Conceptual Model of Underground Brine Formation in the Silty Coast of Laizhou Bay, Bohai Sea, China

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Conceptual Model of Underground Brine Formation in the Silty Coast of Laizhou Bay, Bohai Sea, China

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ABSTRACT

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There were three major transgression-regression events since the late Pleistocene in the southern coast of Laizhou Bay, Bohai Sea, China. The three marine facies were correspondingly formed. Large amount of underground brine has been found in the late Pleistocene aquifers. A multi-source fluvial delta sedimentary system where the processes of replenishment, migration, filtration, storage, capping condition act together may be more dominant for the formation of underground brine. The dissolved salinity in the liquid of surface microtopography such as lagoon, paleochannel and tidal creek was originated from marine sediment and influenced partly by normal seawater over the time interval, which ran across the bar during the process of storm surge sometimes, but the water body in the sediment only came from fresh groundwater in the fluvial delta. In order to better understand and utilize the underground brine resources, we proposed a conceptual model about the formation of underground brine. Underground brine evolution can be categorized into two stages with six processes. The first stage could be the early regression period. Taking microrelief as a control condition of brine generated reactor, dissolved components were fractionated under the effect of pump evaporation combined with ion exchange between sediment and pore water, leading to the mineral composition changes in groundwater. As a result, underground brine was generated in the tidal flat and delta front under the effect of backflow-infiltration. The saline brine flushed into the tidal flat and the delta front under the effect of backflow-infiltration and remained stable with high degree of mineralization for a long time. The second stage would be the end period of regression. Brine-producing reservoir was formed by a combined effect of both long-term evaporation and seasonal fluvial material input. When lateral terrestrial deposit covered the early brine layer, the underground brine was then formed. Therefore, the three underground brine layers were results of three large scale sea-land changes.

ADDITIONAL INDEX WORDS: *Underground brine, conceptual model, silty coast, regression/transgression.*

INTRODUCTION

Underground confined brine within rock or mud sediment usually has high concentration of Total Dissolved Solid (TDS) and trace elements. It is widely used in salt and chemical industry as a raw material to extract salt and elements including Bromine, Iodine, Lithium, Strontium, Barium, Boron, Potassium, Cesium and Rubidium (Zhou *et al.*, 2006; Zhou, 2013). It is generally accepted that groundwater can be classified based on TDS concentrations as follows: fresh groundwater with TDS < 1 g/L, brackish groundwater with TDS between 1 and 3 g/L, saline water with TDS between 3 and 10 g/L, salt water with TDS between 10 and 50 g/L, and brine with TDS > 50 g/L (Yang and Wang, 1989).

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Some scientific studies have been conducted to explain the origin of underground brine in coastal sedimentary environment since late 1960s. Some coastal groundwater researches in advanced coastal countries such as the United States, Netherlands, Japan, and the South Korea have carried out groundwater investigation in the coastal zone, which focused on exploring the interaction of water-sediment by groundwater section monitoring, hydrology geochemistry and isotope method in recent decades. With research development on seawater condensing into salt, some relevant hypotheses about underground brine origin were proposed successively. For example, model studies on carbonate sedimentary facies in the Persian Gulf coast revealed the Evaporite as Sabkha depositional system and the coastal brine was considered as the first step toward the formation of Evaporates (Kinsman, 1966; Purser, 1973; Schreiber *et al.*, 1976). Han *et al.* (1982) proposed the theory of "brine formed by seawater freezing" on the origin of underground brine. Han *et al.* (1996) concluded that the underground brine was derived from seawater which generated in the regressive stage of transgressive period with processes of evaporation-concentration, accumulation and burial activity. Zhang *et al.* (1996) analyzed the chemical characteristics of seawater and considered that underground brine was derived from seawater during the three times transgressions since late Pleistocene, and it was the result of evaporation-concentration, enrichment-infiltration and burial metamorphism, and so on. Han *et al.* (2011) researched groundwater salinisation processes in coastal aquifers through combined hydrochemical and isotopic parameters and inverse hydrochemical modelling, and concluded that the mixing, ion exchange, dissolution of Dolomite and precipitation of Gypsum and Calcite account for the hydrochemical changes of groundwater in the coastal aquifers of Layzhou Bay. The distribution pattern of rare-earth elements showed that origin of underground brines in northeastern Ussuri Bay was most likely related to diagenesis of submarine and freshwater sediments as well as to the interaction with continental carbonate deposits (Chelnokov *et al.*, 2012).

In general, there are three basic environmental conditions for brine formation: 1) arid or semi-arid climate with evaporation greater than precipitation, 2) continuous input of saline water, and 3) enclosed environment for the concentrated brine storage (He, 1978). There were three transgression-regressive events associated with glacial-interglacial climate in Bohai Sea since late Pleistocene, when three marine geological layers were formed (Ma *et al.*, 2006; Qin, 1985; Zhao, 1995). The three marine layers correspond with Cangzhou transgression ($Qp_3{}^1$, 110–70 ka B. P.), Xianxian transgression ($Qp_3{}^3$, 40–21 ka B. P.) and Huanghua transgression (Qh_2, 7–2.5 ka B. P.), respectively. The marine sediment layer contained large amount of brine and was covered by terrestrial sediments formed during regressions and the corresponding marine and land layers were found in the southern coast of Laizhou Bay (Zhang *et al.*, 2005; Zhuang *et al.*, 1999). Peng *et al.* (1992) further confirmed that the sediment of three brine aquifers in Laizhou Bay was associated with corresponding Cangzhou, XianXian and Huanghua transgressive events based on thermoluminescence and geochemical dating techniques. Han *et al.* (1982; 1996) proposed that underground brine was formed in the coastal tidal flat during the regressive stage from sea water through evaporation, concentration and filtration. Similarly, the hydrogeochemical studies conducted by Zhang *et al.* (1996) suggested that underground brine was formed during transgression of the late Pleistocene. Su *et al.* (2011) analyzed the hydrochemical characteristics of underground brine samples collected from the littoral plain in the coast south of Laizhou Bay, and the results showed that the characteristics of underground brine was consistent with those of seawater, and the rNa/rCl, rK/rBr, rMg/rCa coefficients reflected underground brine origination from the paleo sedimentation with strong concentration and weak metamarphism. Based on geological exploration, Wang *et al.* (2008) summarized the geological characteristics and metallogenic regularity of underground brine deposit in the coast of Laizhou Bay. Xue and Ding (2008) built a sedimentary pattern of Weihe-Mihe River delta in the southern coast of Laizhou Bay since the Holocene. However, Zhou *et al.* (1989) found that the concentrations of Cobalt and Uranium in underground brine were 30 and 4000 times higher than that of seawater respectively, and this difference cannot be explained by seawater evaporation alone (Xu *et al.*, 2011). It was suggested that underground brine was generated in a complex geomorphological environment consisting of sand bar, lagoons, tidal creeks as well as tidal flats (Wang and Han, 1998; Wang *et al.*, 2003). The application of hydraulic, geochemical and stable isotope in the study of typical underground saltwater is helpful to understand the relationship between saltwater and seawater as well as their spatial pattern in muddy-silt coast much more accurately (Carol *et al.*, 2009; Hiroshiro *et al.*, 2006; Marimuthu and Reynolds, 2005; Schiavo *et al.*, 2009). Han *et al.* (2014) carried out researches on hydrodynamic and geochemical relationships between freshwater, seawater and brine in the coastal aquifers of Laizhou Bay by means of hydrochemical-isotopic investigation, and the conclusion indicated that the brines could be formed during Holocene as a result of the sequence of transgression-regression and evaporation, which originally reached halite saturation and were subsequently diluted with fresher groundwater over the long-term.

Formation and evolution of underground brine in the silty coast of Laizhou Bay controlled by climate, tidal storm, river delta, sedimentary environment and other factors were revealed and discussed in this paper. Also, the conceptual model about formation of brine was built through the formation processes of replenishment, migration, filtration, storage and capping conditions.

BACKGROUND OF BRINE FORMATION

The study area is located in the southern coast of Laizhou Bay, Bohai Sea (Figure 1). It belongs to the warm temperate zone with continental monsoon climate. Annual average rainfall and evaporation was 559.5 mm and 1936.7 mm, respectively. The meteorological terrain is alluvial and coast plain with the southern part higher than the northern part. The wide tidal flat extend 5 to 20 km into the Laizhou Bay. The climate in the southern coast of Laizhou Bay belonged to subtropical humid climate during Pliocene, alternating glacial and interglacial climate during Pleistocene, and postglacial warm climate during Holocene. Since the late Pleistocene, the climate became drier and drier under the effect of frequent climate fluctuation (Table 1) (Han *et al.*, 1996; Ma *et al.*, 2006). Intense storm events occurred more frequently since the late Pleistocene. Sometimes seawater ran across the sand bar and flowed into adjacent lagoons under the influence of storm surge. High salinity generated in lagoons, channels and tidal creeks recharged into the tidal flat and the delta front under the effect of backflow-infiltration. In addition, high tides and strong evaporation were preferable for the formation of underground brine. Most part of Laizhou Bay has irregular semidiurnal tide. The tidal range becomes bigger towards both sides. Intense storm surge events influenced this area frequently, as more than 70 times records in the past 200 years. The processing tidal line during a strong storm event in 2003 was shown in Figure 2 (Hou, 2010). The highest water level elevated by storm reached 3.0 m an hour after the astronomical ebb tide. And the highest water level during this storm surge which amounted to 6.24 m was observed at Yangjiaogou station three hours after the maximum water increasing. It is the third highest water level in the history (April 23, 1969, 6.74 m and April 6, 1964, 6.26 m) (Hou, 2010).

The Weihe-Mihe River delta was formed by rivers including Xiaoqing River, Mihe River, Bailang River, Yuhe River, Weihe River, and Jiaolai River from west to east (Figure 3). The courses of rivers, especially the Weihe River and Mihe River, drifted frequently in the history. The delta has migrated about 20 − 55 km towards the sea by acceleration since the Late Pleistocene. The fluvial deposit is over 50 m near present coastline already. The silty sand on upper delta was alluvial sediment contributed by rivers. Smaller deltas formed at estuaries of individual rivers were mainly composed of very fine sands. The inner-estuarine sediment was loaded by two adjacent rivers mainly consisting of clayey sand. Due to the drift of river mouth, very fine sand in delta front and clayey sand in inner-estuarine would appear out alternately. As a result, complete stratigraphic sequence consist of predelta, delta front-inner-estuarine, lower deltaic plain and upper deltaic plain of high sequence and tidal flat and continental shelf sediments of the transgression system in relatively deep seawater.

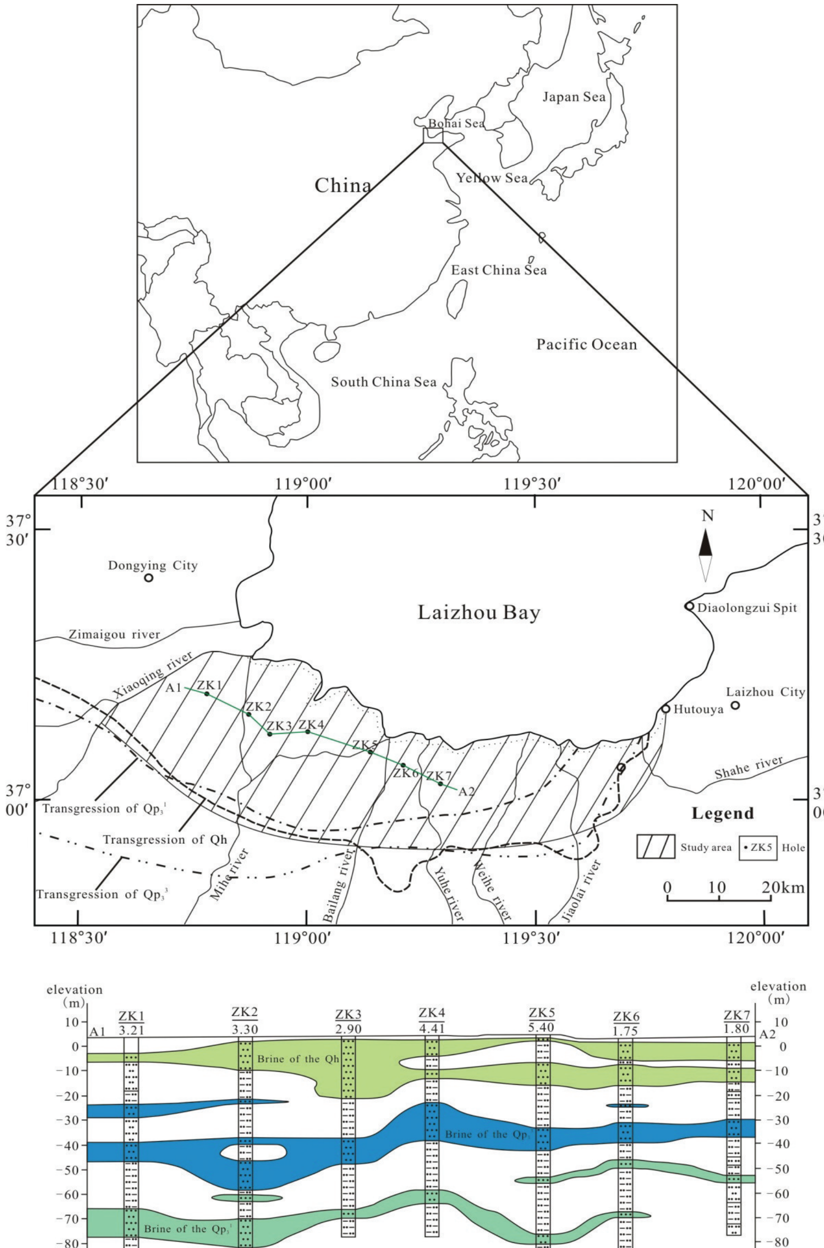

Figure 1. Study area and geologic distribution of underground brine in southern coast of the Laizhou Bay

Table 1. *Paleoclimate change of the Late Quaternary in Bohai Sea* (Han *et al.*, 1996; Ma *et al.*, 2006)

geological times	palaeomagnetic event	glacial epoch		climate	
Holocene		post-glacial period		warm	
Late Pleistocene	Gothenburg Event(12.35-13.75ka)	Wurm glacial stage	Wurm glacial stage Ⅲ	Cold	Cold-dry
	Mungo Event(35-36ka)		Late subinterglacial stage		Warm-humid
			Wurm glacial stage Ⅱ		Cold-dry
	Unknown Event(50-60ka)		Early subinterglacial stage		Warm-humid
			Wurm glacial stage Ⅰ		Cold-dry
		Riss-Wurm interglacial stage		Warm	
Middle Pleistocene	Blake Event(114-108ka)	Riss glacial stage		Cold	

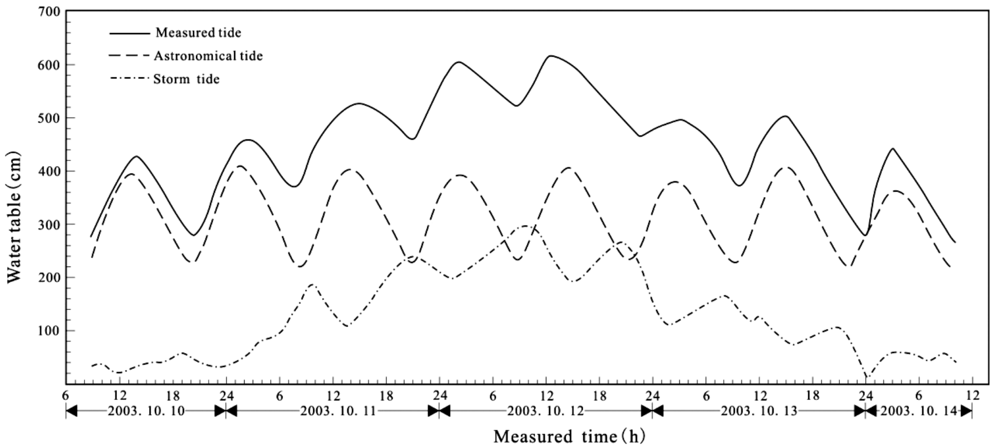

Figure 2. Contrast of measured tide, astronomy tide and surge of storm in southern coast of the Laizhou Bay

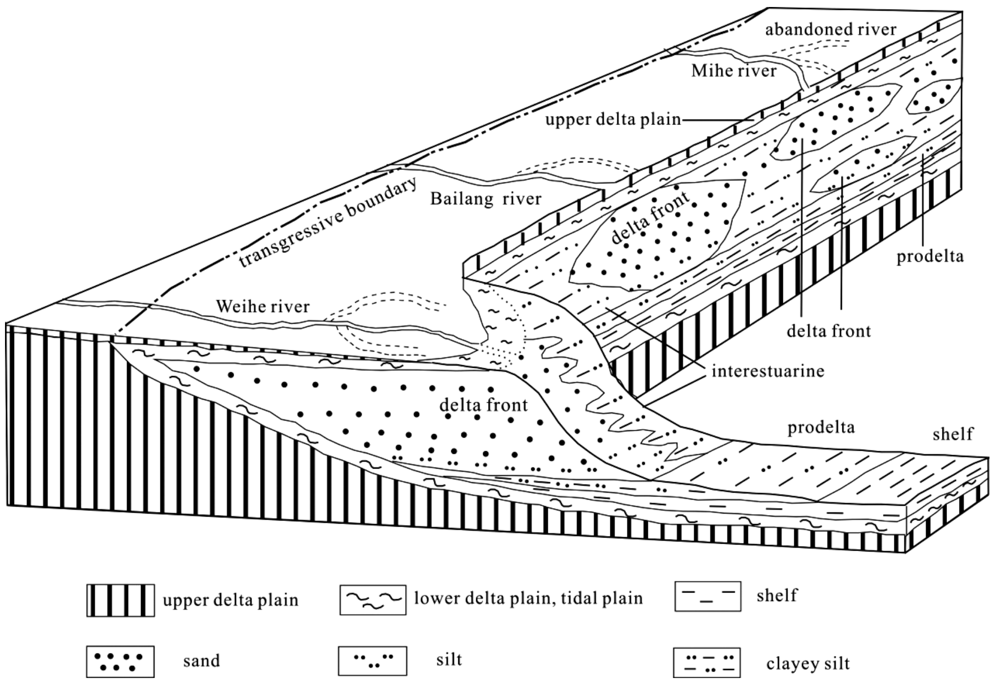

Figure 3. Sediment model of fluvial delta during the Late Pleistocene and Holocene in southern coast of the Laizhou Bay (modified by Xue and Ding, 2008)

DISTRIBUTION OF BRINE

The underground brine in the three marine layers distributes horizontally with zonal distribution (Table 2, Figure 4, Figure 5, Figure 6). Vertically, the upper layer is affected by atmospheric precipitation, while the lower layer is affected by deep groundwater (saltwater and brackish water) input (Feng *et al*., 2010; Wu *et al*., 2006). As a result, it forms special strata with a low salinity in both upper and lower layers, but a high salinity in the middle layer (Zheng *et al*., 2014).

The first marine layer formed in the early stage of late Pleistocene during the Cangzhou transgression with the starfish rotifers fossil and palynological assemblages existing in hot-humid climates. By palaeomagnetic dating, the transgression age was less than 100 ka B. P. belonging to the interglacial period of Rees-Wurm in the early stage of late Pleistocene (Zhang *et al*., 1996). The coastal was clear along Huaguan, south of Wopu town, north of Shouguang city, Weifang farmland and east of Jiaolai River from west to east (Figure 4). The top depth of the confined brine layer is about 30.3 m, and the bottom depth is 32.3–73.2 m and the thickness is about 1–22.6 m. The confined brine aquifers are mainly made of silt and clayey silt, a few fine sands with shell fragments. The value of TDS in the confined brine varies from 50 to 130 g/L. The TDS is more than 100 g/L in the east of Yangkou town-Caiyangzi saltwork, east of Dajiawa town and the north of Liyu-Zaohu saltwork. The value of brine decreases gradually in the west of Dajiawa town, and varies in low-high-low in the east of Dajiawa town from seaward to land.

Large transgression named Xianxian occurred in the sub-interglacial period (40–24 ka B. P.) with the sea level rise again in the late stage of late Pleistocene. The range of Xianxian transgression was much bigger than that of Cangzhou transgression (Figure 5). The Xianxian transgression was ended by the Last Glacial Maximum (Wurm III) as the global sea level dropped (Han *et al*., 1996). The top depth of confined brine layer is about 7–61 m and the bottom depth is 14.8–72.5 m, and the thickness is 2–28.1 m. The second confined brine aquifers mainly composed of silt and fine sand, rarely coarse sand with some shell fragments. The TDS in confined brine varies from 50 to 165g/L. TDS is more than 130 g/L in Zaohu saltwork and the value decreases gradually in the east of Yuhe River. The value varies in low-high-low in west of Yuhe River from sea to land.

Table 2. *Distribution characteristics of the underground brine* (Feng *et al*., 2010; Qin, 1985; Wu *et al*., 2006;)

Time	Transgression	Ranges	TDS(g/L)	Characteristics
Early stage of late Pleistocene (Qp_3^1)	Cangzhou	Length: 60—70 km Width: 15—30 km Thickness: 9.9 m	50—130	Low TDS, decrease gradually from up to down
Late stage of late Pleistocene(Qp_3^3)	Xianxian	Length: 75—90 km Width: 20—35 km Thickness: 16.5 m	50—165	HighTDS, increase gradually from up to down
Holocene(Qh)	Huanghua	Length: 50—85 km Width: 5—35 km Thickness: 7.7 m	50—140	LowTDS, increase gradually from up to down

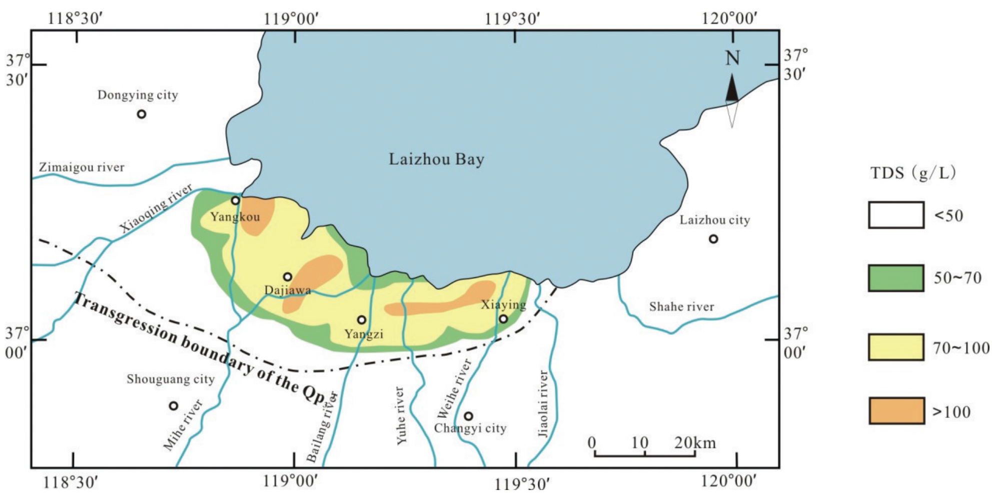

Figure 4. Concentration distribution of underground brine in the early Late Pleistocene

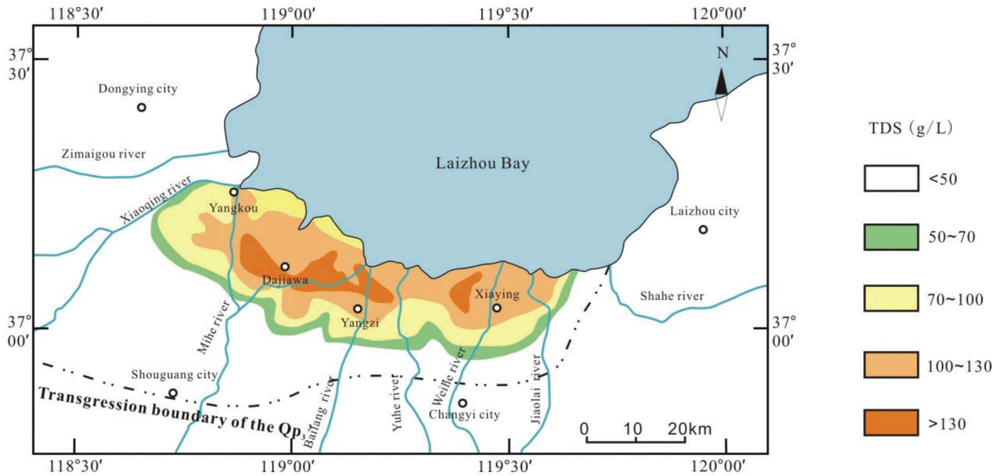

Figure 5. Concentration distribution of underground brine in the late stage of Late Peistocene

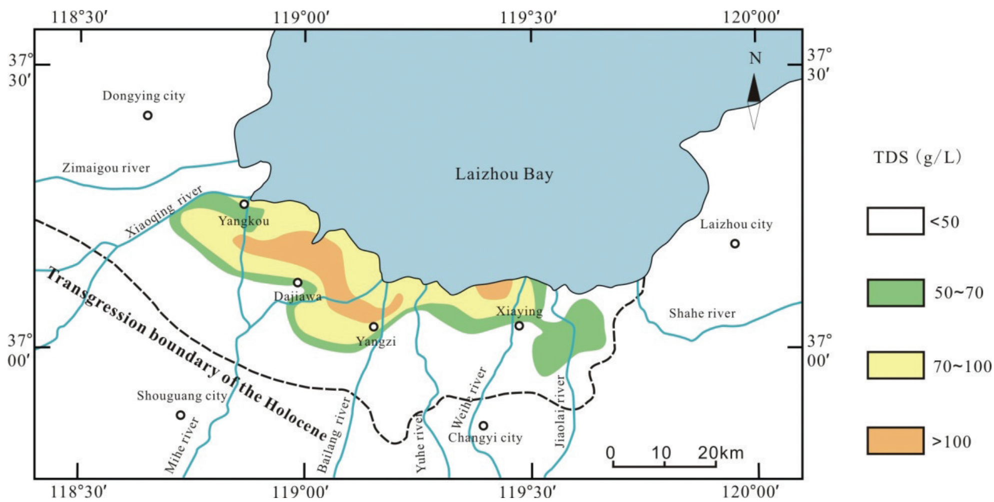

Figure 6. Concentration distribution of underground brine in the Holocene

It was the time that the Bohai Sea began to bring it back to life in the early stage of Holocene, and the Huanghua transgression moved forward to the coastal zone along the depression of Bohai Sea Shelf (Qin, 1985). The coastal range of Huanghua transgression was more than 100 km which was the biggest expansion of the Bohai Sea (7.5–6.5 ka B. P.) (Zhang *et al.*, 1996). Based on the synthesis analysis of dating (Han *et al.*, 1996), the positions of ancient coast lines in post-glacial period and mid-Holocene were given. The third underground brine made of phreatic or micro-confined brine deposited in late Holocene (Figure 6). The phreatic and micro-confined brine aquifers are generally composed of 1–3 stable aquifers with 0.5–6 m single thickness. The top depth of confined brine layer is about 2.8–28.65 m and the bottom depth is 5–31.3 m, and the thickness is 1–14.5 m. The aquifers of confined brine mainly composed of silt and clayey silt with shell fragments. The value of TDS in confined brine varies from 50 to 140 g/L. The TDS is more than 100 g/L in Caiyangzi saltwork, north of Dajiawa town, north of Zaohu saltwork and the value decreases gradually in Xiaying village (east of Yuhe River). And the value varies in low-high-low in west of the Yuhe River from sea to land.

CONCEPTUAL MODEL OF BRINE FORMATION

There are about two formational period of underground brine in the silty coast. At the first, the seawater and salinity flowed into lagoon, abandoned paleochannel, tidal creek and other low site in the transgression period since late Pleistocene. Then during the regression period, strong evaporation was driven by hot and dry climate. Shallow groundwater was drawn toward ground by capillarity force and resulted in increased salt content and the Mg/Ca ratio (Boschetti *et al.*, 2011). During the period without others discharge, more high density saltwater with high concentration of Magnesium flowed downwards due to the effect of backflow-infiltration. Therefore, taken the sand bar-lagoon, abandoned paleochannel, tidal creek and other low site as the brine generated reactor, dissolved components were fractionated under the effect of pump evaporation and ion exchange in sediments, leading to mineral alteration. As a result, underground brine was finally generated in the tidal flat and delta front under the effect of backflow and infiltration.

Underground brine evolution model in the silty coast of Laizhou Bay can be summarized into two evolution stages with six processes (Figure 7). The first stage would be the early period of regression. The brine generated in lagoon, paleochannel and tidal creek, ran into the tidal flat and the delta front under the effect of backflow-infiltration and remained stable with relatively high concentrations for a long time. The second stage would be the end period of regression. Brine-producing reservoir was formed by a combined effect of both long-term evaporation and seasonal fluvial material input. When lateral terrestrial deposit covered the early brine, the underground brine was then formed. Therefore the three brine layers were the result of three large scale sea-land changes.

Figure 7(a) is early brine evolution process and 7(b) is brine evolution process of regression stage in the early late Pleistocene (Qp_3^1). Early brine evolution process and brine evolution process of regression stage in the Late Pleistocene (Qp_3^3) were illustrated in Figure 7(c) and 7(d), respectively. Figure 7(e) shows the early brine evolution process and 7(f) is brine evolution process of regression stage in Holocene(Q_h).

CONCLUSIONS

The dissolved salinity of underground brine in surface microtopography such as lagoon, paleochannel and tidal creek was originated from submarine sediment and influenced partly by normal seawater over the time interval, which ran across the bar during the process of storm surge sometimes, while the water body in the sediment likely most came from fresh groundwater in the fluvial delta.

The formation of underground brine must have special meteorological hydrology, paleogeographic environment, topography and hydrogeology conditions. A multi-source fluvial delta sedimentary system with replenishment, migration, filtration, storage, capping condition may be more dominant for the formation of underground brine. The dissolved components were fractionated under the effect of evaporation pumps, and underground brine was generated in the tidal flat and delta front under the effect of backflow and infiltration.

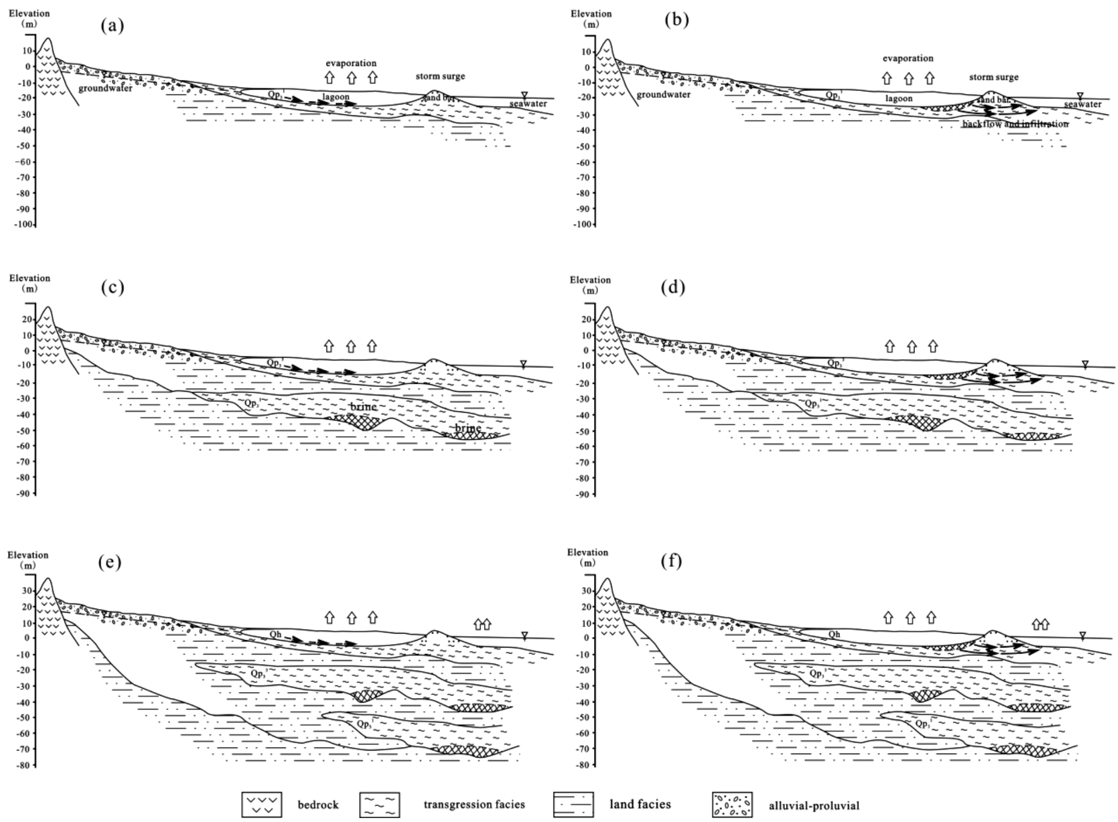

Figure 7. Evolutionary pattern of underground brine in southern coast of the Laizhou Bay. (a) Early brine evolution process. (b) Brine evolution process of regression stage in the early late Pleistocene (Qp_3^1). (c) Early brine evolution process. (d) Brine evolution process of regression stage in the Late Pleistocene (Qp_3^3). (e) Brine evolution process. (f) Brine evolution process of regression stage in Holocene (Q_h)

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