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Research Article

Hydrochemical Characteristics and Quality Assessment of Groundwater under the Impact of Seawater Intrusion and Anthropogenic Activity in the Coastal Areas of Zhejiang and Fujian Provinces, China

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Coastal groundwater is an important resource in the developed region associated with human health and sustainable economic development. To identify the origins of salinity and evaluate the impact of water-rock interactions, seawater intrusion (SWI), and evaporation on groundwater in the coastal areas of Zhejiang and Fujian provinces, a comprehensive investigation was performed. Meanwhile, nitrate and fluoride indicators resulting from the anthropogenic activity and SWI were also considered. At last, the water quality index (WQI) of coastal groundwater was evaluated with geochemical and multivariate statistical methods. The results indicated that (1) the groundwater in coastal areas of Zhejiang and Fujian provinces has been affected by SWI to varying degrees. The analysis of selected ion ratios (Na^+/Cl^- and Br^-/Cl^-) and isotopic compositions showed that SWI is the predominant cause of increasing salinity in the groundwater of Zhejiang Province, while the cause is water-rock interactions (ion exchange and mineral weathering) in Fujian Province. The hydrochemical evolution path of groundwater in Zhejiang Province is Ca/Mg-HCO_3 to Na-Cl, while a different pattern of Ca/Mg-HCO_3 to Na (Mg/Ca)-Cl occurs in Fujian Province. However, the trend of SWI development in both provinces was freshening. (2) Nitrification, sewage infiltration, and SWI increased the NO_3^- content in groundwater. Some of the NO_3^- concentration in Fujian Province exceeds the standard, and the nitrogen pollution was more serious than in Zhejiang Province. The F^- content in coastal groundwater was affected by SWI and mineral dissolution; the F^- content in Zhejiang Province was higher than in Fujian Province, which was close to the groundwater standard limit. The average WQI value of Zhejiang was 103.61, and the WQI of Fujian was 61.69, indicating that the coastal groundwater quality in Fujian Province was better than in Zhejiang Province. The results of the study revealed the impact of SWI and anthropogenic activity on groundwater in the southern coastal zone of China and will be valuable for sustainable groundwater resource management.

1. Introduction

The coastal area is a narrow interface zone between the marine and terrestrial areas, where human population keeps increasing [1]. Although the coastal area covers only 20% of Earth's land area, it is estimated that it is home to 41% of the world's population [2]. The higher population densities in the coastal zone, especially in the urban areas, have caused some negative environmental effects, such as seawater intrusion, depletion of aquifers, land reclamation, wastewater infiltration, land subsidence, and sewage exfiltration [3–6], which induce the coastal area to become the most valuable and vulnerable of Earth's habitats [7–9].

Groundwater is a large active reservoir in the global hydrologic cycle because of its wide distribution and relatively stable water content, and usually, it is the main source of water for industry, agriculture, and domestic use [10, 11]. In recent years, with the ever-increasing demands for the sustainable development of industrial and agriculture in the coastal region, groundwater plays an important role in the Chinese coastal water supply [12]. In general, the groundwater system in the coastal zone is not relatively independent, and it is affected by both ocean dynamics and terrestrial runoff [13]; thus, it is characterized by a transition between marine and terrestrial. What is more, groundwater in coastal areas also suffered the effect of the increasing population. In China, previous studies focused on the northern coastal area, such as Laizhou Bay and Liaodong Bay [3], due to the seawater intrusion caused by the lack of water resources or the rapidly urbanized coastal area in South China [12]. Therefore, the evolution of coastal groundwater chemistry, as well as the water quality, is both determined by the natural processes (such as hydrogeological conditions) and anthropogenic activities (such as agriculture and industry) [14–17].

In recent years, many researchers have devoted to study the negative environmental issues such as seawater intrusion (SWI) and groundwater pollution [18–22]. SWI is a global environmental problem in coastal areas [23, 24] and is associated with water salinization processes such as paleo SWI [3], modern SWI [25], and water-rock interactions [26]. SWI can occur naturally when the sea level rise or is induced by excessive exploitation [27]. According to the IPCC sea-level report, the sea level is predicted to rise along most of the world's coastlines and will inundate large areas of the coastal zone over the next century [28]. As the report, foresight scenario narratives on the sea-level rise will increase by 10 cm until 2030 and 21 cm until 2060 [29], presenting a wide range of hazards for the ~1 billion people living in low-elevation coastal areas by 2050. Moreover, as the most rapidly developing regions, some coastal urban areas have intensive water demands, which lead to the deterioration of groundwater quality, as well as the change in the groundwater evolution and chemical composition. For instance, SWI will directly cause a large increase in total dissolved solids (TDS) and Cl^-. In addition to SWI, other human activities, such as improper wastewater discharge and fertilizer usage [30, 31], will also cause the contamination of coastal groundwater. Therefore, an improved understanding of coastal groundwater evaluation and water quality changes caused by sea-level rise as well as anthropogenic activity is valuable for sustainable groundwater resource management.

Zhejiang and Fujian provinces are located in the southern part of China (Figure 1). Due to the abundant rivers and rainfall, the water supply of the two provinces almost entirely relied on surface water. However, the two provinces have problems with groundwater. The water per capita and availability of Zhejiang is only 1725 m^3/person, far lower than the national average of 2100 m^3/person [32], while the groundwater distribution in Fujian is uneven, with extremely abundant water resources in the northwest but scarce in the southeast coast [33]. The uneven distribution of water resources severely limits the development of coastal areas in both provinces. Therefore, the management of coastal freshwater reserves is increasingly important, not only because water storage in surface water alone is increasingly unable to meet the sustainable development of the coastal regional economy [34], but also because freshwater stored in coastal aquifers is particularly susceptible to degradation by SWI and other groundwater pollution issues. Furthermore, the salinization of aquifer caused by modern SWI, as well as the groundwater pollution, is almost irreversible on human timescales [35, 36]. Therefore, identifying the causes and evolution process of coastal groundwater is essential but difficult for the scientific management of groundwater in the two provinces.

The traditional methods of hydrogeological analysis mainly focused on the description of the groundwater's chemical characteristics, which could not meet the demand of problems about the origin and evolution pattern of the groundwater. Therefore, the method of isotope tracing combined with hydrochemistry was adopted to solve this problem. For instance, the use of multiple indicators such as specific ion ratios (e.g., Na/Cl, Br/Cl, and Na/Ca) is used to describe the composition of groundwater; however, combined with Piper and Gibbs plots, the changes in the dominant chemical compositions in groundwater can be determined [37–39]. In addition, the isotopic analysis has been widely used to identify the status of SWI and the origin of salt and water [40], and the hydrochemical facies evolution diagram (HFE-D) can highlight the succession of different hydrochemical facies developed between the intrusion and freshening phase [41, 42]. The water quality index (WQI) method is usually applied to evaluate groundwater quality [43]. Based on the combination of multiple methods, water quality can be assessed comprehensively.

The objectives of this paper are as follows: (1) identify the distinction between the groundwater and salt source of Fujian and Zhejiang provinces and their geochemical evolution processes; (2) analyze the occurrence and evolution mechanisms of SWI in the two provinces and their impact on groundwater quality; and (3) reveal the influence of anthropogenic activity on groundwater quality in the study area by comparing the distribution of specific pollutants and WQI. The results will improve the overall understanding of evolution as well as the protection and utilization of groundwater resources in different regions and provide environmental protection suggestions for the integrated management of groundwater resources.

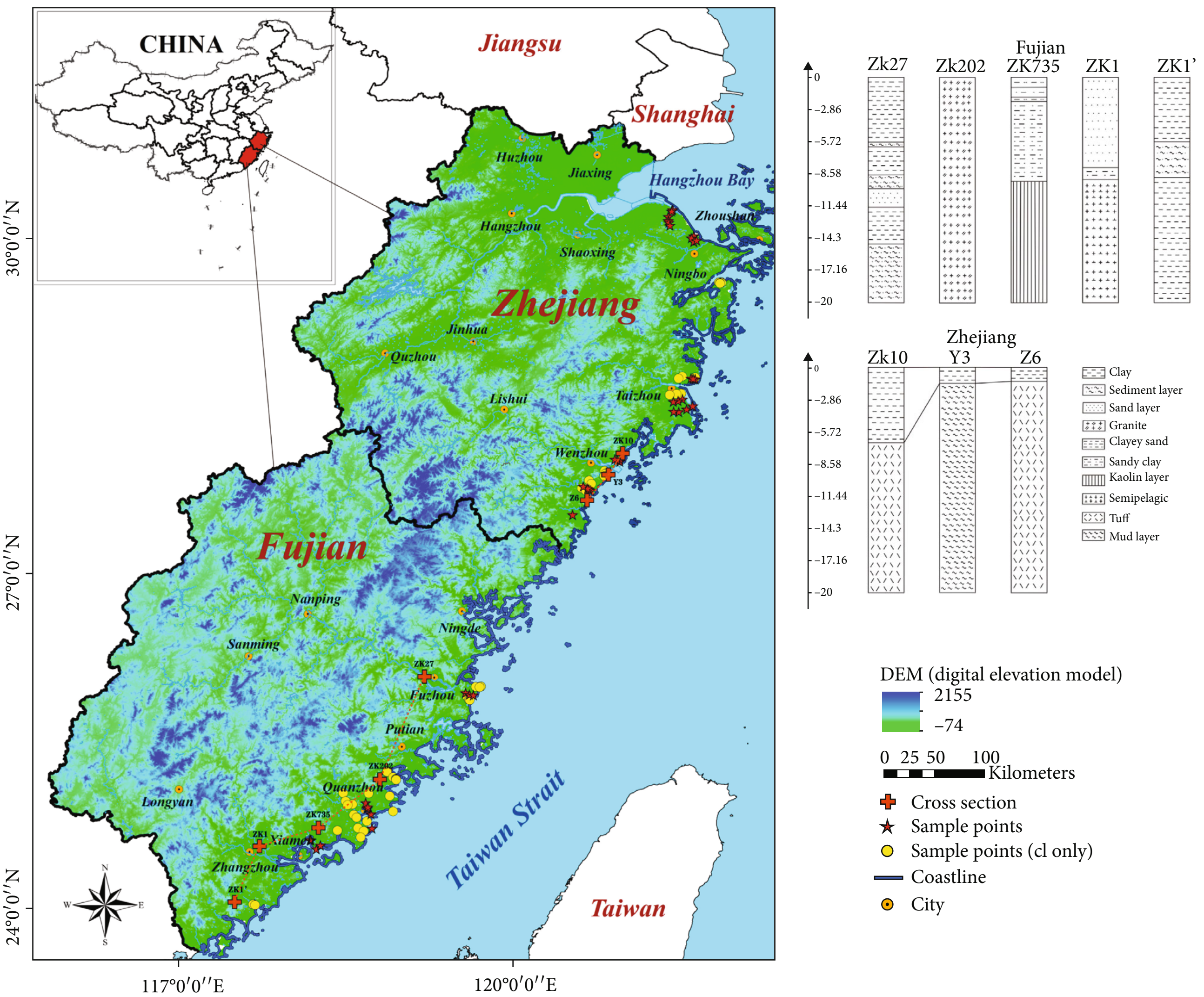


Figure 1: Location map of the study area and sampling points.

2. Description of the Study Area

2.1. Geographical Conditions. Zhejiang Province is located on the southeastern coast of China, which spans latitudes 27°02′ to 31°11′ north and longitudes 118°01′ to 123°10′ east (Figure 1). The topography of Zhejiang is stepped from southwest to northeast, with low flat alluvial plains in the northeast, coastal plains in the east, hills and basins in the central part, and mountains and hills in the southwest. The rock types are mainly magmatic rocks with ages ranging from Silurian to Neoproterozoic. Zhejiang has a subtropical monsoon climate, an annual average temperature of 15-18°C, and an annual average rainfall of 1200-2000 mm. Zhejiang Province has many coastal islands (>3000) and many coastal headlands with many harbors and bays. The abundant precipitation and surface runoff in Zhejiang provide favorable conditions for the formation and enrichment of groundwater, but the magnitude of resources varies greatly among hydrogeological units or groundwater types. The mainland coastline winds northward from the northern part of Hangzhou Bay to Wenzhou in the south [44]. The coastline is 6486.24 km long, accounting for 20.3% of China's total coastline. The coastal areas are highly urbanized and densely populated with a huge amount of domestic sewage generation.

Fujian Province is also located on the southeast coast of China, east of the Taiwan Strait, and its land area is between 23°33′ and 28°20′ north latitude and 115°50′ and 120°40′ east longitude. Mountains and hills account for more than 80% of the province's total area, and the terrain is generally high in the northwest and low in the southeast, with a slightly saddle-shaped cross-section. The stratigraphic development of the Fujian coastal zone is complete, with frequent magmatic activity and complex geological structure; sedimentary rocks and metamorphic rocks, volcanic rocks, and intrusive rocks each account for one-third of the land area, and shallow rock types include mudstone, sandstone, kaolinite, granite, and amphibolite. Except for the absence of

Silurian, Middle and Lower Devonian, and Lower Tertiary, the stratigraphy is relatively well developed from Metasedimentary to Quaternary. The climate of Fujian is subtropical maritime monsoon climate, with abundant precipitation and sufficient light, the average annual rainfall is 1400-2000 mm, and the average annual temperature is 17-21°C. The coastline of Fujian Province is winding, with many coastal ports and islands, and the coastline is 3,752 kilometers long.

2.2. Hydrogeological Setting. Zhejiang coastal groundwater type is mainly loose rock pore water, mainly divided into coastal estuarine plain pore diving and coastal plain pore pressurized water. Pore diving is mainly distributed in the shallow fine-grained strata of the coastal plain, with a total area of about 18,169 km^2. The aquifer is divided into two categories: alluvial lacustrine and lacustrine clay and alluvial marine powder clay and fine sand. The former is about 4~6m thick, with a poor water volume and a single well surge of no more than 5m^3/d. The sea-rushing aquifer is mainly located in the estuary area of major rivers, with a thickness of 5~20 m. The water volume of a single well is 5~50 m^3/d, locally more than 100m^3/d. Influenced by sea flooding, it is mostly brackish water, and the development degree is low. The pore pressurized water layer in the coastal plain is composed of alluvial gravels and sands from various geological and historical periods and is distributed in a band. The aquifer group has a multilayer structure, with the upper part covered by marine silty clay, and the aquifer is separated from each other by a relatively stable water-insulating clay. Due to the different paleogeographic and sedimentary environments, especially the influence of successive sea floods, the water quality of the pressurized water is more complex, with total dissolved solids varying from 0.2 g/L to more than 10 g/L and often showing an interwoven distribution of salty and freshwater. As shown in the borehole information on the right in Figure 1, the shallow coastal layers in the study area are mostly clay, silt layer, and tuff layer, so water content is minimal in the shallow layer, and the aquifer in the study area may exist in the deeper layer or the pressurized aquifer.

The fractured rock aquifer of the Fujian coastal zone is mainly recharged by atmospheric precipitation and discharged by springs, while the pore water of loose rock and carbonate karst, as well as surface water, is the secondary recharge source. The groundwater type in Fujian coastal zone is mainly loose rock pore water, which is mainly stored in the Quaternary sand and gravel pebble layers, mostly distributed in the basin and plain area. The Quaternary sediment sequences are thin, generally, less than 100 m, the maximum thickness of the aquifer is only 20-30 m, and the distribution area is small, only scattered on both sides of the river or mountainous basins. As shown in the borehole on the right side of Figure 1, the stratigraphic sediment type in the study area along the Fujian coast are diverse, including clay, sand, granite, kaolinite, and amphibolite. Most of the estuarine plain aquifers are Holocene or Pleistocene alluvium, with a water volume of 500~1000 m^3/d, which is recharged by river water. The aquifers in the island peninsula area, such as the wind and sea sand layer, are recharged by rainfall, and the water volume is generally 100~500 m^3/d. The water volume in the mountain basins is less than 100m^3/d and is recharged by river water; the water volume in the bay plain is less than 100m^3/d, and the water quality is mostly brackish water.

3. Materials and Methods

3.1. Sampling. A total of 98 groundwater samples were collected from shallow civil wells along the coast of Zhejiang and Fujian provinces in October 2019, and the collection locations are shown in Figure 1. Water samples from Zhejiang Province were distributed in Ningbo, Taizhou, and Wenzhou on the coast; water samples from Fujian Province were distributed in Fuzhou, Putian, Quanzhou, Xiamen, and Zhangzhou along the coast. Samples were collected in situ and below 30 cm of the water table with a plexiglass sampler and then filled into two 250-mL polyethylene bottles for the analyses of trace elements and isotope components. Only one of the bottles was acidified with nitric acid to a pH<2. Conductivity, dissolved oxygen (DO), oxidation-reduction potential (ORP), TDS, pH, hydroxide isotopes, major anion concentrations (F^-, Cl^-, Br^-, and SO_4^{2-}), and cation concentrations (K^+, Ca^{2+}, Na^+, and Mg^{2+}) were examined for 42 of the 98 water samples, and the remaining 56 water samples were examined only for Cl^- concentration. All the samples were stored at 4°C in the laboratory and filtered with the 2 μm membrane on-site until the testing procedures could be performed.

3.2. Analytical Methods. The conductivity, TDS, pH, and other parameters of the groundwater samples were measured in the field using a portable water quality analyzer (YSI Professional Plus, YSI, USA). Groundwater samples were collected and stored in polyethylene bottles after rinsing the sampling bottle with well water three times. The entire bottle was filled and sealed with tape. Samples were collected, stored, and transported in strict accordance with the technical specifications for groundwater environmental monitoring. HCO_3^- was determined by phenolphthalein and standard HCl solution titration. The main anion concentrations (F^-, Cl^-, Br^-, and SO_4^{2-}) and cation concentrations (K^+, Ca^{2+}, Na^+, and Mg^{2+}) were analyzed by ion chromatography (ICS-3000, DIONEX, USA) and inductively coupled plasma emission spectrometry (XSERIES 2 ICP-MS, Thermo Fisher, USA), respectively. The charge balance error of all samples was less than 8%. $d^{18}O$ and dD values were analyzed by LGR liquid water isotope laser spectroscopy (MAT 253 PLUS, Thermo Fisher, USA) from the Institute of Oceanology, Chinese Academy of Sciences, and calculated according to the Vienna Standard Mean Seawater (V-SMOW). The analytical accuracies of the long-term standard measurements of $d^{18}O$ and dD were ±0.2‰ and ±0.6‰, respectively. Based on the concentration of Cl^-, the groundwater samples were divided into three types: freshwater (<250 mg/L), brackish water (250–1000 mg/L), and saline water (>1000 mg/L).

To identify the effect of mixing seawater and groundwater on the chemical composition of groundwater, the end-member mixing model was used to study the mixing of

Table 1: Statistical results of groundwater chemical and physical components.

Components	Zhejiang				Fujian			
	Min	Max	Mean	CV	Min	Max	Mean	CV
Na^+ (mg/L)	52.43	779.90	255.29	0.71	37.05	1321.00	184.15	1.87
Ca^{2+} (mg/L)	10.37	68.25	31.33	0.47	14.54	259.60	63.78	0.97
Mg^{2+} (mg/L)	4.07	67.14	30.13	0.50	4.05	142.50	19.78	1.88
K^+ (mg/L)	6.95	64.89	23.81	0.56	2.17	59.40	17.25	0.93
HCO_3^- (mg/L)	124.90	1026.00	440.06	0.42	58.09	393.53	195.78	0.44
SO_4^{2-} (mg/L)	3.35	107.68	31.92	0.76	11.70	581.38	102.97	1.43
Cl^- (mg/L)	42.26	1773.80	397.89	0.94	29.52	4061.10	433.00	2.53
F^- (mg/L)	0.14	0.94	0.51	0.35	0.15	0.47	0.28	0.39
TDS (mg/L)	312.00	5642.00	1371.27	0.81	271.70	7845.50	1162.27	1.74
DO (mg/L)	0.97	4.09	1.72	0.35	1.42	2.84	1.98	0.23
ORP (mV)	-226.10	20.60	-51.94	-1.26	39.40	440.00	103.14	1.02

groundwater. In the end-member mixing model, inland underground fresh water and seawater were selected as two end-members. Based on the mass balance, the two end-members are mixed in proportion, and a mixing line is drawn according to different mixing proportions that describe the mixing process of groundwater and seawater. The formula of the end-member mixed model is as follows:

$$C_m = C_s \times X + C_f \times (1 - X). \tag{1}$$

where C_m is the concentration of each component in the mixed solution, C_s is the concentration of each component in seawater, C_f is the concentration of each component in freshwater, and X is the mixing ratio of seawater. By setting different X values, the mixing line of seawater and underground freshwater can be obtained, which is called the salt-underground freshwater mixing line.

4. Results and Discussion

4.1. Hydrochemical Characteristics of Groundwater

4.1.1. Hydrochemical Composition. The results of groundwater chemical and physical components are shown in Table 1. The dominant cation in both provinces was Na^+. The mean Na^+ concentration in Zhejiang Province was 255.29 mg/L, higher than the drinking water standard of 200 mg/L [45], with a low coefficient of variation (0.71) indicating that groundwater in most areas of Zhejiang Province has a high concentration of Na^+. The mean value of Na^+ was lower in Fujian Province (184.15 mg/L), but the maximum value was 1321 mg/L, much higher than the standard limit value. The high coefficient of variation (1.87) indicates that Na^+ varies a lot in Fujian Province. The average values of Ca^{2+} and Mg^{2+} in Zhejiang Province are 31.33 mg/L and 30.13 mg/L, respectively, but the Ca^{2+} content (63.78 mg/L) was generally higher than Mg^{2+} (19.78 mg/L) in Fujian Province. The least abundant cation was K^+, with an average concentration of approximately 20 mg/L in both provinces.

The major anion in both provinces was Cl^-, with average values in Zhejiang Province and Fujian Province of 397.89 mg/L and 433.00 mg/L, respectively. The average values in both provinces exceeded the drinking water quality standard value of 250 mg/L [45], inferring the existence of Cl^- pollution of groundwater in the study area. The maximum values and coefficients of variation in Fujian Province were higher than in Zhejiang Province, indicating that there are individual areas in Fujian Province with serious exceedances of groundwater Cl^-. What is more, Cl^- is an indicator of SWI [46], considering the geographical location and the concentrations of Na^+ and Cl^-; SWI may have affected groundwater quality in the two provinces. The average value of HCO_3^- in Zhejiang Province was 440.06 mg/L, which is higher than the average concentration of 195.78 mg/L in Fujian Province. The least abundant anion was SO_4^{2-}, with average values in Zhejiang Province and Fujian Province of 31.92 mg/L and 102.97 mg/L, respectively. The F^- concentration in groundwater was slightly higher in Zhejiang Province that in Fujian Province, but the F^- content in the groundwater samples of both provinces was below the standard limit.

The mean values of total dissolved solids (TDS) in Zhejiang and Fujian provinces were 1371.27 mg/L and 1162.63 mg/L, which were greater than the groundwater quality standard limit value of 1000 mg/L [45]. A positive/negative ORP value means that groundwater is in an oxidizing/reducing environment [47]. The average redox potential in Zhejiang Province was −51.94 mV, with a reductive environment, while the average redox potential in Fujian Province was 103.14 mV, with an oxidizing environment. Besides, ORP is also related to the dissolved oxygen (DO), and the average groundwater DO concentration in Zhejiang Province was 1.72 mg/L, which is lower than the DO of 1.98 mg/L in Fujian Province. Therefore, the groundwater in Fujian Province may be in a relatively open environment.

4.1.2. Major Ion Correlation. Correlation analysis can reveal the similarities and differences of groundwater hydrochemical parameters [48]. To effectively reflect the correlations and differences among groundwater indicators, the Pearson correlation coefficients of each major ion and TDS in groundwater were calculated separately using SPSS software. TDS was significantly positively correlated with all ions

Table 2: Pearson correlation matrix of each indicator.

	Ca^{+}	Mg^{+}	Na^{+}	K^{+}	HCO_3^-	SO_4^{2-}	Cl^-	TDS
Ca^{2+}	1	0.651**	0.659**	0.346*	-0.233	0.882**	0.820**	0.701**
Mg^{2+}		1	0.924**	0.550**	0.405**	0.662**	0.921**	0.912**
Na^{+}			1	0.603**	0.441**	0.612**	0.953**	0.968**
K^{+}				1	0.340*	0.373*	0.581**	0.601**
HCO_3^-					1	-0.210	0.185	0.330*
SO_4^{2-}						1	0.779**	0.636**
Cl^-							1	0.962**
TDS								1

**At level 0.01 (double tail), the correlation was significant. *At level 0.05 (double tail), the correlation was significant.

except HCO_3^-, and the correlation coefficients with Na^+, Cl^-, and Mg^{2+} were all above 0.9 (Table 2), indicating that the increase of TDS was mainly influenced by the three ions. The correlation between HCO_3^- and other ions was weak, and this parameter might be influenced by the background value of the original ion content in the environment. Cl^-, Na^+, Ca^{2+}, Mg^{2+}, and SO_4^{2-} all have correlation coefficients above 0.6. According to the above statistical results, both Na^+ and Cl^- exceed the standard, and their high concentrations may be caused by SWI. Therefore, the contents of Ca^{2+}, Mg^{2+}, and SO_4^{2-} in groundwater may also be related to SWI. Studies have shown that SWI can increase the contents of Na^+, Cl^-, Mg^{2+}, and SO_4^{2-} in groundwater, and the change in Ca^{2+} may be related to changing the hydrodynamic conditions of inland groundwater and the solubility of carbonate minerals caused by SWI.

4.1.3. Hydrochemical Types. Piper diagrams are used to analyze water chemistry characteristics and water samples with similar water chemistry cluster together, allowing simultaneous comparison of the water chemistries of different water samples and revealing water chemistry processes occurring in different regions [49].

Generally speaking, the Mg^{2+}, Ca^{2+}, and HCO_3^- concentrations in fresh groundwater are high, and Cl^- and Na^+ concentrations increase after the salinization of groundwater. According to Figure 2, we conclude that the HCO_3^- concentration in the two provinces was 10–80%, the Cl^- and Na^++K^+ concentrations were 20–90%, the Mg^{2+} concentration was 5–40%, the Ca^{2+} concentration was 5–60%, and the SO_4^{2-} concentration was ≤20%. The points of the two provinces were concentrated in the lower right of the plot, indicating that the content of alkali metal ions was high; during the gradual salinization of groundwater, the ions gradually shifted from carbonate bases to noncarbonate bases with elevated strong acid ions. There was an obvious trend of increasing Cl^- and Na^+ and decreasing Mg^{2+}, Ca^{2+}, and HCO_3^- as the groundwater changed from freshwater to brackish water and then to saltwater. Groundwater in Zhejiang Province evolved from Ca/Mg-HCO_3 type in freshwater to Na-Cl type in brackish and saltwater; while groundwater in Fujian Province evolved from mainly Ca/Mg-HCO_3 type to Na(Ca/Mg)-Cl type in brackish and saltwater.

4.2. Occurrence of Water and Salt

4.2.1. Origin of Groundwater and Salt. The hydrogen and oxygen stable isotope compositions of various water bodies have extensive spatiotemporal heterogeneity [50, 51]. Without high-temperature water-rock interactions and strong evaporation, ^2H and ^{18}O isotopes are considered to be conserved and stable in the environment [52, 53]. Therefore, stable isotopes of hydrogen and oxygen can indicate the formation, migration, and evolution processes of groundwater. The local meteoric water line of Fujian (LWML-FJ, $\delta^2H = 8.52\delta^{18}O + 13.6$, $r^2 = 0.97$) was obtained based on data from the Fuzhou station (13°43′ 48″ N, 100°30′ 0″ E, altitude 10 m) of the International Atomic Energy Agency (IAEA) from 1969 to 2015. And the local meteoric water line of Zhejiang (LMWL-Z, $\delta^2H = 8.29\delta^{18}O + 15.9$, $r^2 = 0.98$) was acquired from the previous study [54]. As Figure 3 shows, the LMWL-ZJ and LMWL-FJ were both above the global meteoric water line (GMWL), reflecting the humid and rainy climate characteristics of the two provinces.

The average values of groundwater $\delta^{18}O$ and δ^2H in Zhejiang Province are −5.57 and −37.12, respectively, and the average values of groundwater $\delta^{18}O$ and δ^2H in Fujian Province are −5.60, and−36.54. Most of the samples fell below the LWML, and the groundwater in the two provinces was mainly recharged by precipitation but also was affected by evaporation, which makes the hydrogen isotope more easily separated from water than the oxygen isotope [55]; in contrast, the influence of seawater on groundwater in some areas made the values of $\delta^{18}O$ and δ^2H closer to those of seawater. The samples from Zhejiang deviated further from the LMWL than the samples from Fujian, indicating that the Zhejiang samples were more affected by evaporation and seawater mixing.

The rock salt dissolution releases equal equivalent amounts of Na^+ and Cl^-, so if the Na^+ and Cl^- in groundwater originate from rock salt dissolution, their equivalence ratios should be approximately 1. When groundwater is invaded by seawater, the Na^+/Cl^- ratio is less than 1 and

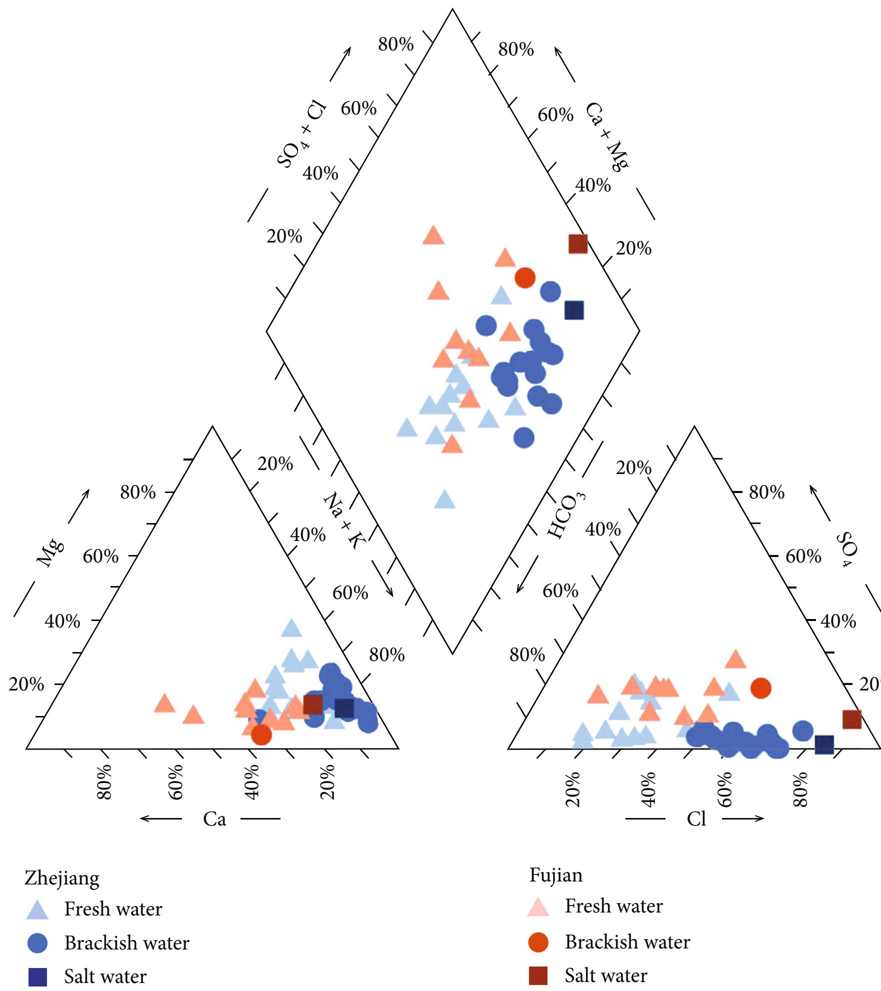


FIGURE 2: Groundwater water chemistry types.

closer to 0.55. The ratio of Na^+/Cl^- in freshwater was >1 (Figure 4(a)), while in brackish water it was approximately 1 and gradually approached the ratio of 0.55 in saltwater. With the increase in Cl^-, the value of Na^+/Cl^- gradually decreased, and most points were below the mixing line because of the large content of Cl^- in seawater invasion; the increase rate of Cl^- was greater than that of Na^+. The mixing lines of the two provinces overlapped. When the Cl^- content was low, the Na^+/Cl^- ratio was far from the seawater standard line and greater than 1, indicating that the groundwater was influenced by silicate weathering and cation exchange; as the Cl^- content increased, the Na^+/Cl^- ratio decreased and converged to the standard line, indicating that the proportion of groundwater salinity derived from SWI increases as the degree of SWI increases.

Most Br^- in nature is stored stably in the ocean, so Br^- can be used to infer the source of salinity in coastal groundwater, and the Br^-/Cl^- ratio is a good indicator that groundwater is affected by seawater. As can be seen from Figure 4 the content of Br^- was between 0.001 mEq/L and 0.1 mEq/L, and the mixing line was closely distributed parallel to the seawater Br^-/Cl^- ratio line ($Br^-/Cl^- = 0.0015$)[40]. Most of the water samples were distributed on the mixing line, indicating that the Cl^- content in the coastal groundwater of the two provinces was closely related to SWI.

4.2.2. Evolution of Groundwater Chemistry. Gibbs diagrams can be used to analyze the main effects of hydrology on the evolution of groundwater chemistry, including precipitation effects, mineral weathering, and evapotranspiration [56]. Brackish and saline water have high Cl^- and Na^+ contents, high salinity, and relatively high TDS. Evaporation and concentration are the main factors affecting the chemical composition of brackish seawater. Figure 5 illustrates that mineral weathering was the main salt source of fresh groundwater, and evaporation also affected fresh groundwater salinity. In contrast, evaporation was dominant in brackish and saline waters. Combined with the isotopic composition, it is clear that SWI and evaporation caused the increase in the salinity of coastal groundwater.

Ion exchange during the mixing of seawater and groundwater affects the deviation of groundwater from the mixing

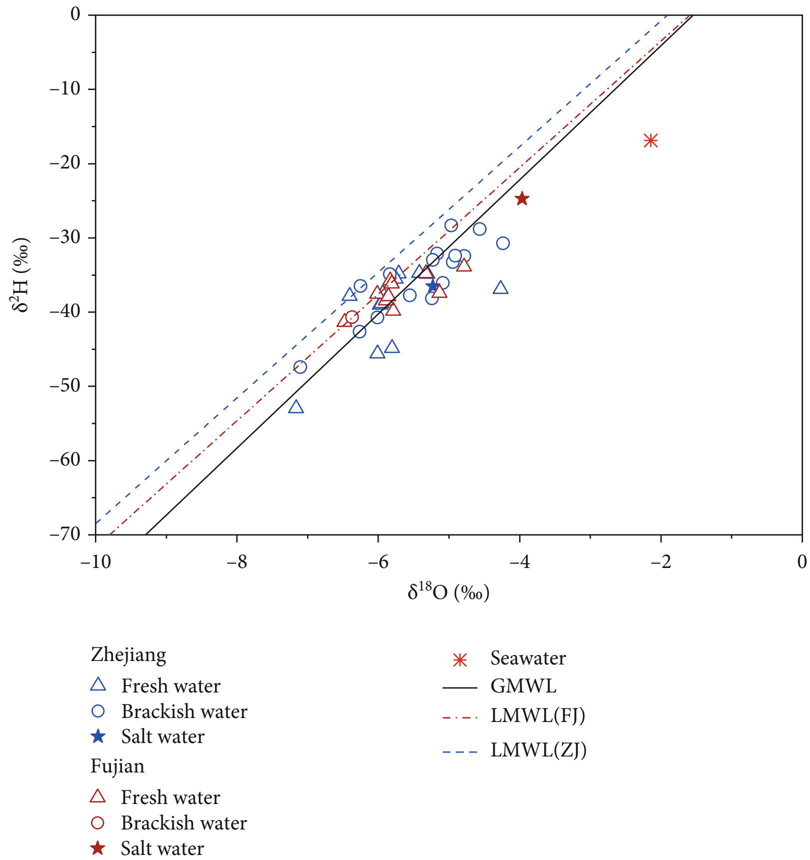


FIGURE 3: Hydrogen and oxygen isotope scatter diagram.

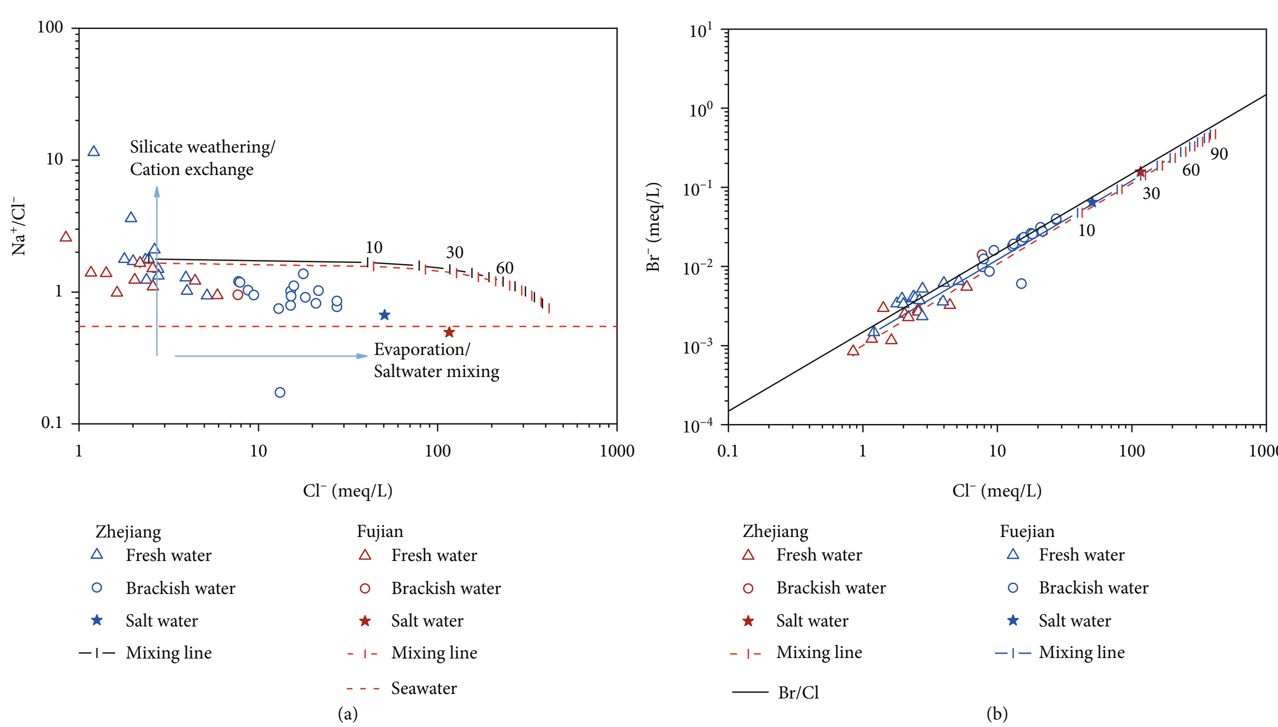


FIGURE 4: Na/Cl and Cl (a) and Br and Cl (b) indicate the source of groundwater salinity.

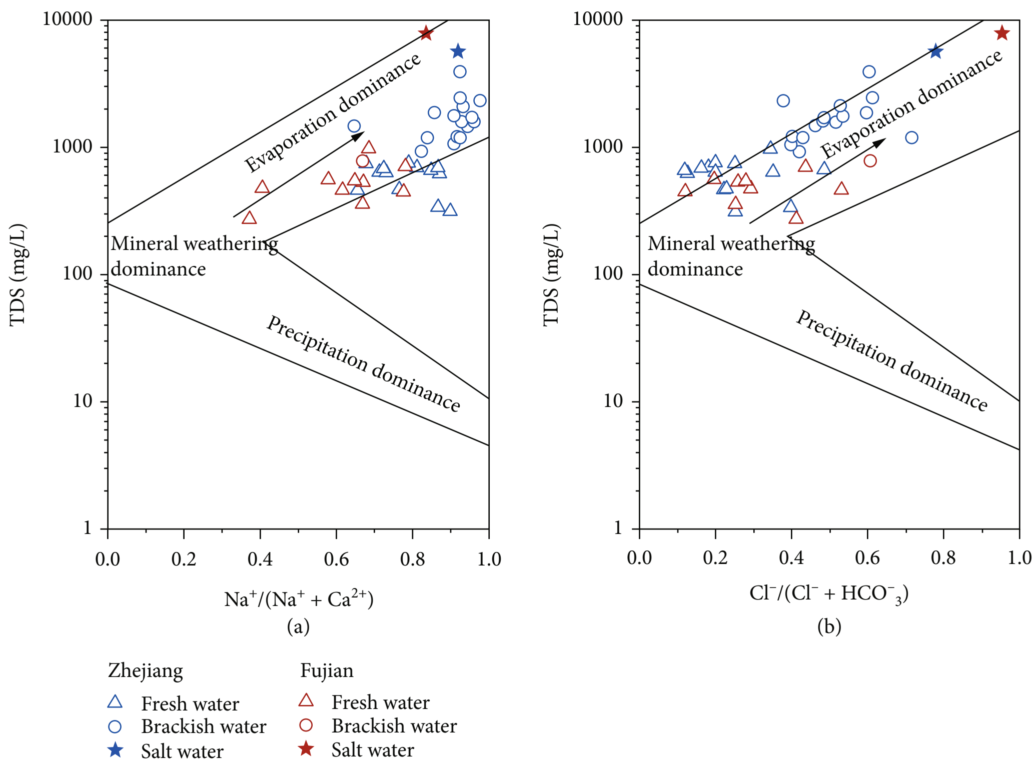


FIGURE 5: Gibbs plot of groundwater in the study area.

line [3]. The ion exchange during mixing occurs mainly between Na^+ and Ca^{2+}/Mg^{2+} by the following mechanisms.

$$\mathrm{Na-Solid} + Ca^{2+}\left(Mg^{2+}\right)\mathrm{aq} = 2Na^{+}\mathrm{aq} + \mathrm{Ca(Mg)-Solid} \tag{2}$$

$$2Na^{+}\mathrm{aq} + \mathrm{Ca(Mg)-Solid} = \mathrm{2Na-Solid} + Ca^{2+}\left(Mg^{2+}\right)\mathrm{aq} \tag{3}$$

The chlor-alkali index (CAI) can further determine the presence of direct cation exchange (6) or reverse cation exchange (7). CAI is calculated by the following equation.

$$\mathrm{CAI} = \frac{Cl^{-} - (Na^{+} + K^{+})}{Cl^{-}} \tag{4}$$

When CAI<0, groundwater Ca^{2+} or Mg^{2+} undergo direct cation exchange with groundwater Na^+ (2); when CAI>0, groundwater Ca^{2+} or Mg^{2+} undergo reverse cation exchange with groundwater Na^+ (3).

We calculated that 33.3% of groundwater water samples in Zhejiang Province have CAI<0 and 66.7% have CAI>0. In Fujian Province, 58.3% of groundwater water samples have CAI<0, and 41.7% had CAI>0. The reaction that occurred in Fujian Province was mainly as per equation (3), the reaction that occurred in Zhejiang Province was mainly as per equation (2), and the groundwater environment in Zhejiang had high Na^+ content, and Fujian had high Ca^{2+} content. We found that the exchange degree of Ca-Na in the groundwater of the study area was higher than that of Mg-Na. With the increased Na^+ content, the Ca^{2+} and Mg^{2+} contents also increased and gradually approached the mixing line, indicating that the exchange rate of freshwater was higher than that of brackish and saline water (Figure 6).

4.3. Salinity Distribution of Regional Groundwater. Conductivity, total dissolved solids (TDS), and Cl^- concentration can be used to determine SWI [57]. In this study, Cl^- concentration was chosen as the standard classification, and the results (Figure 7) indicated that the 49 water samples from Zhejiang Province comprised 22 freshwater samples, 22 brackish water samples, and 5 saline water samples; 35 freshwater samples, 5 brackish water samples, and 9 saline water samples in Fujian Province.

In summary, Zhejiang Province had 44.90% freshwater samples and 44.90% brackish water samples; Fujian Province had 71.43% freshwater samples and 10.20% brackish water samples. The Cl^- content of water samples in Zhejiang Province decreased with increasing distance (Figure 7) and was distributed in strips; however, Cl^- exceeded the standard limits in Fujian Province only in some specific areas (e.g., Zhanggang Town, Changle District, Old Town, and Zhangpu County) and showed a spotty distribution, which was related to the variation in coastal hydrogeological conditions in Fujian Province and also indicated that SWI was more widespread in Zhejiang Province than in Fujian Province.

4.3.1. Effect of SWI on the Regional Groundwater Chemistry. As shown in Figure 8, with the worsening of SWI, the Na^+ content in groundwater continued to increase, and the

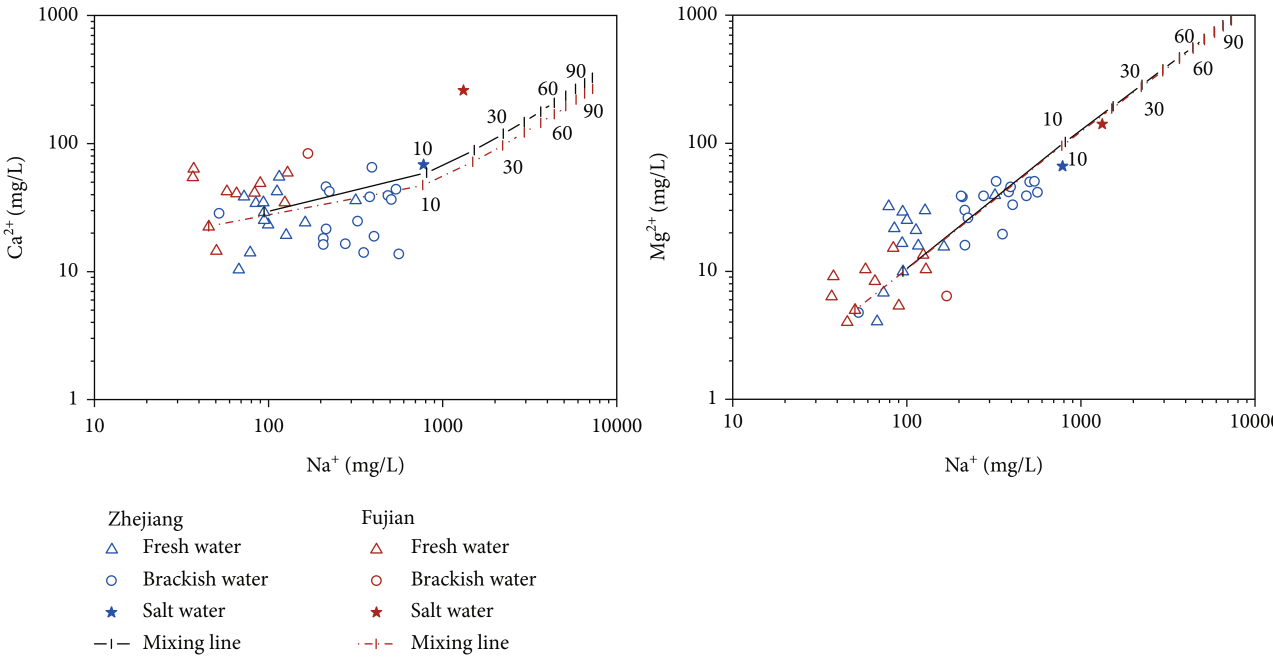


Figure 6: Relationship between Ca and Na(a), and between Mg and Na(b) reflecting the degree of ion exchange.

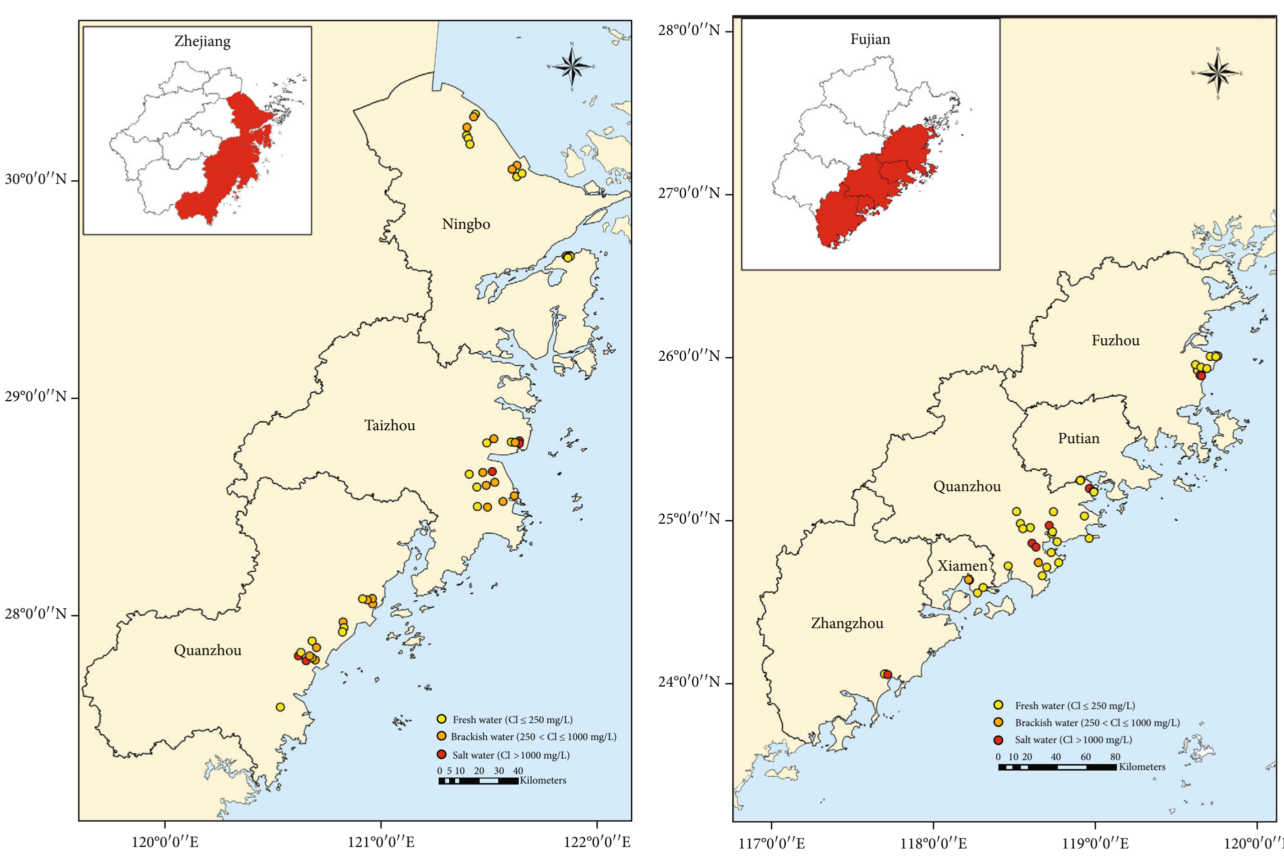


Figure 7: Status of seawater intrusion.

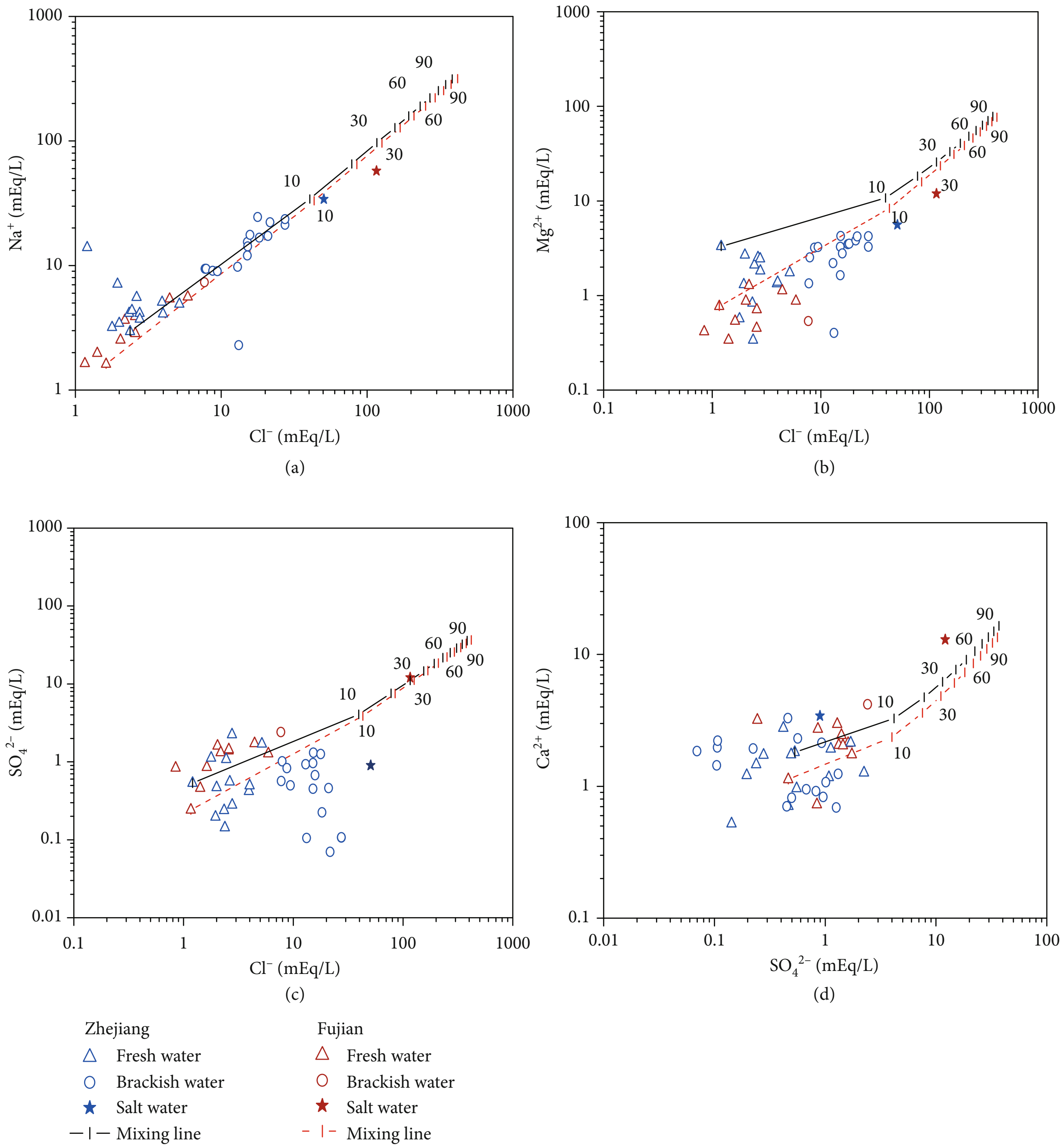
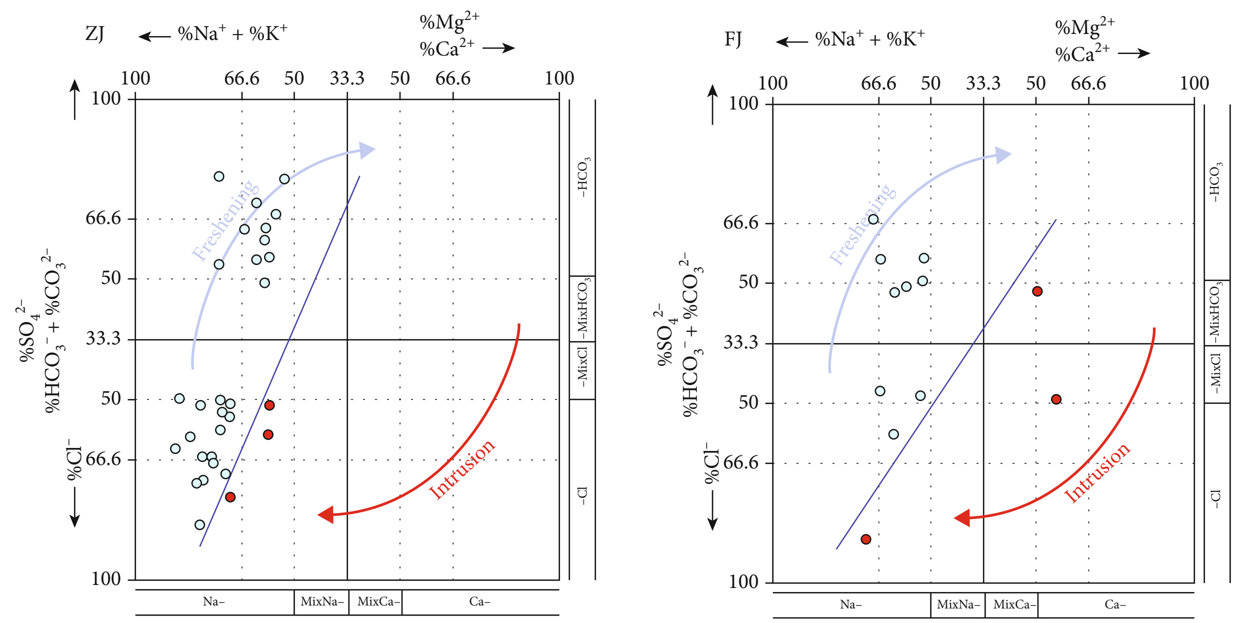


Figure 8: Impact of seawater intrusion on major groundwater ions.

growth trend was nearly linear and coincided with the seawater-freshwater mixing line. Most of the Na^+ mEq ranges in the two provinces were within 10% of the seawater-freshwater mixing line ratio. Most Na^+ contents in Fujian Province were <10 mEq/L, while most of the Na^+ contents in Zhejiang Province were >10 mEq/L, indicating that seawater had a greater impact on groundwater in Zhejiang Province.

The impact of SWI on groundwater also caused the changes in Mg^{2+}, with more than half of the samples with Mg^{2+} content >2 mEq/L occurring in Zhejiang Province and most of the samples with Mg^{2+} content <1 mEq/L occurring in Fujian Province; Mg^{2+} increased with Cl^- content. The SO_4^{2-} contents in the groundwater of both provinces were mostly <1 mEq/L. With the increase of Cl^-, there was an increasing trend of SO_4^{2-} when Cl^- <10 mEq/L and a decreasing trend of SO_4^{2-} content when Cl^- > 10 mEq/L. That is because when the groundwater is saturated with SO_4^{2-}, Ca^{2+} would combine with SO_4^{2-} to form gypsum, although the seawater brought more SO_4^{2-}; the SO_4^{2-} concentration decreased in general. Moreover, Ca^{2+} and SO_4^{2-} originate from gypsum dissolution, and the milligram equivalent ratio of Ca^{2+} to SO_4^{2-} should be equal to 1. The ratio of most water samples from both provinces



(a) (b)

Figure 9: Water chemistry phase division and evolution.

was greater than 1 and close to the seawater-freshwater mixing line, indicating that the main source of Ca^{2+} is not seawater, but the value was influenced by seawater.

4.3.2. Evolution Trend of Seawater Intrusion. The hydrochemical facies evolution (HFE) map is used to identify the evolution process and development of SWI in coastal aquifers [58]. As shown in Figure 9, the HFE-D was divided into 16 parts representing 16 water chemical phases, based on the milligram equivalent percentage values of the major ions in groundwater and using 33% and 50% as the dividing lines. The opposite corners represent two basic water chemical phases: the upper-right Ca-HCO_3/SO_4 phase represents subsurface freshwater, the lower-left Na-Cl phase represents seawater, and the other water chemical phases are the result of the mixing and cation exchange of these two. The line between the lower-left and the upper-right (4-7-10-13) indicates the ideal situation (direct mixing without chemical reaction). If the mixing line between fresh groundwater and seawater is higher than this diagonal line, this indicates that the groundwater is undergoing freshening and there is a tendency to alleviate the SWI; if the mixing line is below the diagonal line, this indicates that the groundwater is undergoing intrusion, there is a tendency to deepen the SWI, and the groundwater will be salted.

As can be seen from Figure 9(a), in Zhejiang Province, the highest proportion of the water chemical phase was Na-Cl (60.00%) followed by Na-HCO_3 (33.33%), with 90% of water samples undergoing a freshening process and only 10% undergoing an intrusion process. In Fujian Province, the water chemical phase with the highest proportion was Na-HCO_3 (33.33%), followed by Na-MixHCO_3, Na-MixCl, and Na-Cl with the same proportion of 16.67%, with 75% of the water samples undergoing a freshening process and 25% undergoing an intrusion process (Figure 9(b)). Although the groundwater in Zhejiang and Fujian Province were affected by SWI, most of the water samples were with a trend toward freshening.

4.4. Groundwater Pollution

4.4.1. Nitrate Pollution. Nitrate is a common contaminant in groundwater and is closely related to human activities such as fertilizer, pesticide use, and industrial and domestic wastewater [3]. Nitrate is less harmful, but it can be converted into nitrite, which is toxic. Nitrite reacts with human blood to form methemoglobin, resulting in a decrease in blood's oxygen-carrying capacity. In addition, when nitrite reaches a certain concentration, carcinogenic, teratogenic, and mutagenic processes will occur in the human body.

According to the National Standardization Management Committee [45], the safe concentration of NO_3^- in drinking water should be less than 20 mg/L. The concentration range of NO_3^- in Zhejiang Province water samples was from 0.05 to 37.20 mg/L, and the average value was 6.42 mg/L. While Fujian Province's concentration range of NO_3^- was from 0.14 to 108.34 mg/L, the average value was 55.68 mg/L. Combined with Figure 10(a), Zhejiang Province water samples were almost all below the standard line, but Fujian Province water samples were almost all above the standard line, indicating that the groundwater in Fujian Province was seriously affected by nitrate. The NO_3^- content in Figure 10 significantly increased when subjected to nitrification and gradually increased with TDS, Cl^-, and Br^- when subjected to sewage infiltration and seawater mixing. Therefore, nitrification, seawater mixing, and sewage infiltration are all sources of NO_3^-. Previous studies suggested that agricultural and aquaculture nitrogen-containing wastewater

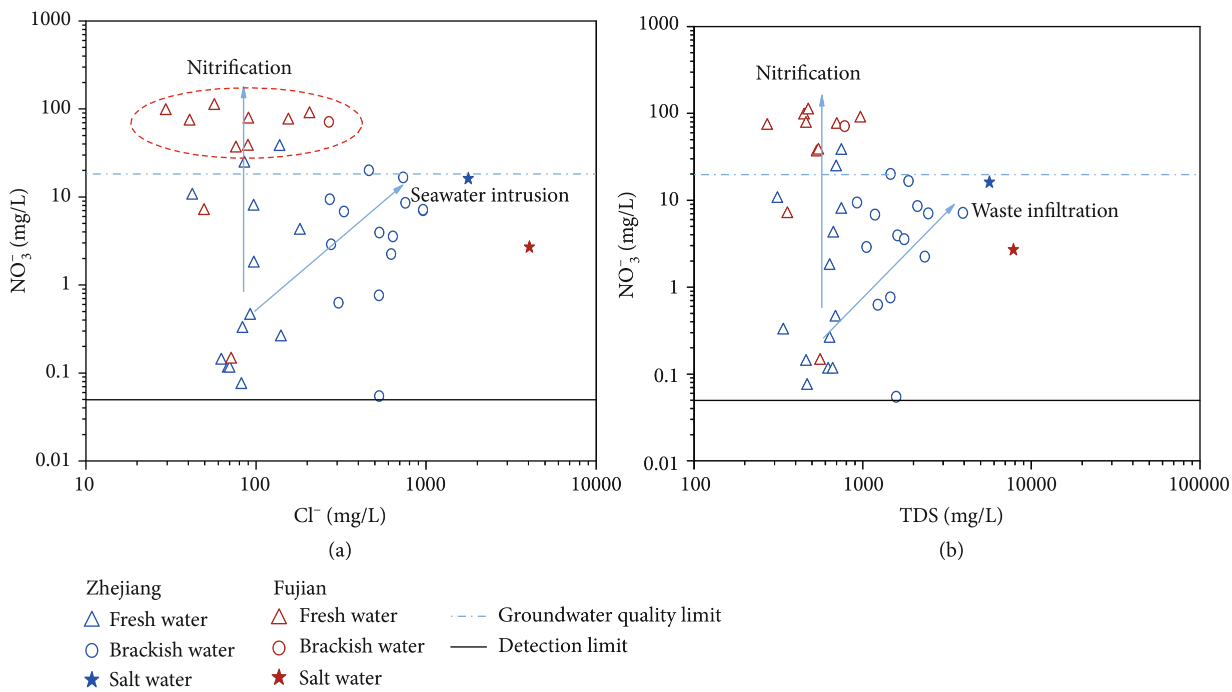


Figure 10: Relationship between NO_3 and TDS (a) and between NO_3, and Cl (b) reflecting the sources of NO_3 in groundwater.

was the main source of nitrate pollution in groundwater in Fujian Province [59, 60], so it is important for the reduction of nitrate pollution in groundwater by treating sewage before the discharge.

4.4.2. Fluoride Pollution. Fluoride is widely distributed in the environment, which is essential to human health; however, the deficiency or excess can both cause serious health problems [40]. If the fluoride content in water is less than 0.5 mg/L, the rate of dental caries will reach 70–90%, while if the local underground water fluoride content exceeds the standard limits, it will cause bone fluorosis, resulting in bone marrow malformation [61].

According to the national drinking water sanitation standard [62], the safe concentration of F^- in drinking water should be less than 1 mg/L. The concentration range of F^- in Zhejiang Province water samples was 0.14–0.94 mg/L, with an average content of 0.51 mg/L. The concentration range of F^- in Fujian Province was 0.15–0.47 mg/L, with an average content of 0.28 mg/L. Although the content of F^- in the water samples of the two provinces did not exceed the standard, the F^- concentration in the Zhejiang was higher than in Fujian in general (Figure 11(a)), and F^- concentrations gradually approached the seawater line with the increase of Cl^-, indicating that the groundwater in Zhejiang Province is at risk of exceeding of F^- under the influence of seawater. As is known that the alkaline environment is more conducive to fluoride enrichment in groundwater [59], groundwater F^-concentration in the study area also has a positive correlation with pH (Figure 11(b)). In addition, Figures 11(c) and 11(d) show that sodium-type and bicarbonate-type groundwater is also characterized by high fluoride content. To sum up, while being subjected to SWI, F^- also increases through evaporation and mineral weathering, indicating that SWI, mineral weathering, evaporation, and concentration are all important sources of F^- in the study area.

4.5. Groundwater Quality Assessment. WQI is an important parameter for establishing groundwater quality and its suitability for drinking purposes [63]. WQI is a technique of rating that provides the composite influence of individual parameters on the overall quality of water for human consumption. The standards for drinking purposes are as recommended by the WHO [64].

The calculation of WQI is in three steps. In the first step, each of the nine parameters was assigned a weight (w_i) according to its relative importance in the overall quality of water for drinking purposes. Maximum weight of 5 was assigned to the parameters, such as the total dissolved solids, fluoride, chloride, and nitrate, due to their importance in water quality assessment. Bicarbonate was given the minimum weight of 1 because it plays an insignificant role in water quality assessment. Other parameters like calcium, magnesium, sodium, and potassium were assigned a weight between 1 and 5 depending on their importance in water quality determination.

In the second step, the relative weight (W_i) was calculated from the following equation:

$$W_i = \frac{w_i}{\sum_{i=1}^{n} w_i}, \tag{5}$$

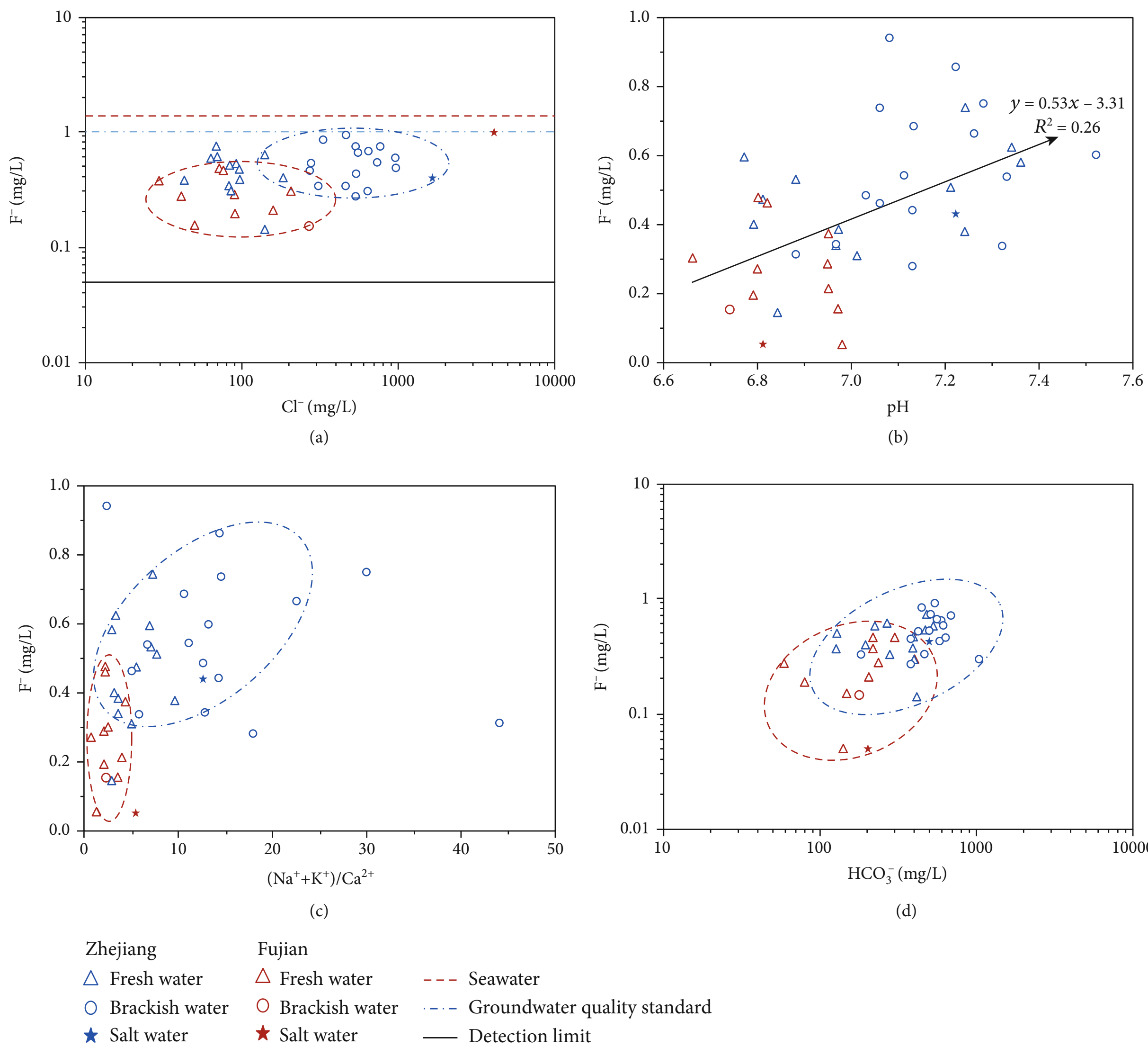


FIGURE 11: F^- content (a) and the relationships between $F^-$and pH (b), HCO_3(c), and (Na + K)/Ca (d).

where W_i is the relative weight, w_i is the weight of each parameter, and n is the number of parameters. Calculated relative weight (W_i) values of each parameter are given in Table 3.

In the third step, a quality rating scale (q_i) for each parameter was assigned by dividing the concentration in each water sample by its respective standard according to the guidelines described by the WHO [64], and the result was multiplied by 100:

$$q_i = \left(\frac{C_i}{S_i}\right) \times 100, \quad (6)$$

where q_i is the quality rating, C_i is the concentration of each chemical parameter in each water sample in milligrams per liter, and S_i is the Indian drinking water standard for each chemical parameter in milligrams per liter according to the guidelines of the WHO [64].

TABLE 3: Relative weights of chemical parameters.

Parameters (mg/L)	WHO standards (2014)	w_i	Relative weight $W_i = w_i / \sum_{i=1}^{n} w_i$
TDS	500	5	0.128
Bicarbonate	500	1	0.0256
Chloride	250	5	0.128
Sulfate	200	5	0.128
Nitrate	45	5	0.128
Calcium	75	3	0.0769
Magnesium	30	3	0.0769
Sodium	200	5	0.128
Potassium	12	2	0.0523
Fluoride	1.5	5	0.128

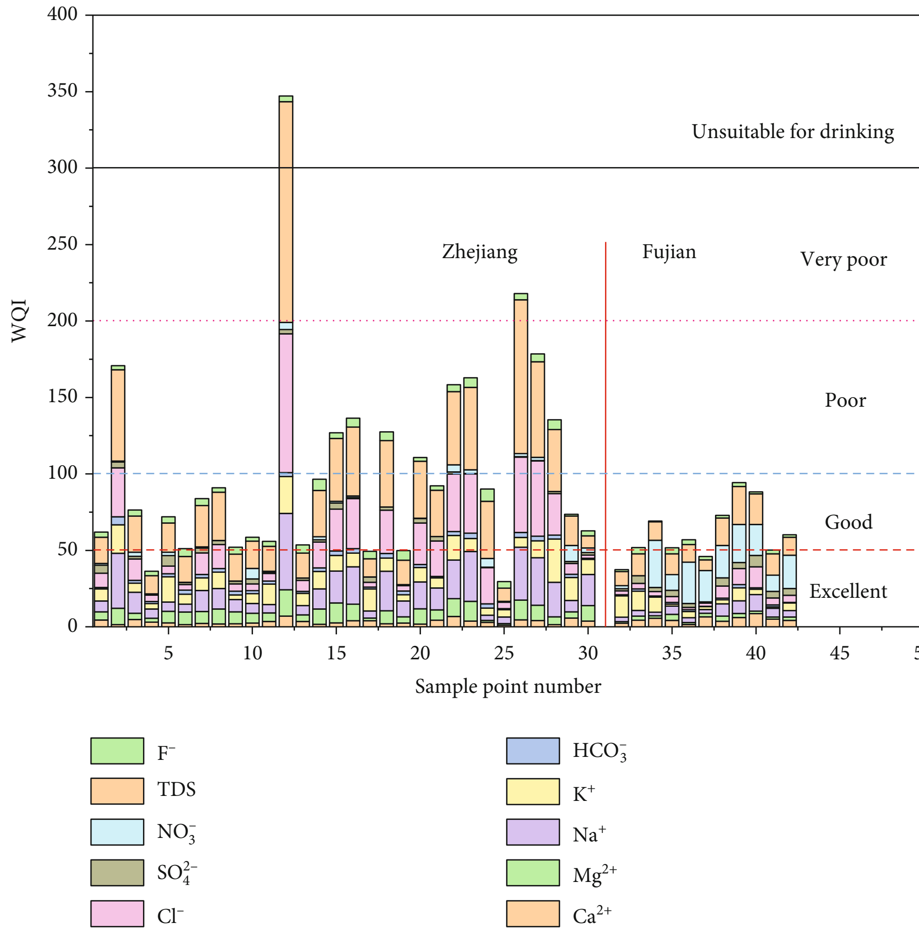


FIGURE 12: Bar graph of calculated WQI values.

The SI was first determined for each chemical parameter, which is then used to determine the WQI as per the following equation:

$$SI_i = W_i \times q_i, \tag{7}$$

$$WQI = \sum SI_i, \tag{8}$$

where SI_i is the subindex of ith parameter.

Water quality types were determined on the basis of WQI. The WQI value and water quality are classified as excellent water<50, 50 < good water<100, 100 < poor water < 200, 200 < very poor water < 300, and 300 < water unsuitable for drinking purposes.

The WQI value of Zhejiang Province ranged from 29.60 to 347.11 (Figure 12), which had 4 excellent water and 15 good water samples, accounting for 63.33% of the total water samples. Meanwhile, there were 9 poor water samples, 1 very poor water sample, and 1 unsuitable for drinking purposes. The WQI value of Fujian Province ranged from 37.33 to 621.78. There were 2 excellent water samples and 9 good water samples, accounting for 91.67%, and only one sample was unsuitable for drinking purposes.

Although there is one water sample unsuitable for drinking in both provinces, the proportion of good water quality was much higher in Fujian Province than in Zhejiang Province. For improving groundwater quality, enhancing the supply capacity of surface potable water and rational utilization of groundwater resources are the priority. Strictly controlling the generation, storage, and treatment of waste along the coast is also important.

5. Conclusion

(1) The hydrogen and oxygen isotopes (δ^2H and δ^{18}O) combined with Br/Cl, CAI, and Gibbs indicate that modern precipitation is the predominant origin of coastal groundwater, which is then affected by seawater mixing and evaporation. The salts originated from mineral weathering, and SWI was the predominant factor in increasing salinity in Zhejiang coastal groundwater. According to the hydrochemical analysis, the evolution path of groundwater in Zhejiang was Ca/Mg-HCO_3 → Na-Cl, and after SWI, the controlling factor of evolution was evaporation, while the evolution path of groundwater in Fujian was Ca/Mg-HCO_3 → Na(Ca/Mg)-Cl and the controlling factors of evolution were water-rock interaction and ion exchange. However, due to the abundant

precipitation and runoff, the evolution trend of SWI in both provinces was freshening

(2) Although groundwater samples of the two provinces did not exceed the standard limit, fluorine concentration in Zhejiang is higher than in Fujian. The increase of Na^+, K^+, and HCO_3^- concentrations in aquifers was caused by seawater intrusion, promoting the release and enrichment of fluorine in groundwater. The NO_3^- pollution in the two provinces was caused by nitrification, sewage infiltration, and SWI. The NO_3^- content in Fujian Province exceeded the fluorine standard because of livestock farming, agricultural activities, and domestic pollution along the coast. At present, groundwater in both provinces is affected to varying degrees by anthropogenic activity, such as excessive groundwater exploitation and unstructured agricultural activities

Finally, the WQI assessment indicated that the water quality in Fujian Province was better than in Zhejiang Province. Although groundwater exploitation in the coastal zone has been effectively controlled by the government, seawater intrusion is still a significant issue in Zhejiang Province and, therefore, the effect of excessive exploitation and seawater level rise should be paid more attention to, while the main issue for groundwater in Fujian Province is nitrate pollution rather than seawater intrusion. Therefore, the first and foremost is to control the generation, storage, and treatment of waste from agricultural and aquaculture in Fujian Province. To sum up, public awareness of the risk posed by SWI and groundwater pollution should be strengthened.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of 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.

Authors' Contributions

Tengfei Fu contributed to conceptualization, formal analysis, funding acquisition, and writing-review and editing. Chen Qi contributed to writing original draft, software, data curation, and visualization. Zhenyan Wang contributed to methodology, supervision, validation, and writing-review and editing. Chenzhe Li: contributed to writing original draft and visualization. Wenquan Liu performed the investigation and methodology. Guangquan Chen contributed to data curation. Qiao Su performed the investigation. Xingyong Xu contributed to funding acquisition and methodology. Hongjun Yu contributed to conceptualization and funding acquisition and writing-review and editing.

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