



Chlorine isotopic constraint on contrastive genesis of representative coastal and inland shallow brine in China

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ABSTRACT

There are great scientific interests to reveal the shallow brine genesis in different geologic environments and also practical significance for resource production and environmental protection, which have not yet gained much attention, however. This study took shallow Quaternary brine in the coastal plain of Laizhou Bay and salt lake brine/intercrystalline brine in arid inland Qaidam Basin in China as two kinds of representative shallow brines, and comparatively analyzed the Cl stable isotopic signatures, coupled with hydrochemistry and water isotopes, aiming at finding out the salinity origin of shallow brines in the coast and inland basin, and further discussing Cl stable isotopic behavior in actual hydrogeological systems. The results indicated that shallow brine in the coastal zone originated from evaporation and precipitation formerly, and halite dissolution by tidewater or rainwater later, as a result of which, most of shallow brine exhibited enriched Cl isotopic signatures. As a contrast in arid inland basin, superficial brine originated from the dissolution of precipitated salt minerals by river water formerly, and intensive evaporation with subsequent precipitation in basin center later, for which all the brines finally exhibited the depleted Cl isotopic signatures. In addition, diffusion and ion filtration in the coast zone, and inflow of deep oilfield water in inland basin, were proposed as potential processes contributing to respective Cl isotopic signatures. In the coast, ^{37}Cl are enriched and ^{81}Br are depleted in most of shallow brines, while all the superficial brine (saline water) samples in arid inland basin exhibit negative $\delta^{37}\text{Cl}$ and positive $\delta^{81}\text{Br}$ values. Precipitation and dissolution of salt minerals were believed to contribute primarily to the contrary isotopic signatures, which would not change the Br isotopic compositions a lot for the solution in which the original Br concentration has been relatively high, which is not the case for Cl isotope, however.

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1. Introduction

Pore water with salinities commonly ranging from 5 to 300 mg/L TDS comprises about 20% of most sedimentary basins, with in-situ temperatures of ~20 to >150 °C and fluid pressure of ~100 to >1000 bar (Kharaka and Honor, 2004). Understanding the origin and controls on the composition of these waters is critical in evaluating their role in burial diagenesis, such as ore-formation, crust cycling, and fluid flow and transport (Hanor and McIntosh, 2007). On the contrast, pore water with high salinity in shallow stratum has not been paid much attention by hydrogeologists or geochemists.

In China, shallow Quaternary brine in the coastal plain of Laizhou Bay (Han et al., 2014) and salt lake brine/intercrystalline brine in inland arid Qaidam Basin (Yu et al., 2013) are two kinds of representative shallow brines, which are exploited to produce salt. In addition, the shallow brine has been also regarded as an important environmental disaster in

the coast of Laizhou Bay when intruding fresh aquifers (Xue et al., 2000; Ma et al., 2007; Han et al., 2011). In order to make salt production more efficient and provide guidance for environmental protection, it is a principal step to understand the origin of salinity. In the aspect of geochemistry, there is also great interest to figure out the difference of these two shallow brines in totally different climatic, geomorphological and hydrological conditions.

Chlorine is a perfect representative of salinity in natural waters, the geochemistry and stable isotope of which can provide valuable insight into origin and evolution of salt and coexisting brine. Chlorine in groundwater has various sources, such as atmospheric deposition of anthropogenic and natural origin, oxidation of chlorine-containing organic compounds, weathering of paleo-seasalts and clay minerals, and discharge of industrial wastes (Koehler and Wassenaar, 2010; Liu et al., 2008; Zhang et al., 2007). Stable chlorine isotope has been widely used to investigate local sources of salinity, mixing of fluids, and interaction between rock and fluids (Stiller et al., 2009; Liu et al., 2008; Godon et al., 2004; Eastoe et al., 2001). Chlorine isotope fractionation can be mainly affected by physical processes such as salt precipitation (Luo et al., 2012; Eggenkamp et al., 1995), ion filtration (Phillips and Bentley,

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1987), ion-exchange (Musashi et al., 2004) and diffusion (Eggenkamp and Coleman, 2009).

The major objectives of the present study were to characterize shallow brines in the coast and inland basin, understand the origin of salinity in two distinct regions, and further discuss chlorine stable isotopic behavior in actual hydrogeological systems using stable Cl isotope coupled with water chemistry. This study is beneficial to enrich related theory of Cl isotope geochemistry and further understand genesis of shallow brines.

2. Geological settings

In this part, the geological background of coastal plain of Laizhou Bay (study area A) and inland arid Qaidam Basin (study area B) were introduced, respectively.

2.1. Geological background of study area A

Laizhou Bay is located in the south of Bohai Sea and the north of Shandong Peninsula (Fig. 1). From south to north in this coastal zone,

the sedimentary types are various with clear transition, from alluvial-proluvial to alluvial-marine, and to marine sediments. The sediment in the upper proluvial fan in the south mainly consists of gravel and coarse sandstone, grading to fine sand, silt, sandy clay, and silty clay towards the coast (Han et al., 2011). The surface elevation ranges from 30 m to 1–2 m along the direction towards the sea (Chen et al., 1997).

The primary aquifers are composed of Quaternary unconsolidated sediments, with the thickness of only about 30 to 50 m in the south, gradually rising towards the north with maximum thickness of >300 m (Xue et al., 2000). The mineralogy of the shallow aquifer includes quartz, anorthite, albite, plagioclase feldspar, picrite, biotite, aragonite, dolomite, calcite, kaolinite, gibbsite and Ca-montmorillonite with minor amounts of evaporites occurred locally (Han et al., 1996; Xue et al., 2000). Gypsum deposits are common in the coastal saltwater zone, while silicate rocks dominate the fresh water zone at the top of the Wei River alluvial fan to the south. The clay layers are composed of illite, kaolinite and chlorite (Zhao, 1996).

In geological history of this area, large-scale seawater intrusion and regression occurred three times since the middle Late Pleistocene. Correspondingly, brines in the north are mainly distributed in three

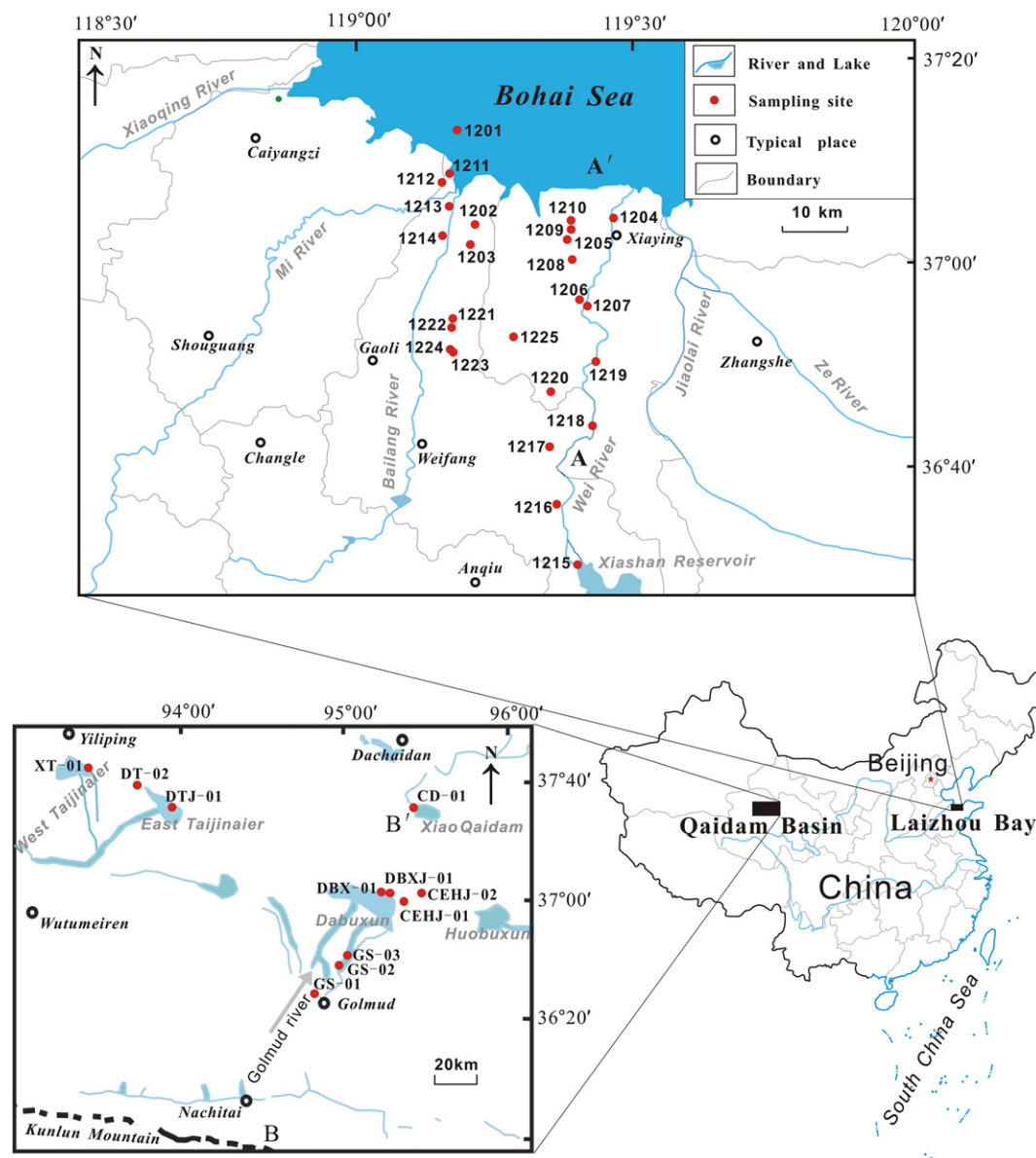


Fig. 1. Map of the southern coastal plain of Laizhou Bay and the typical region of Qaidam Basin, showing the sampling locations and numbers.

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