A Multi–fluid Constrain for the Forming of Potash Deposits in the Savannakhet Basin: Geochemical Evidence from Halite

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Abstract: The Khorat Plateau on the Indochina Terrane is known to have formed during the closure of the Tethys Ocean, although the origin of its potash mineral deposits is a topic of current debate. Data from a borehole on Savannakhet Basin is used in this study to re-define the evaporation processes of the study area. Geochemical analyses of halite from various borehole-derived evaporite strata have elucidated the fluid sources from which these ores formed. Measured δ^{11} B indicated that ore deposits formed primarily due to evaporation of seawater, although non-marine fluids affected the later stages of the evaporation process. Fluctuations in B and Br concentrations in carnallite- and sylvite-rich strata indicate the influence of fresh water. Boron concentration in carnallite unit indicated the influence of hydrothermal fluids. From the relative timings of these various fluid influxes, the evolution of these evaporates can be divided into four stages: (1) an initial marine evaporation at the beginning of the deposit's formation, where seawater (and minor fresh water) trapped on the uplifted Khorat Plateau produced sediments and salts with Br contents lower than those of normal marine-derived evaporites; (2) a transgression stage, where seawater recharged the basin; (3) a hydrothermal infiltration stage, which was coeval with the late Yanshan movement; and (4) a stage of fresh water supply, as recorded by fluctuations in B and Br contents, inferring intermittent fresh water influx into the basin. Thus, although evaporites on the Savannakhet Basin primarily formed via marine evaporation, they were also influenced to a significant degree by the addition of non-marine fresh water and hydrothermal fluids.

Key words: Savannakhet Basin, potash deposit, boron isotopes, geochemistry

1 Introduction

The geological characterization of potash evaporites has become a significant research topic in recent years, because they are of great interest to economic geology, the processes involved in ore deposit genesis, climatology, mineralogy, and sedimentology (Rahimpour-Bonab and Kalantarzadeh, 2005; Cendon et al., 2008; Warren, 2010; Tong et al., 2013; Zhang et al., 2013; Zhou Jiayun et al., 2015; Song Chunhui et al., 2017). Ancient potash deposits of various sizes documented from numerous localities worldwide are believed to represent marine sediments, marine–continental sediments, or continental sediments; however most giant potash deposits formed in marine environments (Liu Chenglin et al., 2010; Zhang Hua et al., 2014). Marine evaporite deposits typically form in response to changing tectonic and/or climatic conditions (Liu Chenglin et al., 2006; Warren, 2010); however, paleogeography and the potassium concentration of seawater also influence this process. The oldest marine potash deposits exposed on the continental crust today are believed to have formed in the Late Neoproterozoic (Liu Chenglin et al., 2015). Numerous continental potash deposits of variable sizes formed during the Cambrian, Permian, Jurassic, and Cretaceous periods, which together represent a prominent era of metallogenesis in the geological record.

The Khorat Plateau, located in central Indochina, contains one of the largest potash evaporites (Maha Sarakham Formation) in the world (Hite and Japakasetr,

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1979; Fan, 2000). Notably, these evaporites are interbedded with red clastic, and seemingly lack the carbonates commonly associated with evaporites. The Maha Sarakham Formation, which has a high escarpment along its western and southern edges, and a low central elevation, occurs within a continental basin that formed during the accretion and breakup of Eurasia (El Tabakh et al., 1999).

There are three analogous potash deposits (Sergipe Basin, Congo Basin and Khorat Basin) which formed during the Cretaceous (El Tabakh et al., 1999). All of the three formations contain the same minerals and lie above the marine and non-marine siliciclastic. However, the lithological origin of the Maha Sarakham is still debatable as there is no specific internal features. Early studies suggested that the Maha Sarakham Formation was of nonmarine (continental) origin, primarily due to the existence of associated red beds (Utha-Aroon, 1993), and the absence of marine-type carbonates and sulfates (Warren, 1999; Lv Zhengxiang et al., 2017). However, many recent studies have favored a marine origin (Tan Hongbing et al., 2010) owing to the ocean-like strontium and boron isotopic compositions in halite sampled from boreholes in the region. In addition to the changing opinions as to the source of the deposition environment, research has also transitioned from interpreting the evaporites as having formed from a single fluid source (marine or terrigenous) to having formed from multiple fluids of different provenances (marine, terrigenous, and hydrothermal) (Smith et al., 1996; El Tabakh et al., 1999). A three-stage model for describing an ore-forming fluid's evolution (based on a multiple-source theory) was introduced by Zhang Xiying et al. (2015), which incorporated the influence of brine on diagenetic processes. While extensive research (Zhang et al., 2013; Saminpanya et al., 2014; Zhang Hua et al., 2015) has confirmed the veracity of multiple-source theory in explaining the origin of potash deposits, considerable difficulty remains in discriminating between terrigenous-sourced and deep hydrothermal-sourced ore-forming fluids.

The study area for this research, Savannakhet Basin, is a sub-basin of Khorat Plateau. It is in central Laos, and belongs to the eastern edge of Khorat Plateau. As a major potash ore deposition, the Savannakhet Basin is an important area for studying the evolution processes of the Maha Sarakham Formation. In this study, we examine evaporite-bearing cores extracted from a borehole located at Nongbok Sag in the Savannakhet Basin, which contains a complete sequence through the Maha Sarakham Formation. We use boron isotopes and trace element concentrations in halite and associated sediments to elucidate the origin of parent fluids for potash deposits within the Khorat Plateau, and the processes by which the evaporites themselves formed.

2 Regional Geology

The Indochina Terrane is a Gondwana-derived crustal block (Thanh et al., 2016) that is composed of numerous micro terranes, metamorphic complexes, possible suture zones, and mylonitic fault zones (Lepvrier et al., 2004). The Khorat Plateau is (Fig. 1) in the center of the Indochina Terrane. During the Devonian/Carboniferous Era, a collision between the South China and Indochina microplates occurred along the Song Ma Suture. The Sibumasu terrane kept drifting northwards during the upper Permian, and collided with Indochina in the middle Triassic (Wakita and Metcalfe, 2005; Metcalfe, 2011). The two sutures, i.e. Nan–uttaradit suture and Song Ma suture, created by these collisions built the western and northern boundaries of the Indochina Terrane (Thanh et al., 2016).

The subsequent Late Triassic tectonic relaxation (extension) created half-graben basins (Hite and Japakasetr, 1979; El Tabakh et al., 1999) and caused the study area to become inundated by seawater. Later tectonic activity (the Yanshan movement) caused the Sibumasu Block to coalesce with some small terranes (Zhang Haifeng et al., 2012), while it rotated clockwise and moved northwards relative to the Indochina Terrane



Fig. 1. Location and general tectonic features and the study region (after Zhang Xiying et al., 2015).

SIT, Sibumasu Terrane; ISZ, Inthanaon Suture Zone; SUT, Sukhothai Terrane; ICT, Indochina Terrane; SDT, Song Da Terrane; SCT, South China Terrane.

(Charusiri et al., 2006). These motions resulted in the uplift of the Khorat sediments and the formation of the NW–SE-trending Phu Phan anticlinorium, which subsequently divided the Khorat Plateau into the southern Khorat Basin and the northern Sakon Nakhon Basin (El Tabakh et al., 1999).

The Savannakhet Basin, the focus of this study, is located east of the city of Thakhek, Laos, and lies within the southeastern part of the Sakon Nakhon Basin. In this area, the Jurassic, Creataceous and Neocene-Quaternary outcropped. The Jurassic sediments are composed of sandstone, mudstone and dolomitic limestone. The Creataceous characterized by lots of potash, gypsumanhydrite, reddish siltstone and mudstone bearing a little of dolomite rock. The Neocene-Quaternary sediments are composed of unconsolidated gravel, sand, silt and clay (Fig. 2). The Cretaceous evaporite sequence of Savannkhet Basin, which is called the Maha Sarakham Formation in Thailand and the Nong Bok Formation in Laos, lies above a thick, non-marine sequence of Mesozoic age within the Khorat Plateau. Constituent ore deposits have a mean thickness of 21.52 m, but show a significant range from 2.06 m to 71.98 m (variation coefficient=40.39%). The main stratum within the Nong Bok Formation is comprised of an evaporite (gypsum and halite) sequence that is interbedded with unconsolidated, fluvial, reddish and greenish silty clay. Borehole data show that this layer ranges in thickness between 500 m and 550 m.

Three depositional cycles have been documented for the Khorat Basin evaporites, with most of the sylvine present in the region belonging to the first cycle, although deposits belonging to the third cycle do not occur at the edge of the Sakon Nakhon Basin. In general, Sakon Nakhon Basin deposits are thinner than their Khorat Basin equivalents, which have been attributed to shrinkage of the former during sedimentation. As the region investigated in this work was located at the northeastern edge of the Khorat Plateau, only cores containing first– and second–cycle deposits could be extracted from most boreholes. Evaporite deposits preserved within the Khorat Basin are exposed at high elevations and generally thin along its eastern and northern margins, and exposed at low elevations and thick along its western and southern edges (Lu Yaozu and Duan Jianhua, 2012).

3 Stratigraphy Patterns of the Borehole– derived Core

The study area contained 66 boreholes; however, only eight penetrated the salt formation, which has been divided into three depositional members (lower, middle, and upper) based on stratigraphic sequencing, sedimentation cycles, and lithologic characteristics. These three members are equivalent to the first-, second-, and third-cycle divisions described above. Each member contains two sublayers: a salt layer and a mud layer. The borehole ZK309, one of the eight boreholes to penetrate the salt formation, has a depth of 309 m. ZK309 contained the lower member with a thickness of 256.7 m and the middle member with a thickness of 33.6m. The lithological characteristics of these memebers are described in detail below.



Fig. 2. Sketch geological map of the study area.

3.1 The first-cycle/lower member (L)

The first-cycle/lower member is the primary ore bed within the study area, and was well preserved in both basins. It comprises a basal anhydrite unit situated above a quartz-rich sand layer, and is readily identifiable in all cores. This stratum contains two distinct sublayers: a lower salt horizon (L_s) and a lower clay horizon (L_c) , and the thicknesses of the two sublayers in ZK309 is 182.3m and 74.4m. The former consists of granular layered and bluish halite, with minor interbedded gypsum, whereas reddish, gray, green-gray, and/or amaranthine mudstones form the major component of the latter. The minerals preserved at different depths within layer L_S were used as a basis for further subdivision, as described below.

3.1.1 The halite L₁ unit

The halite L_1 unit is the base of L_s , and is characterized by an evaporite layer that occurs throughout the basin, which comprises 97% halite, and 3% gypsum and anhydrite. As this horizon reaches a maximum depth of 390 m, only a small number of cores reached the bottom of this unit.

3.1.2 The carnallite L₂ unit

A horizon comprised mostly of layered carnallite (L_2) occurring immediately above halite layer L1. This L2 unit occurs continuously throughout the Basin and provides the largest volumetric contribution of potassium to the entire ore body. This layered unit grades upwards from its base into a massive and poorly bedded carnallite with a granular to sub-rounded texture (El Tabakh et al., 1999).

3.1.3 The sylvinite L₃ unit

The existence of sylvinite deposits throughout the study area was determined from abundant borehole data (El Tabakh et al., 1999). This L_3 unit is laterally discontinuous, thinner than the underlying carnallite layer L₂, and contains several sylvinite phases intermixed with blue-colored halite.

3.1.4 The halite L₄ unit

This halite-rich unit is located at the top of the firstcycle horizon, and comprises off-white halite intermixed with minor gypsum. This L_4 layer has a thickness of 14.8 m, notably thinner than the halite-rich L_1 unit, and is regarded as being transitional from the first-cycle/lower member to the second-cycle/middle member.

3.2 The second-cycle/middle member (M)

The second-cycle/middle member occurred in each borehole throughout the study area, and was divided into two sublayers: a middle salt layer (M_s) and a middle clay

layer (M_c). The thicknesses of the two sublayers are 1.7m and 31.9m, respectively. This middle clay layer contains the same lithological components as the lower clay layer (L_c) ; however, M_s differs from L_s because it is predominantly composed of anhydrite mixed with minor red- and gray-colored mudstones.

Stratum L_s extracted from borehole ZK309 was analyzed to characterize changes in elemental concentrations with depth. Boron isotopes were given particular attention in this work, as these are important criteria that can be used to determine the origin of evaporite deposits. The lithostratigraphy of the core obtained from borehole ZK309 is shown in Fig. 3.

4 Materials and Methods

4.1 Sample collection

Sixty-eight samples were collected from within layer L_S at a sampling interval of 2 m. Eight representative samples from this set were analyzed for their boron isotopic composition. Pre-treatment of these eight samples comprised several steps. Firstly, single halite crystals were separated from their horizons, and then these were cleaned of contaminants that may have adhered to their surfaces. Finally, five-gram masses of individual halite crystals with needle-like or bladed morphologies were then collected for isotopic analysis.

4.2 Analyses of boron isotopes

Boron isotopes were measured at the Qinghai Institute of Salt Lakes, Chinese Academy of Sciences (Table 1). Determination of ${}^{11}B/{}^{10}B$ ratios ($\delta^{11}B$) required the samples to be purified prior to analysis; as such, halite was dissolved in solution and then diluted with low-boron water (pH=6-7). These diluted solutions were passed through an Amberlite resin column at a flow rate of 2.5 ml/min. Boron was then eluted from the resin using 12 ml of 0.1 mol/L HCl. The eluent was evaporated and dried at 60°C, passed through another Amberlite resin column to remove unwanted ions, and then further purified using a low-boron eluent that contained equimolar amounts of mannitol and cesium carbonate (Cs₂CO₃). Finally, the diluted eluents were evaporated and dried at 60°C, and then prepared for isotopic analysis.

All ${}^{11}\text{B}/{}^{10}\text{B}$ ratios were determined using a triton thermal ionization mass spectrometer (Thermo Fisher Company). Our analyses showed a standard deviation of less than 0.2‰ (2σ), as determined by coeval analyses of NIST SRM-951 replicates. Isotope ratios are reported here as a permil deviation (δ^{11} B) in the 11 B/ 10 B ratios relative to those of the standard NBS SRM 951:

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System	Series	Formation	Members	Layers		Layers		Lithological column	Depth (m)	Thickness (m)	Lithology
Neogen-Quarternary						5.0		5.0	gravel, siltstone and mudstone		
	Oligocene	Nong Bok Formation	Middle Member	M _c			32.4	27.4	mudstone		
	ongotene						36.9	4.5	mudstone		
	Eocene						37.4	0.5	anhydrite		
				M _s		ΤΓΓΙΠΙΠΓ	<u>38.4</u> 38.6	1.0	mudstone		
			Lower Member	La		=:: =: =:		0.2	anhydrite		
	Paleocene						100.7	62.1	sandy mudstone		
Palaeogene							113.0	12.3	sandy mudstone		
					L_4	+++	127.8	14.8	halite and gypsum		
					L ₃		134.6	6.8	sylvine		
					L ₂		152.0	17.4	carnallite		
				L _s	L,		291.7	139.7	halite with little gypsum and anhydrite		
							295.3	3.6	basic anhydrite		
Upper Cretaceous							308.1	12.8	quartz sand		

Fig. 3. Lithostratigraphy of core obtained from borehole ZK309.

Table 1 Boron isotopic composition of halite and borate

Sample numbers	Depths (m)	B ¹¹ /B ¹⁰	δ ¹¹ B (‰)	References
Halite of SNB				
ZK309-140	210	4.1303	18.17	
ZK309-161	236	4.1634	26.32	
ZK309-185	260	4.1728	28.65	
ZK309-187	262	4.1702	27.99	
ZK309-205	280	4.1618	25.94	
ZK309-215	290	4.1590	25.24	
ZK309-218	292	4.1516	23.42	
ZK309-220	293	4.1523	23.58	
Borates of SNB				
FJ-01		4.1902	32.94	
FJ-02		4.1430	21.3	
FJ-03		4.1761	29.45	
FJ-04		4.1788	30.13	
FJ-05		4.1697	27.87	
FJ-06		4.1731	28.71	
FJ-07		4.1812	30.72	Zhang et
FJ-08		4.1844	31.49	al., 2013
FJ-09		4.1848	31.59	
FJ-10		4.1859	31.88	
FJ-11		4.1693	27.78	
FJ-12		4.1821	30.94	
FJ-13		4.1817	30.84	
FJ-14		4.1811	30.68	

$$\delta^{11}B(\%) = \left[\left(B^{11}/B^{10} \right)_{\text{sample}} / \left(B^{11}/B^{10} \right)_{\text{standard}} - 1 \right] \times 1000$$

The mean of the absolute ${}^{11}\text{B}/{}^{10}\text{B}$ ratios of NIST SRM– 951 replicates analyzed alongside the samples was 4.05262 ± 0.00077 (2σ =0.02%, *n*=9). The trace elements data in table 2 is taken from Zhang Xiying et al. (2015).

5 Results

5.1 Boron isotope composition

Natural boron occurs as either tetrahedral complexes (i.e., B(OH)₃) or trigonal complexes (i.e., B(OH)₄-)(Barth, 1993). Differences in their reduced partition function ratios and relative masses results in the two stable isotopes ¹¹B and ¹⁰B showing significant isotopic fractionation (Palmer and Swihart, 1996), leading to large variations in natural $\delta^{11}B$ (Fig. 4). The values of $\delta^{11}B$ in modern seawater, continental rainfall, and hydrothermal fluids range from 37.7‰ to 39.61‰, -5‰ to 17‰, and -2.6‰ to 36.8%, respectively. Furthermore, there is a large difference of the δ^{11} B values between marine minerals and non-marine minerals; thus, boron isotopes can readily distinguish marine-derived evaporite deposits from nonmarine evaporite deposits (Vengosh et al., 1991; Xiao et al., 1992; Smith et al., 1996; Liu et al., 2000; Kloppmann et al., 2001).

All eight of the halite samples used in this study originated from the halite L_1 unit and exhibited $\delta^{11}B$ values of 18.17‰–28.65‰, which are different from those

Vol. 92 No. 2

ACTA GEOLOGICA SINICA (English Edition) http://www.geojournals.cn/dzxben/ch/index.aspx

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ample numbers	Depths (m)	Host rocks	K(ppm)	Mg(ppm)	Ca(ppm)	SO4 ² (ppm)	B ₂ O ₃ (ppm)	Br⁻(ppm
ZK309-220	293.00	anhydrite	161.89	64.85	1460.16	3041.81	5.32	95.00
ZK309–218	292.00	anhydrite	146.28	259.26	7878.69	16656.26	15.74	90.00
ZK309–216	291.00	halite	167.51	8.82	249.26	490.92	4.32	22.00
ZK309-215	290.00	halite	156.31	18.66	8780.68	20285.21	3.88	
ZK309-213	288.00	halite	156.02	9.06	3043.06	6857.23	3.55	
ZK309-211	286.00	halite	140.88	66.76	4166.85	8896.34	2.02	10.00
ZK309-209	284.00	halite	128.69	80.42	8736 75	19614 26	1.89	10.00
ZK309_207	282.00	halite	93.58	64 55	5167.67	11403 70	3.88	10.00
ZK309_205	280.00	halite	256.53	26.19	7614 37	18838 32	20.32	
ZK309_203	278.00	halite	285.43	18 19	5490.89	12004.05	20.32	35.00
ZK309_201	276.00	halite	205.45	32.58	3942.68	9720 57	20.75	55.00
ZK309-201 ZK300 100	270.00	halite	248.32	9 60	3026 32	720.57	20.80	
ZK309-199 ZK300-107	274.00	halite	255.69	9.09	2010 72	1202.32	22.75	56.00
ZK309-197	272.00	halita	232.02	19.29	2010.75	4304.32	21.44	20.00
ZK309-193	270.00	halita	243.17	12.99	1402.02	2579.47	23.39	27.00
ZK309-193	268.00	halite	246.22	17.22	1493.92	35/8.4/	15.67	37.00
ZK309–191	266.00	halite	253.37	7.32	591.73	1393.65	21.44	35.00
ZK309–189	264.00	halite	291.99	30.73	2298.63	5414.19	22.95	50.00
ZK309–187	262.00	halite	332.18	87.08	2522.55	5253.30	23.69	91.00
ZK309–185	260.00	halite	279.01	33.12	8197.02	18764.02	17.57	76.00
ZK309–183	258.00	halite	279.70	9.82	193.61	587.69	19.31	104.00
ZK309–181	256.00	halite	280.98	20.44	3189.52	7604.01	21.94	67.00
ZK309–179	254.00	halite	246.24	36.93	4129.10	9335.15	12.87	85.00
ZK309-177	252.00	halite	324.21	11.99	1778.70	4015.94	22.76	116.00
ZK309-175	250.00	halite	336.04	34.68	145.23	343.68	25.28	89.00
ZK309-173	248.00	halite	340.13	104.28	2650.48	6289.33	18.80	92.00
ZK309-171	246.00	halite	342.13	16.06	770.82	1585.26	20.01	57.00
ZK309–169	244.00	halite	313.04	94.18	8010.69	17917.73	33.08	71.00
ZK309–167	242.00	halite	349.42	52.43	2067.82	4304.01	31.61	124.00
ZK309-165	240.00	halite	342.45	36.18	4531.01	10389.02	39.35	85.00
ZK309-163	238.00	halite	390.69	19.57	948 74	1864 37	36 31	116.00
ZK309–161	236.00	halite	347 59	55 37	2151 54	4317 19	40.72	137.00
ZK300_150	234.00	halite	346.61	17.99	4630.85	10623 78	30.11	91.00
ZK300–157	232.00	halite	403 50	20.60	1370 //	2083 71	71.26	154.00
ZK30)-157	232.00	halita	412.24	11.25	274.62	520.59	15.20	211.00
ZK309-155 ZK300_153	230.00	halite	376.01	71.18	274.02	1666.00	43.87	107.00
ZK309-155 ZK200 151	228.00	halite	255.21	19.56	024.24	2171.05	32.39	125.00
ZK309-131 ZK200 140	220.00	halita	220.24	18.30	934.70	21/1.95	24.57	133.00
ZK309-149	224.00	halite	370.74	47.03	204.12	14510.25	32.00	122.00
ZK309-147	222.00	halite	394.57	37.70	394.13	925.25	33.02	140.00
ZK309–145	220.00	halite	381.16	/.18	21.95		27.59	127.00
ZK309–144	218.00	halite	381.34	8.50	53.42	82.24	32.40	129.00
ZK309–143	216.00	halite	368.86	10.55	116.34	226.21	31.39	134.00
ZK309–142	214.00	halite	382.68	9.77	1182.42	2550.03	35.30	140.00
ZK309–141	212.00	halite	335.72	48.90	6954.53	15926.56	33.34	154.00
ZK309–140	210.00	halite	324.75	26.20	4617.26	10263.28	36.26	172.00
ZK309–139	208.00	halite	321.66	11.83	1015.05	2296.03	43.31	159.00
ZK309–138	206.00	halite	317.97	9.02	1731.29	4042.92	39.93	160.00
ZK309–137	204.00	halite	263.09	4.79	258.36	536.75	26.68	158.00
ZK309–136	202.00	halite	181.98	10.54	480.39	1047.67	37.77	205.00
ZK309–135	200.00	halite	387.20	53.77	1031.49	1924.11		108.00
ZK309-134	198.00	halite	386.61	8.56	622.99	1357.03		109.00
ZK309–133	196.00	halite	357.43	1.93	209.47	388.24		116.00
ZK309-132	194.00	halite	398.77	11.77	363.57	745.38		111.00
ZK309–131	192.00	halite	380.04	16.49	1044.67	2238.64		125.00
ZK309-130	190.00	halite	379.76	6.70	912.76	1947.97		106.00
ZK309-129	188.00	halite	382.80	21.32	3025.12	6495.73		106.00
ZK309–128	186.00	halite	403.13	6.77	1014.10	2185.55	59.91	126.00
ZK309-127	184.00	halite	411 37	9.38	180.66	358.09		120.00
ZK309–126	182.00	halite	405 36	5.28	341.82	709.65		138.00
ZK309_125	180.00	halite	424.03	4 52	413.77	837.80		138.00
ZK309_125	178.00	halite	396.07	4 41	610.23	1253 72		135.00
ZK309_124	176.00	halite	30/12	0.83	228.46	431 78		165.00
ZK300-125 ZK300_122	174.00	halita	381 55	8.62	220.40	602 51		103.00
ZK307-122 ZK200 121	172.00	halita	JOI.JJ 405.07	0.05	12.22	271 42		101.00
ZK309-121 ZK300-120	1/2.00	halle	403.07	2.01	120.91	2/1.03		179.00
ZK309-120	1/0.00	halite	413.85	/.25	595.25	1249.62		168.00
ZK309-119	168.00	halite	413.20	12.01	1120.57	2419.40		166.00
ZK309–118	166.00	halite	392.69	1.09	271.38	587.87		197.00
ZK309-117	164.00	halite	400.25	14.03	271.02	553.83		173.00
ZK309-116	162.00	halite	389.58	11.64	1191.87	2594.66		187.00
ZK309–115	160.00	halite	467.78	7.95	439.71	944.91		164.00
ZK309-114	159.00	halite	484.73	6.54	398.26	724.92		129.00
ZK309-113	158.00	halite	652.65	11.40	245.09	494.29		154.00
ZK309-112	157.00	halite	1192.59	7.71	632.44	1378.80		134.00
ZK309-111	156.00	halite	1874 97	7.99	950.24	2045.11		132.00

Apr. 2018

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Vol. 92 No. 2

Sample numbers	Depths (m)	Host rocks	K(ppm)	Mg(ppm)	Ca(ppm)	SO4 ² (ppm)	B ₂ O ₃ (ppm)	Br⁻(ppm
ZK309-110	155.00	halite	1594.42	18.81	596.18	1280.01	48.92	158.00
ZK309-109	154.00	halite	5318.91	275.03	1631.64	3775.03		217.00
ZK309-108	153.00	halite	4759.27	301.52	103.77	148.49		222.00
ZK309-107	152.00	halite	4914.09	173.91	557.31	1257.98		214.00
ZK309-106	151.00	carnallite	6581.76	1210.86	2992.05	7071.32		240.00
ZK309-105	150.00	carnallite	3936.04	190.27	1042.43	2349.43		205.00
ZK309-104	149.00	carnallite	4391.88	378.62	616.18	1380.98		164.00
ZK309-103	148.00	carnallite	5170.16	731.32	729.72	1753.39		150.00
ZK309-102	147.00	carnallite	4389.68	339.53	1598.58	4118.15	18.88	137.00
ZK309-101	146.00	carnallite	8708.84	604.48	1817.28	4511.02	42.30	198.00
ZK309-100	145.00	carnallite	11468.68	798.85	1251.56	2753.93	79.86	179.00
ZK309-99	144.00	carnallite	7886.38	792.98	984.50	2483.25	206.52	204.00
ZK309-98	143.00	carnallite	6053.68	251.86	383.08	783.18	42.26	87.00
ZK309-97	142.00	carnallite	12289.02	1472.49	1680.36	3833.71		109.00
ZK309-96	141.00	carnallite	6982.09	1374.04	1453.62	3298.93	867.07	165.00
ZK309-95	140.00	carnallite	6373.59	943.68	1310.87	3160.35	651.38	102.00
ZK309-94	139.00	carnallite	8440.28	1394.37	1695.46	3626.60	801.46	51.00
ZK309-93	138.00	carnallite	14557.98	2193.94	2325.55	5722.62		95.00
ZK309-92	137.00	carnallite	8921.40	1233.45	960.33	2209.27	537.69	125.00
ZK309-91	136.00	carnallite	11532.42	1201.50	1809.92	4442.53	583.13	94.00
ZK309-90	135.00	carnallite	6518.65	336.11	1120.46	2463.20	28.20	253.00
ZK309-89	134.00	sylvine	35654.22	855.92	1309.67	2825.87	535.62	97.00
ZK309-88	133.00	sylvine	71947.92	795.38	1116.68	2703.27	477.16	64.00
ZK309-87	132.00	sylvine	35119.34	842.37	670.19	1790.15	248.51	105.00
ZK309-86	131.00	sylvine	82382.97	824.08	1194.72	3229.58	595.00	51.00
ZK309-85	130.00	sylvine	24233.51	869.51	831.37	2077.51	626.20	137.00
ZK309-84	129.00	sylvine	39852.10	288.47	1017.90	2366.39	94.57	277.00
ZK309-83	128.00	sylvine	40591.00	360.78	897.50	2215.63	167.66	133.00
Minimum			93.58	1.09	21.95	82.24	1.89	10.00
Maximum			82382.97	2193.94	8780.68	20285.21	867.07	277.00
Averages			5020.33	228.17	1919.47	4379.16	116.63	127.39
		$\delta^{''}$ B curve of seawater	-7~42.7	37.7~39.61 6	- Seawate - River wa	r ater		
z			-2.6~36.	.8	-Hvdroth	ermal fluid		
	щ	-	-(5.1~59	Saline la	ike water		



Fig. 4. Measured δ^{11} B values for different solutions (after Li et al., 2015). SNB, Sakon Nakhon Basin.

of terrigenous halite (Fig. 4). This consistent variation in ${}^{11}\text{B}/{}^{10}\text{B}$ provides strong evidence for a stable depositional environment during the formation of the evaporate sequence in the entire halite L₁ sequence. Boron within halite occurs as fluid inclusions (Fan et al., 2015);

therefore, boron's isotopic ratios are almost the same as the δ^{11} B values of paleo-brines from which the halite precipitated. As Cretaceous seawater had a δ^{11} B value of 40‰ (Fig. 4), which was slightly greater than the δ^{11} B value exhibited by halite in this study, we suggest that

761

early-stage mineralization in the study region was caused by the evaporation of brine (i.e., a seawater and fresh water mixture).

As the δ^{11} B value of halite is similar to that of seawater, it should increase during evaporation (Xiao et al., 2007; Paris et al., 2010). If the sedimentary sequence observed in borehole ZK309 is used as a proxy for evaporation rate, widely variable values for the evaporation curves of seawater (Fig. 5a) and δ^{11} B values (Fig. 5b) are seen. The calculated δ^{11} B value in the halite L₁ column increased with depth up to 260 m, after which it decreased. Zhang et al (2013), reported that the δ^{11} B value of borate (table 1, FJ14-FJ11) in Khorat Plateau had similar variations as compared with this study. Hence, the decreased $\delta^{11}B$ upper than 260 m is not an accidental phenomenon. The extent of boron isotopic fractionation is predominantly controlled by the pH and temperature of its solution (Pagani et al., 2005; Foster et al., 2013), and the lower δ^{11} B value indicated the enrichment of B¹⁰. However, because there was no significant change in the environment during the halite deposit (Zhang Hua et al., 2015a), the Boron fractionation influenced by the temperature can be ignored. Commonly, ¹¹B is enriched in $B(OH)_3$ and ¹⁰B is enriched in $B(OH)_4^-$ (Xiao and Wang, 2001). When the pH of the solution is higher than 10, B $(OH)_4^-$ will be enriched; in other words, the alkaline solution will have a lower δ^{11} B value. Consequently the decreased δ^{11} B value of the evaporites located above 260m can be explained by the increase in pH.

Notably, a sequence of dolomite limestone occurs in the



Fig. 5. Correlations between the δ^{11} B of seawater, its boron content, and the degree of evaporation.

(a), δ^{11} B values of residual evaporated seawater vs. the degree of evaporation: $a_1=0.969$ and $a_2=0.981$ (Vengosh et al., 1992); (b), variation of δ^{11} B in the profile of borehole ZK309; (c), variation of B concentration (ppm) with depth.

northeastern part of the study area (the dash area in Fig. 2). The altitude of the limestone area is about 231 m while the highest altitude of the Savannakhet Basin is 143 m (Kong Xun and Long Qing, 2015). Furthermore, dolomite reacts with CO2-rich water, and the HCO3⁻ created from this reaction acts to increase the pH of a fluid (Cai Jiexing, 1993); hence, we suggest that these evaporites with anomalous predicted source-fluid pH values formed due to an influx of fluids from the northeast. Furthermore, the B-O bond length of 0.137 nm for trigonal coordination is similar to the C-O bond length of 0.128 nm, and the charged $B(OH)_4^{-1}$ is more likely to be attracted to the crystal surface. Hence, the ¹⁰B is likely to be substituting for the CO_3^{2-} in carbonate (Hemming and Hanson, 1992;). Following the reaction between dolomite and water, the ¹⁰B adsorbed by the carbonate would relax back to the water and make the δ^{11} B value decrease.

Boron typically becomes enriched in solutions during evaporation, as it does not participate in significant ion– exchange reactions. As such, the B content of sediments deposited by this fluid increases. Boron concentrations and δ^{11} B values in borehole ZK309 showed a negative relationship above 260 m (Fig. 5), which indicates the influence of seawater influx. This result supports the possibility of a period of marine influence during the Maha Sarakham Formation, as proposed by Zhang Xiying et al. (2015) in their three–stage model. However, our findings disagree with Zhang Xiying et al. (2015) regarding the relative timing (i.e. depth in the sedimentary column) of the marine–derived fluid input.

5.2 Geochemical characteristics of salt rocks

Seawater contains various elements that can form a wide range of minerals during different stages of evaporation, and which may form via different geological processes. Therefore, element concentrations can be used to infer the depositional processes for different evaporite deposits. The elemental concentration data (i.e., trace elements in halite extracted from L_1 , L_2 , and L_3 units) shown in Table 2 were taken from Zhang Xiying et al. (2015b).

5.2.1 Major elements

The K and Mg contents of halite typically increase as the evaporation rate increase (Fig. 6), and these two elements in borehole ZK309 were concentrated in the lower salt layer. The average K contents in halite crystals from the L₁, L₂, and L₃ units were 0.06%, 0.8%, and 4.7%, respectively, and the average Mg contents were 0.0036%, 0.08%, and 0.07%, respectively. The carnallite deposit (L₂) exhibited fluctuating K contents that showed a large relative change over time. We interpret this pattern as the



Fig. 6. Profiles of trace element variation with depth within borehole ZK309 (after Zhang Xiying et al., 2015). (1), halite L_1 unit; (2), carnallite L_2 unit; (3), sylvine L_3 unit; (4), halite L_4 unit. I, II, III, and IV represent various stages of fluid/sediment input.

intermittent input of fresh water, with carnallite being alternately precipitated from brine and fresh water (Lu Yaozu et al., 2015).

5.2.2 Variation of Br in halite

Although natural bromide exists in various oxidation states, it cannot be adsorbed onto mineral surfaces and only participates in redox reactions at high temperatures (Charusiri et al., 2006). The concentrations of Br in solutions and sediments both increase as evaporation progresses, although Br ions only exchange with Cl ions from chloridate lattices during extreme evaporative conditions. As Br behaves in a predictable manner during the evaporation process, it can be reliably used to investigate the distributions, depositional environments, and origins of sylvine deposits.

The average Br content of halite from the lower salt layer in borehole ZK309 was 127.39 ppm, and exhibited a total range of 10–277 ppm. The Br content of halite formed during the initial stages of seawater evaporation is 65–75 ppm (Herrmann et al., 1973; Mccaffrey et al., 1987), though it must reach 320–400 ppm (El Tabakh et al., 1999) before sylvine can precipitate from solution. In this study, the Br content of halite in L₁ increased steadily throughout the stratum from 22 ppm (table2, point ZK309– 107) at its top. These values are lower than the Br content of halite recorded in other marine sediments (Timofeeff et al., 2006), which indicates that sedimentation and evaporite formation on the Savannakhet Basin was influenced by low–Br fresh water.

The lowest Br content recorded in L₁ halite was 10 ppm (table 2, points ZK309-211, 209), which is notably lower than the concentration expected to characterize the basal halite precipitated from seawater. However, the Br content of halite selected from the anhydrite is marginally higher than the initial Br value of halite (65-75ppm). Zhang Xiving et al. (2015) attributed such compositional discrepancies water-rock interaction to and recrystallization. However, an alternative scenario was presented by El Tabakh (1998), who used stratigraphy, petrology, and isotopic analysis to interpret that 'basal anhydrite' typically forms as an insoluble residue that is leached from a sequence of basal evaporites composed of intercalated halite and anhydrite. Notwithstanding, both of these interpretations indicate an event of fresh water supply. These interpretations also fit the variation of the δ^{11} B values described in Section 4.1. Cendon et al (2003) suggested that the partial dissolution/precipitation diagenetic processes in evaporation deposits could be evidenced by petrographic observations of dissolution textures. However, the similar dissolution textures only

occurred at the interface between anhydrite and the L_1 unit. As such, we agree with the fingdings of El Tabakh (1998) that only a part of basal halite and the whole basal anhydrite has been dissolved, and the halite of L_1 unit is the unaltered halites. The lower B content in this unit is thus attributed to an influx of fresh water at the beginning of evaporation, and fluctuating Br contents in the carnallite (L_2) and sylvine (L_3) units are thought to record voluminous (but intermittent) influxes of fresh water during their respective depositional periods.

The partitioning of Br between halite and halitesaturated brine is strongly controlled by variations in the chemistry of the host solution. Hence, there is a correlation between the Br contents of marine basal halite and the $(Mg^{2+}+SO_4^{2-})/(K^++Ca^{2+})$ molar ratio of seawater (Li et al., 2015). However, Fig. 7 shows that L_1 basl halite does not demonstrate this correlation. Instead, the average $(Mg^{2+}+SO_4^{2-})/(K^++Ca^{2+})$ molar ratio of L₁ halite was 0.62, which is similar to the value of 0.5 that characterizes hydrothermal fluids (Siemann, 2003). Due to the influence of a thermal event (Dai Shuang et al., 2016), the Cretaceous paleoseawater had higher concentrations of the Ca ion (Timofeeff et al., 2006). Our value for the $(Mg^{2+}+SO_4^{2-})/(K^++Ca^{2+})$ molar ratio is agreement with that reported by Siemann (2003), suggesting that L_1 unit is comprised of unaltered halite.

5.2.3 Variation of B in halite

Boron is a highly soluble element, and primarily exists on Earth in the hydrosphere and upper lithosphere. Typical B concentrations in seawater and the lithosphere are 4.5 ppm and 3–20 ppm (Eskenazy et al., 1994), respectively. As B has no natural redox chemistry (Gurenko and Kamenetsky, 2011), it occurs in sediments in three forms: incorporated in borate minerals, adsorbed by clay minerals, and as mineral–bearing evaporite deposits (Xiao et al., 1992). The B concentration of sediments typically increases according to the degree of evaporation, being <130 ppm during initial evaporation and reaching 200–



Fig. 7. Relationship between Br content in halite and the $(Mg^{2+}+SO_4^{2-})/(K^++Ca^{2+})$ molar ratio of seawater.

400 ppm in the final stages (Zhang Xiving et al., 2015). The B analyzed in this study is from the halite mineral, so most B exist as mineral-bearing evaporate deposits. The measured B content of L₁ halite in our samples was between 1.9 and 71 ppm (Fig. 6), which is similar to the values that characterize marine evaporation. Halite analyzed from the carnallite (L_2) and sylvine (L_3) layers exhibited major fluctuations in B contents, with concentrations that reached 867 ppm (average = 400 ppm). This indicates that the low-B evaporite-forming water was supplemented at some point by B-enriched water, since the maximum measured B content was much greater than that of marine halite (not higher than 400ppm). Fresh water contains only small amounts of B, while hydrothermal water has a high B content (Yuan et al., 2014); consequently, the Savannakhet Basin was likely influenced by hydrothermal activity during this evaporiteforming stage. The δ^{11} B values reported by Zhang (2013) were found to increase suddenly at two locations (table 1, FJ01 and FJ01). Because hydrothermal water commonly has a low pH (<6), and a high δ^{11} B value, the input of hydrothermal water could explain the variation of $\delta^{11}B$ values at these two points.

6 Discussions

6.1 Origin of the Maha Sarakham Formation

Isotopic and trace element analyses of halite from the ZK309 borehole have shown that the Maha Sarakham Formation within the Savannakhet Basin formed from multiple fluid sources. Element concentrations and $(Mg^{2+}+SO_4^{-2-})/(K^++Ca^{2+})$ molar ratios suggest that the study area was initially inundated by seawater, and then temporarily isolated from further influx due to closure of the Tethys Ocean and localized tectonic uplift. Subsequent influxes of terrestrial water and hydrothermal water led to the early products of evaporation in the basin region formed from three different fluids (seawater, terrestrial water, and hydrothermal water). The relatively minor amount of fresh water involved in this process was insufficient to affect the overall marine characteristics of the evaporite deposit. Upon sedimentary deposits having reached a thickness of 260 m, seawater re-entered the plateau region and caused a minor decrease in the $\delta^{11}B$ of halite. This interpretation matches the first of three evolutionary stages proposed by Zhang Xiying et al. (2015) for evolution of the Khorat Plateau evaporites (the marine input stage).

6.2 The influence of non-marine resources

Numerous investigations of Khorat Plateau sediments have interpreted a marine origin; however, although seawater was indeed the major source of the potash deposits, various non-marine fluid sources also played important roles in their formation. Our analyses have revealed that the formation of evaporites in the lower salt layer (L_S) of borehole ZK309 can be divided into four lithostratigraphic stages:

(1) Seawater-derived evaporite-forming stage (Fig. 6, line I). Variations in the concentrations of B, Br, K, and Mg in halite grains from this stage correlated with changes expected under formation due to seawater evaporation; however, the low Br content and δ^{11} B value exhibited by halite indicated the influence of continental water. Thus, seawater cannot have been the sole evaporite-forming fluid during this stage.

(2) Marine input stage (Fig. 6, line II). Closure of the Tethys Ocean precluded further inundation of the Khorat Plateau region by seawater. During the Late Cretaceous, when sedimentation reached a thickness of ≥ 260 m, the movement of the Indochina plates caused tectonic gaps to re-open and seawater was able to transgress the region. This late-stage influx of seawater is evidenced by the relationship between δ^{11} B and the B content of the halite deposit from this stage, and fluctuations in Ca and SO₄ contents.

(3) Hydrothermal fluid input stage (Fig. 6, line III). Initiation of hydrothermal fluid supply to the Savannakhet Basin region is recorded in the evaporite sequence by carnallite deposition. The halite of the L₃ unit exhibited a decreasing Br concentration and an increasing B concentration in the up-section. Variations in the elemental concentrations of halite with depth indicated that the potash deposit was affected by the addition of Benriched fresh water. The sudden increased in δ^{11} B value reported by Zhang (2013) also indicated hydrothermal fluid input. Boracite and hilgardites reported in the Vientiane Basin indicate the abnormally high B content of the host water (Zhang et al., 2013). Tachyhydrites and saddle dolomites documented within the Khorat Plateau by El Tabakh(1999; 2003), both of which form at high temperatures (>35°C; Huang Keke et al., 2016; Yang et al., 2016), provide evidence for a thermal event during the Cretaceous. Bo (Bo Ying et al., 2015; Bo et al., 2015a) suggested that the Yanjing-Lanping-Simao-northern Laos share geochemical affinities, and the geothermal activity occurred from the north to the south. There are also granite intrusions of an upper Cretaceous extent in the Khorat Plateau (El Tabakh et al., 1999) may indicate the influence of mantle. In this study, the hydrothermal input into the Savannakhet Basin is evidenced by carnallite deposition. However, the driver of these hydrothermal sources is still not fully understood.

(4) Terrestrial water input stage (Fig. 6, line IV). The Br

and B concentrations in halite from the carnallite (L_2) and sylvine (L_3) layers exhibited considerable fluctuations with depth, which are attributed to intermittent influxes of fresh water. The decreasing B concentrations during this stage provide evidence of the low B content in the source fluids, which were likely terrestrial water.

The evolution process of the potash deposition in the Savannakhet Basin can be inferred from the new geochemical data presented here. After initial submergence (Lovatt Smith et al., 1996), inversion of the Khorat group caused the Khorat Plateau to become a restricted continental basin. Trapped seawater and fresh water allowed multiple-source evaporation to take place, although Late Cretaceous seawater influx into the area (through later-formed tectonic gaps) decreased the pH of the evaporation environment. Almost contemporaneously, Yanshan-related tectonic uplift and disturbance (Morley, 2012; Wu Zhenhan et al., 2016; Zhang Jianjun et al., 2017) in the study area allowed fresh water to infiltrate the basin. Faulting associated with this tectonic activity may provid pathways for deep-seated hydrothermal fluids to enter the basin. These interpretations show that evaporite deposits in the region formed via a complex sequence of processes, and thus represent a multi-source ore deposit. Although seawater was the main fluid source, terrestrial water and hydrothermal fluid imparted strong influences on sedimentation, especially during the final stages of evaporation.

7 Conclusions

The boron isotopic compositions and trace element concentrations of halite were studied from multiple strata within borehole ZK309, situated in the southeastern edge of the Savannakhet Basin on the Khorat Plateau. Subsurface exploration in this area was conducted in order to investigate the origin of the Khorat Plateau potash deposit.

(1) The lithology of the study area was predominantly formed by marine evaporation. The ranges of δ^{11} B values and minor element concentrations in these units were similar to those of seawater–derived evaporites. The area was sequentially affected by seawater, terrestrial water, and hydrothermal water influx during different stages of the evaporation process. These non–marine inputs has a strong influence on the crystallization of potash minerals.

(2) The absence of dissolution textures in anhydrite within borehole ZK309 indicates that it is not a basal anhydrite but rather a late–stage residue leached from primary halite by water–rock interaction. According to the $(Mg^{2+}+SO_4^{2-})/(K^++Ca^{2+})$ molar ratios, the halite above the anhydrite is an unaltered halite. The Br content of the

studied halite was much smaller than that expected for deposits from seawater at the beginning of the evaporation process. This phenomenon is attributed to hydrothermal fluid influx, as supported by variations in B concentrations.

(3) From the relative timings at which different fluid sources influenced the Savannakhet Basin, the formation of the evaporites in the lower salt layer can be divided into four evolutionary stages: an initial stage of marine evaporation, a stage of seawater input, a stage of hydrothermal fluid input, and a final stage of terrestrial water input.

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Vol. 92 No. 2

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