

Hydrochemical characteristics and sources of brines in the Gasikule salt lake, Northwest Qaidam Basin, China

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(Received January 1, 2015; Accepted May 19, 2015)

The origin of the abundant salt resources in the Gasikule salt lake in the northwest Qaidam Basin in China remains highly contested. We performed a detailed study of the hydrochemical characteristics and material sources of the Gasikule brines based on their chemical composition and hydrogen-oxygen isotopes, together with high-resolution remote sensing imagery of the stream water, surface brine, and intercrystalline brine. Stream waters within the catchment were found to be weakly alkaline and dominated by HCO_3^- , Cl^- , and Na^+ ions. The Gasikule surface brine was also weakly alkaline and classified as chloride type. The average total dissolved solid (TDS) was 366.19 g L^{-1} ; Cl^- and SO_4^{2-} were the dominant anions, and Mg^{2+} and Ca^{2+} were the dominant cations. The intercrystalline brine was weakly acidic and classified as a magnesium sulfate subtype. The average TDS was 381.80 g L^{-1} ; Cl^- was the dominant anion, and K^+ and Mg^{2+} were the dominant cations. The surface and intercrystalline brines were enriched in K^+ because of the annual formation of “dry salt lake” conditions. Using Enhanced Thematic Mapper (ETM) and Satellite Probatoire Pour l’Observation de la Terre 5 (SPOT5) remote sensing data, two (approximately) north-south trends in salinity in the eastern area of the Gasikule surface brine were identified (referred to as “water swell annular anomalies”). These two anomalies were found to be located (approximately) above an existing buried fault from which Ca-Cl type deep water recharged the surface brine. Three main sources were identified for the recharge of the surface and intercrystalline brines of the Gasikule salt lake: (a) stream water sourced from rainfall and melting ice; (b) leaching of Pliocene salt-bearing host rocks that contain sulfate minerals such as gypsum, mirabilite, and celestite at the northern end of the salt lake; and (c) Ca-Cl type deep water, associated with a buried fault.

Keywords: hydrochemical characteristics, origin of brines, material source, Gasikule salt lake, Qaidam Basin

INTRODUCTION

Hydrochemical characteristics and identification of brine sources underpin the rudimentary understanding of the development and utilization of salt mineral deposits. The study of brine genesis informs us not only on the geochemical evolution of salt lakes (Berner and Berner, 1996; Yan *et al.*, 2002), but also provides important evidence of salt-separating processes during the production of evaporites (Zhang, 1987; Zhang *et al.*, 1999; Zheng *et al.*, 1989). For this reason, the hydrochemical characteristics and origin of brines in salt lakes are currently the focus of worldwide research efforts (Harrington *et al.*, 2008; Jones *et al.*, 1977; Long *et al.*, 1992a, 1992b, 2009; Tweed *et al.*, 2011).

In order to develop the sylvite resources in the salt lakes of China, much research has been performed on the

hydrochemistry and source materials of the brine in the salt lakes of the Qinghai-Tibet Plateau (Casas *et al.*, 1992; Liu *et al.*, 1997; Vengosh *et al.*, 1995; Yang *et al.*, 1995; Zheng *et al.*, 1989; Zheng, 1997; Zheng and Liu, 2009). Zheng and Liu (2009) obtained hydrochemical data from 1956 for various types of salt lake on the Qinghai-Tibet Plateau. Combined with analysis of the tectonic characteristics of the plateau, they discussed the hydrochemical characteristics of the lakes. In the Qaidam Basin salt lakes, different regions have slightly different sources in different periods. However, weathering of the surrounding rock and the circulation of deep water, volcanic hydrothermal water, and paleolake transitive-gathering water were found to be the principal sources (Vengosh *et al.*, 1995; Wei and Wang, 1988; Yang *et al.*, 1995; Yuan *et al.*, 1995; Zhang, 1987; Zhang *et al.*, 1999; Zheng *et al.*, 1989; Zhu *et al.*, 1994).

This study investigates the Gasikule salt lake in the Northwest of the Qaidam Basin, China (Fig. 1). The lake is rich in mineral resources (especially oil, gas, and salt deposits) and it has been the focus of considerable re-

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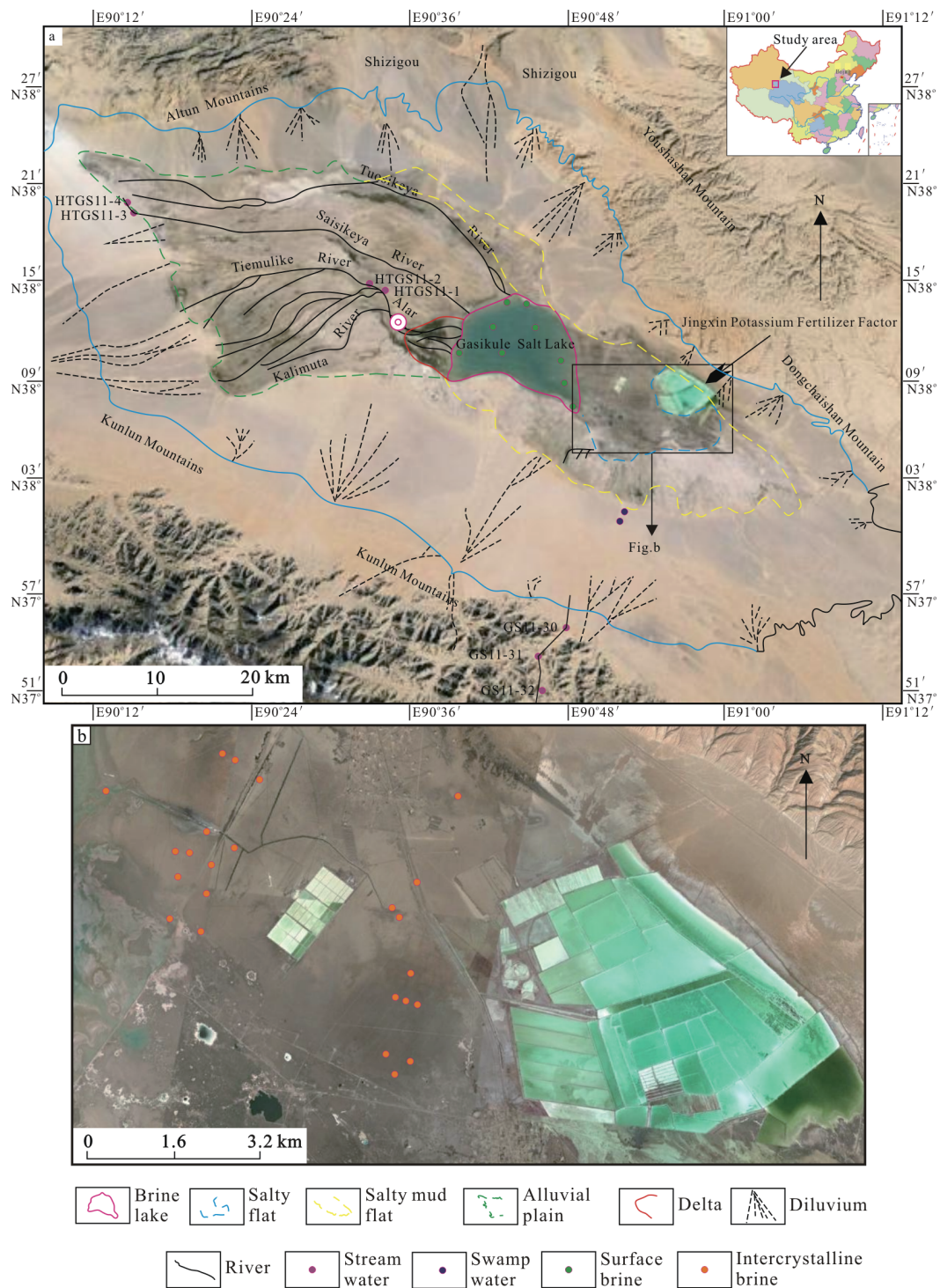


Fig. 1. Lithological sketch map and distribution of sampling locations of the Gasikule salt lake.

search in recent years (Duan *et al.*, 2009; Liu *et al.*, 2013; Zhang *et al.*, 2008a, b; Zhu *et al.*, 2005). For example, the age of the salt formation (Wang *et al.*, 2013), chemical composition, characteristic hydrochemical coefficients, and the origin of the brine in the salt lakes (Fan *et*

al., 2007a, b; Tan *et al.*, 2009, 2011, 2012; Zheng *et al.*, 1989) have all received considerable research interest. However, because of the sampling conditions, the published research has not focused on the brine chemistry of the Gasikule salt lake. Most sampling has been limited to

the surface brine near the lake shoreline, which potentially represents only the lake water quality at this location. The chemistry of the intercrystalline brine has not been explored extensively.

In 2010, we collected samples from the surface brine and streams in the Gasikule salt lake to evaluate uranium resources (unpublished data). It was found that Ca^{2+} concentration in the surface brine was particularly high (Table 1) and it was defined as chloride type with possible deep-water origins. However, previous studies have shown that the concentration of Ca^{2+} in the surface brine of the Gasikule salt lake was lower, which characterized it as magnesium sulfate subtype (Zhang, 1987; Zheng *et al.*, 1989). To explain this apparent contradiction, we collected samples of intercrystalline brine from eastern areas of the surface brine in 2011. Hyperspectral remote sensing images were used to identify the underlying faults in the lake, which confirmed the deep-water sources. Based on analyses of the oxygen ($\delta^{18}\text{O}$) and hydrogen (δD) isotopic compositions, the objective of this study was to identify the sources of two types of natural brines (surface and intercrystalline brines), particularly with consideration of the possible deep-water supply to the surface brine.

STUDY AREA

The Gasikule salt lake (2870 m asl) was formed by tectonic movement of the Himalayas during the Tertiary, and it is today located in the northern part of the Tibetan Plateau (37°57′–38°11′ N, 90°40′–91°10′ E) in the northwest of the Qaidam Basin (Fig. 1). The climate of this area is classified as arid continental. Average annual rainfall is just 13.4 mm, whilst the average potential evaporation rates are high at 30 mm yr⁻¹. Mean annual air temperature is 3°C, exceeding 30°C in the boreal summer (July) and reaching lows of less than -20°C in the winter (January). The primary wind direction is NW and NNW with a maximum wind velocity of 40 m s⁻¹. The ancient lake basin comprises a large modern saline lake in the western part, where water depth reaches 1.3 m in the south-central areas. The modern saline lake has a SE-NW orientation, is about 12.87-km long, and approximately 10.21-km wide in the most eastern part. The surface area of the lake is 117.5 km² and the watershed has a total drainage area of 5000 km², with a watershed to lake area ratio of 42.55. In contrast, the dry saline pan is widespread in the east (Chen and Bowler, 1986; Zhang, 1987). The dry saline pan consists of pressure ridges with well-developed polygonal honeycomb-shaped structures surrounded by a dry saline mudflat and floored by layered halite with permanent groundwater brine located at a depth of 0–1.2 m. The surface salt crust is commonly rugged and consists predominantly of a chaotic mixture of fine-

grained halite crystals and mud. Vadose diagenetic features, such as dissolution pipes and cavities and pendant cements, occur where the surface salt crust lies above the water table.

Five streams drain into the lake bringing meteoric water from the western mountains (including the seasonal Tiemulike, Kalimuta, Saisikeya, and Tuosikeya streams) and snowmelt from the southern Kunlun Mountains; almost no streams drain from the northern and eastern areas of the Gasikule salt lake (Fig. 1). The Tiemulike and Kalimuta streams converge at the village of Alar as the perennial Alar stream. Inflowing streams in the southern margin of the Gasikule salt lake are short and steep, while streams in the west are relatively long and flow through vast flat areas. The groundwater around the Gasikule salt lake is characterized as phreatic pore water confined in loose Quaternary sediments (Tan *et al.*, 2009).

The catchment comprises varied geology. On the southern margin of the salt lake, the Kunlun Mountains are mainly composed of strongly metamorphosed Paleozoic epimetamorphic rocks, where strata ranging from Sinian to Triassic in age are well exposed. The Youshashan Mountain in the north consists of strongly folded Neogene halite-bearing strata and the weathered crust of an epigenetic zone, which often contains salt. The Altun Mountain in the northwest developed as a Paleozoic intrusive body and it has exposures of old gneiss and sporadic ultrabasic rocks. In the east, the Dongchaishan Mountain contains Late Neogene and Early Pleistocene clastic rocks (Wang and Ye, 1992).

According to Zhang (1987), in the central area of the western Qaidam Basin, the lower Youshashan Formation (N_2^1) is a set of brownish-grey and greyish-green mudstones with clear features of gypsification and salinization, interbedded with thin layers of marlstone, gypsum, halite, earth yellow catfish-shaped rocks, and gypsum-cemented fine sandstone. The Upper Youshashan Formation (N_2^2) in the same area is mainly greyish-green and greyish-brown mudstones, interbedded with thin layers of marlstone, limestone, and gypsum-cemented sandstone, celestite, gypsum, mirabilite, and halite. The Shizigou Formation (N_2^3) is mainly grey and greyish-green sandy mudstones, interbedded with a large amount of gypsum, halite, gypsum-cemented sandstone, oolite, and small amounts of mirabilite and bloedite. These strata are widely distributed in the area of Shizigou and Youshashan on the northern side of the Gasikule salt lake, and the lithological assembly is mainly of sulfate (gypsum, mirabilite, and celestite) with very little chloride.

SAMPLING AND ANALYSIS

Field sampling techniques

In all, 34 brine samples were collected between July

and September 2010 and in August 2011. Seven samples of stream water and two of swamp water were collected for comparison. All sampling locations are depicted in Fig. 1 and briefly summarized in the following:

(1) Stream water: two samples were obtained from each of the Saisikeya and Alar streams and three samples from a stream on the northern slope of Kunlun Mountain.

(2) Swamp water: two samples were obtained from the salt mudflat swamp at the southeastern end of the salt lake, which contains residual rainwater from seasonal rainfall that collects on the surface.

(3) Surface brine: nine samples were collected from the modern saline lake using a rubber dinghy and a depth sampler.

(4) Intercrystalline brine: overall, 25 samples were collected, 13 of which were obtained manually by digging to a depth of 0.5 m and storing the samples in sample bags. The remaining 12 samples were obtained as cores by drilling to a depth of up to 2 m under the salt crust of the dry saline pan at the southeastern end of the lake using an American Sauer single backpack type STS-2 corer.

Measurements of electrical conductivity, total dissolved solid (TDS), oxidation-reduction potential, pH, dissolved oxygen, and temperature were undertaken in the field using a HACH digital and portable multi-parameter meter. All samples were filtered through a 0.45 μm glass fiber membrane. Samples for cation analysis were acidified with HNO_3 to $\text{pH} < 2$ and stored in 50 mL or 100 mL polyethylene bottles. The bottles were sealed and sent to the Analytical Laboratory of Beijing Research Institute of Uranium Geology for isotope and ions analysis.

Laboratory analytical techniques

Undiluted freshwater samples (stream and swamp water) were analyzed using ion-chromatography (DIONEX, DX-500 ion chromatography). Standard analytical protocols were followed for cations (National Analysis Standard DZ/T0064.28-93) and anions (National Analysis Standard DZ/T0064.51-93). Alkalinity was measured by automatic titration using a 785DMP™. Analytical precision was 3% of the concentration based on the reproducibility of the samples and standards and the limit of detection was 0.1 mg/L.

Brine samples were diluted $\times 100$ to bring the total salt load to below seawater concentrations for the analysis of cations and anions by ion chromatography. Analysis of the major cations (K^+ , Na^+ , Ca^{2+} , Mg^{2+}) of the brines was performed using a Dionex ICS-1100 ion chromatograph with an IonPac CS12A column, and using 20 mM methanesulfonic acid eluent and a C1RS500 suppresser. Anions (Cl^- , SO_4^{2-} , and HCO_3^-) were analyzed using a Dionex ICS-500 ion chromatograph with an IonPac AS14

column, and sodium carbonate and sodium bicarbonate eluents and an AIRS suppresser. Analysis of field blanks showed that contamination during the sampling procedure, transportation, and treatment was not significant. HCO_3^- and CO_3^{2-} were analyzed by general titration. As the pH of the freshwater and brine was generally < 8.8 , CO_3^{2-} could be considered negligible. The precision for the analysis of the major cations and anions was better than $\pm 2\%$. Charge balance was excellent with all brines showing less than 5% difference between the cations and anions. TDS was calculated by summing all the ions and subtracting half of the HCO_3^- (as per Shen *et al.*, 1993; Wang *et al.*, 2010).

Oxygen and hydrogen isotope compositions were determined using standard methods (Coleman *et al.*, 1982; Epstein and Mayeda, 1953) on a Finnigan MAT 252 stable isotope ratio mass spectrometer (Thermo Scientific). Oxygen was liberated for isotopic analysis using BrF_3 and the resultant oxygen reacted with graphite rods to produce CO_2 , the isotopic composition of which was determined with analytical uncertainty of $\pm 0.2\%$. Hydrogen isotope analyses were performed on aliquots of the samples for oxygen isotopes by the following procedure. Aliquots were enclosed in a molybdenum tube, dried for 12 h at 150°C under vacuum (10^{-3} mbar), to eliminate adsorbed moisture from the air, and then heated in an induction oven to approximately 800°C in quartz for 45 min or until gas release had ceased. The resultant H_2O was reduced to H_2 by hot chromium at 800°C . The results were reported relative to V-SMOW with analytical uncertainties of $\pm 2\%$.

RESULTS AND DISCUSSION

Chemical compositions of stream and swamp waters and surface and intercrystalline brines

All chemical analyses of the stream and swamp water samples are summarized in Table 1. The Gasikule salt lake stream waters were found to be weakly alkaline, i.e., with a pH range of 7.25–9.24 and an average of $\text{pH} = 8.51$. Cation concentrations in stream samples were found to decrease in the order of $\text{Na}^+ > \text{Ca}^{2+} > \text{Mg}^{2+} > \text{K}^+$ with average concentrations of 98.29, 46.57, 29.00, and 4.43 mg L^{-1} , respectively. Anion concentrations in the stream waters decreased in the order of $\text{HCO}_3^- \geq \text{Cl}^- > \text{SO}_4^{2-}$ with averages of 165.57, 164.57, and 101.71 mg L^{-1} , respectively. The average concentration of TDS was 0.53 g L^{-1} with a range of 0.31–0.75 g L^{-1} . The most dominant ions in the stream waters were HCO_3^- , Cl^- , and Na^+ , which differs from other lakes on the Tibetan Plateau, such as Nam Co (Wang *et al.*, 2010) and Pumoyum Co (Ju *et al.*, 2010). Samples GS11-30, GS11-31, and GS11-32 were obtained from the upstream, middle reaches, and downstream areas, respectively, of a stream on the Kunlun

Table 1. Geochemical composition (cations, anions, and TDS) of the stream water, swamp water and brine in the Gasikule salt lake

Sample	Water type	Cl ⁻ (mg L ⁻¹)	SO ₄ ²⁻ (mg L ⁻¹)	HCO ₃ ⁻ (mg L ⁻¹)	Na ⁺ (mg L ⁻¹)	K ⁺ (mg L ⁻¹)	Mg ²⁺ (mg L ⁻¹)	Ca ²⁺ (mg L ⁻¹)	TDS (g L ⁻¹)	pH
GS11-32	Stream water	80	76	99	37	2	12	50	0.31	8.98
GS11-31		95	79	104	40	4	16	64	0.35	7.86
GS11-30		196	80	146	73	4	43	43	0.51	7.25
HTGS11-4		244	180	152	188	7	37	17	0.75	9.36
HTGS11-3		183	121	155	109	5	26	49	0.57	9.24
HTGS11-2		222	100	289	142	5	39	59	0.71	8.64
HTGS11-1		132	76	214	99	4	30	44	0.50	8.24
GS11-18	Swamp water	82	85	102	55	4	13	39	0.33	5.77
GS11-33		64	71	112	55	4	17	35	0.30	5.98
Surface brine (9)	Minimum value	205756	30864	593	2833	4860	65650	19350	335.97	7.32
	Average value	227770	35414	642	3451	5304	72719	21213	366.19	7.63
	Maximum value	247884	40374	665	3918	6101	76960	22760	390.12	8.53
Surface brine (Zhang, 1987)		175757	45235	190	76948	4511	29592	399	332.54	7.56
Intercrystalline brine (25)	Minimum value	143430	20090	371	31130	3586	15548	171	279.99	6.34
	Average value	218652	45578	676	62853	5188	48784	403	381.80	6.89
	Maximum value	306188	85058	912	103778	7895	73854	701	554.19	7.37

Mountain (Fig. 1). All ions (Na⁺, Mg²⁺, K⁺, HCO₃⁻, Cl⁻, SO₄²⁻, except Ca²⁺) and TDS increased progressively downstream from the upper reaches; however, ion concentrations in the southern stream were generally lower than the western stream (Table 1). The reasons for this are likely related to glacial melting, which feeds into the southern stream, together with the relatively short runoff path due to the increased gradient, which results in reduced contact time between the southern stream water and surface debris. Moreover, pH in the streams reduced progressively downstream from the upper reaches in both the southern and the western streams.

Ion concentrations in the two swamp water samples were lower than observed in the stream water samples (Table 1; Fig. 2). This is most likely because the stream water receives rainwater runoff affected by bedrock contact and associated leaching, whereas the swamp water is not influenced by such runoff.

Surface brines were weakly alkaline with a pH range of 7.32–8.53 and an average of pH = 7.63. Cation concentrations in the surface brine decreased in the order of Mg²⁺ > Ca²⁺ > K⁺ > Na⁺ with average concentrations of 72719, 21213, 5304, and 3451 mg L⁻¹, respectively. Anions decreased in the order of Cl⁻ > SO₄²⁻ > HCO₃⁻ with averages of 22770, 35414, and 642 mg L⁻¹, respectively. The average concentration of TDS was 366.19 g L⁻¹ with a range of 335.97–390.12 g L⁻¹. The dominant anions in the surface brines were Cl⁻ and SO₄²⁻, whilst the dominant cations were Mg²⁺ and Ca²⁺, with anomalously low concentrations of Na⁺ in comparison with all other cations

(Fig. 2). The ordering of cation concentrations was similar to those observed previously in Dongling Lake (Zhang, 1987) and some of the Nanyishan Oilfield brines (Tan *et al.*, 2011) in the western Qaidam Basin, which themselves differ from oceanic water and most salt lake brines throughout the Qaidam Basin.

Generally, there are three stages to brine evaporation and Ca²⁺ concentration changes during the different stages (Zhang, 1987). In the first stage, as carbonate and sulfate of Ca²⁺ have lower solubilities, they are precipitated in solid phase when the brines become concentrated, which limits the accumulation of Ca²⁺ in the brines, whilst K⁺ concentration is gradually increased. Secondly, with further evaporation, K⁺ concentration becomes higher than both Ca²⁺ and Cl⁻, i.e., it becomes saturated for chloride, which is precipitated as halite, resulting in increased Mg²⁺ concentration relative to Na⁺. Thirdly, because of the extremely high solubility of CaCl₂ during the process of brine evaporation, SO₄²⁻ loss results in large accumulations of Ca²⁺ in the latter stages of brine evolution. According to Valyashko's classification (Valyashko, 1965), all the surface brines belong to the chloride type, which shows they have reached the latter stages of evolution. Thus, it is reasonable that there are comparatively high concentrations of Ca²⁺ in the surface brines.

The intercrystalline brines were near pH-neutral with a pH range of 6.34–7.37 and an average of pH = 6.89. The cations in the intercrystalline brine decreased in the order of Na⁺ > Mg²⁺ > K⁺ > Ca²⁺ with average concentrations of 62853, 48784, 5188 mg, and 403 mg L⁻¹, respec-

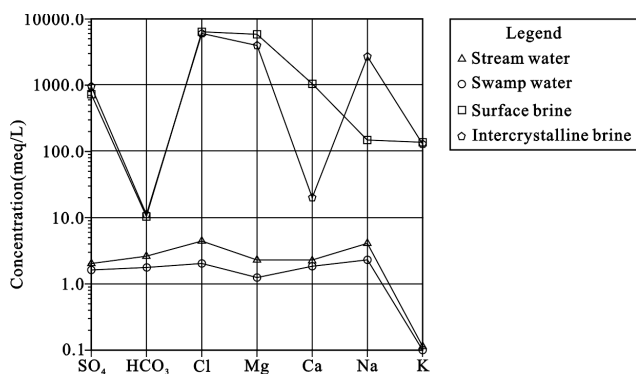


Fig. 2. Schoeller diagram of the median concentrations of anions and cations of stream water, swamp water, surface brine, and intercrystalline brine in the Gasikule salt lake.

tively. The anions decreased in the order of $\text{Cl}^- > \text{SO}_4^{2-} > \text{HCO}_3^-$ with average concentrations of 218652, 45578, and 676 mg L^{-1} , respectively. The average concentration of TDS in the intercrystalline brine was 381.80 g L^{-1} with a range of 279.99–554.19 g L^{-1} (Fig. 2). Compared with the surface brines, the intercrystalline brines were similar in K^+ , enriched in Na^+ , and depleted in Ca^{2+} and Mg^{2+} , possibly because of brine evolutionary processes. For example, as carbonate and sulfate of Ca^{2+} are less soluble and precipitate in the solid phase as the brines become concentrated, the accumulation of Ca^{2+} becomes limited, which results in the gradual increase of K^+ . As the brine becomes even more concentrated, K^+ increases to concentrations higher than Ca^{2+} , and Cl^- becomes saturated and is precipitated as halite. Concentrations of Mg^{2+} exceed those of Na^+ (Zhang, 1987). Because of intense weathering of underlying bedrocks, Na^+ concentrations in the intercrystalline brines are typically higher than observed in the surface brines (Fontes and Matray, 1993a, 1993b). The considerable enrichment of K^+ in the intercrystalline brines might be due to K^+ accumulation processes that operate in “dry salt lakes,” as defined by Zhang *et al.* (1993). Furthermore, the K^+ accumulation process can be divided into two stages: (1) open system in surface brines and (2) “dry salt lakes”. The two stages are connected but have their own distinct features. In the open system, K^+ accumulation depends mainly on the recharge water and water lost by evaporation. This accumulation process progresses slowly and K^+ accumulation is small. The so-called “dry salt lake” stage consists of a surface dry salt crust underlain by salt. The various depths of the intercrystalline brine and silicate detrital deposits might reflect this. The water level of intercrystalline brines in “dry salt lakes” is close to the surface because of capillary action and evaporating pump suction. Intercrystalline brines enter into a stage of self-separation, which accelerates K^+ accumulation. Ac-

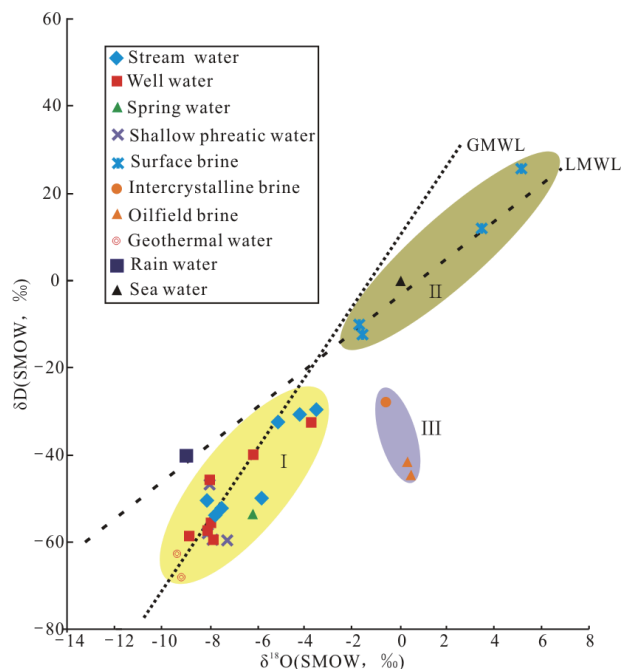


Fig. 3. Stable H and O isotopic compositions of Gasikule salt lake brines (surface and intercrystalline) and fresh water samples (stream, well, spring, shallow phreatic, geothermal, rain and sea). Modern worldwide precipitation produces the Global Meteoric Water Line (WMWL); local precipitation produces the Local Meteoric Water Line (LMWL) (Zhang *et al.*, 1999).

ording to Valyashko’s classification (Valyashko, 1965), all the intercrystalline brines belong to the magnesium sulfate subtype.

Hydrogen and oxygen isotope analysis

To analyze the origin and reason for the formation of the brine in the Gasikule salt lake, comparative analyses of stream water, well water, spring water, shallow phreatic water, surface brine, and intercrystalline brine in the Gasikule salt lake, and geothermal water and rain water in Da Qaidam were performed (Table 2). Some of the $\delta^{18}\text{O}$ and δD data of the stream water, groundwater, surface brine, intercrystalline brine, oilfield brine, and rain water in the Gasikule salt lake have been reported previously and discussed by Tan *et al.* (2009, 2011) and Zhang *et al.* (1993). These data are incorporated into this paper to further the discussion.

Hydrogen and oxygen isotope compositions vary between the different water types of the Gasikule salt lake (as shown in Fig. 3). However, the hydrogen and oxygen isotope compositions in the stream water, well water, and phreatic water are less variable than the surface brines. In the shallow phreatic water, well water, stream water, and surface brine, δD and $\delta^{18}\text{O}$ increase gradually, indicating that δD and $\delta^{18}\text{O}$ became heavier in the recharge,

Table 2. $\delta^{18}\text{O}$ and δD compositions of the water of the Gasikule salt lake

Sam. No.	Sam. site	Sam. type	$\delta\text{D}[\text{‰}]$	$\delta^{18}\text{O}[\text{‰}]$	Data source
1	Gasikule	Stream water	-50.1	-5.81	This study
2	Gasikule	Stream water	-29.4	-3.50	This study
3	Gasikule	Stream water	-30.5	-4.20	This study
4	Gasikule	Stream water	-32.3	-5.10	This study
5	Gasikule	Stream water	-54.5	-7.74	Tan <i>et al.</i> (2009)
6	Gasikule	Stream water	-52.8	-7.50	Tan <i>et al.</i> (2009)
7	Gasikule	Stream water	-51.0	-8.07	Tan <i>et al.</i> (2011)
8	Gasikule	Well water	-40.0	-6.10	This study
9	Gasikule	Well water	-32.6	-3.60	This study
10	Gasikule	Well water	-55.7	-7.86	Tan <i>et al.</i> (2009)
11	Gasikule	Well water	-58.8	-8.77	Tan <i>et al.</i> (2009)
12	Gasikule	Well water	-57.2	-8.00	Tan <i>et al.</i> (2009)
13	Gasikule	Well water	-45.9	-7.90	Tan <i>et al.</i> (2009)
14	Gasikule	Well water	-59.6	-7.78	Tan <i>et al.</i> (2009)
15	Gasikule	Spring water	-53.5	-6.07	Tan <i>et al.</i> (2011)
16	Gasikule	Shallow phreatic water	-59.8	-7.17	Tan <i>et al.</i> (2009)
17	Gasikule	Shallow phreatic water	-58.0	-8.00	Tan <i>et al.</i> (2011)
18	Gasikule	Shallow phreatic water	-46.8	-7.90	Tan <i>et al.</i> (2011)
19	Gasikule	Shallow phreatic water	-59.6	-7.78	Tan <i>et al.</i> (2011)
20	Gasikule	Surface brine	25.6	5.30	This study
21	Gasikule	Surface brine	-12.5	-1.44	Zhang <i>et al.</i> (1999)
22	Gasikule	Surface brine	-10.2	-1.59	Zhang <i>et al.</i> (1999)
24	Gasikule	Surface brine	11.9	3.60	This study
25	Gasikule	Interstalline brine	-28.2	-0.46	Zhang <i>et al.</i> (1999)
26	Gasikule	Oil-field brine	-41.6	0.47	Tan <i>et al.</i> (2011)
27	Gasikule	Oil-field brine	-44.6	0.62	Tan <i>et al.</i> (2012)
28	Dachaidan	Geothermal water	-68.0	-9.21	Zhang (1987)
29	Dachaidan	Geothermal water	-62.7	-9.39	Zhang (1987)
30	Dachaidan	Rain water	-40.6	-8.80	Zhang <i>et al.</i> (1999)
31		Sea water	0	0	Tan <i>et al.</i> (2011)

runoff, and discharge systems of the Gasikule salt lake.

Most δD and $\delta^{18}\text{O}$ of the Gasikule salt lake waters (Fig. 3) were on the global meteoric waters line ($\delta\text{D} = 8$, $\delta^{18}\text{O} = +10$ per mille) (Craig, 1961) or local meteoric waters line of the Qaidam Basin ($\delta\text{D} = 4.4$, $\delta^{18}\text{O} = -4.0$ per mille) (Zhang, 1987). Moreover, δD and $\delta^{18}\text{O}$ of all waters can be divided into three groups:

Group I: stream, well, spring, ground, and geothermal waters. In this group, δD and $\delta^{18}\text{O}$ values are the least variable and concentrate close to the global meteoric water line. Spring water, groundwater, geothermal water, and some stream and well waters have lower δD values than meteoric waters, indicating that they originate from melting snow or ice. Other stream and well waters have higher δD values than meteoric waters, which indicate that these represent meteoric waters that were not only confined to the rainfall within the Gasikule catchment, but also from large drainage basins.

Group II: surface brine with values close to those of

the local meteoric waters line for the Qaidam Basin, and away from the global meteoric waters line, implying that these waters originate mainly from local meteoric water. Their δD and $\delta^{18}\text{O}$ values are higher than Group I, which has undergone strong evaporation.

Group III: intercrystalline brine and oilfield brines have distinctive δD and $\delta^{18}\text{O}$ values that plot away from Groups I and II. The δD and $\delta^{18}\text{O}$ values in the water samples of this group might reflect either a strong water-rock exchange in an environment of high pressure and temperature, as has been observed previously (Banner *et al.*, 1989; Birkle *et al.*, 2009; Gregory *et al.*, 1989; Land, 1995), or the joint influence of evaporation in arid regions and a deep-water mixing effect (Zhang, 1987). The distinct values of $\delta^{18}\text{O}$ in the oilfield brines monitored by Tan *et al.* (2011) were attributed to the strong water-rock exchange in the observed environment of high pressure and temperature. However, at this site, intercrystalline brines are found within the salts and silicate clastic

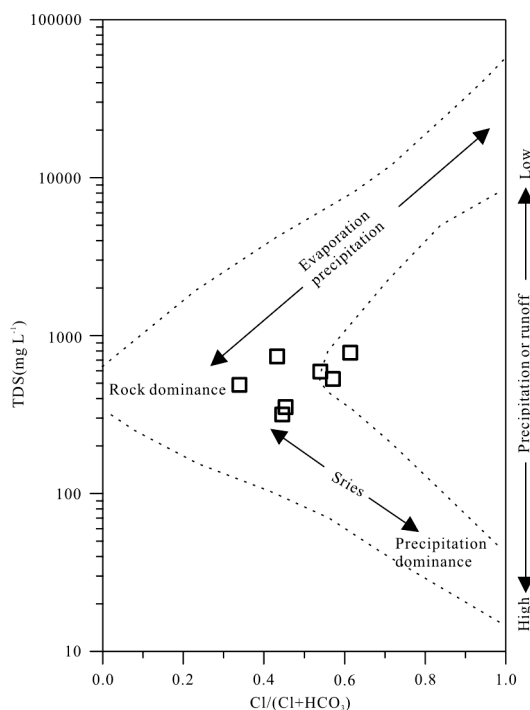


Fig. 4. Gibbs plot of the stream water at Gasikule salt lake. Dashed line represents the diagram range presented in Gibbs (1970); it shows some stream water samples lie outside the diagrammatic range.

sediments underlying the dry salt crust to depths of 70 m (Ye *et al.*, 2013, 2014) and thus, a high-pressure and high-temperature environment would not exist there. Therefore, the distinct intercrystalline brines are likely due to the joint effects of evaporation and deep-water mixing.

Origin of stream water samples

Gibbs (1970) suggested that a simple plot of TDS versus the weight ratio of $\text{Cl}/(\text{Cl}+\text{HCO}_3)$ could provide information on the relative importance of three major natural mechanisms controlling surface water chemistry: (a) atmospheric precipitation, (b) rock weathering, and (c) evaporation and fractional crystallization. From Gibbs plots of TDS versus the ion ratios $\text{Cl}/(\text{Cl}+\text{HCO}_3)$ of stream water in the Gasikule salt lake (shown here in Fig. 4), we know that most stream waters are characterized by relatively high Cl^- concentrations and fall within the rock-weathering domain. This suggests that their chemical compositions are controlled mainly by rock weathering and that evaporation crystallization is not significant.

Weathering of different parent rocks (e.g., carbonate, silicate) yields different combinations of dissolved cations and anions in solution (Stallard and Edmond, 1983). Furthermore, ratio graphs of ions can also help determine the sources of the material feeding the stream waters of

the Gasikule salt lake. All stream waters were plotted above the isoline of HCO_3^- vs. $\text{Cl}^- + \text{SO}_4^{2-}$ in Fig. 5a, indicating that the ion equivalent of $\text{Cl}^- + \text{SO}_4^{2-}$ is higher than HCO_3^- for all stream waters. As the $\text{Cl}^- + \text{SO}_4^{2-}$ is derived mainly from sulfate and chloride evaporite rock series, and the HCO_3^- is derived from carbonate rock series, it is clear that the eluviation of sulfate and chloride evaporite rock series contributes more than the carbonate rock series around the Gasikule salt lake. In evaporites, the main components of halite are Na^+ and Cl^- ; the main components of sylvite are K^+ and Cl^- , whereas SO_4^{2-} is distributed as sulfate type evaporites. $\text{Na}^+ + \text{K}^+$ are correlated with Cl^- and SO_4^{2-} (Figs. 5b and 5c), which implies that Na^+ and K^+ are derived mainly from dissolution of chloride-type evaporates (halite or sylvite) and sulfate-type evaporates (gypsum or mirabilite). $\text{Mg}^{2+} + \text{Ca}^{2+}$ are correlated with HCO_3^- (Fig. 5d) and therefore, Mg^{2+} and Ca^{2+} are sourced from carbonate rock series. In short, the chemical composition of the streams in the Gasikule salt lake is deeply influenced by the lithology and mineralogy of the surrounding mountains.

Origin of surface and intercrystalline brines

The brine $C_{\text{Na}}/C_{\text{Cl}}$ ratio (a molar concentration ratio) is an important indicator of brine evaporation-concentration and halite dissolution-leaching processes (Davis *et al.*, 1998; Sun *et al.*, 2002). Previous research using TDS- $(C_{\text{Na}}/C_{\text{Cl}})$ diagrams has shown that if a line's $C_{\text{Na}}/C_{\text{Cl}}$ ratio is equal to one, this represents a halite dissolution line (Anders *et al.*, 2014; Davis *et al.*, 1998; Fan *et al.*, 2007a, b; Han *et al.*, 2014; Llewellyn, 2014). If a brine sample plots near the halite dissolution line in the TDS- $(C_{\text{Na}}/C_{\text{Cl}})$ diagram, it indicates that the brine is strongly influenced by halite dissolution, and vice versa. The Gasikule salt lake has been a continental environment since the Paleogene and Neogene. If these brines were formed by halite dissolution and evaporation concentration, its hydrochemical evolutionary path during evaporation concentration should be similar to continental water. Therefore, we compared the evaporation curves of the Gasikule salt lake brine with the Qinghai Lake, which is the largest inland saltwater lake in China (Sun *et al.*, 2002).

With increasing salinity, the $C_{\text{Na}}/C_{\text{Cl}}$ ratio in the Qinghai Lake water initially shows no obvious change but then decreases sharply (Fig. 6). The $C_{\text{Na}}/C_{\text{Cl}}$ ratios in the intercrystalline brine of the Gasikule salt lake are scattered, but decrease with increasing TDS concentration. Therefore, the change trend of the $C_{\text{Na}}/C_{\text{Cl}}$ ratios in the intercrystalline brine becomes the same as the Qinghai Lake water evaporation line (Fig. 6). All intercrystalline brines plot below both the Qinghai Lake water evaporation line and the halite-dissolution line, as shown in Fig. 6. This indicates that evaporation and dissolution have joint control over the evolution of the intercrystalline

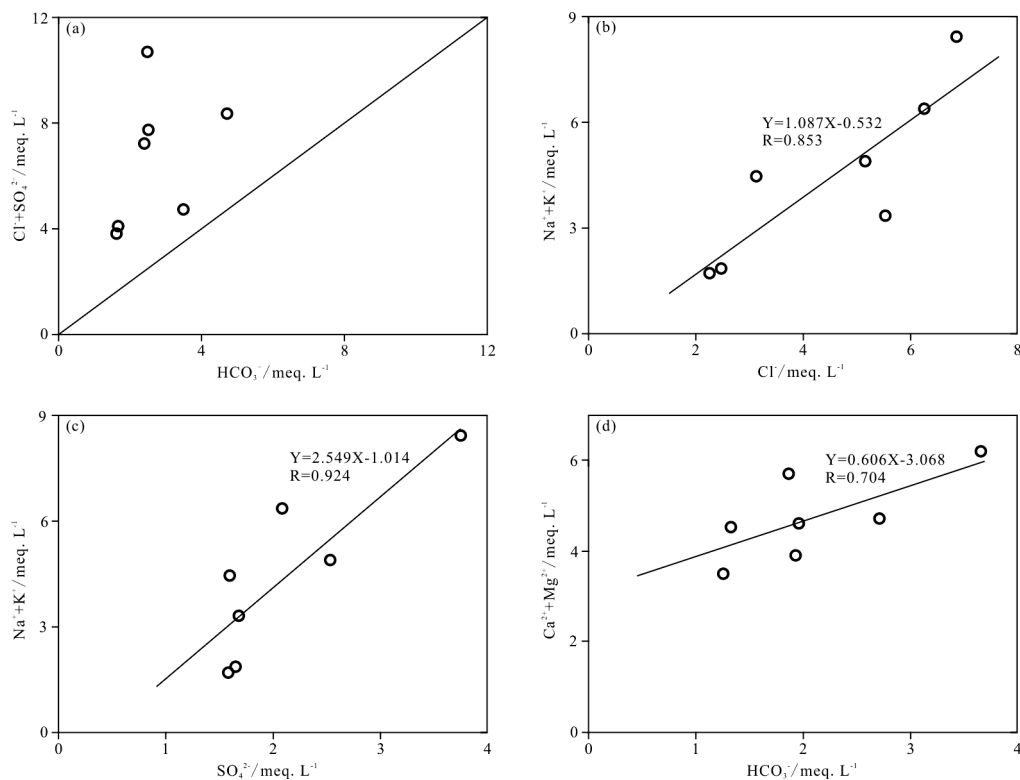


Fig. 5. Relationship graphs of ions in the stream water of the Gasikule salt lake basin: (a) plot of HCO_3^- vs. $(\text{Cl}^- + \text{SO}_4^{2-})$; (b) plot of Cl^- vs. $(\text{Na}^+ + \text{K}^+)$; (c) plot of SO_4^{2-} vs. $(\text{Na}^+ + \text{K}^+)$; (d) plot of HCO_3^- vs. $(\text{Ca}^{2+} + \text{Mg}^{2+})$.

brine; however, evaporation is the dominant factor.

For the surface brine, the $C_{\text{Na}}/C_{\text{Cl}}$ ratios plot in the center of Fig. 6 and far below the Qinghai Lake water evaporation line and halite dissolution line. This indicates that evaporation and halite dissolution do not have much impact on the surface brine. The Gasikule salt lake has a continental arid climate and the surface brine has had longer exposure to air; thus, it is more evaporated than the intercrystalline brine. The suggestion that the surface brine is not strongly influenced by evaporation, based on the $C_{\text{Na}}/C_{\text{Cl}}$ ratios, is because the recharge of Ca-Cl deep water causes higher Cl^- concentrations in brine and it lowers the $C_{\text{Na}}/C_{\text{Cl}}$ ratios. Thus, the recharge of Ca-Cl deep water dilutes the impact of the evaporation process on surface brine. In fact, the surface brine is deeply influenced by evaporation.

From the analysis of the $C_{\text{Na}}/C_{\text{Cl}}$ ratios, it is known that the dissolution of sulfate minerals such as gypsum, mirabilite, and celestite in the Pliocene strata of Youshashan and Shizigou is an important recharge source for the intercrystalline brine. However, halite dissolution is not the main recharge source of the surface brine. Furthermore, the process of formation of the intercrystalline and surface brines is strongly influenced by evaporation.

In the surface brine in the Gasikule salt lake, Ca

equivalent is higher than $\text{SO}_4 + \text{HCO}_3 + \text{CO}_3$ equivalent, which is quite different from the leachates of the parent rocks due to atmospheric weathering. The Ca-Cl zone in the triangular diagram of Ca- SO_4 - HCO_3 (Fig. 7) belongs to Ca-Cl type brine (Lowenstein *et al.*, 1989). According to the long-term research of Hardie (1984), Lowenstein *et al.* (1989) and Lowenstein and Risacher (2009), the chemical composition of Ca-Cl type brine reflects the high-temperature reaction of underground water with rocks or sediments. Following this, the Ca-Cl type brine reaches the earth's surface by convection-driven or terrain-driven cycling and it moves along faults in the form of spring water or oozing flow. In tectonically active closed basins, Ca-Cl type brine is probably an important recharge source (Lowenstein and Risacher, 2009) that reflects a deep source.

In the Qaidam Basin, magnetotelluric and seismic prospecting have demonstrated the presence of a high-conductivity and low-velocity zone in the middle part of the continental crust (20-km below the surface) (Xu, 1996; Zhang *et al.*, 2008). Following long-term research, Zheng *et al.* (1989) believed that lithium and boron in special salt lakes of the Qinghai-Tibet Plateau are likely sourced from geothermal water, which in turn, is sourced from magmatic evolution. Zhu *et al.* (1989) found active

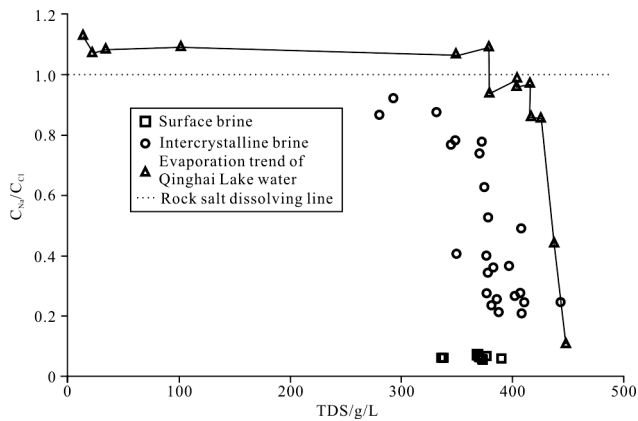


Fig. 6. Distribution of C_{Na}/C_{Cl} ratio and TDS of surface brine and intercrystalline brine in Gasikule salt lake.

volcanism on the collision margin of the East Kunlun Mountains and Altun Mountains, and the eruption zones where volcanoes erupted in 1952 and 1984 are close to the Gasikule salt lake. All this demonstrates the occurrence of strong and frequent magmatic hydrothermal activities around the Gasikule salt lake and thus, it is entirely possible that these activities could cause deep recharge to the Gasikule salt lake.

Effects of tectonic structure on surface brine

According to hydrogen-oxygen isotopes, the intercrystalline brines of the Gasikule salt lake are recharged to some extent by deep water. The surface brine is also thought to be recharged by deep water, based on the relative concentration of ions. However, it was necessary to establish how the deep water influenced these two natural brines and therefore, this was considered in relation to the location of faults within the region.

Faults distributed at the bottom of a salt lake introduce highly mineralized water at depth, which eventually mix with the salt lake water. Taking a linear fault zone as the center, the highly mineralized water brought into the lake will spread extensively and form an annular anomaly belt in which salinity changes gradually from high to low, which would be revealed as an obvious “water swell” phenomenon. By means of remote sensing data, the salinity information of the surface brine was inverted; thus, identifying the central area of the annular anomaly belt of mineralization in the water. This central area would be just the place where a buried fractured fault might exist.

Based on the above hypothesis, we used multi-source, multi-temporal remote sensing data of the Gasikule salt lake surface brine zone, obtained by the Enhanced Thematic Mapper (ETM) at 12:00 on November 10, 2000 and the Satellite Probatoire Pour l’Observation de la Terre 5

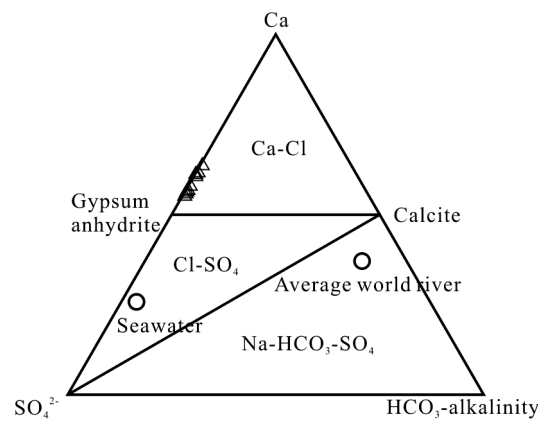


Fig. 7. Ternary $Ca-SO_4-HCO_3$ phase diagrams for surface brines in the Gasikule salt lake (modified from Lowenstein *et al.*, 1989).

(SPOT5) at 11:00 on January 16, 2010. The processing steps were as follows.

1. Radiation correction of ETM and SPOT5 remote sensing images by radiation regression analysis method, and geometric correction of above-mentioned images by polynomial correction method;
2. Accurate extraction of the water profile from ETM and SPOT5 data by the mid-inferred band method and unsupervised classification method (ISODATA method);
3. Extraction of the water spectrum information in different monitoring sites;
4. Establishment of the water mineralization remote sensing indices;
5. Extraction of the water mineralization remote sensing information;
6. Abnormal information analysis of the water mineralization.

After the above data processing steps were performed, ETM and SPOT5 qualitative estimation figures (Figs. 8a and 8b, respectively) of the salinity of the Gasikule salt lake surface brine were produced. They clearly show that the salinity is not evenly distributed, but shows an obvious linear “water swell” phenomenon with the center of the “water swell” located in the east of the surface brine. From the center of the “water swell” to the surroundings, an annular anomaly zone is formed where the salinity changes gradually from high to low. Generally, the surface brine nearer the recharging stream waters has lower salinity, and the surface brine with higher salinity is far from the stream waters and closer to the bank sides and surface salt deposits (Zhang, 1987). Furthermore, only the western streams and southern phreatic water recharge the surface brine, and there is no stream flow into the surface brine from the north or east of the lake. With only stream water recharge, the surface brine salinity would

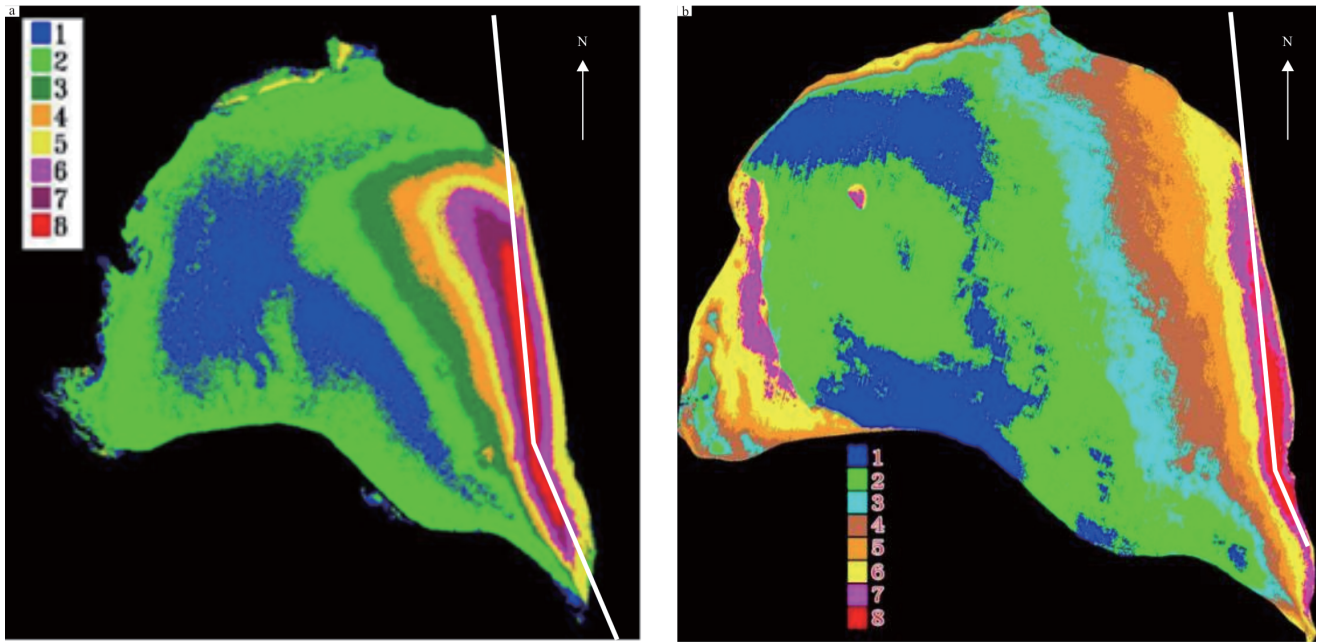


Fig. 8. Buried fault structure in the Gasikule salt lake surface brine area, based on inversion of ETM and SPOT5 remote sensing image data. Notice: (a) ETM qualitative estimation map of salt lake salinity; (b) SPOT5 qualitative estimation map of salt lake salinity. White line represents inferred buried fault structure.

increase gradually from the southwest to the northeast, and the “water swell” phenomenon would not occur. However, the obvious linear “water swell”, located in the east of the surface brine, indicates that apart from stream water recharge, there must be highly mineralized brine recharge to the surface brine from a fault buried in the lake bottom. The long axis position of the maximum value of the linear “water swell” annular anomaly (white line in Fig. 8) is just where the buried fault structure exists. Because of the long-term existence of the fault, highly mineralized water from the lake bottom forms the ring-shaped anomaly of salinity, which changes gradually from high to low values. It is thus confirmed that there exists a buried fault structure with an approximate N-S orientation in the east of the surface brine area.

SUMMARY

Stream waters around the Gasikule salt lake were weakly alkaline and HCO_3^- , Cl^- , and Na^+ were the most dominant ions. The concentrations of ions in the stream waters to the south were lower than in the west. Moreover, both the western and the southern streams flow into the brine area and Mg^{2+} , Cl^- , and K^+ concentrations increased sharply. Compared with the western tributaries, ion concentrations decreased in the southern tributaries, associated with a distinct host rock. Weathering of this host rock was the dominant driver of stream water chemi-

cal composition, whereas evaporation crystallization showed negligible impact. Cations in the surface brine and intercrystalline brine had contrasting size orders, while the anion size order for both types of brine was $\text{Cl}^- > \text{SO}_4^{2-} > \text{HCO}_3^-$. The average K^+ concentration in the surface brine and intercrystalline brine was 5304 and 5188 mg L^{-1} , respectively. This was identified as having great significance for resource exploration. The large enrichment of K^+ is another notable characteristic of the intercrystalline brine. The K^+ accumulation process in “dry salt lakes” likely explains the high K^+ concentration in the intercrystalline brine.

Hydrogen and oxygen isotope compositions of the different types of water of the Gasikule salt lake showed considerable differences; however, most δD and $\delta^{18}\text{O}$ were on the global meteoric waters line or local meteoric waters line of the Qaidam Basin. The combined influence of evaporation and deep-water doping could be the reason for the positive excursion in $\delta^{18}\text{O}$ of the intercrystalline brine.

Ions are uniquely sourced from their mother rock under the leaching scenarios around the Gasikule salt lake. Here, we identified that Na^+ and K^+ were mainly sourced from chloride-type evaporates (halite and sylvite) and sulfate-type evaporates (gypsum and mirabilite); Cl^- from chloride-type evaporates (halite and sylvite), SO_4^{2-} from sulfate-type evaporates (gypsum and mirabilite), and Mg^{2+} , Ca^{2+} , and HCO_3^- from carbonate rock series. Dis-

solution of sulfate minerals such as gypsum, mirabilite, and celestite in the Pliocene strata of Youshashan and Shizigou was the dominant source of recharge of the intercrystalline brine. However, halite dissolution was found not to be the main recharge source of the surface brine. Moreover, evaporation deeply influenced the process of formation of the intercrystalline and surface brines.

Two approximately N-S-trending “water swell” annular anomaly salinity belts were found in the east of the surface brine area, identified by inversion of ETM and SPOT5 remote sensing data. The absolute position of the two “water swell” annular anomaly salinity belts approximately corresponded with the location of buried fault structures.

We conclude therefore that three main sources recharge the surface and intercrystalline brines of the Gasikule salt lake:

(a) Stream water from rainfall and melting ice;

(b) Leaching of the Pliocene salt-bearing rock series to the north of the salt lake, which contains sulfate minerals such as gypsum, mirabilite, and celestite from the Pliocene strata of Youshashan and Shizigou;

(c) Ca-Cl type deep water from buried fault structures.

Although the surface and intercrystalline brines are in part recharged from deep water and the Pliocene salt-bearing rock series, respectively, evaporation of the influent stream waters is the main recharge source.

Acknowledgments—This research was supported by the China Geological Survey (Grant No. 1212011220791 and 12120115027801). We are grateful to vice Prof. J. X. Xu, Doctor T. W. Li, Doctor X. L. Yuan, and Doctor W. L. Miao from the Institute of Salt Lakes, Chinese Academy of Sciences, for their assistance in the field sampling. We also wish to express thanks to Prof. Y. Hu who helped improve the manuscript.

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