 mg/L to 1521.5 mg/L; thus, 65.22% of the samples were freshwater (TDS < 1000 mg/L). In Fig. S1(a), the abundance of major cations in groundwater followed the sequence: Ca > Na > Mg > K; the abundance of major anions was in the order of $HCO_3^- > SO_4^{2-} > Cl^- > NO_3^-$. In addition, the TH (as $CaCO_3$) fell in the range of 180.1–895.2 mg/L with an average of 516 mg/L. The diagram of TH vs. TDS (see Fig. S2(a)) presented that most of the groundwater belonged to hard and very hard water except for 2 groundwater samples. Based on the Piper plot (see Fig. S2(b)), the main hydrochemical type of groundwater in the study area was Ca+Mg−HCO_3, accounting for 91.3%.

The data of trace elements in the groundwater in Rugao city were listed in Table S5. Three trace elements (Cd, Hg, and Ag) were not detected. According to Table 2 and Fig. S1(b), Sr had the highest concentration (0.1814–0.9882 mg/L, with an average of 0.5544 mg/L), followed by Br^-, F^-, and B with a mean of 0.2489 mg/L, 0.2362 mg/L and 0.1695 mg/L, respectively. Furthermore, some elements like Ni, Sb, Pb, Mo, Co, Be, and Se, had very low concentrations; their mean values were all below 0.001 mg/L.

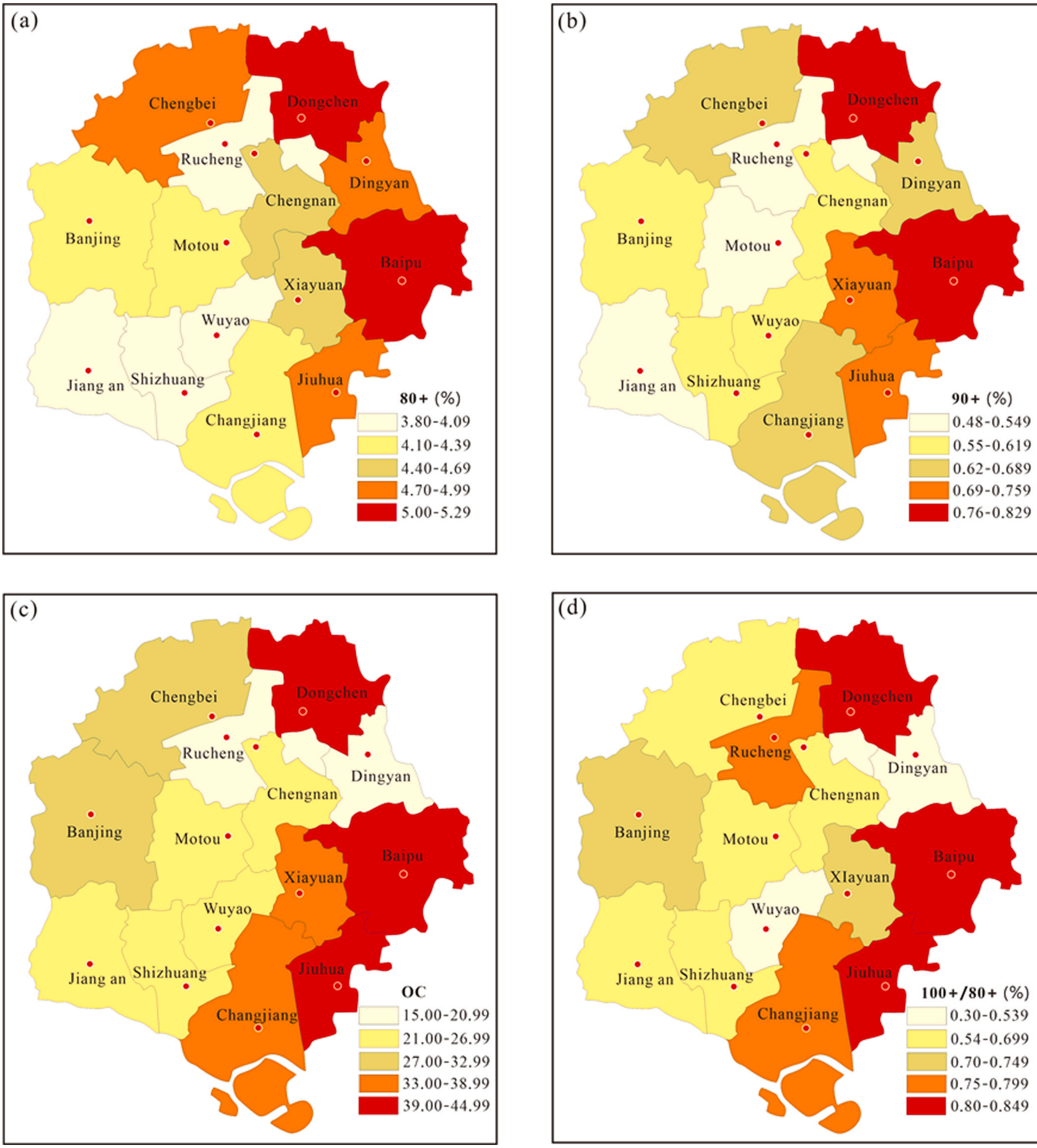

Fig. 2. Spatial distribution of longevity indicators in Rugao city: (a) 80+(%); (b) 90+(%); (c) OC; (d) 100+/80+(%).

3.3. Groundwater quality assessment and health risk assessment in Rugao city

As presented in Fig. S3, the measured groundwater quality parameters were compared with the corresponding values in China National Standard for Drinking Water Quality (GB5749-2006). TDS, TH, and some of the major elements in some groundwater samples exceeded the national standard (Efs value was above the line in Fig. S3). More specifically, there were 15 samples above the standard in TH, accounting for 65.2%; the maximum value of Efs (Efs_{max}) was 1.99. As to TDS, there were 8 samples, accounting for 34.8% and Efs_{max} was 1.52. For Na, Cl^-, NO_3^-, and SO_4^{2-}, there were 1, 1, 2, and 3 samples, respectively. The excessive trace elements were As and Mn. Among them, there were 5 samples exceeding the As standard with the over-standard rate of 21.7%; to Mn, there were 4 samples exceeding the standard ($Efs_{max} = 3.73$) with the over-standard rate of 17.4%.

The application of WQI can help us to understand the groundwater quality comprehensively. The WQI values of the groundwater samples are shown in Table S6 and Fig. 3(a). In general, the WQI values fell in the range of 16.29–50.48 with a mean of 30.19. Among them, only one

Table 1
Statistical analysis of major elements concentration in groundwater in Rugao city.

Unit	Element	Minimum value	Maximum value	Mean value	Median value	SD	Kurtosis Test	Skewness Test
–	pH	6.90	7.50	7.15	7.10	0.19	-1.32	0.29
mg/L	TDS	521.0	1521.5	886.2	841.7	299.19	-0.79	0.56
	Na	24.00	244.17	90.68	80.39	52.54	2.10	1.20
	Mg	23.95	86.20	50.33	44.00	18.93	-1.08	0.49
	K	4.23	20.01	9.98	8.81	4.62	-0.38	0.74
	Ca	22.46	277.49	123.57	116.17	50.13	3.49	0.92
	HCO_3^-	326.59	751.15	525.19	481.71	120.94	-0.69	0.45
	Cl^-	20.50	284.40	101.68	87.44	64.22	1.98	1.41
	NO_3^-	0.03	139.77	35.29	22.31	38.18	2.84	1.71
	SO_4^{2-}	25.83	271.90	119.92	106.50	78.05	-0.37	0.76

Note: SD: standard deviation; –: not applicable;

Table 2
Statistical analysis of trace elements concentration in groundwater in Rugao city.

Items	Cr	Ni	As	Cd	Sb	Hg	Pb	Al	Mo	Sr	Li	Co
MDL	0.00001	0.00002	0.00002	0.00004	0.00001	0.00001	0.00002	0.00006	0.00005	0.00002	0.00001	0.00004
Minimum	< MDL	< MDL	< MDL	n.d.	< MDL	n.d.	< MDL	0.0077	< MDL	0.1814	0.0026	< MDL
Maximum	0.0044	0.0015	0.0407	n.d.	0.0003	n.d.	0.0001	0.1251	0.0021	0.9882	0.0366	0.0003
Mean	0.0009	0.0007	0.0076	n.d.	0.0001	n.d.	< MDL	0.0505	0.0004	0.5544	0.0166	0.0001
Median	0.0005	0.0007	0.0030	n.d.	0.0001	n.d.	< MDL	0.0404	0.0002	0.4962	0.0160	0.0001
SD	0.0010	0.0004	0.0104	n.d.	0.0001	n.d.	< MDL	0.0346	0.0005	0.2055	0.0080	0.0001
Items	Mn	Ag	Be	Cu	Se	Ba	Fe	Zn	B	F^-	Br^-	
MDL	0.00006	0.00001	0.00002	0.00002	0.00001	0.00005	0.00006	0.00004	0.00004	0.0001	0.0001	
Minimum	< MDL	n.d.	< MDL	< MDL	< MDL	0.0074	0.0017	0.0020	0.0285	0.0240	< MDL	
Maximum	0.3732	n.d.	0.0007	0.0024	0.0033	0.1428	0.0804	0.0755	0.5438	0.4428	0.8527	
Mean	0.0386	n.d.	0.0001	0.0011	0.0002	0.0525	0.0213	0.0204	0.1695	0.2362	0.2489	
Median	0.0025	n.d.	< MDL	0.0011	< MDL	0.0425	0.0161	0.0129	0.1556	0.2335	0.1990	
SD	0.0907	n.d.	0.0002	0.0007	0.0008	0.0282	0.0185	0.0204	0.1041	0.1002	0.1984	

Note: The unit is mg/L; Minimum: Minimum value; Maximum: Maximum value; Mean: Mean value; Median: Median value; SD: standard deviation; MDL: Method detection limit; n.d.: Not detected.

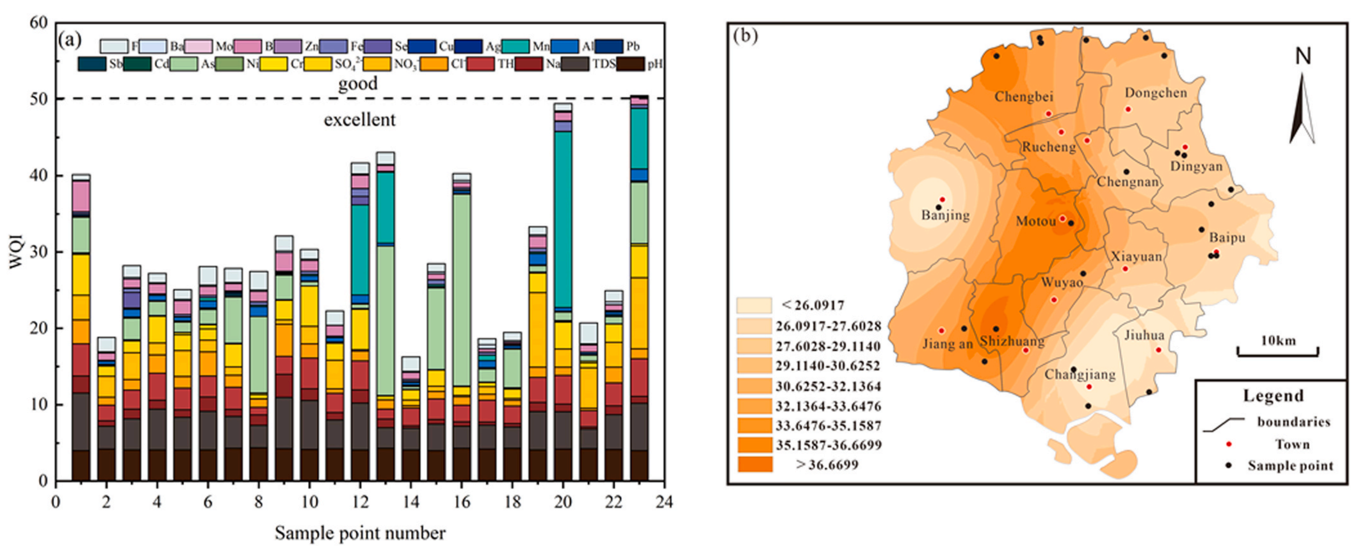

Fig. 3. (a) Bar graph of calculated WQI values and (b) spatial distribution of WQI values of groundwater in the study area.

sample (No. 23) from Shizhuang town had the highest value above 50, which meant the groundwater quality in there was good. The rest part of Rugao city had excellent groundwater quality. The WQI value of the No. 23 sample was ascribed to the high concentrations of As, Mn and NO_3^- in the water sample, which made a significant contribution to the WQI calculation. As shown in Fig. 3(b), the towns with high WQI values were mainly situated in the northwest, central and southwest of Rugao city; whereas, the towns with low WQI values were located in the west, northeast, east and southeast of Rugao city. Except for Banjing town, the place with low WQI presented a curved shape.

The HI values of groundwater samples were shown in Table S6 and Fig. 4. For both children and adults, there were 8 samples whose HI values were above 1. This suggested that the groundwater in these towns had adverse effects on both children's and adults' physical health. Meanwhile, the HI value of children was obviously higher than that of adults, which meant that, compared with an adult, children had a relatively higher risk. According to Fig. 4(a, b), the high concentration of As made the greatest contribution to the high HI value. Fig. 4(c, d) showed that the groundwater samples with high HI values were distributed in the northwest and southwest of Rugao city.

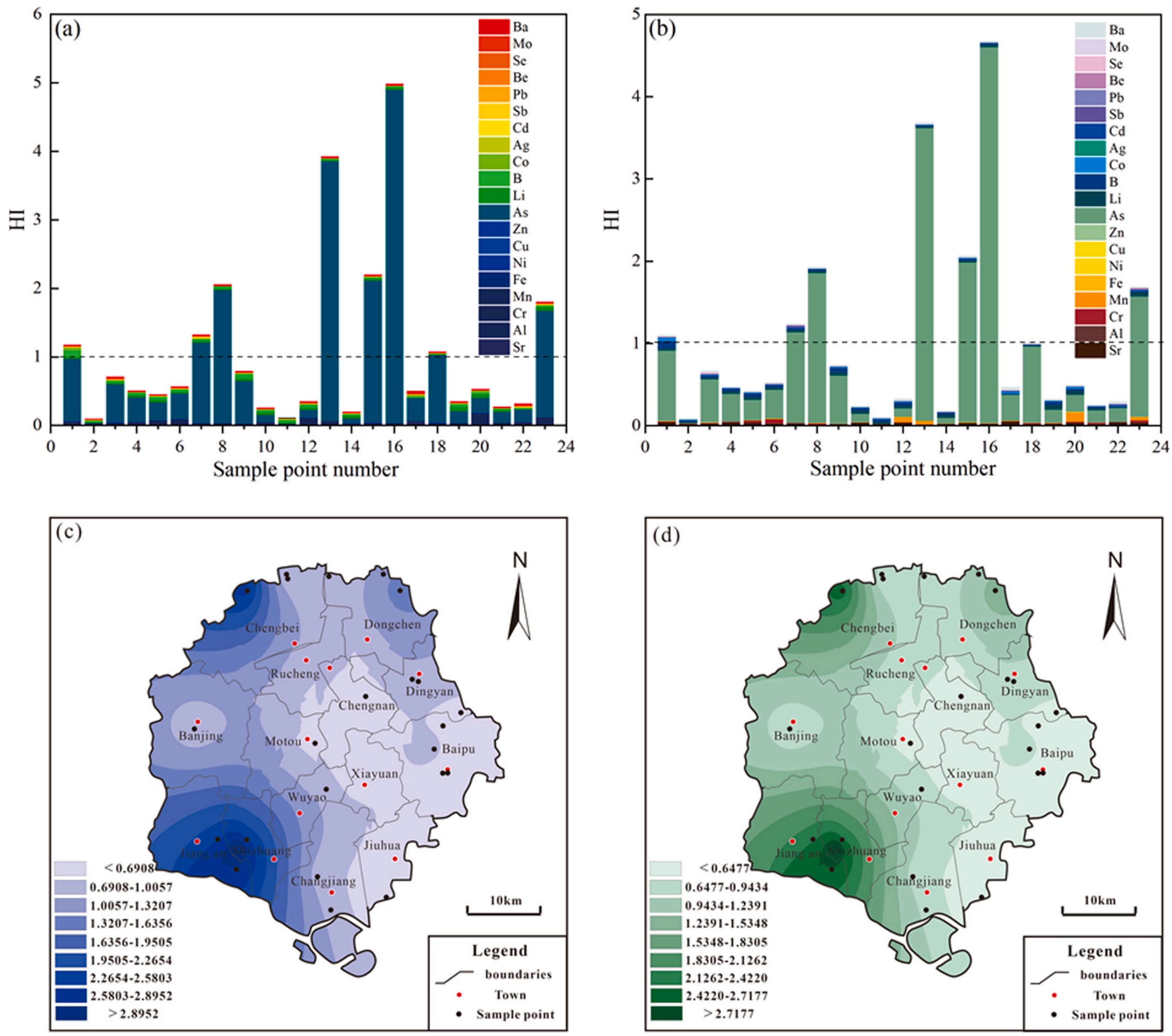

Fig. 4. Calculated HI values of groundwater samples: (a) Children and (b) Adults; spatial distribution of HI in Rugao city: (c) Children and (d) Adults.

4. Discussion

4.1. Causes of groundwater chemistry in Rugao city

The evolution of the chemical composition of groundwater is influenced by factors such as water-rock interaction, evaporation, precipitation and anthropogenic activities (Vasanthavigar et al., 2012). The degree of influence of these factors could be well determined by the Gibbs diagram. The Gibbs diagram was obtained based on the relationships between Na/ (Na+Ca) and TDS as well as $Cl^-/(Cl^-+HCO_3^-)$ and TDS (as shown in Fig. S4). In this research, most of the water samples fell in the water-rock dominance section, which indicated that the water-rock interaction was the main mechanism affecting groundwater chemistry. Some samples located in the evaporation dominance section revealed that the groundwater in these places was affected by evaporation and seawater intrusion (Samsudin et al., 2008). Finally, the groundwater samples situated near the section boundary were probably influenced by anthropogenic activities (Srinivasamoorthy et al., 2008).

In order to further understand the differences in groundwater chemistry resulting from mineral weathering of rocks during the water-rock interaction process, the effects of weathering of carbonate rocks, silicate rocks and evaporative rocks were studied. As Fig. S5 illustrated, the groundwater composition in Rugao was influenced by the weathering of silicate rocks; at the same time, the weathering of carbonate rocks also made a contribution. To further recognize the contribution of weathering of different rocks, the ratios of different ions in groundwater were studied. If the dolomite, calcite, and gypsum are the main weathered minerals, the ratio of Ca+Mg to $HCO_3^-+SO_4^{2-}$ should be close to 1: 1 (Kumar et al., 2006). Therefore, Ca+Mg and $HCO_3^-+SO_4^{2-}$ ratios can be used to indicate weathering and cation exchange adsorption of silicate (Barzegar et al., 2016). In Fig. S6(a), most of the groundwater samples fell on both sides of the 1:1 line and only a few samples were on the line, which indicated that the weathering and cation exchange adsorption of silicate had a significant influence on the groundwater chemistry. From Fig. S6(b), it is noted that the weathering of silicate rocks was dominated by carbonic acid.

In addition, according to Table S7 showing the correlation coefficients between different ions, Sr was positively correlated with Ca ($r = 0.817$, $p < 0.01$) and HCO_3^- ($r = 0.689$, $p < 0.01$), whose correlation coefficients were larger than that with Mg ($r = 0.588$, $P < 0.01$) and

SO_4^{2-} ($r = 0.565$, $p < 0.01$). Thus, the relatively high concentration of Sr in groundwater results from the dissolution of $SrCO_3$ in carbonate rocks. Besides, the correlations between Cl^-, Na, Br^- and B were obviously positive, and the concentration of Br^- and B in seawater was relatively high. We inferred that Br^- and B in groundwater in Rugao city came from seawater intrusion. Consequently, we compared the Cl^-/Br^- ratio in seawater and groundwater. As shown in Fig. S6(c), when the Cl^- concentration increased, the Cl^-/Br^- ratio in groundwater and that in seawater had a linear relationship, indicating that seawater intrusion did affect the chemical composition of groundwater in Rugao city. Furthermore, assuming that the Na and Cl^- in the water sample with the lowest TDS were from rock weathering, we drew a line linking the water sample with the lowest TDS and seawater sample in the Na/Cl^- ratio scatter plot (Argamasilla et al., 2017) (see Fig. S6(d)). The values of Na/Cl^- ratios of groundwater samples were mainly distributed near the connection line, and they were closer to the connecting line as the Na and Cl^- concentrations increased. Thus, when the Na and Cl^- concentrations in groundwater are high, the seawater intrusion is their main source.

It is then necessary to evaluate the degree of seawater intrusion through seawater fraction calculation. Because Cl^- is recognized as a conservative tracer that is hardly influenced by ion exchange, Cl^- is often used to calculate the seawater fraction (f_{sea}) by the following equations (Argamasilla et al., 2017):

$$f_{sea} = \frac{C_{Cl,\ sample} - C_{Cl,\ fresh}}{C_{Cl,\ sea} - C_{Cl,\ fresh}} \quad (6)$$

Where $C_{Cl,\ sample}$ is the Cl^- concentration in the water sample; $C_{Cl,\ sea}$ is the Cl^- concentration in the seawater; $C_{Cl,\ fresh}$ is the Cl^- concentration in freshwater.

It is assumed that the water sample with the lowest TDS is the freshwater sample. Due to the high solubility of Cl^-, the inputs are either from the dissolution of aquifer bedrock or from a salinization source such as seawater intrusion (Argamasilla et al., 2017). Fig. S7(a) showed the distribution of f_{sea} vs. TDS. The f_{sea} was between 0% and 3%, indicating that the groundwater in the study area was suffered from mild seawater intrusion. Besides, the relationship between f_{sea} and TDS was not linear, meaning that the study area's groundwater quality was controlled by both seawater intrusion and water-rock interaction.

After obtaining the f_{sea}, it is possible to calculate the theoretical concentration ($C_{i,\ max}$) of each ion in the mixture of seawater and freshwater as follows (Appelo and Postma, 2004).

$$C_{i,mix} = f_{sea} \times C_{i,sea} + (1 - f_{sea}) \times C_{i,fresh} \quad (7)$$

Where $C_{i,\ sea}$ and $C_{i,\ fresh}$ are the ion i concentration in seawater and freshwater, respectively.

Base on the method mentioned above, we obtained the theoretical concentrations of Br^-, and then compared them with the corresponding measured concentration of Br^-. As shown in Fig. S7(b), it could be found that the ratios of calculated concentrations to measured concentrations were approximately 1: 1; however, most of the points did not fall on the 1: 1 line, indicating that the ion concentration is affected by the mixture of seawater and freshwater.

The research area is located in the Yangtze river delta plain in which dense population and developed industrial and agricultural activities are found. Therefore, human activities are expected to affect shallow groundwater quality. For example, in some groundwater samples, high concentrations of NO_3^- were detected. The range of NO_3^- concentration was from 0 to 139.77 mg/L with a standard deviation of 38.18 mg/L, which meant large differences in the spatial distribution of NO_3^- exist. In Wuyao and Shizhuang towns where the NO_3^- concentration far exceeded the standard, rice cultivation is widespread; thus, the wide application of nitrogen fertilizer is probably the reason for the higher NO_3^- concentration in shallow groundwater. In addition, some studies reported that As in groundwater in the Yangtze river delta plain was produced by the natural hydrogeochemical process (Shao et al., 2018) and man-made pollution (Yin et al., 2017). Migration and transformation of As in groundwater are under the control of pH and redox conditions. Usually, As(III) is the dominant species of As and is positively correlated with Fe/Mn (Zhou, 2017). However, according to Table S7, As had a poor correlation with other ions. As a result, the high As concentration in this area is not only caused by the natural condition but also by industrial and agricultural activities, like pesticide pollution and industrial wastewater discharge (Smedley and Kinniburgh, 2002).

In summary, groundwater formation in Rugao city is controlled by the water-rock interaction of silicate and seawater intrusion. Nevertheless, anthropogenic activities affect shallow groundwater quality as well. Therefore, the groundwater components in Rugao city are quite complex since they are influenced by natural and anthropogenic processes.

4.2. Effects of shallow groundwater on physical health in Rugao city

Comparing the spatial distributions of longevity population, WQI and HI in Rugao city (Figs. 2, 3(b), and 4(c, d)), the apparent inverse relationship is clear. For instance, the towns, Baipu, Jiuhua, and Changjiang, had more longevity populations; while, the WQI and HI values were relatively lower in these towns. Furthermore, we compared the WQI and HI values of children in the study area with those in non-longevity areas (see Table S8). The average WQI and HI values of children in non-longevity areas fell in the range of 42–77 and 1.47–60.6, respectively, which was well above the mean values of WQI and HI in Rugao city (30.19 and 1.00). Therefore, the groundwater in the longevity area is usually characterized by low WQI and HI values. The water samples with high WQI were caused by the high concentrations of As and Mn; moreover, high As concentration was also the cause of the high HI value. In Fig. 5(a, b), the northwest and southwest part of the research area with high As concentrations, as well as the central part of the research area with high Mn concentrations had a relative low longevity index; conversely, the east part with a high longevity index had low As and Mn concentrations. Therefore, As and Mn have an adverse effect on physical health. Based on the HI values, the potential health risk of drinking water in this area was caused by As. Chronic exposure to As will have potential carcinogenic effects and lead to diseases like high blood pressure, skin lesions, diabetes, neuropathy, and so on (Mohammadi et al., 2019). The highest As concentration in Rugao city was 0.0407 mg/L, which is 4.07 times higher than the upper limit of As in the China National Standard for Drinking Water Quality (GB5749–2006). As a result, attention should be paid to the health risk caused by high As drinking water. Unlike As, Mn in drinking water is an essential trace element for human health (Aschner and Aschner, 2005). It is of great significance in immune function, cell energy, blood sugar regulation, lipid and carbohydrate metabolism, skeletal cartilage development and brain function (Aschner, 2000). However, a prolonged exposure to high concentrations has shown that Mn behaves as a neurotoxin. (de Carvalho et al., 2018).

WQI and HI can comprehensively reflect the groundwater quality in the study area in terms of drinking water safety. Towns with high longevity usually have low WQI and HI values in Rugao city; that is, groundwater is safer to drink. However, this rule does not work for Banjing town. As a result, low WQI and HI values are necessary but not sufficient conditions for longevity. The possible reasons are as follows: on the one hand, longevity is the result of the combination of environment, health care, lifestyle, heredity, and psychological factors; thus, safe drinking water cannot guarantee regional longevity (Kim and Kim, 2016; Lv et al., 2011). On the other hand, major elements such as Ca, Mg and trace elements such as Sr, Mn in drinking water play important roles in maintaining human health (Rosborg and Kozisek, 2016); however, WQI and HI do not reflect the effects of these nutritional elements on physical health.

In order to compare and analyze the differences in the nutrient

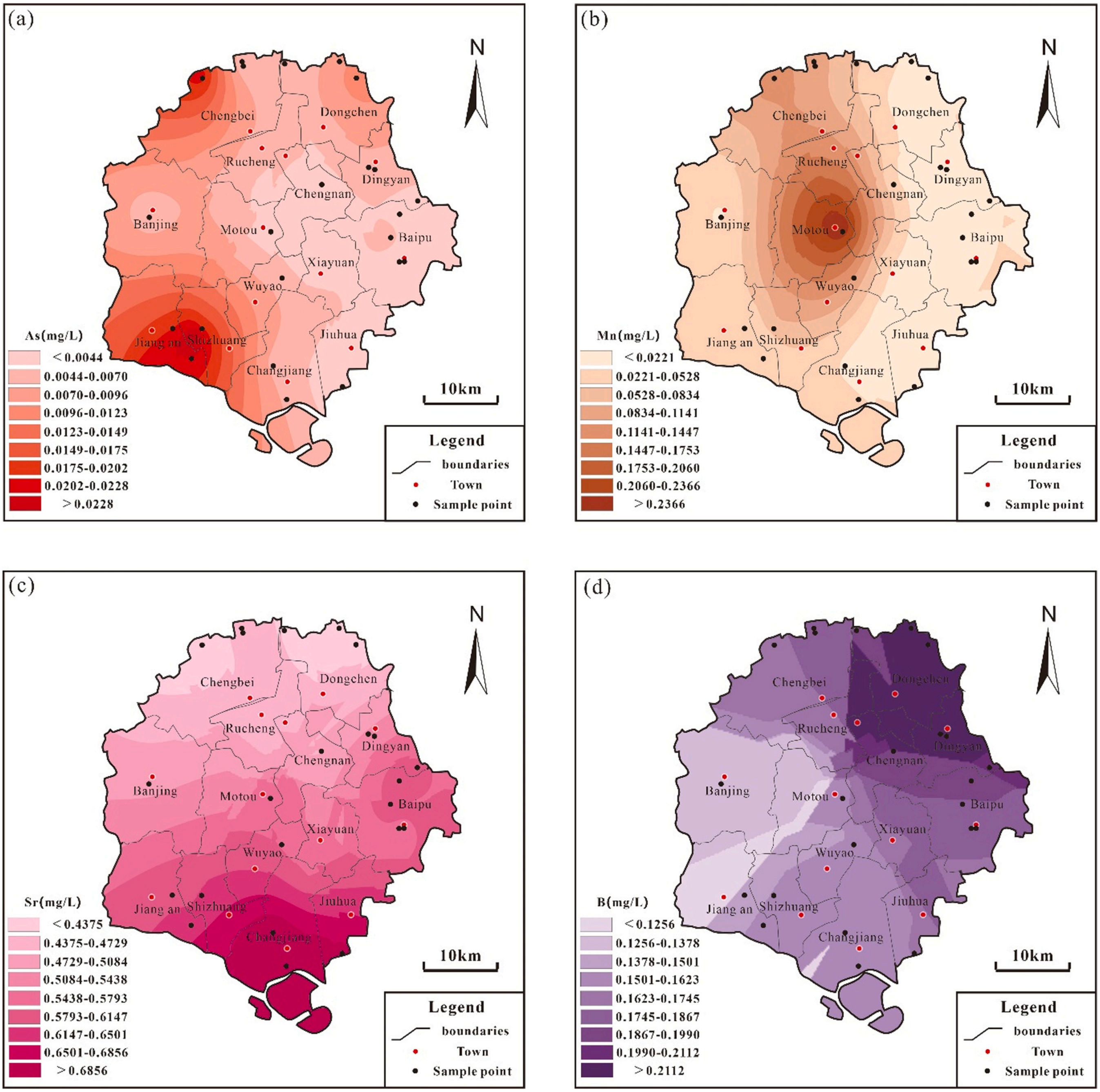

Fig. 5. The spatial distribution of concentration of As (a), Mn (b), Sr (c), and B (d) in groundwater in the study area.

indicators of groundwater between Rugao city and adjacent non-longevity areas, the major and trace elements in groundwater in Qidong city, Suzhou city, as well as the Su-Xi-Chang region were listed in Table S9. Compared to these places, the concentrations of Ca, Mg and SO_4^{2-} were all higher in Rugao city. Ca and Mg are known to be beneficial to human health: Ca plays an important role in bone mineralization (Rosborg and Kozisek, 2016), and Mg is essential for preventing atherosclerosis and inappropriate coagulation, maintaining vascular tone and electrolyte balance, which is crucial for physiological processes in our body (Seelig, 2012). The Ca concentration in groundwater in the study area was from 22.46 to 277.49 mg/L, with an average of 123.57 mg/L; the concentration of Mg in groundwater in this area fell in the range of 23.95–86.20 mg/L, with a mean of 50.33 mg/L. It is assumed that the daily consumption of drinking water is 2 L, so the Mg intake from drinking water in Rugao city is between 47.9 and 172.4 mg/day. The recommended daily intake of Mg is 450 mg (Dietary Reference Intakes for Calcium, Phosphorus, Magnesium, Vitamin D, and Fluoride); thus, the Mg in drinking water can meet 10.6–38.3% of the daily Mg requirement, with an average of 22.4%. Similarly, the Ca intake from drinking water is in the range of 44.92–554.98 mg/day. According to the recommended daily intake of Ca (1000 mg/day), the amount present in drinking water can account for 4.5–55.5% (mean value 12.4%) of the daily demand of Ca. These are crucial for elderly people who have insufficient intake of Ca and Mg. Due to the high concentrations of Ca and Mg in groundwater, the TH in Rugao city fell in the range of 179.62–894.47 mg/L with a mean of 515.27 mg/L. The TH in 65.2% of the groundwater samples exceeded the limit value in China National Standard for Drinking Water Quality (GB5749–2006). A large number of studies have revealed the relationship between TH and diseases. For example, it is generally recognized that high values for TH can help prevent cardiovascular disease (Rubenowitz-Lundin and Hiscock, 2013). Cardiovascular disease is the main ailment affecting the health of the elderly. The relationship between SO_4^{2-} concentration and physical health has rarely received much attention. The limits set for SO_4^{2-} in drinking water standard mainly consider how the high SO_4^{2-} concentration will affect the water taste (World Health Organization, 2011). However, a study reported that SO_4^{2-} could decrease the health risk brought by heavy metals intake (Rosborg and Kozisek, 2016). As a result, the high SO_4^{2-} concentration in Rugao city may reduce the health risk caused by As.

Due to the lack of data about the trace element concentrations, such as Sr and B, in the groundwater in these places, it is difficult to directly observe the differences in trace element concentration between Rugao and adjacent non-longevity regions. Nevertheless, compared with the Sr concentration in national public drinking water (0.3604 mg/L) (Peng et al., 2021), we found that the Sr concentration (0.1814–0.9882 mg/L with a mean of 0.5544 mg/L) in Rugao city was at a high level. Based on previous studies, the Sr in drinking water has a positive correlation with longevity (Liu et al., 2018). Sr is known to promote bone growth and development and maintain bone health in the elderly (Peng et al., 2021). Similarly, the concentration range of B in the groundwater of Rugao city was 0.0285–0.5438 mg/L, with an average of 0.1695 mg/L. B has been reported to enhance bone development, antioxidant defense systems, mineral and hormone metabolism, wound repair, energy metabolism, and so on (Abdik et al., 2019). According to Fig. 5(c, d), the towns with a high Sr concentration were mainly located in the south and east part of the study area; and the towns having a high B concentration were situated in the northeast and east part of the study area. Based on the spatial distribution of the longevity population, all aforementioned towns are longevity areas in Rugao. As a result, the trace elements, like Sr and B, in groundwater may have a positive correlation with longevity.

4.3. Strengths and limitations

This study discussed the influence of groundwater quality on longevity in terms of safety by comparing the spatial distribution of WQI and HI to that of the longevity indices. In addition, we compared nutrient indicators, such as Ca and Mg levels, in groundwater of longevity and non-longevity areas to reveal the impact of mineral nutrition on health. However, there are some limitations and uncertainties in this study. Firstly, longevity is the result of a combination of genes, natural environment, and socioeconomic status. Therefore, other factors might interfere with the discussion about the relationship between groundwater environment and longevity. Moreover, it is unknown whether the conclusions obtained from Rugao city can be applied to other longevity areas due to the different geological conditions, climatic conditions, and so on. Hence, it is of great significance to study the effects of groundwater environment on longevity based on the common characteristics of groundwater quality in multiple longevity areas. Secondly, although we tried to evaluate groundwater quality from the perspective of nutrition, such as the discussion about the concentrations of Ca and Mg as well as other minerals in groundwater, we still failed to comprehensively evaluate the nutritional properties of drinking water. In the following studies, more nutritional indicators are needed to establish a holistic evaluation method. Lastly, the different components in drinking water have both synergistic and antagonistic effects on human health. For instance, Zn and Sb presumably reduce the toxicity of As (Rahman et al., 2019); the ratios of Ca/Mg and Cu/Zn exert great influence on maintaining physical health as well (Khandare et al., 2018). In this study, even though we did not discuss the synergistic and antagonistic effects of all elements, which could affect the accuracy of groundwater quality assessment, the conclusions from this research still reveal the genesis of the groundwater environment in a longevity area and are able to provide a reference for establishing the drinking water health standard.

5. Conclusions

Rugao city is a typical longevity area with a developed economy and is located in the Yangtze River delta plain. According to the assessment of the OC, 90+(%), 80+(%), and 100+/80+(%) indices, the longevity towns in Rugao city exhibit a bow shape distribution. The elderly people in longevity areas live on shallow groundwater as the main drinking water resource, whose chemical components are under the control of water-rock interaction, specifically the dissolution of silicate rock. Meanwhile, Na, Cl^-, Br^-, B, etc. in groundwater emanate from seawater intrusion. The abnormal concentrations of NO_3^- and As also indicate that anthropogenic activities have posed significant influences on groundwater quality in Rugao city. The average WQI value is about 30.19; only one groundwater sample's WQI value exceeds 50, which indicates that the overall groundwater quality in Rugao city is pretty good. However, the concentrations of As and Mn in parts of groundwater samples are relatively high, with 4 and 5 water samples exceeding the China National Standard for Drinking Water Quality (GB5749–2006), respectively. There are 8 water samples whose HI value is above 1, which indicates that these groundwater samples pose a threat to physical health. The main reason for the high HI value is that a high As concentration is detected in these samples. In terms of the spatial distribution of WQI and HI, the distribution of longevity populations has an inverse relation with them. That is, the region with a large longevity population usually has low values of WQI and HI. In addition, the regions with high concentrations of As and Mn have poor longevity. Thus, As and Mn have an adverse effect on longevity. It is necessary to remove the As and Mn in groundwater in the study area. Enough attention should be paid to the protection of shallow groundwater in Rugao city.

Compared with adjacent non-longevity areas, the potable groundwater in Rugao city has the characteristics of high Ca, high Mg and high SO_4^{2-}. Due to the high concentrations of Ca and Mg, 65.2% of groundwater samples exceed the limit of TH in the China National Standard for Drinking Water Quality (GB5749-2006). The average concentrations of Ca and Mg are 123.57 ± 50.13 mg/L and 50.33 ± 18.93 mg/L, respectively, which could meet the 12.4% and 22.4% of daily Ca and Mg requirements through drinking water for local people in Rugao city. In addition, the areas where the Sr and B contents are higher in groundwater usually have a larger longevity population. In summary, we put forward that the high concentrations of Ca, Mg, SO_4^{2-}, Sr and B in drinking water with low WQI and HI values probably contribute to physical health and longevity. Certainly, more similar studies should be carried out in different longevity areas in the future, which will help to refine the environmental characteristics of groundwater in the longevity area. In addition, both the safety and nutritional properties of drinking water will affect human health; however, we often overlook the mineral nutrition in drinking water when we conduct the drinking water quality assessment. Therefore, it is necessary to establish drinking water nutritional indicators based on the synergistic and antagonistic effects of each component in drinking water. Then, based on these nutritional indicators, as well as safety indicators, such as WQI and HI, it will be possible to comprehensively evaluate the quality of drinking water in terms of both safety and nutrition.

CRediT authorship contribution statement

Hao Peng: Conceptualization, Methodology, Investigation, Writing - original draft preparation; **Pengfei Zou**: Funding acquisition, Supervision; **Chuanming Ma**: Data curation, Resources; **Shuang Xiong**: Investigation, Software; **Taotao Lu**: Formal analysis, Writing - original draft preparation, Writing- review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2021.112279.

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