Groundwater Chemical Characteristics and Salinization Mechanism in the Coastal Plain of the South Bank of Laizhou Bay

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Abstract. Based on 59 groundwater samples which were collected on the south bank of Laizhou Bay, the main ionic ratios analysis and the main ionic Piper three-line diagram was done. The results show that from offshore to nearshore, groundwater hydrochemical is mainly freshwater, brackish water, salt water, and brine. And the ion type transforms from HCO₃⁻•Ca to HCO₃⁻•Cl⁻•Ca, then converted to HCO₃⁻•Cl⁻•Na and further transformed into Cl⁻•HCO₃⁻•Na, then changed to Cl⁻•Na regularity. By calculating some conventional water ion ratios which include Na/CL, K/CL, Ca/CL, SO₄/CL, the hydrogeochemical effects of water-rock interactions, the source of groundwater salinity and the mechanism of groundwater salinization were investigated in this study. The anion-cation exchange reaction in the groundwater of the brackish water transition zone in the northern part of the monitoring section is more obvious than that in the southern underground freshwater. The main hydrogeochemical processes occurring in the brackish water transition zone are the dissolution of minerals (calcite, anhydrite and rock salt), precipitation of gypsum and dissipation of carbon dioxide. And the main cation exchange in this area is Ca-Na exchange.

1. Introduction
Salinization of groundwater is the most common form of groundwater pollution and has become a global environmental problem that needs to be solved urgently. Excessive exploitation of groundwater in coastal areas has resulted in seawater invasion, salinization of groundwater and serious deterioration of water quality, which has aggravated the contradiction between supply and demand of water resources in the region [1-3]. Studying the chemical characteristics of groundwater and analyzing the mechanism of groundwater salinization are beneficial to understanding the evolution law of groundwater environment and rational planning, utilization and management of groundwater resources [4-5]. Laizhou Bay, located on the southern coast of Bohai Bay, is a relatively developed area of China's economy and one of the most serious areas of groundwater environment in China. With the rapid growth of regional economy and population expansion, the demand for water resources has increased sharply. Water resources have become a bottleneck restricting the rapid economic development of the region. It is of great practical value to study the hydrochemical characteristics of groundwater and the mechanism of salinization for...
the rational development and utilization of water resources and the formulation of effective measures for the protection of water resources [6-7]. However, the research on the south coast of Laizhou Bay mainly focuses on the types and mechanism of seawater intrusion, while the research on groundwater chemistry and salinization mechanism is relatively less. Therefore, in this paper, the chemical characteristics and salting mechanism of groundwater in the southern coast of Laizhou Bay are deeply analyzed by correlation analysis, Piper diagram and main conventional ion ratios, which provide scientific basis for the rational development and utilization of water resources in this area.

2. Overview of Study Area
The coastal plain on the south coast of Laizhou Bay is a fragile ecological area under the common influence of land-sea interaction and human activities. The overall terrain of the study area is high in the South and low in the north, and inclines to the ocean. The ground elevation is below 5 m. In the east and west direction, there are some changes due to the accumulation of rivers, that is, the river belt and the paleochannel belt are slightly higher, and the depression is formed between them. Under natural conditions, underground fresh water is mainly supplied by infiltration of atmospheric precipitation, and the overall direction of underground fresh water is from south to north. Due to over-exploitation of underground fresh water and brine resources for many years, two freshwater mining funnels were formed in Hanting and Changyi in the south, respectively, and brine descending funnels were formed in Zaohu and Xili Yu in the north [8-9].

![Location map of study area.](image)

**Figure 1.** Location map of study area.

3. Materials and Methods
A total of 59 groundwater samples were collected from the southern coast of Laizhou Bay for water quality detection. In the process of sample collection, the interfaces representing different degrees of saline water intrusion are fully covered, including freshwater/brackish water (salinity is less than 3g/L, chloride ion content is less than 100mg/L), saline water (salinity is higher than 3g/L, chloride ion content is 100-1000mg/L), brine (salinity is more than 50g/L, chloride ion content is higher than 1000mg/L). In addition to conventional pH value, conductivity EC and salinity TDS, the contents of analysis and testing include Cl\(^{-}\), SO\(_4\)\(^{2-}\), HCO\(_3\)\(^{-}\), Na\(^{+}\), K\(^{+}\), Ca\(^{2+}\), Mg\(^{2+}\), Br\(^{-}\), I\(^{-}\), F\(^{-}\), Sr. Cl\(^{-}\) was determined by silver nitrate volumetric method; Flame Atomic Absorption Spectrophotometry was used to determine K\(^{+}\) and Na\(^{+}\); The determination of Ca\(^{2+}\) and Mg\(^{2+}\) was carried out by atomic absorption spectrometry; The determination of SO\(_4\)\(^{2-}\) was carried out by BaCrO\(_4\) spectrophotometry; HCO\(_3\)\(^{-}\) was determined by titration; Br\(^{-}\), I\(^{-}\) and F\(^{-}\) were determined by ion chromatography. The calculation of hydrochemical parameters and the drawing of hydrochemical maps were respectively completed by EXCEL, piper three-line maps and Mapgis software.

4. Analysis of Hydrochemical Characteristics of Groundwater

The test data of some groundwater samples are shown in table 1. According to the content of chloride ions, the samples were divided into three groups, among which 13 samples with chloride ions less than 100mg/L were in group A, 28 samples with chloride ions between 100mg/L and 1000mg/L were in group B, and 18 samples with chloride ions higher than 1000mg/L were in group C. Figure 2 is a piper three-line map, figure 3-8 shows the relationship between the ratios of SO\(_4\)\(^{2-}\), Ca\(^{2+}\), K\(^{+}\), Mg\(^{2+}\), Na\(^{+}\), HCO\(_3\)\(^{-}\) and Cl\(^{-}\) (measured in mg/L value) and Cl\(^{-}\), which are expressed in logarithmic coordinates. The horizontal line in figures represent the theoretical seawater dilution line of the corresponding ions, the ratio of ions and Cl on the line is equal to the Marine ratio, so it is also called the seawater ratio line [10-11]. Figure 9 shows the relationship between halogen elements Br and Cl. The three oblique lines in the figure from top to bottom represent the Br/Cl ratio of 1×10\(^{-3}\), 3.47×10\(^{-3}\) and 1×10\(^{-2}\), respectively. Br/Cl=3, 4.7×10\(^{-3}\) represents the dilution line of seawater, Br/Cl=1×10\(^{-3}\) represents the dissolution line of stone salt, and Br/Cl=1×10\(^{-2}\) represents the high value of organic pollution (such as livestock sewage) [12-14].

| number | coordinate | position | Welldepth(m) | HCO\(_3\) | Cl\(^{-}\) | K\(^{+}\) | Na\(^{+}\) | Ca\(^{2+}\) | Mg\(^{2+}\) | SO\(_4\)\(^{2-}\) | F\(^{-}\) | Br\(^{-}\) | I\(^{-}\) | Sr | TDS |
|--------|------------|----------|--------------|----------|---------|---------|---------|---------|---------|---------|-------|-------|-------|    |
| 120039 | 6355400491299 | Sumiaocun | 400 | 363.1 | 24.8 | 1.8 | 39.5 | 85.7 | 21.4 | 51.5 | 0.34 | 0.1 | <0.05 | 0.43 | 596.2 |
| 120041 | 63731404106510 | Hanjia | 480 | 326.5 | 42.5 | 2.7 | 95.4 | 47.2 | 25 | 84.9 | 0.32 | 0.1 | <0.05 | 0.48 | 624.2 |
| 112142 | 7097540477990 | Chenjiawazi | 100 | 238 | 46.1 | 0.6 | 28.6 | 56.2 | 14 | 9.2 | 0.64 | 0.1 | <0.05 | 0.56 | 392.7 |
| 112139 | 7693804126685 | Shabuzhuang | 40 | 189.2 | 65.6 | 0.8 | 28.1 | 81.9 | 14.3 | 52.5 | 0.29 | <0.1 | <0.05 | 0.89 | 432.4 |
| 111197 | 7083260474714 | Wanghucunnan | 30 | 265.4 | 78 | 0.6 | 38.1 | 146.9 | 42 | 47.9 | 0.76 | 0.4 | <0.05 | 1.27 | 910.2 |
| 120042 | 63731404106510 | Mahoucun | 280 | 308.2 | 85.1 | 2.3 | 92.3 | 59.8 | 37.8 | 112.6 | 0.4 | 0.4 | <0.05 | 0.81 | 698.1 |
| 111194 | 7081680475048 | Ximabucun | 60 | 302 | 92.2 | 0.7 | 44.4 | 78.8 | 29.2 | 37.7 | 1.54 | 0.3 | <0.05 | 0.83 | 585 |
| 120040 | 6357180491469 | Sumiaocundong | 100 | 378.3 | 92.2 | 1 | 29.2 | 113.8 | 29.1 | 14.7 | 0.32 | 0.3 | <0.05 | 0.46 | 658.3 |
| 111193 | 7083390481674 | Nanxucunnan | 30-40 | 341.7 | 97.5 | 2.4 | 76.1 | 63.4 | 38.5 | 46 | 0.82 | 0.6 | 0.08 | 0.85 | 665.5 |
| 120027 | 7085970480097 | Houbucunbei | 45 | 256.3 | 156 | 3.1 | 52.3 | 154.6 | 47.4 | 43.1 | 0.82 | 0.3 | <0.05 | 1.13 | 968.4 |
| 111192 | 71109104807104 | Dongpengcun | 45 | 482.1 | 230.4 | 12.1 | 300.9 | 28 | 27.3 | 121.4 | 1.12 | 0.5 | <0.05 | 0.34 | 1202.2 |
| 112135 | 7673320437320 | Dufengcun | 30 | 314.3 | 234 | 0.8 | 68.3 | 172.5 | 33.3 | 88.1 | 0.41 | 0.4 | <0.05 | 1.47 | 911.3 |
| 112140 | 7086050480395 | Taipingcun | 30 | 280.7 | 285.4 | 3.2 | 135.2 | 129 | 63.6 | 55.9 | 0.67 | 0.6 | <0.05 | 1.63 | 1170.8 |
| 120030 | 7080770482659 | Beichucunbei | 28 | 527.8 | 285.4 | 4.2 | 208.9 | 145 | 72.5 | 219.5 | 0.79 | 0.4 | <0.05 | 1.55 | 1463.3 |
| 120029 | 7080790482659 | Beichucundong | 46 | 543.1 | 299.6 | 1.9 | 182.1 | 169.2 | 89.9 | 278.2 | 0.57 | 0.6 | <0.05 | 1.85 | 1563.9 |
The analysis of the ratio of main ions and the drawing of Piper's three-line map of main ions are carried out for the test data of groundwater samples (figure 2). Piper triangle is composed of an equilateral parallelogram and two equilateral triangles. The unit of concentration is the percentage of milligram equivalent per liter of water. In mapping, the position of water point on two triangles is determined by the percentage of milligram equivalent of each cation and anion, and then the extension line parallel to the scale line is made through the point. The intersection point of the two extension wire on the parallelogram is the position of the water point on the parallelogram.

The greatest advantage of this map is that a large number of water analysis data are plotted on the map. According to its distribution, many hydrogeochemical problems can be explained [15]. The three-line diagram method can be used to determine whether a water is the result of a simple mixing of two other types of water. If the water sample C is a mixing of seawater sample A and underground freshwater sample B (no reaction occurs during mixing), then the mixed water C will fall on the line of three-line diagram A and B (figure 2). In figure 2, none of the groundwater samples collected falls on the line between A and B. Therefore, no simple mixing or mixing of seawater and fresh groundwater has been responded in the groundwater samples. In figure 2, when the chloride ion content is less than 1000mg/L, the sample points go down from the top on the three-line graph, indicating that cations change from calcium and magnesium ions to potassium and sodium ions, and cation exchange takes place. From the three-line diagram, it can be seen that the main hydrochemical types of underground freshwater areas in the south are Ca-Na-HCO$_3$-SO$_4$, Na-Ca-Cl-SO$_4$ Na-HCO$_3$-SO$_4$-Cl. The hydrochemical types are complex and changeable, mainly the interaction of cationic alkali metal Na$^+$, alkaline earth metal Ca$^{2+}$, anion Cl$^-$, HCO$_3^-$ and strong acid is weaker than weak acid. The chemical types of groundwater in northern saline water area mainly include Na-Ca-Cl, Na-Cl-SO$_4$, Na-Cl, Na-Mg-Cl and a small amount of Ca-Na-Cl. Most of the groundwater in this area is non-carbonate alkalai $>$ 50%. The chemical types of groundwater in brine area are mainly Na-Mg-Cl and Na-Cl, and Mg-Na-Cl-SO$_4$ occurs in some spots. The concentration of Mg$^{2+}$ in brine is the second most important ion except Na$^+$, Cl$^-$ component, which may come from silicate alteration or dolomitization in deep underground. Cl$^-$ content is mainly caused by chloride dissolution and release in transgressive strata. Cl$^-$ is the main anion (the average percentage of chloride ion in freshwater to total anion content is 15.64%; brackish water is 25.19%; saline water is 84.85%; brine is 91.48%). From the relationship between chloride ion and conductivity, EC and Cl$^-$ content are also regular, which can be seen with groundwater. The EC increased exponentially with the increase of Cl$^-$ content, especially for fresh water and brackish water, indicating that Cl$^-$ plays a decisive role in TDS in this area.

Through the analysis of the main ion ratios, the SO$_4$-Cl ion ratios on the comparison map show that the main points on the sea water dilution line are Beichucun and Sumiaocun Rubber Factory (figure 3); The Ca-Cl ion ratios maps show that only one point on the sea water dilution line is located in the south of Zebucun (figure 4); The K-Cl ion ratio maps show that the points on the sea water dilution line are the south of Zengbu Village, the north of Beipu Village, the south of Nanbu Village, the rear village of Daquan Town and Houguan Village (figure 5); the points on the Mg-Cl ion ratio maps are Bailang River (surface water) sample points (figure 6); and the Na-Cl ion ratio maps show the positions. The points on the sea water dilution line are Shuinan Zongcun, Pinglihe Village, Chen Jiawazi, etc. (figure 7). No points on the sea water dilution line can be seen on the HCO$_3$-Cl ion ratio map (figure 8).

In figure 3, when the chloride ion concentration is below 1000mg/L, the SO$_4$/Cl$^-$ ion ratios are basically above the sea water dilution line, indicating that the local acid rain is relatively strong. When the chloride ion concentration is higher than 1000mg/L, the SO$_4$/Cl$^-$ ion ratio fluctuates near the sea water dilution line, which indicates that this part of the sample may have been diluted by river water earlier,
and the reduction of SO₄ in admixing water may also occur.

Figure 2. Piper three-line maps of major ions in groundwater samples from the southern coast of Laizhou Bay.

When the concentration of chloride ion is low, from the analysis of chemical composition formation of groundwater samples, atmospheric precipitation first dissolves some pollutants and evaporative salts in the atmosphere and the ground. In the process of infiltration into the ground, it dissolves a large amount of CO₂ in the soil zone, and dissolves weathered minerals at the same time. The weathered minerals do not produce Cl, and the dissolved marine faces evaporative salts are mainly chlorides of K, Na and Mg. However, the ratios of HCO₃/Cl, Ca/Cl and Na/Cl deviate from the seawater dilution line, mainly due to the dissolution of weathered minerals and the addition of pollutants in groundwater (fresh water), resulting in the growth of HCO₃, Ca²⁺ and Cl.

The average Na/Cl coefficient of standard seawater is 0.85 [16]. If the marine sedimentary water is in the course of geological history, the Na ions in the marine sedimentary water exchange cation exchange with the exchangeable calcium ions in the formation, the sodium ion content decreases, and the Na/Cl coefficient is less than 0.85. If groundwater is mainly formed by dissolution and filtration of salt-bearing strata, Na/Cl should be close to 1. In figure 7, the groundwater sample with a Na/Cl coefficient close to 0.85 is one piece with a value of 0.83, which is close to the average Na/Cl coefficient of standard seawater. The sample is located in Taiping Village, Changyi City, with a chloride ion content of 1134 mg/ L and the degree of mineralization is 3.09 g / L. The location of this point is just at the junction of the brine distribution area and the underground freshwater area, so the sample is mainly affected by the underground brine. In samples with a chloride ion content greater than 1000 mg/L, the
Na/Cl coefficients were all less than 0.85, indicating that sodium ions in the water produced cation exchange with the exchangeable calcium ions in the formation, resulting in a decrease in sodium ion content. Na/Cl coefficients of most samples with chloride ion content of 100-1000mg/L are less than 0.85, and a small number of samples are close to 1, indicating that the groundwater of this part of the sample is mainly composed of rock salt formation.

![Figure 3](image1.png)  
**Figure 3.** The diagram of SO4/Cl and Cl.

![Figure 4](image2.png)  
**Figure 4.** The diagram of Ca/Cl and Cl.

![Figure 5](image3.png)  
**Figure 5.** The diagram of K/Cl and Cl.

![Figure 6](image4.png)  
**Figure 6.** The diagram of Mg/Cl and Cl.
In figure 8, the sample points are basically much higher than the seawater dilution line. In the soil zone, CO$_2$ dissolves in water to form carbonic acid, and carbonic acid dissociates H$^+$ and HCO$_3^-$, and H$^+$ reacts with weathered minerals of granite clasts in the soil zone, making the components such as Ca, K, Na, and Mg in the mineral enter the water body together with the remaining HCO$_3^-$ in the form of ions of Ca$^{2+}$, K$^+$, Na$^+$, and Mg$^{2+}$, respectively. This process will greatly increase the concentration of HCO$_3^-$ so that HCO$_3^-$/Cl is much higher than the seawater dilution line [17].

However, with the increase of Cl concentration, the ratio of HCO$_3^-$ to Cl$^-$ decreases gradually and approaches the seawater dilution line, which indicates that the solubility of weathered minerals from soil zone has stabilized, and the ratio of HCO$_3^-$ to Cl$^-$ decreases with the increase of Cl concentration.
Since both chlorine and bromine are halogen elements, the physical properties are similar, they exist simultaneously in seawater, and in general freshwater water, the content of Br is very small, so the Cl/Br coefficient is a characteristic value of seawater. In ocean water, Cl/Br is about 300 (except for local seawater); if it is residual seawater, because of the precipitation of sodium chloride produced by concentration, and the solubility of bromide is larger than that of NaCl, so the residual bromine in seawater is relatively concentrated, Cl/Br <300; If it is leachate water from rock salt-bearing strata with poor bromine content, Cl/Br>300. Therefore, Cl/Br coefficients are often used not only to determine the origin of deep water (brine and brine), but also to determine the extent and extent of seawater intrusion into freshwater layer according to Cl/Br coefficients [18]. In figure 9, only one sample has a Cl/Br coefficient close to 300, which is 304.28. The sample is a mixture of river water and seawater at the estuary of Weihe River. The Br content of the sample is up to 30, which indicates that a large number of the samples are seawater. In other groundwater samples, Cl/Br coefficients were not close to 300, indicating that no seawater was found in other groundwater samples. The Cl/Br coefficients of a few samples tend to be less than 300, which may be related to the surface saline pollutants (such as municipal sewage and solid landfill leachate) and the absorption of some Br by surface vegetation. The Cl/Br coefficients of most groundwater samples tend to be more than 300, which indicates that it is related to the dissolution of organic pollutants in the area (such as livestock and poultry manure and septic tank sewage, etc.).

5. Conclusion
Based on the above analysis, the groundwater chemical characteristics of the monitoring section from south to north mainly from fresh water, brackish water, salt water to brine, and the change of groundwater chemical type is mainly from $\text{HCO}_3^-\cdot\text{Ca} \rightarrow \text{HCO}_3^-\cdot\text{Cl} \cdot \text{Na} \rightarrow \text{Cl} \cdot \text{HCO}_3^-\cdot\text{Na}$ showed a regular change. Some water chemical ion ratios during salt water intrusion, such as Na/Cl, K/Cl, Ca/Cl, SO$_4$/Cl, etc., can indicate the hydrogeochemical role in the interaction between water and rock, and evaluate the source of salt. The anion-cation exchange reaction in the groundwater in the northern brackish water transition zone is more obvious than that in the southern underground freshwater. The main hydrogeochemical processes occurring in the brackish water transition zone are calcite, anhydrite, dissolution of rock salt, precipitation of gypsum, dissipation of CO$_2$ gas, The main cation exchange is Ca-Na exchange. The high concentration of Na$^+$ and Mg$^{2+}$ in the groundwater of the
transition zone is adsorbed by the aqueous medium, while the Ca^{2+} adsorbed by the aqueous medium is replaced into the mixed water, resulting in Na^{+} and Mg^{2+} in the groundwater of the saltwater intrusion area relatively reduced, Ca^{2+} is enriched, which changes the chemical composition and redox environment of groundwater in the salt water intrusion area.

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