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Isotope and chemical constrains on the hydrogeology of the low-temperature geothermal system in Shandong Peninsula, China

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ABSTRACT

Utilization of low temperature geothermal fluids is an alternative energy source to fossil fuels and plays an increasingly important role in space heating in Northern China. There are about 16 local low temperature geothermal systems that have been discovered in Shandong Peninsula by following the hot springs in the surface. However, the origin of geothermal water, fluid mixing, water-rock interaction processes, reservoir temperatures are still largely unexplored. In order to answer these questions, major, trace elements and water isotope analysis were performed for borehole and natural groundwaters in the Shandong Peninsula area. 16 geothermal water samples were collected and analysed in this study during Sep.,30th to Dec. 17th, 2018. Another 48 groundwater data points are collected from a database of former studies in the Shandong Peninsula. The analysis result showed that geothermal fluid temperatures ranging from 28 to 83°C, pH from 6.84 to 8.54, Cl from 41.4 to 5554 ppm, Na from 110 to 2781 ppm, SiO2 from 34.2 to 137 ppm, B from 0.04 to 0.91 ppm, CO_2 from 29.4 to 522 ppm, δD from -72.0 to -44.3‰ and $\delta^{18}O$ from -9.95 to -6.06‰. Based on the δD, $\delta^{18}O$, B, Na and Cl contents, it is shown that the groundwaters were mainly of modern meteoric origin, with some seawater mixing in the coast areas. The fluid chemical concentrations have been modified by primary rock leaching. The extent of reaction (ξ) is between 0.001 and 0.01 kg of rock dissolved per kg of geothermal water. A SiO_2 geothermometor might be used better in this study, which shows the reservoir temperature range from around 80 °C to 130 °C.

1. INTRODUCTION

Low and medium temperature geothermal fields are widely distributed in Northern China. Shandong Providence is one representative place, which is located between Beijing and Shanghai and extends out to sea as the Shandong Peninsula. It has a population of more than 100 million and has a large demand of space heating during winter time. Geothermal fields with intrusion rock reservoirs are mainly distributed in the Shandong Peninsula, famous for the hot springs, producing geothermal fluids within a depth around 200~300m, which is much shallower compare to the other two types of sedimentary geothermal fields. It turns out that these shallower reservoirs are suitable for the local people to utilize, but also can be disturbed easily. Long monitoring data shows that there are some environmental problems occurring due to uncontrolled geothermal water productions, such as water level decline, water temperature dropping, geochemical components changing, etc. (Kang, 2013; Zheng and Kang,2015).

Geochemistry is an efficient method to understand the geothermal systems and help to ensure sustainable use of the geothermal resources. There are different processes in hydrothermal systems which can be determined by chemical compositions and isotope contents of hydrothermal water. Nonreactive elements or conservative elements (such as Cl and B) and stable isotopes ($\delta^{18}O$ and δD) are often carried out to determine the origins of the fluids and mixing processes (Craig, 1961; Friedman et al., 1964; Árnason, 1976; Árnason, 1977). Some "mobile elements" can help determine the process of water rock interaction which controls the water chemistry and indicates the geochemical evolutionary processes in the hydrothermal systems. Geothermometers, based on assumptions of chemical reactions equilibrium, can provide ideas about reservoir temperatures (Fournier, 1979; Arnórssonl et al., 1983; Giggenbach, 1988; Fournier et al., 1973; Arnórsson, 1975; Fournirer, 1982; Reed, 1984).

In general, this study focuses on the intrusion rock reservoirs in Shandong Peninsula, aiming to assess the origin of geothermal waters, find out the effects of mixing and water-rock interaction and estimate the reservoir temperatures.

2. STUDY AREA

Shandong Peninsula is a geographical concept which means the eastern land area from Tanlu fault zone. The geographic range of Shandong Peninsula is around E 118°30′~122°40′, N 34°40′~37°50′ (Figure 1). Administratively, this area belongs to Shandong Province. The size of the peninsula is about 40000km^2. The main prefecture-level cities in this area are Yantai, Weihai, Qiangdao, eastern Weifang, most parts of Rizhao and eastern Linyi, with a population of about 37 million. In topography, Shandong Peninsula is affected by tectonic denudation, characteristically a low mountain and hill region, cut by plenty of faults. Elevation of most areas are about 100m~300m.

Tectonic units of this area are magmatic intrusion uplift areas and sedimentary depression areas. Due to Multi-stage magma intrusions, geologic structure is very complex in this area. The strata of Shandong Peninsula are intrusive rocks, metamorphic rocks, cretaceous sediments, volcanic accumulation and loose deposits of the Quaternary period. Large and deep faults were formed, which caused the formation of a tectonic framework in Shandong Peninsula with mainly NNE trending, NE trending and EW trending (Figure 1).

The earliest rock unit exposed in the centre area is the Archean tonalitic–trondhjemitic–granodioritic (TTG) gneisses basement, with 207Pb/206Pb ages of 2.69 Ga (Tang et al., 2007). Paleoproterozoic lithology units are assigned to the Fenzishan Group and Jingshan Groups, which is a series of high grade metamorphic complexes, from amphibolites to granulite facies (Tam, 2012). The major lithologies from the bottom to the top of the Jingshan Group formation are: garnet sillimanite biotite schist, dolomitic marble, diopside biotite granulite, another layer of marble, graphite biotite granulite and garnet sillimanite biotite schist again. The major lithology from the bottom to the top of the Fenzishan Group formation are: biotite granulite, leucoleptite, dolomitic marble, tremolite schist, graphite biotite granulite and garnet sillimanite biotite schist.

The Neoproterozoic rocks in the study area are mainly two groups. In the north, it’s called Penglai Group and the major lithology are conglomerate, slate, marble, and limestone, which are all low or none metamorphic rocks. In the south, the major Neoproterozoic rock is granitic gneiss, the protolith of which are monzonite granite, granodiorite etc. Mesozoic cretaceous rocks are the major sedimentary rock in the study area. They are mainly conglomerate, sandstone, shale, tuff, andesite and so on.

Mesozoic intrusive granite sequence is widely distributed in the north of the study area. The rock types are: mantle-derived granites (0.23-0.20 Ga), crustal-derived granites (0.16-0.14 Ga) and crust-mantle mixed sourced granites (0.135-0.10Ga). The lithology of each sequence shows a trend of evolution from meso-base to meso-acid and alkaline to meso-acid magma (Ding et al., 2015). Mesozoic tectonic-magmatic activities provided superior conditions for the mineralization of gold deposits and also geothermal resources.

So far, 16 hot spring areas were found in Shandong Peninsula and that is accounting for about 70% of the total hot spring areas in Shandong Province. All the hot spring fields have a total geothermal water production of 2.11 million m^3 (in 2008). Each hot spring indicated a small geothermal field. 9 of them are distributed in Weihai, 5 are in Yantai and 2 are in Qingdao. From 1990s, lots of researchers had been to this area to discover the mechanism of geothermal reservoir formation. Previous researchers concluded that there are mainly two abnormal heat flow areas. They are both along the two regional anticlines in the uplift areas. Fifteen of the springs come out from these anticline areas and the lithology of most geothermal reservoirs are the monzonitic granite of Mesozoic or Archaean periods, the other one comes out from Cretaceous quartz sandstone.

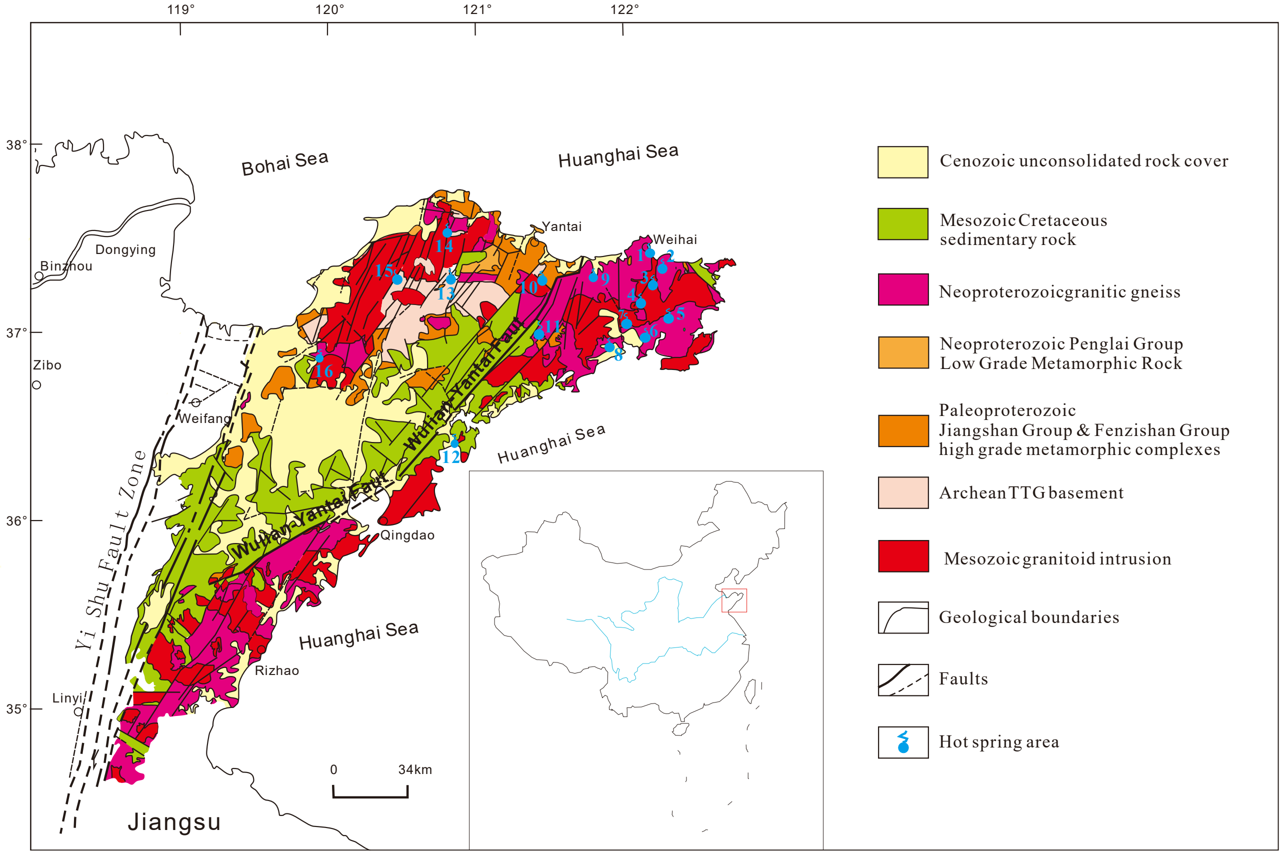

Figure 1: Geological and tectonic map of Shandong Peninsula

These geothermal systems are all low temperature fracture controlled convection type, with belt shaped geothermal reservoirs. The heat in these geothermal systems is "gathered" by deep circulated groundwater. So the main keys to form one of these convection geothermal system are dominated geological structure and plenty of water source (Wang et al., 1993 and Li et al., 1997). Secondary fractures broke up the formation, controlling where the hot springs came up. All the hot springs flow out at the higher permeability cross areas of the secondary faults. The strikes of faults that controlled the formation of hot springs occur in four types: NE, NW, NNE and NNW. The NE strike faults are considered to be heat transfer faults, which are connected to deep heat sources (could be other large deep faults or a dyke approach to magmatic heat). The NW strike faults are considered to be water transfer faults, which have high permeability, gathering recharge water and getting it to the system.

3. SAMPLING AND ANALYSIS OF MAJOR ELEMENTS AND ISOTOPES

Major, trace elements and δ D and $\delta^{18}O$ water stable isotope analysis were performed for borehole and natural groundwaters (non-thermal and thermal) in the Shandong Peninsula area. 16 geothermal water samples(AST1-BQT1) were collected and analysed in this study during Sep., 30th to Dec., 17th, 2018. Another 48 groundwater data (with major, trace elements, stable and radioisotopes) are collected from SDGM(Shandong Provincal Bureau of Geological and Mineral Resources) database of former work in the Shandong Peninsula. Sampling locations are shown in Figure.1.

Samples for pH were collected into amber glass bottles and analyzed using pH electrode and alkalinity titration. Samples for major elements analysis were filtered into PP bottles, filtered by 0.2 μm membrane. Samples for cation elements were acidifed with ultra-purifed HNO_3. Samples for anions were collected with no further treatment. Cation elements were analyzed by ICP-OES and anions were analyzed using IC, both at the University of Iceland. Raw water were collected and stored in PP bottles for hydrogen (δD) and oxygen ($\delta^{18}O$) isotope analyses. The analyses were performed on IRMS at the University of Iceland.

4.RESULTS

4.1 Chemical and isotope compositions

The results of all 64 samples showed that geothermal fluid temperatures ranging from 28 to 83°C, pH from 6.84 to 8.54, Cl concerntration was from 41.4 to 5554 ppm, Na concerntration was from 110 to 2781 ppm, SiO_2 concerntration was from 34.2 to 137 ppm, B concerntration was from 0.04 to 0.91 ppm, DIC concerntration was from 29.4 to 522 ppm, δD content was from -72.0 to -44.3‰, $\delta^{18}O$ from -9.95 to -6.06‰, $\delta^{13}C_{DIC}$ was -17.1 to -2.2‰ and $^{14}C_{DIC}$ was 7 to 132 pMC. The results of chemical and isotope analysis of 16 samples collected in 2018 were shown in Table 1.

Table 1. The results of chemical and isotope analysis of 16 samples collected in 2018

#	ID in Fig. 1	t°C	pH/°C	SiO_2	B	Na	K	Ca	Mg	Cl	CO_2	SO_4	δD	$\delta^{18}O$	Water Type
				ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	‰	‰	
AST 1	13	38	8.18/21	65.8	0.08	212	6.91	14.2	0.82	70.3	217	139.3	-68.1	-9.59	HCO_3-Na
TDQ 1	15	83	8.09/20	98.2	0.16	1030	112	118	3.42	1645	142	133.8	-62.7	-8.42	Cl-Na
TDQ 2	15-1	57	7.40/20	34.2	0.04	158	16.1	15.8	12.2	205	305	48.2	-53.8	-7.62	HCO_3-Na
YJT 1	10	42	8.54/20	75.3	0.07	113	4.69	11.0	0.73	41.4	112	73.8	-63.0	-9.25	HCO_3-Na
HLT 1	5	80	8.31/21	111	0.18	287	13.6	34.0	0.25	175	51.0	378.5	-64.6	-9.22	SO_4-Na
HLT 2	5-1	60	8.35/21	100	0.16	278	12.6	35.3	0.34	171	53.1	365.0	-65.4	-9.07	SO_4-Na
DYT 1	7	66	8.08/21	70.1	0.10	411	13.3	213	0.64	857	29.4	266.9	-62.0	-8.84	Cl-Na
TCT 1	6	35	8.03/21	63.6	0.30	1264	56.3	656	2.79	3063	32.7	302.3	-56.8	-8.22	Cl-Na
WST	14	45	7.05/20	97.4	0.12	368	13.9	45.1	3.81	91.8	522	298.6	-71.1	-9.95	HCO_3-Na
DWQ 1	12	75	7.04/21	78.1	0.48	1874	107	843	16.0	4254	58.4	524.1	-56.5	-7.95	Cl-Na
XT 1	8	50	8.00/21	70.1	0.18	654	18.1	222	2.44	1250	33.6	205.5	-61.6	-8.61	Cl-Na
HSLT 1	3	75	7.33/21	100	0.10	177	12.3	24.2	1.55	54.4	240	148.5	-59.8	-8.36	HCO_3-Na
QLT	4	60	7.83/21	105	0.09	173	8.81	18.2	0.67	43.4	147	176.7	-64.9	-9.11	SO_4-Na
LQT	9	46	8.31/20	62.4	0.07	155	3.46	9.8	0.51	53.8	157	78.8	-64.0	-9.36	HCO_3-Na
WQT 1	2	38	7.21/21	86.0	0.12	331	19.9	48.2	4.12	382	169	147.1	-62.2	-8.80	Cl-Na
BQT 1	1	52	6.84/21	95.7	0.59	2648	13.7	919	51.7	5554	156	409.6	-44.3	-6.06	Cl-Na

All the 64 samples were initially classified in 4 groups, based on the sampling location and depth of aquifers. From a Piper diagram (Figure 2), it can be shown that the sampling distance from sea doesn't present much regularity of water types. Samples from deeper aquifers present a sodium chloride type. Most samples from shallower aquifers do not present a dominant type, a small part of them are calcium chloride type. All the samples demonstrated that strong acids exceed weak acids. It also can be determined that alkaline earths exceed alkalis in samples from shallower aquifers and alkalis in samples from deeper aquifers exceed alkaline earths.

In conclusion, geothermal waters (deeper aquifer) and colder groundwaters (shallower aquifer) have different water types. The main factor here is the circulation depth. Samples don't present a significant difference of types whether they are near a sea area or not.

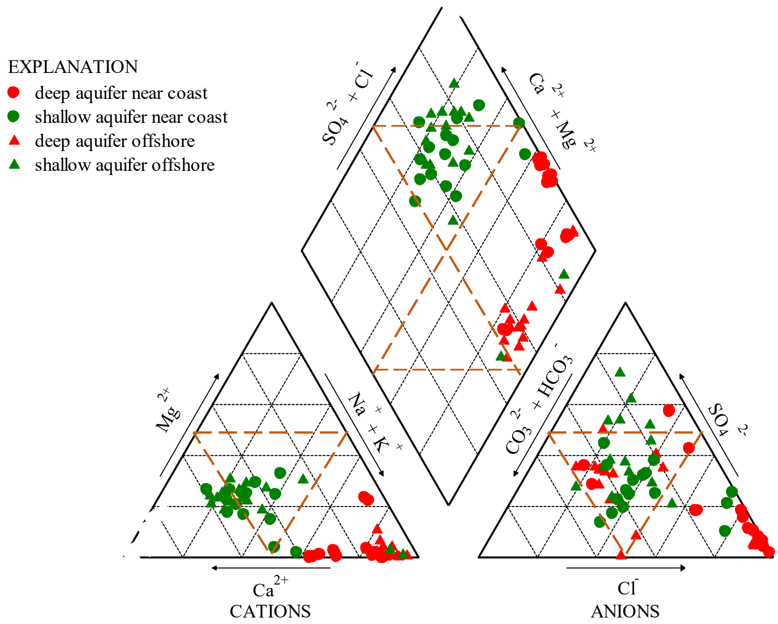

Figure 2: Piper diagram of waters in Shandong Peninsula

5. DISCUSSION

5.1 Water origin and progressive water rock interaction

Origin of water is an important item to show the recharge conditions of geothermal systems. It can be traced by hydrogen (δD) and oxygen ($\delta^{18}O$) stable isotopes and also can be indicated from relationships among various elements; in particular, some relatively incompatible (conservative) elements, such as boron (B) and chlorine (Cl).

The concentrations of δD and $\delta^{18}O$ of groundwater in the study area vary from -72.0 to -44.3‰ and -9.95 to -6.06‰, respectively. The δD values of the local precipitation are between ~-105 to ~ -34‰, according to the data base of GNIP (Global Network of Isotopes in Precipitation, IAEA) and early research (Tian, 2012).

In general, all the groundwater samples lied along the GMWL line and within the range of local rainwater δD values. This showed that the origins of these water samples are mainly from precipitation (Figure 3). The samples from near coastal areas showed there was a slight mixing with seawater. Cold water samples (~<17 °C) of shallow aquifers were overall with higher δD values when comparing with geothermal waters. This meant that in the recharge area the elevations of geothermal water recharge might be higher with longer circulations.

To understand more about the origins of the groundwaters and evolution processes of the cold groundwaters and geothermal waters, a relationship between B-Cl in the samples is shown in Figure 4.

A first impression from figure 4 is: the origin of the groundwaters in the study area is mainly meteoric water, with some degree of water-rock interaction. However, both cold groundwater and thermal water have a clear Cl concentration shift from the precipitation-rock interaction region. This indicated that there might be an addition of a Cl source in the regional groundwater system. It might be because there are high Cl-bearing minerals existing in the system, such as amphibole, biotite, scapolite and apatite.

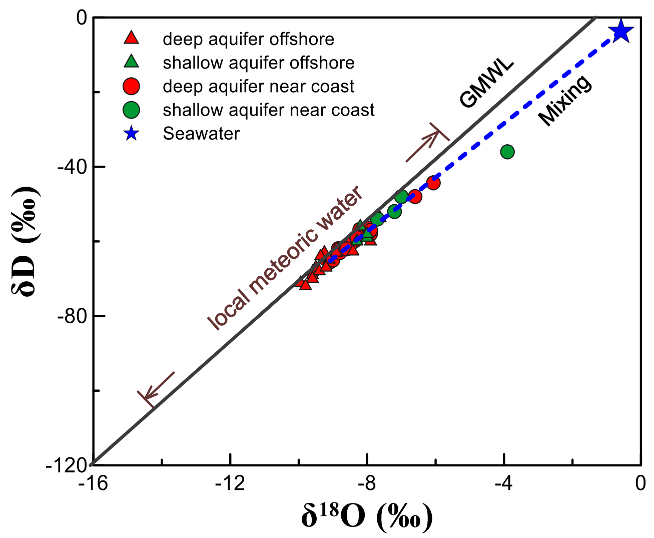

Figure 3: The relationship between δD and $\delta^{18}O$

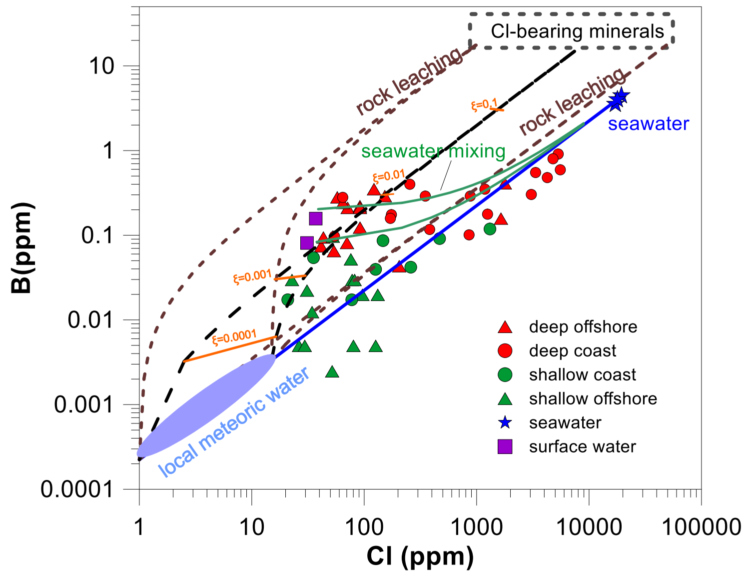

Figure 4: The relationship between B and Cl

The extent of reaction (ξ) is between 0.001 and 0.01 kg of rock dissolved per kg of geothermal water, while for the colder groundwater it is between 0.0001 and 0.001. This might indicate the geothermal water has a longer runoff path and slower circulation, compared with colder groundwater.

In addition, the coastal samples demonstrate a clear sign of seawater mixing. Some of them only have a very slight mixing, with a ratio less than 2%. But some of the mixing can be larger than 10%. There are also samples that have higher Cl concentration, that might be because of minerals, such as halite, dissolving.

5.2 Reservoir fluid composition and temperatures

Geothermal reservoir temperature is an important parameter for studying geothermal activity and evaluating geothermal resource potential. Chemical geothermometers can be used to estimate the reservoir temperature when there is a lack of borehole logging data. Cation geothermometers and silicon geothermometers are usually used for low temperature geothermal reservoirs.

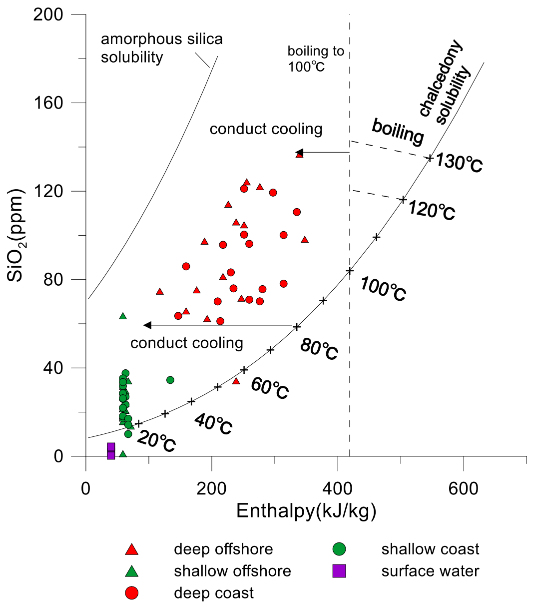

Figure 5: Silica-enthalpy plot of water samples

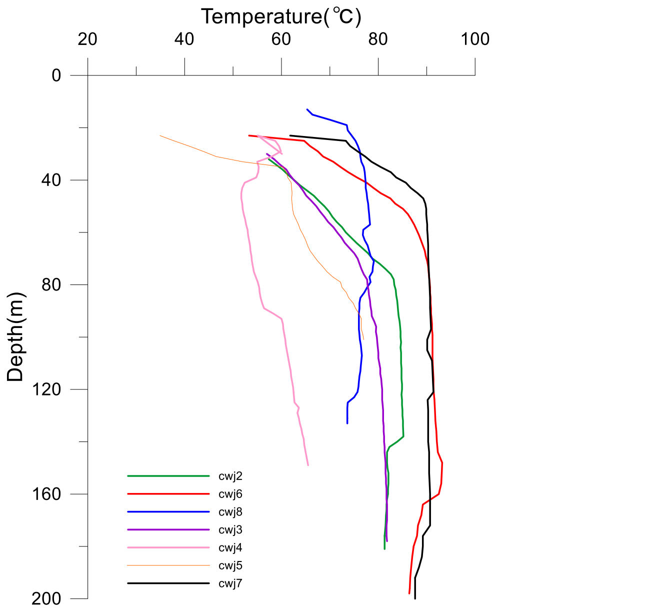

Figure 6: Downhole temperature logging curve in Zhaoyuan

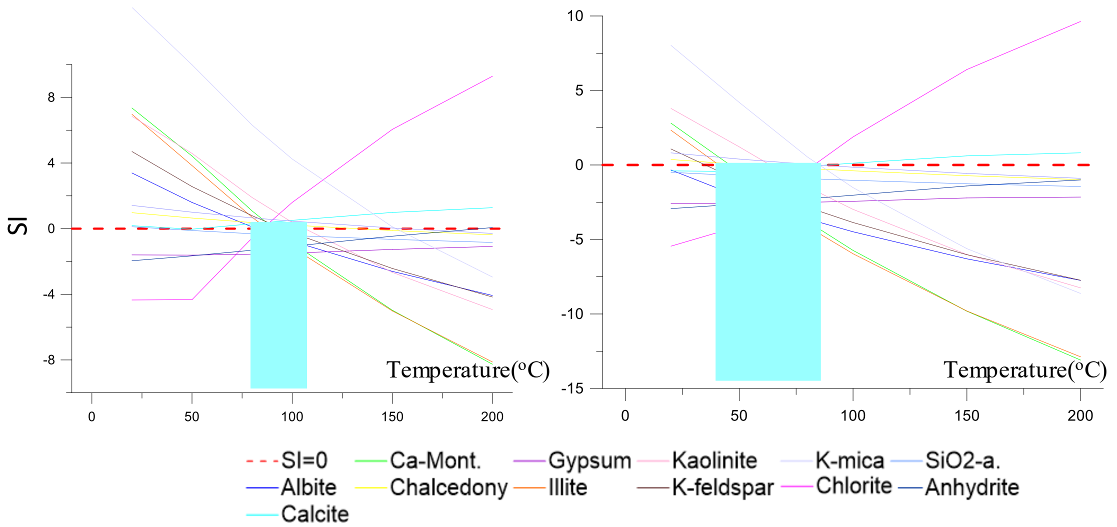

Figure 7: The relationship between saturation index of selected minerals and temperature (JDR 15 and TDQ 2)

The silica concentrations of different waters were plotted against their corresponding enthalpies (Figure 5). It can be seen that all the geothermal samples were distributed at the left of the 100℃ boiling line. That means the most likely process happening in all geothermal reservoirs sampled here were conductive cooling. This can be proven by the downhole temperature logging curve of Zhaoyuan geothermal field (JDR15, Figure 6). It showed that the downhole temperature curve is far below the boiling curve, indicating that there is no boiling happening downhole. The silica-enthalpy plot showed that the reservoir temperatures of Shandong Peninsula ranged from 80~130℃.

Multiple mineral equilibrium calculation, as a geochemical thermodynamic modeling method, is used to estimate a more realistic reservoir temperature. The PhreeqC program (Parkhurst and Appelo, 1999) was used to calculated the aqueous speciation and mineral saturation index (SI) of the geothermal samples. The minerals choosen for the calculations are: Ca-montmorillonite, gypsum, kaolinite, K-mica, amorphous silica, albite, chalcedony, illite, K-feldspar, chlorite, anhydrite, calcite. These minerals were usually present in the spring sediments occurring in the previous studies (Database of SDGM).

Based on the assumption that all the reactions between minerals and fluids should be in equilibrium, the SI of these selected minerals should have a mineral SI convergence point at SI=0, and this point indicates the reservoir temperature. However, that might not be suitable for all the samples. Figure 7 showed two samples with highest and lowest sampling temperature, respectively. This method coincides well with sample JDR 15(Figure 7 left plot), with the estimated reservoir temperature of about 80~110 ℃. However, the sample TDQ 2(Figure 7 right plot) had a large range of temperatures, that might be because not all the reactions were in equilibrium.

Estimated reservoir temperatures with different methods are shown in Table 2. The downhole temperature measurement had much lower temperature than the other ones, that maybe because the measurements were taken before the borehole finished warming up.

Table 2 Estimated reservoir temperatures with different geothermometers

#	Location	Sampling	Downhole measurement (Shallow)	Chalcedony	Na/K*	Multiple mineral equi.
		T (°C)				
BQT 1	Baoquan tang	52	40-45	108	--	70-100
JDR1		67		93.5	120	80-110
WQT 1	Wenquan tang	38	30-45	101	150	60-80
JDR2		55		99	159	
HSLT 1	Hongshuilan tang	75	40-60	110	164	50-100
JDR3		71		120	174	65-115
QLT	Qili tang	60	61-64	113	136	70-100
JDR4		66		122	139	
HLT 1	Hulei tang	80	65-70	114	128	55-90
HLT 2		60		108	125	60-90
JDR5		60		121	135	80-100
TCT 1	Tangcun tang	35	51-53	83.1	123	65-85
JDR6		51		81.9	111	70-100
DYT 1	Daying tang	66	48-57	88.2	101	50-95
JDR7		62		89.7	86.9	65-95
XT 1	Xiao tang	50	70-75	88.5	90.6	60-100
JDR8		56		93.2	88.8	70-90
LQT	Longquan tang	46	39-43	81.3	78.6	50-100
JDR9		59		89.5	89.8	70-80
YJT 1	Yujia tang	42	40-64	90.9	120	65-100
JDR10		57		112	137	50-110
JDR11	Xingcun tang	28	18-26	91.0	93.4	60-110
DWQ 1	Dongwenquan (Jimo)	75	70-84	95.2	143	70-80
JDR12		62		107	137	80-110
AST 1	Aishan tang	38	26	84.7	102	75-100
JDR13		52		96.4	103	65-95
WST	Wenshi tang	45	55-56	109	112	55-95
JDR14		54		118	132	70-85
TDQ 1	Tangdongquan (Zhaoyuan)	83	50-90	106	210	70-105
TDQ 2		57		53.4	204	30-80
JDR15		81		129	180	80-108
JDR16	Jiudian	61	--	122	164	80-120

*** Calculated by WATCH program, developed by Iceland Geosurvy (ISOR), Arnórsson et al.,1982**

6. CONCLUSION

Chemical compositions and isotope (δD, $\delta^{18}O$) of low temperature geothermal water from Shandong Peninsula were investigated in order to delineate the origin, mixing, and water-rock interaction of the waters. From the geothermal fluid temperatures ranging from 28 to 83°C, pH from 6.84 to 8.54, based on the δD, $\delta^{18}O$, B, Na and Cl contents, it is shown that geothermal waters were mainly of modern meteoric origin, with certain seawater mixing in coastal areas, and modified by primary rock leaching. The extent of reaction (ξ) is between 0.001 and 0.01 kg of rock dissolved per kg of geothermal water. A SiO_2 geothermometer might be used better in this study, which shows the reservoir temperature range from around 80 °C to 130 °C.

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