



# Evolutionary process of saline-water intrusion in Holocene and Late Pleistocene groundwater in southern Laizhou Bay



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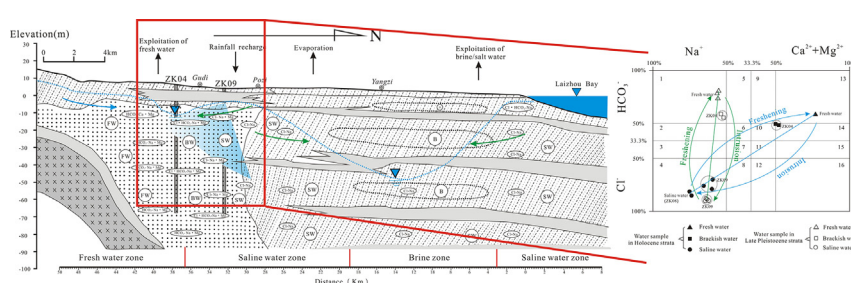
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## HIGHLIGHTS

- Structure of sedimentary layer is the dominant controlling factor of groundwater distribution.
- The salt of saline water (brine) is from the dissolution of evaporates.
- Saline water intrusion presents wedge-shaped intrusion pattern.
- Combined of HFE-D, there are differences of saline water intrusion process between Holocene groundwater and Late Pleistocene groundwater.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Saline water intrusion is one of the most serious groundwater problems in southern Laizhou Bay. In this study, formation of groundwater with different qualities and saline water intrusion were analyzed using hydrochemical and stable isotopic methods, and the Hydrochemical Facies Evolution Diagram (HFE-Diagram). The results demonstrate that the structure of the sedimentary layer in this area is the dominant controlling factor of groundwater distribution. From the south (land) to the north (sea), the hydrochemical distribution presents a regular changing pattern following the order:  $\text{HCO}_3\text{-Na}\cdot\text{Mg}$  and  $\text{HCO}_3\cdot\text{Cl}\text{-Mg}\cdot\text{Ca}$  (fresh water),  $\text{HCO}_3\cdot\text{Cl}\text{-Na}\cdot\text{Mg}$  (brackish water),  $\text{Cl}\text{-Na}\cdot\text{Mg}$  (saline water),  $\text{Cl}\text{-Na}$  (saline water) and  $\text{Cl}\cdot\text{HCO}_3\text{-Na}$  (brackish water). Hydrochemical data show that saline water and brine are not the result of evaporation or the concentration of seawater. Brackish water and saline water with low mineralization in Holocene groundwater are formed by the mixing of fresh water and highly mineralized saline water, dissolution of evaporates by meteoric water, and water/salt interaction. And the saline water formed through dissolution of evaporates in Holocene and Late Pleistocene groundwater. Isotopic results reveal that the main recharge of saline water in Holocene groundwater and Late Pleistocene groundwater is a combination of the meteoric water and lateral recharge from rivers. Saline water intrusion was found to follow a wedge-shaped intrusion pattern. Significant variations in  $\text{Cl}^-$  and  $\text{Na}^+$  indicate saline intrusion in the southern area. The degree of saline water intrusion in Holocene groundwater was found to be more serious than that in Late Pleistocene groundwater. Hydrochemical data and HFE-Diagram show that there is an intrusion process in Holocene groundwater. In this process, it is accepted the fresh water recharge, such as meteoric water and lateral recharge from rivers. In Late Pleistocene groundwater, it presents a simple intrusion process from saline water to fresh water.

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## 1. Introduction

Residential coastal zones have the fastest growing economy and most dense population in the world, increasing the local demands for groundwater and pressure on the environment (Gao and Zhu, 2006; Han et al., 2014). To meet the demands, groundwater resources have been extensively exploited in coastal zones, which has led to a series of environmental problems. Among several problems, seawater (saline water) intrusion is the most prominent, and it leads to various other environmental issues, such as soil salinization and groundwater pollution (salinization), which negatively affect agricultural production and cause shortage of drinking water (Barker et al., 1998; Huang and Guo, 2008; Barlow and Reichard, 2010; Li et al., 2011). Since the 1970s, some groundwater depression cones have been forming because of excessive groundwater (fresh water and brine) exploitation in southern Laizhou Bay. This has induced a complex hydrodynamic field and caused the intrusion of saline water to fresh water aquifers, leading to fresh water salinization (Zhang et al., 1997; Xue et al., 1997, 2000; Feng et al., 2006). Furthermore, groundwater salinization involves evaporation condensation, hydrolysis, dissolution of evaporated salts and the mixing of groundwater of different qualities and hydrochemistry (Matthess, 1982; Drever, 1997; Clark and Fritz, 1997; Mazor, 2004; Applo and Postma, 2005; Edmunds et al., 2006; Ma et al., 2007; Gu, 2011; Salama et al., 1999; Yang et al., 2016a, 2016b; Todd and Mays, 2005).

Southern Laizhou Bay has a wide distribution and high concentration of brine, and is one of the main brine storage areas in China (Zheng et al., 2014). The unique meteorological and hydrological conditions as well as ancient geographical environment and topographic features of southern Laizhou Bay provide suitable conditions for the formation of local brine (Gao et al., 2015, 2016). Stable isotopic and hydrochemical analyses have revealed that the salt in brine (saline water) formed by evaporation of seawater left behind during transgression and regression in the Late Pleistocene and Holocene. Subsequent supply of moisture through precipitation, recharge from piedmont groundwater, and lateral recharge from rivers resulted in the saline water (Han et al., 2011, 2014; Yang et al., 2016a, 2016b).

Underground saline water in southern Laizhou Bay has formed under the background of transgression and regression during Late Pleistocene and Holocene, and its salinity is higher than that of seawater (Gao et al., 2015, 2016). This study was conducted to investigate the evolutionary process of saline water intrusion of Late Pleistocene and Holocene groundwater. For this purpose, groundwater monitoring wells of different depths were set and groundwater samples were collected to obtain basic data for analysis and discussion. Stable isotopic and hydrochemical methods were employed to analyze the origin of moisture and salt of groundwater. Furthermore, the Hydrochemical Facies Evolution Diagram (HFE-Diagram) was used in discussing the mixing process and mechanism of saline water intrusion in different aquifers of Holocene and Late Pleistocene sedimentary strata (80 m depth above) (Edmunds, 1996; Edmunds et al., 2006; Hiroshiro et al., 2006; Gates et al., 2008; Schiavo et al., 2009; Giménez-Forcada, 2010, 2014).

## 2. Background of study area

### 2.1. Location and climatic conditions

The study area is located north of Shandong Peninsula and southern Laizhou Bay, Bohai Sea, China (Fig. 1(a)). The slope of the terrain is gentle and flattens gradually from south (land) to north (sea). The study area also lies in the downstream basin of Bailanghe River, where saline water is widely distributed and saline intrusion is more serious. Bailanghe River is 127 km long with an average flow rate of  $3.88 \times 10^7 \text{ m}^3/\text{a}$ , and is the one of the main recharge resource of shallow groundwater within the study area. One half of the study area belongs to the warm temperate zone, and the other half to the moist monsoon climatic region. The

mean annual temperature is 53.9 °F, and the average annual precipitation is 731 mm. Most of the total annual precipitation, 70%–80% occurs during July–September. The mean annual evaporation capacity is 1648 mm, with 50% of the total evaporation capacity attributed to the months from March to June (Bi et al., 2012; Han et al., 2014; Gao et al., 2015; Liu et al., 2016).

### 2.2. Geology

According to previous research, on the sedimentary strata of this area, several transgression layers and saline water sedimentations formed in the Early and Middle Pleistocene in the study area (Gao et al., 2015). Since the Late Pleistocene, the study area has experienced many glacial–interglacial climatic alternations. In particular, three obvious temperature fluctuations occurred, namely from 85 to 76 ka BP, 50 to 24 ka BP, and 10 to 4 ka BP. These three periods were accompanied by three transgressions (Cangzhou transgression, Xianxian transgression, and Huanghua transgression), known as the marine–continental transition depositional environment (Fig. 1(a)). Between these periods, two cold dry periods (Early Dali ice period and the Late Dali ice period) occurred from 76 to 50 ka BP, and from 24 to 10 ka BP, providing a continental sedimentary environment (Qin, 1985; Zhuang et al., 1987; Zhang et al., 2005). The marine–continental transition sedimentary strata formed as a result of transgression and regression. From top to bottom, the sedimentary layers are as follows: the Late Holocene continental layer, Holocene Huanghua transgression layer, Early Holocene continental layer, Late Pleistocene glacial continental layer, Xianxian transgression layer from the later part of the Late Pleistocene, continental layer from the later part of the Late Pleistocene and Early glacial age, and Cangzhou transgression layer from the early part of the Late Pleistocene (Qin, 1985).

Overall, three massive sea level fluctuations occurred and three marine sedimentary strata formed: the first, second and third marine sedimentary strata. Masses of seawater collected in these marine sedimentary strata and were occupied by continental deposits formed during regression. The amount of silt in continental deposits was correspondingly increased, along with the weakening of the river carrying capacity in the marine zone. This served the role of closing or closed covering on marine sedimentary layers (Gao et al., 2015, 2016).

### 2.3. Hydrological conditions

Fresh water, brackish water, saline water and brine aquifers are distributed throughout the study area (Fig. 1(c)) (Han et al., 2014; Liu et al., 2016). The shallow freshwater aquifers mainly comprise fine sand and medium fine sand. The shallow fresh water contains  $\text{Cl} \cdot \text{HCO}_3 \cdot \text{SO}_4 \cdot \text{Ca} \cdot \text{Na}$ ,  $\text{Cl} \cdot \text{Ca} \cdot \text{Na}$ ,  $\text{HCO}_3 \cdot \text{Ca} \cdot \text{Na}$ , and  $\text{HCO}_3 \cdot \text{SO}_4 \cdot \text{Cl} \cdot \text{Na}$  (Bi et al., 2012) and the value of total dissolved solids (TDS) is  $< 1 \text{ g/L}$ . Brackish water and saline water aquifers mainly comprise silty sand, fine sand, clay silt, and their TDS values range from 1 to 50 g/L. Brine aquifers are composed of phreatic to confined aquifers (1st Marine brine layer, TDS = 50–130 g/L), the first and the second brine aquifers (2nd Marine brine layer, TDS = 50–165 g/L), and the third and fourth brine aquifers (3rd Marine brine layer, TDS = 50–140 g/L) (Qin, 1985; Zheng et al., 2014; Gao et al., 2015).

The study area features an ancient buried channel with coarse sediments, allowing ample groundwater movement (Liu et al., 2003). Under natural conditions, the flow direction of groundwater is from south (piedmont area) to north (sea area). The shallow groundwater is mainly recharged by meteoric water and lateral recharge of rivers, and it is discharged mainly through atmospheric evaporation and groundwater exploitation. In contrast, the recharge source of deep groundwater aquifers is groundwater from other aquifers. Nevertheless, they are discharged mainly through artificial exploitation. Near the coastal line, groundwater is influenced by tides and waves, but their influence is limited within the context of this study, and thus they were not considered.

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