Geoscience Frontiers 10 (2019) 253–262

Contents lists available at ScienceDirect

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Geoscience Frontiers

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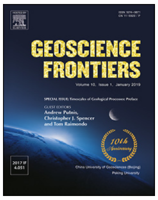

Research Paper

Evaluation of boron isotopes in halite as an indicator of the salinity of Qarhan paleolake water in the eastern Qaidam Basin, western China

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ARTICLE INFO

Article history:
Received 15 September 2017
Received in revised form
12 January 2018
Accepted 27 February 2018
Available online 3 April 2018
Handling Editor: Nick M W Roberts

Keywords:
B isotope compositions
Brine
Chemical concentrations
Qarhan salt lake
Western China
Salinity of paleolake water

ABSTRACT

In this study, nineteen brine samples from the Qarhan Salt Lake (QSL) in western China were collected and analyzed for boron (B) and chlorine (Cl) concentrations, total dissolved solids (TDS), pH values and stable B isotopic compositions. The B concentrations and δ^{11}B values of brines in the QSL range from 51.6 mg/L to 138.4 mg/L, and from +9.32‰ to +13.08‰, respectively. By comparison of B concentrations and TDS of brines in QSL with evaporation paths of brackish water, we found that B enrichment of brines primarily results from strong evaporation and concentration of Qarhan lake water. Combining with comparisons of B concentrations, TDS, pH values and δ^{11}B values of brines, previously elemental ratios (K/Cl, Mg/Cl, Ca/Cl, B/Cl) and δ^{11}B values of halite from a sediment core (ISL1A), we observe good correlations between B concentrations and TDS, TDS and pH values, pH and δ^{11}B values of brines, which demonstrate that higher B concentrations and more positive δ^{11}B values of halite indicate higher salinity of the Qarhan paleolake water as well as drier paleoclimatic conditions. Based on this interpretation of the δ^{11}B values of halite in core ISL1A, higher salinity of the Qarhan paleolake occurred during two intervals, around 46–34 ka and 26–9 ka, which are almost coincident with the upper and lower halite-dominated salt layers in core ISL1A, drier climate phases documented from the δ^{18}O record of carbonate in core ISL1A and the paleomoisture record in monsoonal central Asia, and a higher solar insolation at 30°N. These results demonstrate that the δ^{11}B values of halite in the arid Qaidam Basin could be regarded as a new proxy for reconstructing the salinity record of paleolake water as well as paleoclimate conditions.

1. Introduction

The high geochemical activity of boron (B) and the large relative mass difference between ^{10}B and ^{11}B lead to significant B isotope fractionation in nature (Xiao et al., 2013). In the past, B isotopic compositions of evaporites and brines have been used to trace paleosalinities and to reconstruct marine and non-marine depositional environments (Palmer and Slack, 1989; Qi et al., 1989; Vengosh et al., 1991, 1992, 1995; Xiao et al., 1992; Palmer and Swihart, 1996; Jiang, 2000; Liu et al., 2000; Kloppnam et al., 2001; Paris et al., 2010; Tan et al., 2010; Zhang et al., 2013). Similarly, the origin and geochemical evolution of brines, groundwater, hot springs, and salt sediments have been evaluated based on B isotopic compositions and elemental concentration ratios (Vengosh et al., 1991, 1995, 1998, 1999, 2007; Xiao et al., 1992; Li et al., 2013; Lv et al., 2014; Boschetti et al., 2015; Ma et al., 2015; Awaleh et al., 2017). In addition, over the past two decades, B isotopic compositions of marine biogenic carbonates (e.g., corals and planktonic foraminifer shells) have been regarded as an indicator to reconstruct the pH value of paleoseawater (Hemming and Hanson, 1992;

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Peer-review under responsibility of China University of Geosciences (Beijing).

https://doi.org/10.1016/j.gsf.2018.02.016

Hönisch and Hemming, 2004; Hönisch et al., 2004; Ke et al., 2015). Meanwhile, δ^{11}B values and δ^{18}O values of terrestrial carbonates from a Lop Nor profile have been used successfully as an index to identify the lacustrine and fluvial-eolian depositional environment of Late Miocene episodic lakes in the arid Tarim Basin, western China (Liu et al., 2014). In addition, Wei et al. (2014) reported that the more negative δ^{11}B values of an acid-soluble silicate phase from a sediment core in the Dongtai Salt Lake in the central Qaidam Basin indicate the influence of intensive weathering. These studies have provided insight into the application and future possible uses of B isotope geochemistry in regard to sediments under marine and terrestrial conditions.

However, research on B isotope geochemistry and its geologic implications for salt minerals from a sediment core taken from an arid salt lake is very rare. Liu et al. (2000) stated that the δ^{11}B values of halite might record the evolution of salt lake brines based on an investigation of B isotope geochemistry of artificial brines produced in an evaporation experiment and halite and coexisting brines from seven salt lakes in the Qaidam Basin, western China. This work provides a sound basis for reconstructing the evolution of paleobrines according to B isotopic values of halite in salt lakes. Their conclusions also included the hypothesis that a minor isotopic fractionation (1–5‰) between the halite and brine phases exists in the non-marine salt lakes. Later, this hypothesis was verified in a marine study, which determined that δ^{11}B values of modern halite from salt marshes along the Atlantic and Sicilian coasts are comparable to that of modern seawater, suggesting seawater evaporation and halite formation do not induce B isotope fractionation (Paris et al., 2010). In order to systematically evaluate B isotopic fractionation between halite and brines from drill cores in the typical salt lakes, Fan et al. (2015) conducted an investigation on the B isotope fractionation between halite from sediment core ISL1A from the Qarhan Salt Lake (QSL) and brines from salt lakes in the Qaidam Basin (Fig. 1). The results show that the B^{3+} values in halite are mainly derived from fluid inclusions and no or only minor B isotope fractionation between the halite and brines occurred in the Qarhan playa, which confirmed the viewpoints of Liu et al. (2000) and Paris et al. (2010). These studies provide a connection between B isotopes from halite and brines in the salt lakes and suggested that the δ^{11}B values of halite might record the evolution of salt lake brines. However, the evaluation of B isotope geochemistry of halite from a sediment core in combination with B isotope values, pH values, and total dissolved solids (TDS) of brines in an identical salt lake has yet to be reported.

Over the past three decades, salt sediments have been used to reconstruct late Quaternary environmental and climatic conditions on the Tibetan Plateau (TP) (Chen and Bowler, 1986; Zhang et al., 1993; Han et al., 1995; Wang et al., 1995; Yang et al., 1995; Liu et al., 1998, 2008; Fang et al., 2008; Li et al., 2010). Based on conventional chemical concentrations, mineral assemblages, saline rhythm, and thickness of salt-bearing sediments in the Chaka, Qarhan, Kunteyi, and Dalangtan salt lakes in the Qadiam Basin, paleoclimatic patterns and possible forcing mechanisms on the northeastern TP since the Plio–Pleistocene have been reconstructed and discussed (Chen and Bowler, 1986; Han et al., 1995; Fang et al., 2008; Liu et al., 2008; Li et al., 2010). Relative to studies involving conventional elemental analysis, mineralogy, and sedimentology of salt deposits in salt lakes, research involving isotope geochemistry of salt deposits from salt lakes on the TP has been limited (Spencer et al., 1990; Zhang et al., 1993; Lowenstein et al., 1994; Wang et al., 1995; Yang et al., 1995; Liu et al., 1998). For example, three paleoclimatic periods in the QSL during the past 50 ka were reconstructed based on major elements (Mg^{2+} and Na^{+}) and isotopic compositions (δD and δ^{18}O) of fluid inclusions in halite and ^{230}Th dating of halite from two drill cores (Ck8801 and Ck8904): (1) a period of salt-precipitation due to higher temperatures (2 °C higher than the present temperature) between 50 ka and 30 ka; (2) a dry period with lower temperatures (4 °C lower than the present temperature) between 30 ka and 15 ka; and (3) a dry period with temperature similar to present temperatures after

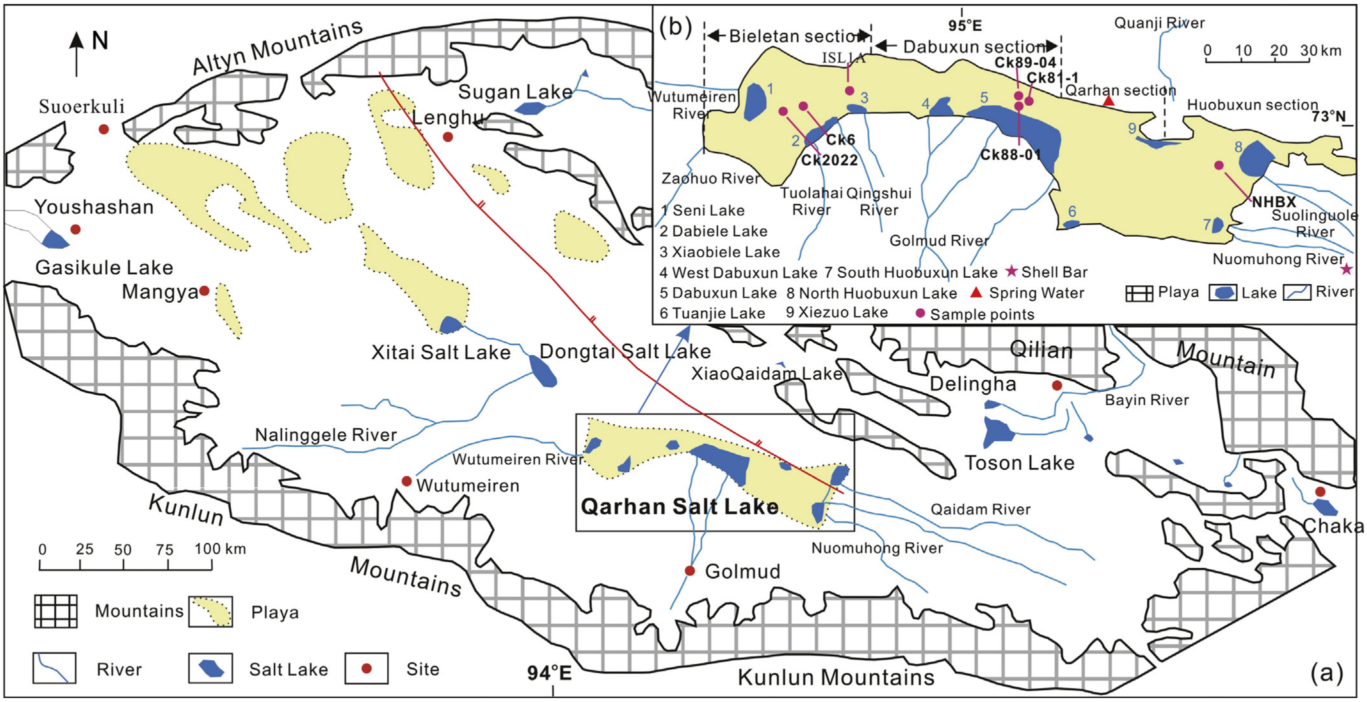

Figure 1. Map showing the location of drilling cores and salt lakes in the Qaidam Basin. (a) Map showing the location of ISL1A in the Qarhan Salt Lake and other salt lakes in the Qaidam Basin. (b) Map showing the drilling cores and the salt lakes in the Qarhan Salt Lake area.

15 ka (Spencer et al., 1990; Zhang et al., 1993; Lowenstein et al., 1994; Yang et al., 1995). Liu et al. (1998). Wang et al. (1995) and Luo et al. (2016) also determined stable chlorine (Cl) isotopic compositions of halite from cores Zk3208, Zk8904 and ISL1A in the Kunteyi and Qarhan salt lakes in the Qaidam Basin, respectively. They concluded that the variation in δ^{37}Cl values of halite is representative of the evolution of paleobrines of salt lakes. These studies have provided insight into the evolution of salt lakes in the Qaidam Basin based on the isotope geochemistry of salt minerals. However, the evaluation and geologic implications of the B isotopic composition of halite from a sediment core in a salt lake has yet to be reported.

To remedy this, we present B and chlorine (Cl) concentrations, pH values, TDS, and stable B isotopic values of brines from the QSL in the eastern Qaidam Basin, combining with previously elemental ratios (K/Cl, Ca/Cl, Mg/Cl and B/Cl) and stable B isotopic values of halite from a sediment core (ISL1A) in order to decipher the relevant geochemical information recorded in the sediments. In addition, the relationships between pH and TDS, the pH and δ^{11}B values, and B/Cl ratio and TDS of brines, and the geological implications of the B isotopic composition of halite in the QSL are discussed in this work.

2. Description of the study area

The Qaidam Basin is one of the largest basins on the northeastern Tibetan Plateau (NETP), covering an area of 1.2×10^5 km^2. It is surrounded by the Kunlun Mountains to the south, the Altyn Mountains to the west, and the Qilian Mountains to the north and east (Fig. 1a). When the mountains were higher and the basin was lower (Yuan et al., 1983), 27 salt lakes developed. Thus, deposits of thick salt beds and abundant brine resources are common in this basin (Chen and Bowler, 1986; Zhang, 1987).

The QSL (36°37′36″N–37°12′33″N, 94°42′36″E–96°14′35″E) is the largest playa in the eastern Qaidam Basin (Fig. 1a). It has an extremely arid desert climate. The mean annual temperature is 5.33 °C, the mean annual precipitation is approximately 24 mm, and the annual evaporation is about 3564 mm (Qarhan meteorologic station). The average wind speed is 4.3 m/s, and the relative moisture is 27.7% (Yu et al., 2009). The QSL extends about 168 km from west to east and 20–40 km north to south, covering an area of 5856 km^2 (Huang and Han, 2007). It contains ten shallow perennial and ephemeral saline lakes, including the Seni, Dabiele, Xiaobiele, West Dabuxun, Dabuxun, Tuanjie, Xiezuo, South Huobuxun, Nouth Huobuxun lakes and Shell Bar (Fig. 1b). Dabuxun Lake is the largest saline lake in the Qarhan playa (Zhang, 1987). These saline lakes are characterized by thick salt-bearing layers and brines (Chen and Bowler, 1986; Zhang, 1987; Casas et al., 1992). These salt-bearing layers mainly contain halite and minor gypsum (Gao, 1987; Li, 1987). Most of the brines are enriched in K$^+$ and the TDS of the brines is generally more than 300 g/L (Yu et al., 2009). This makes it the largest base for potash fertilizer production in China, having a total reserve of 194×10^6 t of KCl (Cao and Wu, 2004). From east to west, the QSL can be divided into four sections, i.e., the Huobuxun, Qarhan, Dabuxun, and Bieletan sections (Fig. 1b). The evaporitic sequence of the entire Qarhan playa is generally comprised of three halite-dominated evaporite units separated by clastic-dominated sediment units (Yu et al., 2013). The Bieletan section is located in the westernmost part of the QSL, which contains a complete sequence of evaporite deposition (up to 70 m) over an area of 1500 km^2. The Dabuxun and Qarhan sections are located in the central part of the QSL where the thickness of the halite layers ranges from 35 to 55 m over an area of 1120–1250 km^2. The Huobuxun section is located in the easternmost part of the QSL where the thickness of the halite layers ranges from 15 to 20 m over an area of 835 km^2 (Zhang, 1987; Yu et al., 2009).

Table 1
The pH, TDS and δ^{11}B values of brines in Qarhan playa.

Sample code	B (mg/L)	TDS (g/L)	pH	δ^{11}B (‰)	Error (‰)
ISL-01	63.77	308.3	6.50	11.00	0.14
ISL-02	118.85	338.6	6.10	9.90	0.17
ISL-03	91.37	314.6	6.46	9.67	0.10
ISL-04	138.44	346.9	6.00	10.04	0.10
ISL-05	123.84	331.5	6.22	9.32	0.18
ISL-06	120.11	325.8	6.29	10.18	0.13
ISL-07	89.71	329.9	6.31	13.08	0.22
ISL-08	132.88	356.8	5.95	9.87	0.12
ISL-09	98.53	330.6	6.25	10.43	0.05
ISL-10	90.12	312.4	6.24	10.10	0.10
ISL-11	88.64	314.2	6.25	9.60	0.22
ISL-12	57.24	309.9	6.50	9.56	0.11
ISL-13	56.96	309.0	6.49	11.23	0.13
ISL-14	56.90	311.0	6.47	10.22	0.05
ISL-15	52.15	311.8	6.33	11.39	0.17
ISL-16	51.59	313.6	6.40	11.29	0.05
ISL-17		215.4	6.42	9.75	0.17
ISL-18		320.4	6.03	10.66	0.02
ISL-19		352.2	5.72	11.89	0.13

There are perennial rivers and springs in the QSL region, which are the two main sources of water for the salt lake (Lowenstein et al., 1989; Zhang et al., 1993). The Golmud River, located in the southern Qarhan playa, flows from the eastern Kunlun Mountains into the QSL. It is the main water source with an annual average discharge of 19.2×10^9 m^3 (Yu et al., 2009). Other rivers, including the Qingshui, Tuolahai, Zaohuo, Wutumeiren, Nuomuhong, and Halawusu rivers, with various average discharges also originate from the south and southwest and flow into the Qarhan playa. The geochemistry of the rivers in this region is similar to that of world average river water (Lowenstein et al., 1989; Lowenstein and Risacher, 2009). The springs emerge at the northern margin of the Qarhan playa along a linear fault zone (Fig. 1a) (Zhang, 1987). This spring water dissolved salt minerals and contains more Ca on an equivalent basis than $SO_4+HCO_3+CO_3$. The spring water is classified as a $CaCl_2$ brine (Lowenstein et al., 1989; Zhang et al., 1993; Lowenstein and Risacher, 2009).

3. Samples and analytical methods

3.1. Sample collection

A 102 m long sediment core (ISL1A) was drilled from the Bieletan section of the QSL in the eastern Qaidam Basin (Fig. 1b). Twelve clay samples containing dark organic matter were collected from the upper 54.5 m of the core for accelerator mass spectrometry (AMS) ^{14}C dating. Eight halite samples were collected from the upper 46.0 m of the core for ^{230}Th dating. AMS ^{14}C and ^{230}Th dating results of upper 46.0 m from ISL1A in this study was shown in Supplementary Tables 3 and 4. An assessment and comparison of these ^{230}Th and AMS ^{14}C ages for core ISL1A have been reported in previous works (Fan et al., 2014a, b).

For this study, we collected nineteen brines from QSL (Table 1). At each sample site, brines were collected in acid-washed 500 mL low-density polyethylene (LDPE) bottles, rinsed with sample water. Within 48 h of collection, each sample was filtered through a 0.45 μm Luerloch syringe filter (polypropylene membrane) into an acid-washed 125 mL LDPE bottle.

3.2. Analytical methods

100 mL brine samples were collected for analysis of elemental concentrations and isotopic compositions. The analyses of Cl$^-$ and

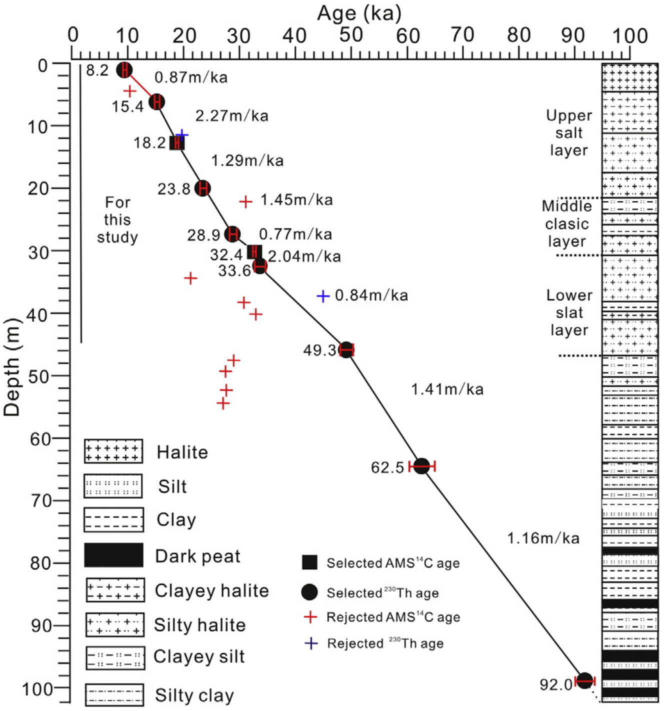

Figure 2. Age framework and lithology of ISL1A (Fan et al., 2014a, b).

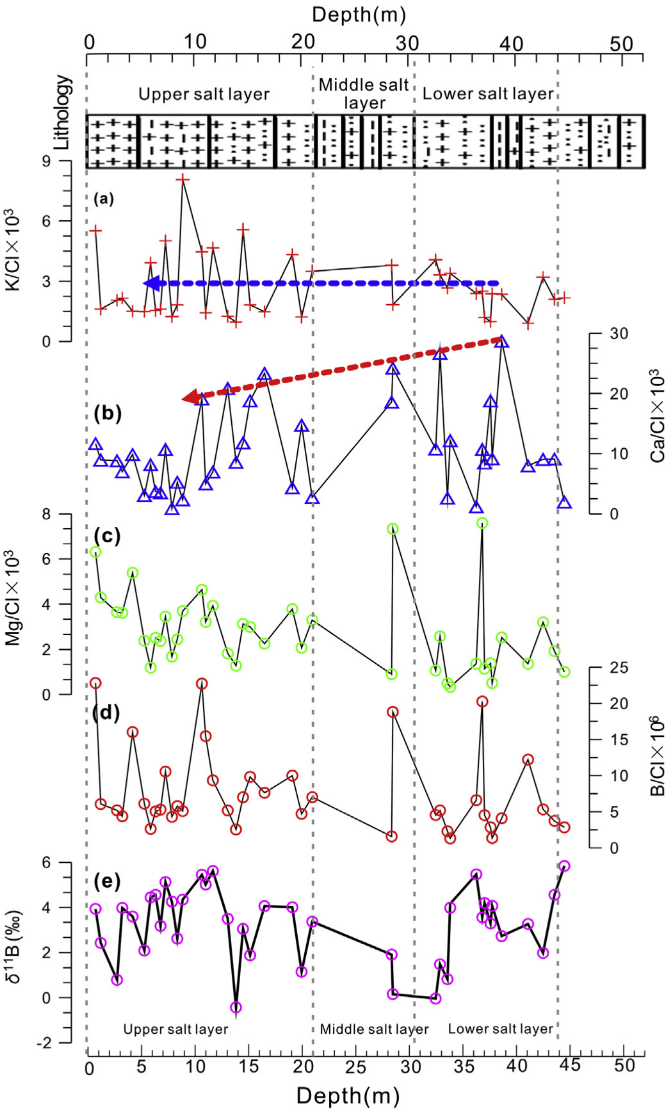

Figure 3. The variation of (a) K/Cl × 10^3, (b) Ca/Cl × 10^3, (c) Mg/Cl × 10^3, (d) B/Cl × 10^6 and (e) $\delta^{11}B$ values of halite with depth in ISL1A.

B^{3+} concentrations were performed in the Qinghai Institute of Salt Lakes, Chinese Academy of Sciences. The Cl^- concentrations were determined by $AgNO_3$ and $BaCl_2$ potentiometric titration. The B^{3+} concentration was determined by absorption photometry.

To determine the $^{11}B/^{10}B$ ratio, the halite was dissolved in low-B water and centrifuged for approximately 20 min to remove the small amount of detrital impurities. These solutions of halite from core ISL1A and brines from the Qarhan playa, contained about 10 μg B (pH 6–7). They were passed through a 0.8 mL Amberlite resin column (120–200 mesh) using low-B water and 2 M NH_4OH to remove the cations. Then, 12 mL of 0.1 M HCl was used to elute the B from the resin. The eluate was evaporated at 65 °C by adding an equal amount of mannitol to suppress volatilization of B. Then, the residue was dissolved in low-B water and passed through a 0.15 mL Amberlite resin column (120–200 mesh) to further purify the sample. After adding more mannitol, the eluate was again evaporated at 60 °C for mass spectrometry analysis. The low-B water used in the experiment was obtained by passing pure water through a column loaded with Amberlite IRA 743 (a boron-specific resin). The boron blank, as determined by isotope dilution mass spectrometry, was less than 40 ng (Liu et al., 2000). The B isotopic measurement procedures are the same as those described elsewhere (Xiao et al., 1988, 2003; He et al., 2013). A single tantalum filament (1.5 cm × 0.098 cm × 0.0025 cm) was degassed in a vacuum for 1 h at a current of 3.0 A, was first treated with 2.5 μL of graphite slurry, and was then successively loaded with sample solution when the slurry was almost dry. Isotopic measurements were performed on a Triton thermal ionization mass spectrometer (TIMS; Thermo Fisher Scientific Inc., U.S.A.) at the Qinghai Institute of Salt Lakes. The instrument was equipped with nine Faraday cups The instrument was retrofitted with a double Faraday collector array with fixed spacing, enabling more precise measurements using a static multi-collector mode for $^{133}Cs_2^{10}BO_2^+$ (mass 308) and $^{133}Cs_2^{11}BO_2^+$ (mass 309) ions. The intensity of the ion beams at masses 308 and 309 ($R_{309/308}$) were calculated. The $^{11}B/^{10}B$ ratio was calculated as $R_{309/308}$–0.00078. The B isotopic composition was expressed as the $\delta^{11}B$ value according to the following formula:

$$\delta^{11}B\left(‰\right) = \left\{\left[\left(^{11}B/^{10}B\right)_{sample} - \left(^{11}B/^{10}B\right)_{stand}\right]\left(^{11}B/^{10}B\right)_{stand}\right\} \times 1000$$

where the standard material was NIST SRM 951. The measured $^{11}B/^{10}B$ value of NIST SRM 951 is 4.0500 ± 0.0002 (2δ, $n = 6$) with an analytical uncertainty of 0.3‰ (He et al., 2013).

4. Results

4.1. The stratigraphy and geochronology of core ISL1A

Lithologic and sedimentologic characteristics indicate that there were two large units represented in core ISL1A. The upper part of core ISL1A (0–51.0 m) is composed of white and white-gray halite layers, silty sand, and clay layers; the lower part of core ISL1A (51.0–102.0 m) is composed of silty sand and clay with dark organic matter (Fig. 2). The upper 44.0 m of sediment in core ISL1A was divided into three salt-bearing layers (0–21.0 m, 27.0–28.2 m, and 31.5–44.0 m) (Fig. 2).

In this study, the age vs. depth framework of core ISL1A was established based on the stratigraphic order as reported by Fan et al. (2014a, b). By comparing the ^{230}Th ages of the halite with the AMS ^{14}C ages of the TOC in core ISL1A, we found that the upper four AMS ^{14}C ages from 4.65 to 30.29 m are in good agreement with the upper six ^{230}Th ages from 0.35 to 32.29 m in this core. The AMS ^{14}C ages at 30.29–54.50 m were younger with increasing depth due, in part, to ages that are greater than the upper limit of the radiocarbon ^{14}C dating technique (Fan et al., 2014a, b). The ^{230}Th ages from the upper 54 m of sediment from core ISL1A are in stratigraphic order (Fig. 2). Therefore, we selected six ^{230}Th ages of halite and two AMS ^{14}C ages of TOC to construct an age model of the salt layer in core ISL1A. In this study, this age framework of the upper 44.0 m of the sediment core to discuss the paleoclimatic implication of the B isotopes of halite.

4.2. Elemental concentrations and B isotopic values of brines in QSL

The Cl^- and B^{3+} concentrations, TDS and pH values of brines in the QSL are listed in Table 1. The B concentrations of brines in the QSL range from 51.6 to 138.4 mg/L, which are higher than those (0.77–13.08 μg/g) of halite in ISL1A (Fan et al., 2015). TDS and pH values of these brines in the QSL are varied from 215.4 to 256.8 g/L and 5.72 to 6.50, respectively.

The δ^{11}B values of brines from the QSL are also listed in Table 1. The result shows that δ^{11}B values range from +9.32‰ to +13.08‰. These values are obviously lower than those of marine evaporites (+18.2‰ to +31.7‰) and fall within the range of non-marine evaporites (−30.1‰ to +10.2‰) (Barth, 1993).

5. Discussion

5.1. B concentrations of river/spring water and brines in the QSL

B is a conservative element and is relatively enriched in the upper part of the lithosphere, volcanic outgassing, and thermal springs (Zhang, 1987). The B concentration of seawater is 4.5 mg/L, while those of brines and hot springs from salt lakes on the TP are higher (Zhang, 1987). Previous studies indicate that rivers and springs are the two main water sources in the QSL (Zhang, 1987; Lowenstein et al., 1989; Yang et al., 1993; Zhang et al., 1993; Yuan et al., 1995). The geochemistry of the Golmud River indicates that B concentrations vary from place to place, and average values along the Golmud River at the Gobi Desert, alluvial fan, alluvial plain, and lacustrine plain are 0.274, 0.340, 0.618, and 0.721 mg/L, respectively (Tan et al., 2001). Other rivers (including the Wutumeiren, Qingshui, Tuolahai, Xiangride, and Qaidam rivers) have similar B concentrations to that of the Golmud River (Zhang, 1987), suggesting that the rivers, originated from eastern Kunlun Mountain, basically supplied similar or consistent B concentrations to the QSL. In addition, another source for the QSL is spring waters which located on the northern margin of the Qarhan playa alone a linear fault zone. Lowenstein et al. (1989), Spencer et al. (1990), and Zhang et al. (1993) reported that the chemical composition of this spring water is quite different from that of the rivers. The spring water contains more Ca on an equivalent basis than $SO_4+HCO_3+CO_3$ and is classified as a $CaCl_2$ brine (Zhang, 1987). They proposed that brines in the QSL consist of a mixture of about 40 parts river water and 1 part spring water (Lowenstein et al., 1989; Zhang et al., 1993; Lowenstein and Risacher, 2009). The B concentrations of three springs are ∼50 mg/L (in this study), which is obviously higher than those of the rivers. The mixing pattern between the river water and spring water (Lowenstein et al., 1989; Zhang et al., 1993) dominates the chemical concentrations (including the B concentrations) of the original lake water and the brines in the study area.

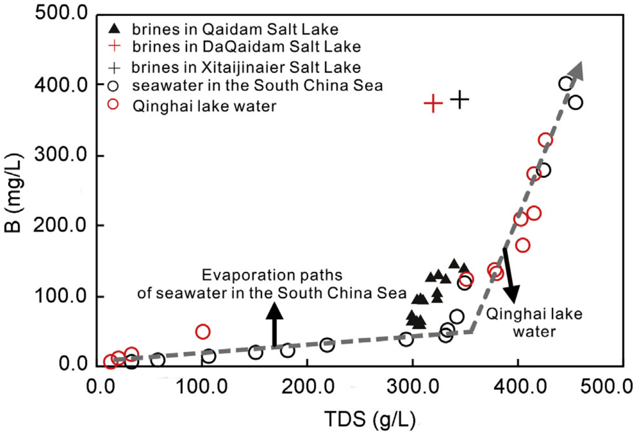

Figure 4. The diagram between B concentrations and TDS from evaporation experiments of seawater in South China Sea (Song and Li, 1994) and brackish water in Qinghai Lake (Sun et al., 1995), and brines from Qarhan (in this study), DaQaidam and Xitaijinaier salt lakes in the Qaidam Basin (Zhang, 1987). The slope variation point indicates the precipitation of the halite.

Based on field investigations and chemical analyses of brines in salt lakes, Zhang (1987) concluded that two zones of higher B concentrations brines (ranging from 200 to 500 mg/L) existed in the Qaidam Basin. One is the Taijinaier salt lake area (including the Dongtai and Xitai salt lakes, and Yiliping playa) with B concentrations ranging from 200 to 300 mg/L in the central Qaidam Basin; and the other is the Qaidam salt lake area (including the Da Qaidam and Xiao Qaidam salt lakes) with B concentrations ranging from 300 to 500 mg/L in the northern Qaidam Basin. The geochemical evidence indicates that the higher B values in those two zones are mainly supplied through river water that originated from B-rich hot springs or geothermal fluids from deep faults (Xiao et al., 1992; Tan et al., 2012; Yu et al., 2013). Except these salt lakes, Zhang (1987) argued that the enrichment of B in other salt lakes in the Qaidam Basin resulted from the solar evaporation of salt lakes. In order to verify the effect of evaporation on the B concentrations of brines in the Qarhan playa, we plotted the B concentrations and TDS of brines from the Qarhan, Da Qaidam, and Xitaijinaier salt lakes (Zhang, 1987) (Fig. 4) and found that the B concentrations of the Da Qaidam and Xitaijinaier salt lakes obviously fell outside the evaporation paths of brackish water in Qinghai Lake on the northeastern TP (Sun et al., 1995) and seawater in the South China Sea (Song and Li, 1994). On the contrary, the B concentrations and TDS of brines from the Bieletan, Dabuxun, Qarhan, and Huobuxun sections of the QSL fell around the evaporation paths of brackish water and seawater (Song and Li, 1994; Sun et al., 1995). This comparison demonstrated that the B enrichment of brines in the QSL primarily results from stronger evaporation and concentration of lake water. Therefore, the higher B concentrations of the brines from the QSL imply that stronger concentration of brines or lake water occurred under conditions that favored stronger evaporation.

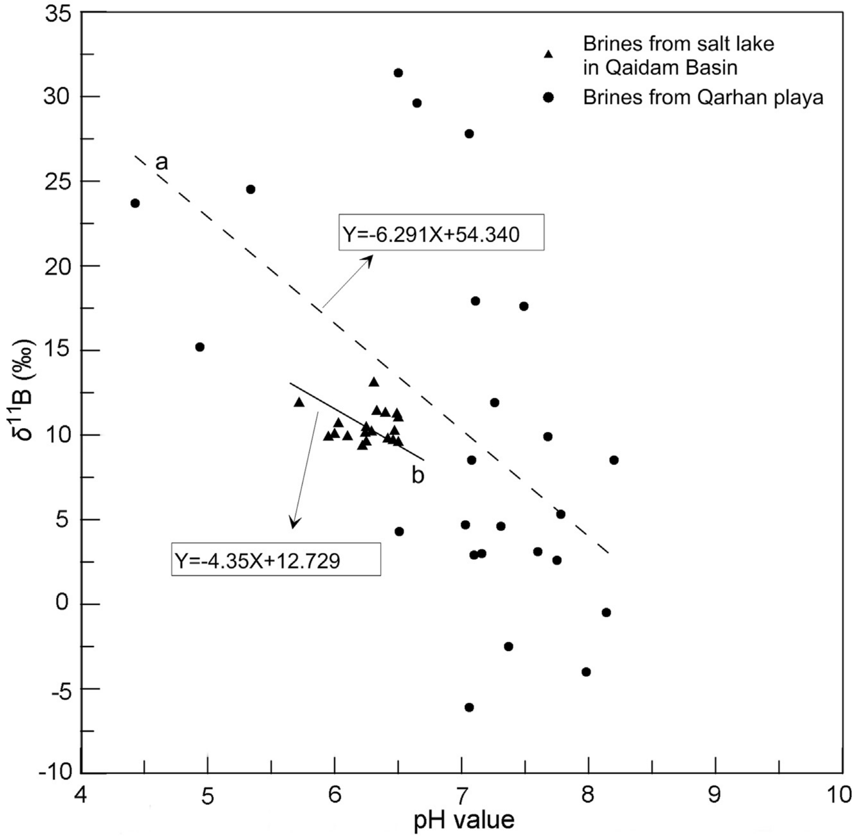

Figure 5. The diagram between pH and $\delta^{11}B$ values of brines from salt lakes in the Qaidam Basin. Line a: The relationship between pH and $\delta^{11}B$ values of brines (Liu et al., 1999; Xiao et al., 1999); Line b: The relationship between pH and $\delta^{11}B$ values of brines in Qarhan playa (this study).

5.2. B concentrations of halite from core ISL1A in the QSL

Combining with previously chemical concentrations of halite in ISL1A (Fan et al., 2015), the comparisons of K/Cl, Mg/Cl, Ca/Cl, B/Cl ratios and $\delta^{11}B$ values of halite in ISL1A have been shown in Supplementary Table 1 and Fig. 3. On the whole, K/Cl values frequently fluctuant with a relatively high average value in the upper salt layer. In contrast, the Ca/Cl values gradually decrease from the lower salt layer to the upper salt layer over the entire 44.0 m (Fig. 3a and b). In addition, the Mg/Cl and B/Cl values of the halite exhibit a synchronous variation pattern in Fig. 3c and d. The B concentrations in the halite from core ISL1A range from 0.77 to 13.08 μg/g (Supplementary Table 1) (Fan et al., 2015), which is lower than those of the brines in the QSL. Previous studies show that B concentrations in the halite are mainly derived from brine fluid inclusion in the halite (Dong, 1984; Liu et al., 2000; Fan et al., 2015). This conclusion can also be confirmed by the roughly synchronous oscillating trends of the B/Cl ratio and the $\delta^{11}B$ value of halite from the lower, middle, and upper salt layer of core ISL1A (Fig. 3d and e). However, the B/Cl ratio does not seem to correlate with the K/Cl and Ca/Cl ratios of the halite from core ISL1A (Fig. 3a and b). In addition to a prominent peak, the K/Cl ratio of the halite exhibits a slight increase from 44.0 to 7.0 m (Fig. 3a), while the Ca/Cl ratio of the halite from core ISL1A gradually decreases with depth (Fig. 4b). The X-ray diffraction (XRD) results of core ISL1A indicate that more sylvite (KCl)/carnallite ($MgCl_2 \cdot KCl \cdot 6H_2O$) and the less gypsum ($CaSO_4$) were present in the upper salt layer than in the middle and lower salt layers (An, 2012). The high consistent of potash minarels in upper salt layer should led to an increase in the value of K/Cl, but in this study, the value of K/Cl in upper layer did not changed obviously. The decreasing Ca/Cl values in this study and less abundant gypsum (an early precipitating mineral), indicated by the XRD results (An, 2012), show that paleobrines from Qarhan Salt Lake were concentrated from the lower salt layer to the upper salt layer of core ISL1A.

The values of K/Cl should be primarily derived from fluid inclusions in the halite. From Fig. 3a and d, it is difficult to match the peaks and valleys of the K/Cl and B/Cl values at every depth node. The relationships between K/Cl and B/Cl values and K/Cl and $\delta^{11}B$ values (Fig. 3a, b, and e) in the salt layers are quite weak. Zhang (1987) reported that B concentrations co-varied with TDS for brines in the Qaidam Basin, while the K^+ concentrations of the brines steadily increased at first and then suddenly decreased with increasing TDS due to the precipitation of potash deposits (such as sylvite or carnallite). Therefore, the precipitation of sylvite and carnallite incorporates more K^+ from the brines into solid minerals while the B concentrations of the brines increased by water concentration. This accounts for the weak relationships between the K/Cl and B/Cl values and the K/Cl and $\delta^{11}B$ values of core ISL1A. This result indirectly confirms that the B/Cl values of the halite in core ISL1A might be regarded as a good index of the evolution of the paleobrines in the study area.

5.3. Paleoclimatic interpretation of $\delta^{11}B$ values of brines and halite in the QSL

B has two stable isotopes, ^{10}B and ^{11}B, which make up approximately 19.8% and 80.2% of total boron, respectively. There are two main dissolved species of B, $B(OH)_3$ (boric acid, trigonal species) and $B(OH)_4^-$ (borate anion, tetrahedral species), whose relative abundance is pH dependent. ^{11}B is usually enriched in $B(OH)_3$, while ^{10}B is usually more abundant in $B(OH)_4^-$ (Swihart et al., 1986;

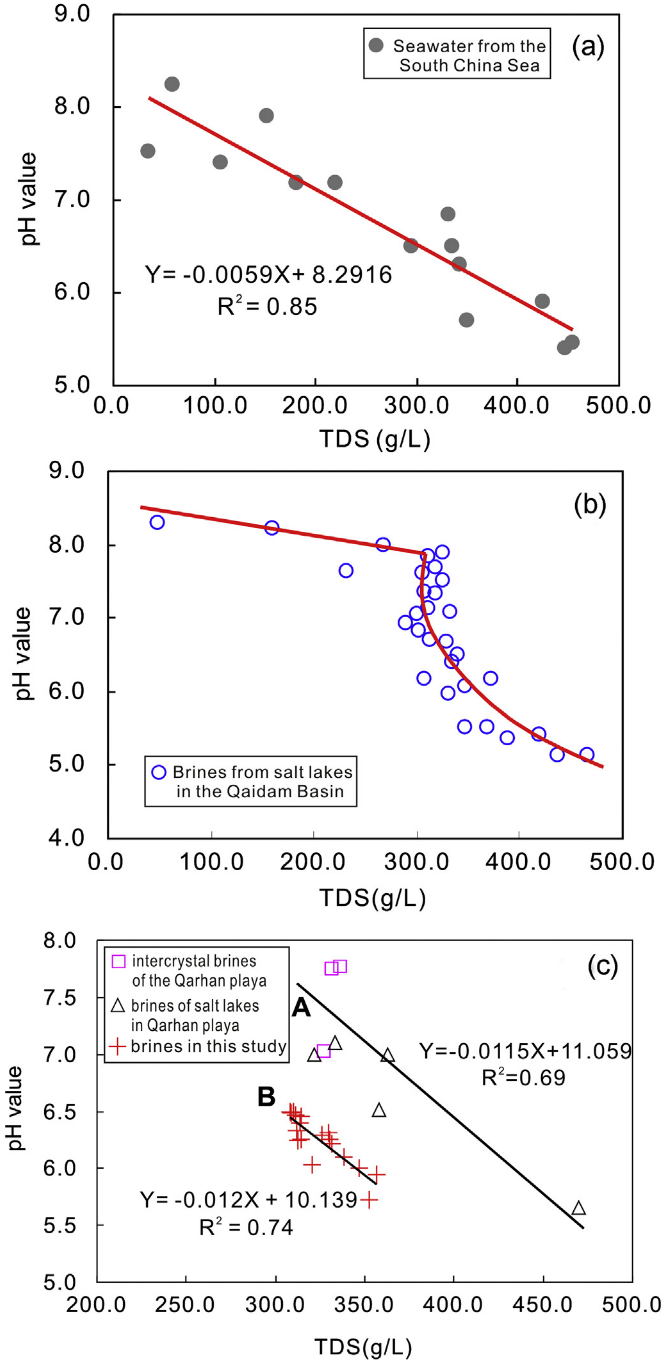

Figure 6. The diagram between pH and TDS values of seawater and brines. (a) The pH and TDS values from evaporation experiment of seawater in South China Sea (Song and Li, 1994); (b) The pH and TDS values of brines from salt lakes in the Qaidam Basin (Zhang, 1987); (c) The pH and TDS values of brines in Qarhan playa (Line A: previous data; Line B: in this study).

Palmer et al., 1987; Qi et al., 1989; Vengosh et al., 1991, 1992; Barth, 1993; Palmer and Swihart, 1996). Therefore, the pH of brines dominates the variation in δ^{11}B values of brines in salt lakes. Evaporation experiments on Qinghai lake water, artificial brines, salt lake brines in the Qaidam Basin, and normal seawater indicate that the δ^{11}B values of brines increase with decreasing pH of the brines or the parent solutions (Vengosh et al., 1992; Sun et al., 1995; Liu et al., 2000). This negative correlation between pH and δ^{11}B values of brines in the QSL and other salt lakes in the Qaidam Basin can also be observed in Fig. 5. This is due to the fact that B(OH)_4^- becomes more depleted in salt lake brines with decreasing pH, which results in more abundant ^{11}B in the brines (Vengosh et al., 1992; Xiao et al., 1992; Liu et al., 2000), and thus, increasing δ^{11}B values with decreasing pH.

What factor dominated the pH of brines in salt lakes? Evaporation experiments on Qinghai lake water, seawater, and salt lake brines indicate that the pH and TDS of the parent solutions decreased and increased, respectively, with steady evaporation (Vengosh et al., 1992; Sun et al., 1995; Liu et al., 2000). This obviously negative correlation between pH and TDS can also be seen in the evaporation of seawater from the South China Sea (Song and Li, 1994) (Fig. 6a; $R^2 = 0.85$, $n = 14$), salt lake brines from the Qaidam Basin (Zhang, 1987) (Fig. 6b), and surface brines (Line A in Fig. 6c; $R^2 = 0.69$, $n = 8$) (Xiao et al., 1999) and intercrystalline brines from the Qarhan playa in this study (Line B in Fig. 6c; $R^2 = 0.74$, $n = 19$). All of these results demonstrate that higher TDS of brines corresponds to lower pH of brines in salt lakes (Fig. 6).

Based on the discussion of the relationships between B concentrations and TDS, pH and TDS, and pH and δ^{11}B values in brines and halite in sections 5.1, 5.2, 5.3 we conclude that higher B concentrations correspond to higher TDS and more positive δ^{11}B values for brines under conditions that favor stronger evaporation. This conclusion is supported by the comparison of B concentrations and δ^{11}B values for halite from core ISL1A in Fig. 7a and b. In addition, these results imply that the more positive δ^{11}B values of the halite in core ISL1A indicate higher salinity Qarhan paleobrines or paleolake water.

5.4. *Paleoclimatic records and regional comparisons on the NE QTP*

In this study, based on the paleoclimatic interpretation of B isotope values of halite from core ISL1A, three phases (I, II, and III) have been identified as shown in Fig. 7. In addition, paleoclimatic interpretation of the fine-grained carbonate form core ISL1A will be discussed in this study. The XRD results indicate that the fine-grained (<38 μm) carbonate minerals are mainly dominated by calcite, and the concentration of dolomite is very low in core ISL1A (Fan et al., 2014b), implying that the fine-grained carbonates contain a large amount of authigenic carbonate that precipitated from surface lake water (Fan et al., 2014b). Therefore, the δ^{18}O values of the fine-grained carbonate from core ISL1A are a useful proxy for determining the balance between precipitation and evaporation in the Qarhan region. Positive δ^{18}O values of the carbonate from core ISL1A would indicate dry paleoclimate conditions. In contrast, negative δ^{18}O values of the carbonate from core ISL1A would indicate large amounts of runoff draining into the QSL and a lower relative salinity of Qarhan paleolake water (Fan et al., 2014b). The δ^{18}O value of the fine-grained carbonate is also used as a good reference for comparison with the δ^{11}B values of halite from the same core in this study.

Phase I (44.0–31.5 m, corresponding to 46–34 ka): On the whole, the δ^{11}B values of halite from core ISL1A during this phase (corresponding to higher solar insolation at 30°N; Fig. 7e) are more positive than those during phase II (corresponding to lower solar insolation at 30°N; Fig. 7e). Coincidently, the more positive δ^{11}B values of the halite correspond to the lower salt layer and the more positive δ^{18}O values of the fine-grained carbonate from core ISL1A (Fig. 7c), suggesting that positive δ^{11}B values of the halite from core ISL1A indicate dry paleoclimate conditions. Some of the details of the δ^{11}B values of the halite from this phase can also be observed in Fig. 7b. At the beginning of this phase, the positive δ^{11}B values of the halite correspond to the slightly positive δ^{18}O values of the carbonate from core ISL1A. These positive isotopic values also correspond to a less paleomoisture record in monsoonal central Asia

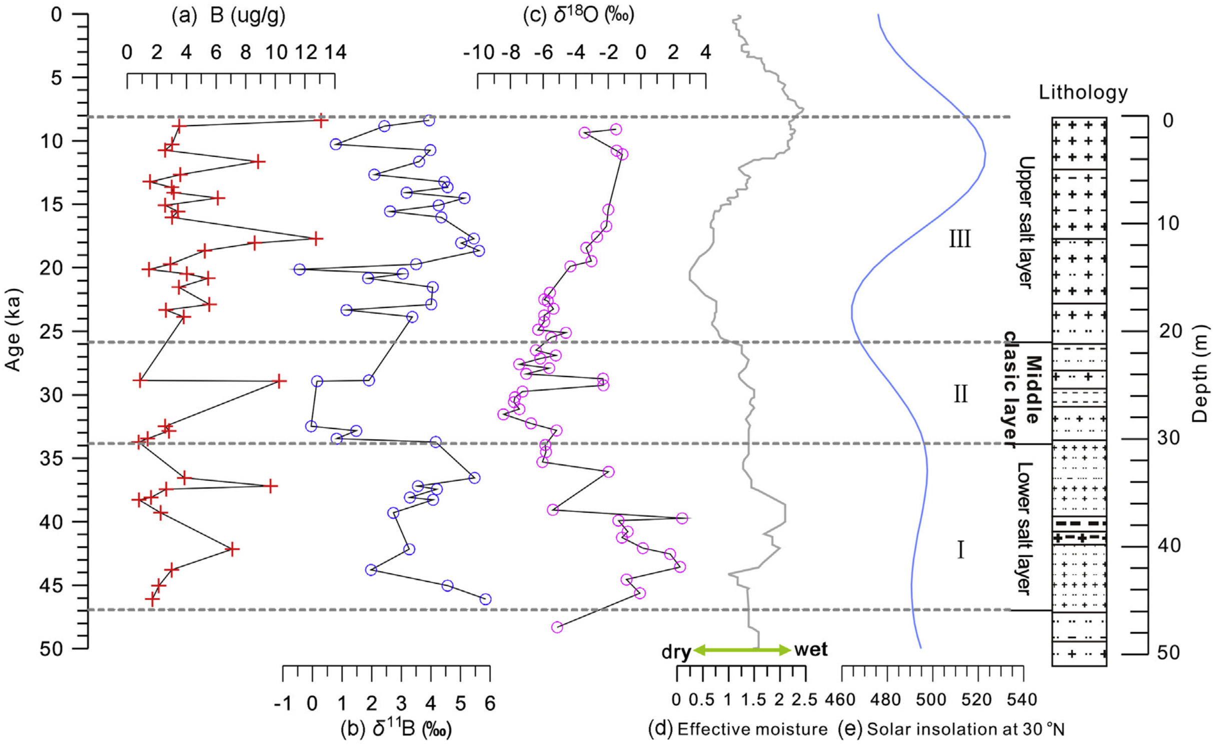

Figure 7. The boron concentrations, $\delta^{11}B$ values of halite and $\delta^{18}O$ values of lake carbonates in ISL1A, effective moisture and Northern Hemisphere solar insolation at 30°N. (a) B concentrations of halite in ISL1A (this study); (b) $\delta^{11}B$ values of halite in ISL1A (this study); (c) $\delta^{18}O$ values of lake carbonates in ISL1A (Fan et al., 2014b); (d) synthesized effective moisture record in monsoonal central Asia (Herzschuh, 2006); (e) Northern Hemisphere solar insolation at 30°N (Berger and Loutre, 1991).

during the last 50,000 years (Fig. 7d) (Herzschuh, 2006). From 42 ka to 38 ka, the more negative $\delta^{11}B$ values of the halite (Fig. 7b), the more positive $\delta^{18}O$ values of the carbonate (Fig. 7c), and the lower B concentrations of the halite from core ISL1A (Fig. 7a) correspond to a more paleomoisture record in monsoonal central Asia (Fig. 7d). After which, the increasing B concentrations and $\delta^{11}B$ values of the halite once again correspond to a less paleomoisture record in monsoonal central Asia (Fig. 7d).

Phase II (31.5–21.0 m, corresponding to 34–26 ka): As a whole, less salt minerals and more clastic sediments precipitated during this phase, suggesting a relatively wetter climate than phases I and III. This wet paleoclimate can also be verified by the more negative $\delta^{11}B$ values of the halite and the more negative $\delta^{18}O$ values of the carbonate from core ISL1A, corresponding to lower solar insolation at 30°N (Fig. 7e). This comparison demonstrates that the negative $\delta^{11}B$ values of the halite from core ISL1A indicate either lower salinity of paleolake water or a wetter paleoclimate. This conclusion has been confirmed by the $\delta^{11}B$ values of salt minerals from the Dongtai Salt Lake where more negative $\delta^{11}B$ values correspond to the mudstone strata deposited under warm-humid climate conditions (Wei et al., 2014). In addition, a synchronous positive shift of the B concentrations and $\delta^{11}B$ values of the halite and the $\delta^{18}O$ values of the carbonate from core ISL1A can also be observed at 27–28 m (phase II).

Phase III (21.0–0 m, corresponding to 26–9 ka): This phase corresponds to the upper salt layer, indicating a dry paleoclimate during this period. The progressively increasing $\delta^{18}O$ values of the carbonate from core ISL1A correspond well to the sedimentary deposition of a thick salt layer and the formation of the Qarhan playa. The relatively positive $\delta^{11}B$ values of halite during this phase compared to those seen during phase II demonstrate that the higher salinity of the Qarhan paleolake water corresponds to increasing solar insolation at 30°N (Fig. 7e). This dry paleoclimate interpretation for the Qarhan region does not seem to match the progressively more paleomoisture record in monsoonal central Asia (Fig. 7d; Herzschuh, 2006). This might be due to the topography in the Qaidam Basin influencing the balance between precipitation and evaporation during the interglacial period (Zhao et al., 2007; Fan et al., 2014c). Except for these comparisons, we observe that the B concentrations and $\delta^{11}B$ values of the halite from core ISL1A during this phase show frequent fluctuations. Although we cannot explain this phenomenon at present, partly due to a low resolution B isotope proxy and uncertainties in the ages, this comparison suggests the possibility that refined paleoclimate information could be obtained from further research of B isotope values of salt minerals.

The generally synchronous variation of the $\delta^{11}B$ values of halite and the $\delta^{18}O$ values of carbonates from core ISL1A corresponds well to the paleomoisture record in monsoonal central Asia and solar insolation at 30°N. These results imply that the paleoclimate implications of the B isotopic analysis of halite are reasonable in this study. In addition, the B isotope values of salt minerals, especially in the salt lake region, can be used as a new proxy to reconstruct the salinity record of paleolake waters or brines as well as paleoclimate conditions, although some details, such as those in phase III cannot always be explained.

6. Conclusions

B concentrations and its isotope compositions of brines from Qarhan Salt Lake, western China have been analyzed in this study. By comparisons of B concentrations and total dissolved solids (TDS) of brines in Qarhan Salt Lake (QSL) with evaporation path of brackish water, we found that B enrichment of brines primarily results from strong evaporation and concentration of Qarhan lake water. Good relationships between B concentrations and TDS, pH and TDS, and pH and δ^{11}B values of brines demonstrate that higher B concentrations correspond to higher TDS and more positive δ^{11}B values for brines under conditions that favor stronger evaporation. Combining with previously elemental ratios (K/Cl, Mg/Cl, Ca/Cl and B/Cl) and δ^{11}B values of halite from a sediment core (ISL1A), comparisons show that higher B concentrations and more positive δ^{11}B values of halite can indicate higher salinity of Qarhan paleolake water as well as drier climate conditions. The higher salinity and drier climate conditions of the Qarhan paleolake occurred in two intervals, around 46–34 ka and 26–9 ka, which almost coincide with the upper and lower halite-dominated salt layers in core ISL1A. Drier climate phases documented by the δ^{18}O record of carbonate from core ISL1A, the paleomoisture record in monsoonal central Asia, and higher solar insolation at 30°N, suggest that δ^{11}B values of halite in the arid Qaidam Basin can be regarded as a new proxy for reconstructing the salinity record of Qarhan paleolake water as well as paleoclimate conditions.

Acknowledgements

We are grateful to editor and two anonymous reviewers for their constructive comments and suggestions. Thanks to Dr. Yunqi Ma for isotope analyses in the laboratory. This research was financially supported by the National Natural Science Foundation of China (Grant Nos. 41872093, 41502096), Foundation of Qinghai Science & Technology Department (2016-ZJ-715), One-Thousand Innovative Talent Project of Qinghai Province (Grant to QS Fan).

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.gsf.2018.02.016.

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