Hydrochemical characteristics and quality assessment of shallow groundwater in Yangtze River Delta of eastern China

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Received: 26 September 2021 / Accepted: 20 March 2022 / Published online: 28 March 2022
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Abstract
Water resource is in high demand within the Yangtze River Delta, given its developed economy. Long-term exploitation of this resource has posed risks of artificial pollution and seawater intrusion to the shallow groundwater. This study aims to reveal the hydrochemical characteristics and health risks of shallow groundwater in the coastal plain of the Yangtze River Delta, as well as to discuss the possible factors affecting groundwater quality. Standard methods for hydrochemical parameter measurements, water quality assessment, and health risk models were applied to fulfill the objectives of the study. The results showed that the shallow groundwater was slightly alkaline, and the average values of total dissolved solids (TDS) and total hardness (TH) were 930.74 mg/L and 436.20 mg/L, respectively. The main hydrochemical types of groundwater were HCO3–Ca·Mg and HCO3–Ca·Na, accounting for 44.3% and 47.5%, respectively. In addition, As concentration was generally high, with a mean value of 0.0115 mg/L. The principal factors affecting the groundwater components include water-rock interactions (especially silicate), cation exchange, seawater intrusion, and human activities. The data also showed that As is strongly influenced by the redox of Fe, Mn, and NO3−. The results of the groundwater quality evaluation indicated that the shallow groundwater in some regions was unsuitable for drinking and agricultural irrigation. Health risk assessment showed that 44.3% of the water samples had significant health risks, which was attributed to the high As concentration. Therefore, it is urgent to establish long-term As monitoring to maintain sustainable groundwater management and drinking water safety. The results of this study provide essential data for water resource management and human health security in the Yangtze River Delta.

Keywords Yangtze River Delta · Hydrochemistry · Shallow groundwater quality · Health risk assessment

Introduction

Shallow groundwater is a crucial water source for industrial and agricultural production as well as daily life due to its shallow buried depth and easy accessibility (Li et al. 2020; Liu et al. 2021c). As a result, its quality plays a critical role in urban development and resident health. However, a growing number of studies have proposed that shallow groundwater in many places have become unsuitable for drinking and production, because of the natural environment and the impact of human activities (Hao et al. 2020; Lima et al. 2020). Water salinization and the enrichment of some harmful trace elements, like As, F, etc., in shallow groundwater pose a danger to both ecological safety and physical health (Liu et al. 2021c; Long et al. 2021; Wang et al. 2020; Wu and Sun 2016). In recent years, groundwater quality and health risk assessment have become an essential part of research on medical geology (Selinus et al. 2016). The
distributions, sources, and contamination levels of inorganic components in shallow groundwater are the foundation for pollution evaluation and management.

The evolution of shallow groundwater in estuaries and river deltas is quite complex. Due to the influence of weather, depositional environment, and anthropological activities, the shallow groundwater environment is fragile (Zhi et al. 2021). Cao et al. (2020) found that the groundwater salinization and nitrate contamination in estuaries and river deltas in Qinhuangdao city in northern China were caused by seawater infiltration and agricultural pollution. Hou et al. (2020) proposed that the discharge of domestic and industrial wastewater during urban development had led to increased Mn concentration in shallow groundwater in the Pearl River Delta plain. More and more studies have revealed growing deterioration of shallow groundwater quality in estuaries and river deltas (Li et al. 2018). These studies analyzed the hydrogeochemical features and the evolution of groundwater quality. However, less attention has been paid to the impact of groundwater quality on human health. Currently, some studies have obtained great achievements in qualitative and quantitative evaluation of groundwater quality using WQI and HI assessments which make it possible to evaluate the impact of shallow groundwater in the estuaries and river deltas on human health and industrial production in terms of drinking suitability and irrigation suitability (Liu et al. 2021b; Peng et al. 2022).

The Yangtze River Delta plain is one of the fastest growing urban areas with the most developed economy in China. Groundwater exploitation has been increasing year by year as a result of rapid population growth. Due to its unique geographical and geological environment, previous studies mainly focused on ground subsidence caused by groundwater extraction (Ma et al. 2018). Even though the Yangtze River Delta plain has abundant groundwater resources, human activities have led to a remarkable deterioration in water quality (Wu et al. 2014). In addition, the groundwater buried depth is relatively small in this area; thus, high TDS in groundwater was found due to the intense evaporation, with the TDS value on an upward trend, even tending towards salinization (Zhao et al. 2017). Meanwhile, the research on the variation of shallow groundwater quality in the Yangtze River Delta coastal plain proposed that seawater intrusion contributed to groundwater salinization and exerted a substantial influence on groundwater quality according to the analysis of Sr, O, and H isotopes in groundwater (Mao et al. 2020). Although the deterioration of water quality in the Yangtze River Delta has attracted much attention, there are still no studies on shallow groundwater quality and its health risks. In particular, the spatial distributions and sources of trace elements, the groundwater quality, and its related controlling factors in the Yangtze River Delta plain are, thus far, still unknown.

Based on these considerations, shallow groundwater samples were taken from the coastal plain in the Yangtze River Delta; then the dissolved inorganic ingredients in the groundwater were systematically analyzed to achieve the following targets: (1) identify the hydrochemical characteristics of shallow groundwater in this area and (2) evaluate shallow groundwater quality and possible impacts on physical health. These findings provide valuable insight for water resource management and human health security in the Yangtze River Delta.

Materials and methods

Study area description

The study area, located in Jiangsu province, China, is flanked by the Yellow Sea to the east and the Yangtze River to the south. It is a typical estuarine delta plain. The topography of this area is generally flat, with a slight decrease in altitude from west to east. The regional has a humid subtropical monsoon climate with average annual precipitation and evaporation of 1050.8 mm and 877.2 mm, respectively.

The shallow aquifers in the study area consist of Quaternary Holocene estuarine and coastal unconfined aquifer groups. The porous media are composed of gray, gray-green silt, and silty sub-sandy loam. The lower bedding layer is comprised of shallow marine silty sub-clay deposits, with its bottom slab buried at a depth of about 30 m with an average thickness of 27 m. The aquifers change from unconfined aquifers to slightly confined aquifers towards the east; simultaneously, the groundwater quality gradually becomes saline (Mao et al. 2020). The primary recharge sources of the shallow groundwater are atmospheric precipitation and inflow of the Yangtze River and its tributaries. Since the terrain is relatively flat, the groundwater flow in aquifers is sluggish under the control of topography. Groundwater discharge mainly includes near-surface evaporation and artificial exploitation. It is reported that the annual groundwater extraction in the study area is about $1.1 \times 10^7$ m$^3$, which makes shallow groundwater an important water source for agricultural irrigation and industrial production (Mao et al. 2020). Overall, the water quality of shallow groundwater in the research area is quite complicated because the groundwater was salinized by Holocene transgression and then gradually desalinated by the large quantity of fresh water from the Yangtze River and atmospheric precipitation. In addition, the Yangtze River Delta is one of the most densely inhabited regions in China. Seawater intrusions resulting from a large amount of groundwater extraction as well as industrial and agricultural pollution have caused a significant impact on the shallow groundwater in this area (Zhao et al. 2017).
Groundwater sample collection and measurement

In this study, 61 water samples, all of which were shallow groundwater, were taken from the domestic wells in the study area in August 2019. The depth of these wells was less than 10 m. In addition, 1 seawater sample was obtained from the coast near the study area, and 1 rainwater sample was taken in the middle of the study area (see Fig. 1). The specific sampling procedure based on our previous studies (Peng et al. 2021a; Peng et al. 2021b) can be found in

Fig. 1 Spatial distribution of groundwater samples in the study area
the supplementary material. The concentrations of trace elements and major elements in the water samples were obtained through ion chromatography, inductively coupled plasma emission mass spectrometer and standard HCl titration. The measurement process was provided in the supplementary material. In addition, other parameters like pH and TDS were measured on-site by a portable analyzer.

The statistical analysis of obtained data was carried out using SPSS software (IBM 23). For the correlation analysis of each component in groundwater, the data was first assessed to determine whether it follows normal distribution or not. To do this, the Kolmogorov-Smirnov (K-S) test and Shapiro-wilk test were applied separately. All hydrochemical indicators, except for pH, were not in accordance with the normal distribution; thereby, the Spearman’s model was adopted for correlation analysis. Lastly, the spatial distribution of hydrochemical indicators was drawn by ArcGIS software (version 10.5) according to the inverse distance weighting interpolation.

**Groundwater quality assessment**

**Groundwater quality assessment for agricultural irrigation**

Sodium adsorption ratio (SAR) and sodium percentage (Na %), which can be calculated by the equations (Eqs.) (1–2), are usually used to estimate whether groundwater is suitable for irrigation (Long and Luo 2020). Na plays a vital role in irrigation water classification because of its interaction with the soil. Na with a high concentration in irrigation water is readily adsorbed on clay minerals in the soil, replacing Mg and Ca. Besides, the ion exchange of Na with Mg and Ca reduces soil permeability. SAR is considered to be an essential indicator for the evaluation of irrigation water quality. When this indicator is high, the groundwater is considered unsuitable for irrigation.

\[
\text{Na}\% = \left( \frac{\text{Na} + \text{K}}{\text{Ca} + \text{Mg} + \text{Na} + \text{K}} \right) \times 100\%
\]  
\(\text{(1)}\)

\[
\text{SAR} = \frac{\text{Na}}{\sqrt{\text{Ca} + \text{Mg}}}
\]  
\(\text{(2)}\)

**Groundwater quality assessment for drinking purpose**

The water quality index (WQI) has been widely used for drinking water quality assessment since Horton proposed it in the 1960s. The critical point of WQI calculation is determining the weight of each water quality indicator because minor changes in weight will change the assessment result (Zhang et al. 2020). The methods that are commonly used to determine the weight are the objective weighting method (e.g., entropy-weighted method and criteria importance though inter-criteria correlation method) (Zhang et al. 2021a) and the subjective weighting method (e.g., order relation analysis method) (Gao et al. 2020). The subjective weighting method is based on the investigator’s actual experience, which brings multiple uncertainties to the result due to the different preferences of different investigators (Islam et al. 2020). Although the objective weighting method does not rely on subjective judgment and has a solid mathematical basis, it fails to reflect the degrees of importance of different indicators to the decision-maker (Narayananamoorthy et al. 2020). Therefore, this study used the integrated-weight method, which combines the subjective and objective weighting methods.

In the subjective weighting part, the weighting value \(w_{ij}\) of the water quality indicator is determined by its relative perceived effect and importance on human health, which can be calculated by the following Eq. (3):

\[
w_{ij} = \frac{P_j}{\sum P_j}
\]  
\(\text{(3)}\)

where \(P_j\) is the importance score of water quality indicator \(j\), which varies in the range of 1–5 (1 shows the minimal impact on water quality; 5 indicates the most significant impact on water quality), for example, heavy metals, such as As, Cd, Cr, and Pb, are harmful to physical health, so their \(P\) values are 5. The importance score of the different hydrochemical indicators can be found in Table S1.

In the objective weighting part, the weight of water quality indicator \(j\) \(w_{ij}\) is determined using the entropy weight method. The specific procedures are as follows:

In the first place, the initial data are standardized to obtain initial matrix \(X\) (Eq. (4)).

\[
X = \begin{bmatrix}
    x_{11} & \cdots & x_{1n} \\
    \vdots & \ddots & \vdots \\
    x_{m1} & \cdots & x_{mn}
\end{bmatrix}
\]  
\(\text{(4)}\)

Where \(m\) is the total number of groundwater samples; \(n\) is the total number of measured hydrochemical indicators in one sample.

After that, the initial matrix \(X\) is reversely normalized with Eq. (5) to get the standard matrix \(Y\) (Eq. (6)).

\[
y_{ij} = \frac{(x_{ij})_{\text{max}} - x_{ij}}{(x_{ij})_{\text{max}} - (x_{ij})_{\text{min}}}
\]  
\(\text{(5)}\)

\[
Y = \begin{bmatrix}
    y_{11} & \cdots & y_{1n} \\
    \vdots & \ddots & \vdots \\
    y_{m1} & \cdots & y_{mn}
\end{bmatrix}
\]  
\(\text{(6)}\)
At last, the $w_{j2}$ are obtained by Eqs. (7–9) (Islam et al., 2020):

$$y_j = \frac{y_j + 10^{-4}}{\sum_{j=1}^{m} (y_j + 10^{-4})}$$

$$e_j = -\frac{1}{\ln m} \sum_{i=1}^{m} y_j \ln y_j$$

$$w_{j2} = \frac{1 - e_j}{\sum_{j=1}^{n} (1 - e_j)}$$

Based on the Eqs. (3 and 9), the integrated weight can be calculated by the following equation:

$$W_j = \frac{\sum_{j=1}^{n} w_{j1} \times w_{j2}}{\sum_{j=1}^{n} w_{j1} \times w_{j2}}$$

After calculating the weights ($w_{j1}$, $w_{j2}$, and $W_j$ values are provided in Table S1), the WQI value is calculated by Eq. (11) (see Table S2).

$$\text{WQI} = \sum_{j=1}^{n} \left[ W_j \times \left( \frac{C_j}{S_j} \right) \times 100 \right]$$

where $C_j$ is the value of water quality indicator $j$ in the water sample; $S_j$ is the upper limit of the water quality indicator $j$ in the World Health Organization Guidelines for Drinking-water Quality. The groundwater can be classified into five categories based on the calculated WQI value: (1) excellent ($\text{WQI} < 25$); (2) good ($25 < \text{WQI} < 50$); (3) medium ($50 < \text{WQI} < 100$); (4) poor ($100 < \text{WQI} < 150$); and (5) extremely poor ($\text{WQI} > 150$).

Hazard index (HI) is the most commonly used method to evaluate the risk of trace elements in groundwater to human health. Because children are the most sensitive to exposed heavy metals, the analysis in this study focused on the children’s HI (Chen et al. 2020). There are two main exposure pathways to contaminants in drinking water: dermal absorption (e.g., bathing) and oral ingestion (e.g., drinking water). Therefore, the health risk evaluation of components in groundwater was estimated based on these two main pathways. The specific equations to calculate the HI value can be found in the supplementary material. The calculated HI values are presented in Table S2. Overall, when HI is less than 1, there is no health hazard. Even if a health hazard is present, it is negligible and difficult to be detected. In contrast, HI > 1 means an adverse effect on health.

### Results and discussion

#### Hydrochemical features of shallow groundwater in the study area

The statistical characteristics of the main components in shallow groundwater in the study area are shown in Table 1 (the complete data set can be found in Table S3). The pH of the groundwater samples varied between 6.90 and 7.90, with a mean of 7.31, indicating that the groundwater in the study area is slightly alkaline. The TDS fell in the range of 411.45–2361.77 mg/L, with an average of 930.74 mg/L. A total of 67.21% of the groundwater samples were freshwater (TDS < 1000 mg/L). The TH (CaCO$_3$) was in the range of 181.63–897.74 mg/L, with a mean of 436.20 mg/L. Based on the TDS and TH values, the groundwater in the study area fell into moderately hard, hard, and very hard water (see Fig. S1(a)). From Fig. S2, it could be found that the contents of major cations followed the order of $\text{Na}^+ > \text{Ca}^{2+} > \text{Mg}^{2+} > \text{K}^+$, with mean values of 124.08 mg/L, 87.46 mg/L, 52.21 mg/L, and 22.80 mg/L, respectively. Among them, the maximum Na content was 482.79 mg/L, which exceeded its upper limit (250 mg/L) in the World Health Organization Guidelines for Drinking-water Quality. Similarly, the contents of main anions followed the sequence of $\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-} > \text{NO}_3^-$, with average values of 543.87 mg/L, 117.99 mg/L, 96.12 mg/L, and 32.29 mg/L, respectively. The maximum values of $\text{Cl}^-$ and $\text{NO}_3^-$ contents were 806.00 mg/L and 139.77 mg/L, all of which exceeded the upper limits in the World Health Organization Guidelines for Drinking-water Quality. In addition, the Piper diagrams indicated that the shallow groundwater in the study area had four different types (see Fig. S1(b)). The main types were $\text{HCO}_3^-–\text{Ca-Mg}$ and $\text{HCO}_3^-–\text{Ca-Na}$, accounting for 44.3% and 47.5%, respectively, while the remaining ones were $\text{HCO}_3^-–\text{Na}$ and $\text{Cl–Na}$ type.

By analyzing the spatial distribution of hydrochemical indicators, the features of groundwater hydrochemistry can be more clearly understood in the study area. As indicated in Fig. 2a, the TDS values of shallow groundwater ranged from 400 to 1000 mg/L; and the groundwater samples with high TDS were distributed in the middle part of the study area and coastal regions. In terms of TH (see Fig. 2b), groundwater with relatively high TH was located in the west part of the study area, such as Rugao city and Nantong city; in contrast, the groundwater in the east part had low TH. The spatial distributions of $\text{Na}^+$ and $\text{Cl}^-$ were similar to that of TDS, as shown in Fig. 2c and d, with the highest concentrations in the middle part of the study area and coastal regions, while their concentrations were low in the west. The spatial distribution of $\text{SO}_4^{2-}$ was different
from that of Na\(^+\) and Cl\(^-\), with relatively high levels in the west and relatively low levels in the east (see Fig. 2e).

The spatial distribution of NO\(_3^-\) varied irregularly; the relatively high NO\(_3^-\) levels were found in the agricultural regions of Rugao city, Haimen city, and Qidong city (see Fig. 2f).

The statistical characteristics of trace elements in shallow groundwater can be found in Table 1 and Fig. S2(b). The average concentrations of trace elements followed the below order: Sr > Br\(^-\) > F\(^-\) > B > 0.1 mg/L > Ba > Al > Fe > Zn > Mn > Li > As > 0.01 mg/L > Cu > Se > Cr = Ni > Mo > 0.001 mg/L > others (Co, Ag, Cd, Sn, Sb, W, Tl, Pb, and Be). Sr, Br\(^-\), F\(^-\), and B were the main trace elements and their average values (maximum values) were 0.4975 mg/L (0.9882 mg/L), 0.3729 mg/L (2.6350 mg/L), 0.2888 mg/L (1.0065 mg/L), respectively. It is also worth noting that the maximum concentrations of Fe, As, Se, and Cr exceeded the upper limits in the

### Table 1 Chemical composition and their statistical characteristics in the shallow groundwater in the study area

| Unit | Project | Detection limit | Minimum value | Maximum value | Average value | Median | Standard deviation | Shapiro-Wilk test |
|------|---------|-----------------|---------------|---------------|--------------|--------|--------------------|-------------------|
| pH   | --      | --              | 6.90          | 7.90          | 7.31         | 7.30   | 0.23               | 0.967 0.104       |
| mg/L | TDS     | --              | 411.45        | 2361.77       | 930.74       | 841.68 | 370.26             | 0.907 0          |
| mg/L | TH      | --              | 181.63        | 897.74        | 436.20       | 418.98 | 144.02             | 0.959 0.041       |
| mg/L | Na      | 0.002           | 24.00         | 482.79        | 124.08       | 99.90  | 92.52              | 0.828 0          |
| mg/L | Mg      | 0.01            | 20.11         | 106.15        | 52.21        | 48.96  | 19.73              | 0.961 0.047       |
| mg/L | K       | 0.001           | 2.61          | 221.00        | 22.80        | 11.93  | 31.75              | 0.517 0          |
| mg/L | Ca      | 0.02            | 22.46         | 277.49        | 87.46        | 81.55  | 45.27              | 0.905 0          |
| mg/L | HCO\(_3^-\) | 0.01          | 292.47        | 860.20        | 543.87       | 504.65 | 135.43             | 0.944 0.008       |
| mg/L | Cl\(^-\) | 0.004           | 11.18         | 806.00        | 117.99       | 88.20  | 114.16             | 0.648 0          |
| mg/L | NO\(_3^-\) | 0.0006         | 0.03          | 139.77        | 32.29        | 22.90  | 32.34              | 0.854 0          |
| mg/L | SO\(_4^{2-}\) | 0.002        | 6.57          | 271.90        | 96.12        | 87.79  | 63.07              | 0.906 0          |
| μg/L | F\(^-\)  | 0.02            | 24.00         | 1079.00       | 347.00       | 297.70 | 210.30             | 0.867 0          |
| μg/L | Br\(^-\) | 0.02            | 0.00          | 2635.00       | 372.90       | 259.80 | 411.40             | 0.215 0          |
| μg/L | Sr      | 0.1             | 181.40        | 988.20        | 497.50       | 453.20 | 180.00             | 0.958 0.037       |
| μg/L | Al      | 0.04            | 0.70          | 229.10        | 52.60        | 41.00  | 44.30              | 0.840 0          |
| μg/L | Cr      | 0.05            | 0.00          | 10.40         | 1.40         | 0.60   | 2.00               | 0.606 0          |
| μg/L | Mn      | 0.02            | 0.00          | 373.20        | 20.40        | 21.0   | 58.80              | 0.388 0          |
| μg/L | Fe      | 0.06            | 1.70          | 677.40        | 48.40        | 22.40  | 103.20             | 0.345 0          |
| μg/L | Ni      | 0.02            | 0.00          | 20.50         | 1.40         | 0.70   | 3.60               | 0.306 0          |
| μg/L | Cu      | 0.02            | 0.00          | 73.60         | 3.00         | 1.50   | 9.40               | 0.229 0          |
| μg/L | Zn      | 0.04            | 0.90          | 530.00        | 33.90        | 14.40  | 78.70              | 0.358 0          |
| μg/L | As      | 0.02            | 0.00          | 77.80         | 11.50        | 3.70   | 17.90              | 0.653 0          |
| μg/L | Li      | 0.06            | 2.10          | 36.60         | 12.10        | 12.10  | 7.70               | 0.938 0.004       |
| μg/L | B       | 0.04            | 28.50         | 1006.50       | 288.80       | 203.00 | 219.01             | 0.789 0          |
| μg/L | Se      | 0.04            | 0.00          | 135.70        | 2.70         | 0.00   | 17.30              | 0.184 0          |
| μg/L | Mo      | 0.02            | 0.00          | 7.60          | 1.10         | 0.60   | 1.40               | 0.746 0          |
| μg/L | Ba      | 0.02            | 7.40          | 282.80        | 68.50        | 60.40  | 39.70              | 0.628 0          |
| μg/L | Ag      | 0.002           | 0.02          | 0.03          | 0.02         | 0.02   | 0.00               | 0.500 0          |
| μg/L | Cd      | 0.004           | 0.00          | 0.06          | 0.03         | 0.03   | 0.02               | 0.837 0          |
| μg/L | Sn      | 0.002           | 0.00          | 0.10          | 0.01         | 0.00   | 0.02               | 0.477 0          |
| μg/L | Sb      | 0.002           | 0.00          | 1.70          | 0.10         | 0.07   | 0.22               | 0.331 0          |
| μg/L | W       | 0.01            | 0.00          | 1.27          | 0.09         | 0.04   | 0.17               | 0.366 0          |
| μg/L | Ti      | 0.005           | 0.00          | 0.02          | 0.00         | 0.00   | 0.01               | 0.659 0          |
| μg/L | Pb      | 0.002           | 0.00          | 0.92          | 0.04         | 0.01   | 0.14               | 0.286 0          |
| μg/L | Be      | 0.006           | 0.00          | 1.40          | 0.10         | 0.00   | 0.23               | 0.510 0          |
| μg/L | Co      | 0.002           | 0.00          | 0.30          | 0.08         | 0.06   | 0.06               | 0.789 0          |

Note: The concentration of 0.00 μg/L means below the detection line. -- represents not applicable
World Health Organization Guidelines for Drinking-water Quality.

The spatial distributions of typical trace elements are illustrated in Fig. 3. The groundwater samples from coastal regions contained higher F$^-$ levels than non-coastal regions in the study area. Similarly, the groundwater samples with higher B content were distributed in the coastal regions; the highest B content was found in the western part of Haimen city. The groundwater samples containing the highest Al, Fe, and Ni concentrations were from the central and southeastern regions. In addition, groundwater having the highest Mn level was found in Rugao city. The As level in shallow groundwater was generally high; in particular, Nantong city and the southeastern part of Qidong city had higher As concentrations than other regions. Except for one water sample from the north of Qidong city containing high Se levels,
Factors controlling the water chemistry of shallow groundwater and trace element sources

Governing factors of groundwater chemistry

Effect of water-rock interaction  In order to obtain information on the shallow groundwater genesis, the ratios of different components in groundwater were calculated and analyzed. The Gibbs diagram can reflect the degree of influence of evaporation, water-rock interaction, and precipitation on the evolution of groundwater hydrochemistry (Gibbs 1970). In this study, most groundwater samples were found in the rock dominance and evaporation zones (see Fig. S3), which means groundwater hydrochemistry is mainly influenced by rock weathering, with evaporation and concentration also playing a crucial role (Samsudin et al. 2008).

Different rocks will lead to varying levels of ions in the groundwater after weathering. In the study area, carbonate rock, silicate rock, and evaporite rock are the primary weathered materials. The molar ratios of Ca$^{2+}$/Na$^+$ versus HCO$_3^-$/Na$^+$ and Ca$^{2+}$/Na$^+$ versus Mg$^{2+}$/Na$^+$, as shown in Fig. 4a and b, indicated that the water-rock interaction in the study area was largely controlled by the weathering of silicate rocks. The molar ratio of Na$^+$/Cl$^-$ ranged from 0.89 to 6.28, with a mean value of 1.91. Even in 91.80% of groundwater samples, the ratio was larger than 1, which is above the seawater (Na$^+$/Cl$^-$ = 0.87) and halite dissolution (Na$^+$/Cl$^-$ = 1.00) lines (Fig. 4c), indicating that silicate weathering is one of the important factors resulting in the excess release of Na$^+$ into the groundwater. In addition, the ratio of (Ca$^{2+}$+Mg$^{2+}$)/(HCO$_3^-$+SO$_4^{2-}$) could denote the effect of dissolution of carbonate rocks and gypsum on the water quality (Liu et al. 2021b). According to Fig. 4d, some points fell on the 1:1 line, suggesting that the dissolution of carbonate rocks and gypsum also contributed to the evolution of groundwater quality. Meanwhile, some points were distributed below the 1:1 line, which means that Na$^+$ is required to ensure the conservation of charge in addition to the dissolution of carbonate and gypsum. This process may include the dissolution of silicate minerals and cation adsorption. As illustrated in Fig. 4e, Ca$^{2+}$ and Mg$^{2+}$ in most groundwater samples are derived from dolomite and calcite dissolution. At the same time, silicate weathering also has an important effect on the contents of Ca$^{2+}$ and Mg$^{2+}$ in some water samples (Argamasilla et al. 2017). The ratio of Mg$^{2+}$ to HCO$_3^-$ can indicate the sources of Mg$^{2+}$. In Fig. 4f, the primary source of Mg$^{2+}$ is from the weathering of dolomite, with magnesium silicate weathering contributing less. Using PHREEQC, we calculated the saturation indices (SI) of calcite, dolomite, and gypsum of the water samples. The results showed that the gypsum was undersaturated. On the other hand, calcite and dolomite were supersaturated in all water samples (see Fig.4g), which indicates an abundance of calcite and dolomite present in the aquifers. This also supports the fact that the dissolution of gypsum contributes less to Ca$^{2+}$ content (Argamasilla et al. 2017).

In addition, the ratio between Na$^+$+K$^+$–Cl$^-$ and HCO$_3^-$+SO$_4^{2-}$–Ca$^{2+}$–Mg$^{2+}$ is commonly utilized to identify the influence of the cation exchange in groundwater systems (Liu et al. 2021a). The ratios of most water samples were near the 1:1 line, and Na$^+$+K$^+$–Cl$^-$ was positively correlated with HCO$_3^-$+SO$_4^{2-}$–Ca$^{2+}$–Mg$^{2+}$, as shown in Fig. 4h, revealing that the cation exchange process significantly affects groundwater hydrochemistry. Thus, the Chlor-alkali index (CAI) calculated by Eqs. (14–15) was used to determine the cation exchange patterns.

\[
\text{CAI} - 1 = \frac{\text{Cl}^-(\text{Na}^+ + \text{K}^+)}{\text{Cl}^-} 
\]

\[
\text{CAI} - 2 = \frac{\text{Cl}^- (\text{Na}^+ + \text{K}^+)}{\text{HCO}_3^- + \text{SO}_4^{2-} + \text{CO}_3^{2-} + \text{NO}_3^-} 
\]

Based on Fig. 4(i), the CAI-1 and CAI-2 values of most groundwater samples in the study area were less than 0, suggesting that Na$^+$ on the surface of clay minerals replaces Ca$^{2+}$ in groundwater. In the west part of the study area, there was a high amount of Ca$^{2+}$ in the groundwater due to carbonate dissolution. Meanwhile, the aquifer sediments near the shoreline contained a lot of Na$^+$ because of seawater intrusion. Na$^+$ in sediment-substituted Ca$^{2+}$ in groundwater during groundwater runoff, resulting in the gradual decrease of Ca$^{2+}$ concentration along the flow direction. This is also the reason for the distribution of TH value which was high in the west and low in the east (see Fig. 2b).

Effects of seawater intrusion  Groundwater in the coastal plain of the Yangtze River Delta has been affected by seawater intrusion for a long time (Zhao et al. 2017). For the purpose of evaluating the extent of seawater intrusion, seawater fraction ($f_{\text{sea}}$) is introduced. Because Cl$^-$, a stable tracer, is less affected by ion exchange, the Cl$^-$ concentration is usually utilized to calculate $f_{\text{sea}}$ value through Eq. (12) (Argamasilla et al. 2017; Mountadar et al. 2018).

\[
f_{\text{sea}} = \frac{C_{\text{Cl, sample}} - C_{\text{Cl, fresh}}}{C_{\text{Cl, sea}} - C_{\text{Cl, fresh}}} 
\]
Where \(C_{\text{Cl, sample}}\), \(C_{\text{Cl, sea}}\) and \(C_{\text{Cl, fresh}}\) are the \(\text{Cl}^-\) concentrations in groundwater sample, seawater, and freshwater, respectively.

It is assumed that the groundwater sample with the lowest TDS is freshwater that has not been polluted by seawater (Han et al. 2015). Because of the high solubility of \(\text{Cl}^-\), the dissolution of aquifer bedrock and seawater intrusion are its primary source (Argamasilla et al. 2017). Figure 5a showed that the \(f_{\text{sea}}\) varied with TDS, with \(f_{\text{sea}}\) in the range of 0–6.42% and a mean of 0.86%, suggesting that moderate seawater intrusion has impacted the shallow groundwater. Moreover, linearity between TDS and \(f_{\text{sea}}\) was not clearly apparent \((R^2 = 0.71)\), which also implies that water-rock interaction also controls the groundwater quality (Najib et al., 2016).

In addition, the theoretical concentration of ion \(i\) \(C_{i,\text{mix}}\) in the combination of seawater and freshwater can be calculated with \(f_{\text{sea}}\) value by Eq. (13) (Appelo and Postma 2005).

\[
C_{i,\text{mix}} = f_{\text{sea}} \times C_{i,\text{sea}} + (1-f_{\text{sea}}) \times C_{i,\text{fresh}}
\]  (13)

Where \(C_{i,\text{sea}}\) and \(C_{i,\text{fresh}}\) are the concentrations of ion \(i\) in the seawater and freshwater.

\(\text{Br}^-\) present in groundwater of coastal regions is mainly from seawater (Zhao et al. 2017), so the accuracy of the \(f_{\text{sea}}\) can be assessed by comparing the calculated

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Fig. 4 Ratio graphs of different indicators in shallow groundwater in the study area
Br\(^{-}\) concentration with its measured concentration. As seen in Fig. 5b, it can be found that the calculated and measured concentrations of Br\(^{-}\) were approximately distributed around the 1:1 line, indicating that the calculated \(f_{\text{sea}}\) is reliable. Nevertheless, most points failed to lie on the 1:1 line, which resulted from the effect of seawater and freshwater mixture. In Fig. S4, the relatively high \(f_{\text{sea}}\) was mainly distributed in the coastal regions and the middle part of the study area. This implies that seawater intrusion is more severe in these regions. Also, the distribution of \(f_{\text{sea}}\) was in accordance with that of Cl\(^{-}\) and Na\(^{+}\), proving that the high Na\(^{+}\) and Cl\(^{-}\) levels in the study area result from the sea. By comparing the \(f_{\text{sea}}\) distribution map (Fig. S4) with the land-use type map (Fig. S5), it is clear that \(f_{\text{sea}}\) is significantly higher in urban areas than arable areas. In particular, \(f_{\text{sea}}\) in the eastern part of Nantong city was even higher than that of the coastal regions. This is because urban areas are the most densely populated region in the study area, with high water consumption for industry; thus, the massive exploitation of shallow groundwater makes this area suffer from the most severe seawater intrusion. Meanwhile, the \(f_{\text{sea}}\) is lower in the areas near Yangtze River than the areas far from Yangtze River.

**Fig. 5** Plots of a \(f_{\text{sea}}\) vs. TDS, b Br\(^{-}\) measured vs. Br\(^{-}\) calculated, c Cl\(^{-}\)/Na\(^{+}\) vs. NO\(_3\)^{-}/Na\(^{+}\), and d NO\(_3\)^{-}/Na\(^{+}\) vs. SO\(_4\)\(^{2-}\)/Na\(^{+}\) for groundwater samples
which shows that the recharge of shallow groundwater from Yangtze River reduces the impact of seawater intrusion.

Effects of human activities For areas with extensive agriculture and industry, anthropogenic input is another important factor affecting the chemical composition of shallow groundwater (Zhang et al. 2021b). $SO_2^{2-}$ in groundwater may come from the dissolution of evaporites, oxidation of sulfides, or human industrial activity, and $NO_3^{-}$ in groundwater mainly results from agricultural activities and domestic sewage. The molar ratio of $NO_3^{-}/Na^+$ is generally high in groundwater contaminated by agricultural activities and domestic wastewater, while groundwater polluted by industrial activities has a high molar ratio of $NO_3^{-}/Na^+$ (Liu et al. 2021b). As shown in Fig. 5c and d, agricultural activities and domestic discharges have significantly impacted shallow groundwater quality. According to Fig. 2f and Fig. S5, $NO_3^{-}$ content was higher in the agricultural area, and there were no elevated levels of $NO_3^{-}$ content in urban areas. Thus, agricultural activities are the main source of $NO_3^{-}$.

Identification of sources of trace elements by principal component analysis

Principal component analysis was used to analyze the origins of trace elements. The Kaiser-Meyer-Olkin (KMO) value was 0.66, and the significance of Bartlett’s sphericity test was 0, indicating the data in this study were suitable for PCA analysis. In Fig. S6, PC1 accounted for 33.97% of the variance and indicated high squared loadings of Fe, Ni, Zn, Ba, Al, and Cr. Ni, Fe, Zn, and Cr are usually from human activities; for example, vehicle exhaust and coal combustion are the main sources of Cr in the environment; while Zn, Fe, and Ni are mainly from vehicle exhaust and industrial waste. As illustrated in Table S4, Al showed good positive correlations with Zn, Fe, and Ni ($r_{Al-Zn} = 0.617^{**}$, $r_{Al-Fe} = 0.609^{**}$, $r_{Al-Ni} = 0.563^{**}$), but no correlations with Ca and Mg ($p < 0$, Table S4), which indicates that Al has a similar origin with Ni, Fe, Zn, and Cr, rather than from the weathering of silicate rocks. Ba often results from the complex combinations of natural processes and human inputs (Xiao et al. 2019). In summary, PC1 mainly shows the input from human activities such as industrial emissions. PC2 contributed 18.63% of the variance and included Se, Mo, and Rb. Se, Mo, and Rb are usually derived from soil and bedrock weathering. Se and Rb were positively correlated ($r_{Se-Rb} = 0.936^{**}$, Table S4), suggesting that they are derived from the weathering of the same mineral. Mo is positively correlated with Se and Rb, as well as has good positive correlations with B and Br ($r_{Mo-B} = 0.637^{**}$, $r_{Mo-Br} = 0.537^{**}$, Table S4), indicating that Mo originates from a combination of mineral weathering and seawater intrusion. PC3 explained 11.52% of the variance and had high loadings of As, Li, Ba, and Mn. As, Mn, and Li mainly come from bedrock weathering and/or soil leaching due to their similar sorption and desorption behavior (Long et al. 2020). In addition, As, as an impurity in phosphate fertilizer, can be leached into the groundwater during fertilizer application (Du et al. 2018). As was also positively correlated with Fe with correlation coefficients of 0.399 ($p < 0.01$). The redox of Fe strongly influences the geochemical cycling of As in groundwater (Saha and Rahman 2020). Under oxic conditions, As is adsorbed on Fe oxides and hydroxides; while under anoxic conditions, the reduction of oxides of Fe decreases the adsorption sites for As on their surface; at the same time, the As(V) is reduced to As(III) which has weak adsorption, thus promoting the release of As from sediment to groundwater (Duan et al. 2017). Furthermore, As was apparently negatively correlated with $NO_3^{-}$ ($r_{As-NO_3^{-}} = -0.290^*$, Table S4). It has been proposed that the reduction of $NO_3^{-}$ occurs in the presence of denitrifying bacteria; this process can provide As(V) with enough electrons, accelerating the reduction of As(V) to As(III); thus, more As ions are released into the groundwater (Xie et al. 2018). PC1, PC2, and PC3 accounted for 64.2% of the total variance.

Shallow groundwater quality assessment in the study area

For irrigation purposes

Yangtze River Delta is one of the major agricultural bases in China. Therefore, shallow groundwater is an important water source for agricultural irrigation. The Na% ranged from 12.30 to 81.06%, with an average value of 38.05%; and the SAR varied from 0.53 to 11.96 with a mean value of 2.70. According to the Wilcoxon diagram (see Fig. 6a), most groundwater samples were in the good to permissible section (68.85%), while 22.95% of the samples were in the doubtful to unsuitable section and 4.92% of the samples belonged to the unsuitable section, indicating that the groundwater is unsuitable for irrigation in some regions with high Na content. In addition, based on the classification proposed by the US Salinity Laboratory (USSL) (see Fig. 6b), the majority of the groundwater samples were located in the C3S1 zone (72.13%). In comparison, 27.87% of the samples were situated in the C4S1, C4S2, C4S3, and C3S2 zones, suggesting that nearly 30% of the shallow groundwater is not suitable for irrigation in the study area. In summary, the Na% and SAR values reveal that shallow groundwater in regions with severe seawater intrusion (e.g., Nantong and Haimeng cities) is unsuitable for irrigation. The people in these places should pay special attention to it in agricultural production.
For drinking purposes

Figure S7 illustrates the Efs values, the ratios of measured values of hydrochemical indicators to their corresponding upper limits in the World Health Organization Guidelines for Drinking-water Quality. Among them, the indicators with Efs > 1 were TDS, Na, Cl−, NO3−, Fe, As, Cr, and Se, with the exceeding rates of 32.79%, 18.3%, 8.20%, 22.95%, 3.28%, 26.23%, 6.56%, and 1.64%, respectively. Se and As had the highest exceedance levels, with maximum Efs values of 13.57 and 7.78, respectively. Only 36.07% of water samples met the guidelines for all the measured indicators. In addition, WQI values of shallow groundwater varied from 12.78 to 141.34, with an average value of 46.49. According to their WQI values, the percentage of groundwater with excellent, good, medium, and poor quality were 14.75%, 52.46%, 27.87%, and 4.92%, respectively. Thus, most shallow groundwater can be utilized for drinking in the study area. Figure 7a presented the spatial distribution of WQI values in the study area. It could be found that the groundwater samples with excellent and good quality were from the eastern and northern parts of the study area. Besides, groundwater samples with medium and poor quality were sporadically distributed in the central part of the study area like Nantong city, and in the southeastern part of the study area like Qidong city.

The HI values of shallow groundwater in the study area were in the range of 0.264–9.555, with a mean of 1.795. The percentage of groundwater samples with HI > 1 in the study area was 44.3%. Overall, groundwater in some regions may pose a significant non-carcinogenic risk to the health of local residents. Figure 7b showed that the groundwater in
the southwestern part of Rugao city, Nantong city, and the northern and southeastern coastal regions of Qidong city had HI > 1. Except for Rugao city, the spatial distribution of HI was consistent with that of As (see Fig. 3f). Thus, As level is the main factor affecting the HI values.

Figure 8 displayed the non-carcinogenic risks of different trace elements obtained by oral intake and dermal absorption in children. The non-carcinogenic risk of each trace element obtained through dermal absorption was 2 orders of magnitude less than that obtained through the oral intake, and HQ\textsubscript{dermal} was far less than 1, indicating that oral intake is the main health risk exposure pathway and the non-carcinogenic health risk through dermal absorption is negligible. With the exception of As, the maximum HQ\textsubscript{oral} value of each trace element was less than 1. The mean and maximum values of HQ\textsubscript{oral} for As were 1.371 and 9.284, showing significant health risks. The average HI value of each trace element was in the order of As (1.374) > Li (0.217) > B (0.052) > Sr (0.030) > Se (0.020) > Cr (0.019) > Ti (0.019) > Ba (0.013) > Sb (0.011) > others. It is worth noting that the maximum $E_{fs}$ value of Se was 13.57, and its maximum HI value was 0.973, which is close to the threshold value of the non-carcinogenic health risk. The HI values of Al, Mn, Fe, Ni, and B, which exceeded the national drinking water standard, were low, with maximum values of 0.008, 0.096, 0.035, 0.037, and 0.180, respectively. These trace elements have no significant non-carcinogenic health risks.

In Fig. S8, As in shallow groundwater made the greatest contribution to the HI calculations, with a mean value of 54.1% and a maximum value of 97.4%; followed by Li, with a mean value of 24.9% and a maximum value of 74.2%; the average contribution of the remaining elements to total HI was less than 10%. In general, the contribution (mean value) of each element to HI in shallow groundwater in the study area has the following order: As (54.1%) > Li (24.9%) > B (5.0%) > Ti (4.0%) > Sr (3.5%) > Cr (2.2%) > Sb (1.5%) > Ba (1.4%) > Co (1.0%) > others. Long-term ingestion of high As groundwater will lead to serious diseases such as skin, hematological, and renal diseases (Wu et al. 2020). As should be the focus of attention in drinking water risk management in the study area.

As mentioned above, the results of WQI and HI assessments are broadly consistent. For example, in the north part of the study area, the groundwater had good quality with low WQI and HI values; in contrast, the groundwater quality in the middle and southeast parts was poor with high WQI values.
and HI values. However, some areas had opposite WQI and HI values; for instance, the WQI value was low, but the HI value was high in southwest Rugao city. This is because WQI chooses the restrictive indicators such as TDS, TH, pH, etc., in the national drinking water standards for water quality evaluation. However, the HI assessment includes some trace elements such as Sr and Li, which are not restricted in national drinking water standards. Therefore, combining the results from these two assessments provides a more comprehensive understanding of the effects of shallow groundwater quality on health. Taken together, the shallow groundwater in some regions may pose significant health risks to local residents. For example, the shallow groundwater in southwest Rugao city, Nantong city, and parts of Qidong city was no longer suitable as drinking water due to the high As concentration which was a severe threat to the health of local residents. High arsenic groundwater is a current environmental and health problem worldwide (Podgorski and Berg 2020). Numerous studies on the causes and health risks of high arsenic groundwater have been conducted in the Hetao plain and Jianghan plain in China, while the investigations in the Yangtze River Delta have not been reported (Mao et al. 2018; Zheng et al. 2020). Unlike Hetao plain and Jianghan plain, the Yangtze River Delta is one of the most densely populated areas in China. Thus, the health risks associated with high arsenic groundwater require more attention.

Since shallow groundwater is an important water source for irrigation, heavy metals in the water can be enriched through crops. However, in this study, the health risk assessments did not consider the elements from food, resulting in an underestimation of health risk. In addition, the relative sparsity of sampling points could lead to the uncertainty of the spatial distribution of groundwater quality. More detailed research should be carried out in regions with severe seawater intrusion and high As concentrations in the future.

Conclusions

In this study, the shallow groundwater in the coastal plain of Yangtze River Delta was sampled and analyzed. On this basis, groundwater quality was evaluated using methods such as WQI and HI. The results showed that the shallow groundwater was slightly alkaline in the study area. The mean values of TDS and TH were 930.74 mg/L and 436.20 mg/L, indicating that the groundwater was moderately hard, hard, and very hard. Na and HCO₃⁻ had the highest concentrations in the shallow groundwater. The percentages of groundwater samples with Ca+Mg–HCO₃ type and Ca/Na–HCO₃ type were 44.3% and 47.5%, respectively. Sr, Br, F, and B were the major trace elements with mean values of 497.5 μg/L, 372.9 μg/L, 347.0 μg/L, and 288.8 μg/L, respectively. Meanwhile, As had a relatively high concentration in shallow groundwater (its average value was 11.5 μg/L). Factors like rock weathering (silicate), cation exchange, seawater intrusion, and industrial and agricultural activities played an important role in the chemical composition of shallow groundwater in the Yangtze River Delta plain. The massive exploitation of shallow groundwater has caused the most severe seawater intrusion in Nantong city. Fe, Ni, Zn, Al, and Cr are mainly from industrial pollution; Se, Mo, and Rb are derived from weathering of soil and bedrock; As, Mn, and Li are largely controlled by soil-water interaction; meanwhile, agricultural pollution is also a source of As. According to the water quality assessment, shallow groundwater in some regions was unsuitable for irrigation. At the same time, TDS, Na, Cl⁻, NO₃⁻, Fe, As, Cr, and Se in some groundwater samples exceeded their corresponding upper limits in the World Health Organization Guidelines for Drinking-water Quality. Only 36.07% of water samples met the guidelines for all measured indicators. According to the classification of WQI values, excellent, good, medium, and poor groundwater samples accounted for 14.75%, 52.46%, 27.87%, and 4.92%, respectively. The analysis of health risk showed that 44.3% of the water samples had significant health risks, with As playing the most important role, with a mean contribution of 54.1% to the total HI; therefore, As should become the focus in drinking water management in the study area. The shallow groundwater quality in the Yangtze River Delta, especially the concentrations of trace elements and their health risk assessment, has been neglected for a long time. It is necessary to conduct long-term monitoring of As contamination in the Yangtze River Delta and to take measures to prevent and control groundwater contamination. In the meantime, new high-quality water sources should be sought for the regions, such as Nantong city, where seawater intrusion is serious; and shallow groundwater extraction should be reduced to maintain sustainable groundwater resource management.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s11356-022-19881-w.

Author contribution TL: Conceptualization, methodology, investigation, writing-original draft preparation. RL: Methodology and investigation. ASNF: Resources. SX: Software and investigation. PZ: Supervision and funding acquisition. HP: Formal analysis, writing-original draft preparation, and writing-reviewing and editing.

Funding This project was supported by New Era Health Industry (Group) Co. Ltd. (ZAT2019X01002) and the China Scholarship Council (201708420145).

Availability of data and materials Supplementary Material data to this article can be found in the online version of this article.
Declarations

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent to publish Not applicable.

Competing interests The authors declare no competing interests.

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