Proceedings World Geothermal Congress 2010
Bali, Indonesia, 25-29 April 2010

The Isotopic and Chemical Characteristics of Geothermal Fluids from Two Selected Hot Spring Areas in Jiangxi Province, SE-China

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Keywords: Isotopic and chemical characteristics, Hengjing, Gangxinan, low temperature geothermal areas

ABSTRACT

Two typical low temperature geothermal areas, the Hengjing and Gangxinan areas from Jiangxi province in SE-China, were selected for chemical and isotope studies of selected warm and cold groundwater circulating in the areas. Thermal waters from both the Hengjing and Gangxinan hot spring areas are classified as HCO_3 type water. Na-K-Mg triangular diagrams indicate that the thermal waters from the Hengjing area are partially equilibrated or mixed waters, whereas those from the Gangxinan area are immature. The Na-K-Mg-Ca diagram indicates that the temperature within the Hengjing geothermal reservoir is in the range of 130~180°C. Oxygen and hydrogen isotope study of the thermal waters suggest localized precipitation origin. The recharge altitudes are around 350 m~800 m and 530 m~1460 m for hot springs in the Hengjing and Gangxinan areas, respectively. Helium and light carbon isotopes from Hengjing geothermal fluids show that they could partly be derived from the mantle, whereas those from Gangxinan are considered to be derived from the upper most crustal radiogenic production and also by addition of much depleted ^{13}C.

1. INTRODUCTION

In the Jianxi province, SW-China, hydrogeological isotope studies of ground waters have been conducted in many hot spring areas (Li et al., 1992; Sun et al., 1992; Sun and Li, 2001). Local meteoric water line, origins, recharge areas and the ages of the geothermal waters have been reported (Sun and Zhang, 2005). Particularly, in recent years, the provenance of geothermal fluids has been suggested and a new geothermal models have been established by applying gas compositions and carbon isotopes in some hot spring areas, as well as some noble gas isotopes, such as ^3He and ^4He (Sun and Li, 2001; Zhou and Zhang, 2001). In this paper, two typical hot spring areas in the Jiangxi province were selected for study. Based on varied geological and isotopic and chemical survey, the isotopic and chemical characteristics of the springs are studied. The analyzed results are illustrated in this paper.

2. GEOLOGICAL BACKGROUNDS

Jiangxi province, SE-China, is one of the provinces in which hot springs are most widely distributed in China (Sun, 1998). Two important hot spring areas in the Jiangxi province, namely Hengjing and Gangxinan, where selected for this study. Both have typical hot springs, some with remarkable amounts of gas escaping, and higher temperature hot springs are found within these two areas. Two investigations performed in 1997 and 2003 have been done to study the occurrences of geothermal fluids and assessment of geothermal reservoirs (Sun and Zhang, 2005).

2.1 The Hengjing Hot Spring Area

The Hengjing Hot Spring Area located in Xunwu County, South Jiangxi province, SE-China, has more density of hot springs than any other area in the province. Many of the hot springs have CO_2 and other gases escaping. Their temperatures are in the range of 26°C to 73°C at the surface. The rock matrix consists mainly of the upper Cretaceous system (K_2) characterized by red sandstone and conglomerate, the lower Jurassic system (J_1) and the upper Jurassic system (J_3) by sandstone and conglomerates, the Sinian-Cambrian system(Z-∈) by metamorphic sandstone, and migmatite as well as the Yanshannian granite (γ_5^2) and biotite-granite($\pi\gamma_5^2$). Since 1960's, the geological and hydrogeological surveys have identified 13 thermal springs in the area, located along NE faults, but also controlled by NW faults (Sun and Zhang, 2005). Those faults are developed well in the area. During 1974 to 1992, more than 30 earthquakes have occurred in the area. One of the biggest earthquakes occurred in August, 1987, with magnitude up to 5.7. It has been proposed that the earthquake activity indicates that these faults are still active. The average terrestrial heat flow of 74.10 mW/m^2 is somewhat higher than the average value of 69.79mW/m^2 for the whole province (Li and Zhou, 1992).

2.2 The Gangxinan Hot Spring Area

The Gangxinan Hot Spring Area is located in South-Western Jiangxi province, SE-China, covering almost five counties including Dayu, Chongyu, Shuicuan and Shanyu County. The rock types consist largely of the Cretaceous system (K) characterized by sandstone and conglomerates, the Jurassic system (J) by sandstone, conglomerates and tuff, and migmatite as well as the Yanshannian granite. Hydrothermal manifestations are widespread in the area and the faults exerting localized control surface distribution of hot springs. The faults involve two major fault trends NNE and NEE. More than 10 hot springs have been found with temperature ranging from 37.7°C to 83.8°C at the faults southern end. Five of the springs have gases escaping.

3. SAMPLING AND ANALYTICAL METHODS

3.1 Sampling

The types of samples used in this study are water samples and gas samples from cold and hot springs and from hot water wells. The collection of representative gas samples for isotopic analysis from a discharging spring involves the collection of dry gas (volatile). In this paper, several data sets were used which were made available for the present study. These waters for Hengjing and Gangxinan Hot Springs were sampled in 1997 and 2003, respectively, under the project which was funded by a grant from the National Natural Science Foundation of China (40472147). Some of them have been published (Sun and Li, 2001; Sun and Zhang 2005; Zhou and Zhang, 2001).

3.2 Analytical Methods

The analytical methods, including water chemistry, isotopes used to obtain data for the report are presented in Table 1.

Table 1: Analytical methods for geothermal fluids and Isotopes

Composition	Sample fraction	Methods of analysis
CO_2	Ru	Alkalinity-titration
H_2S	Ru	Titration
SiO_2	Rd	Spectrophotometry
Na	Fa	Atomic absorption spectrometry
K	Fa	Atomic absorption spectrometry
Ca	Fa	Atomic absorption spectrometry
F	Ru	Ion selective electrode
Cl	Ru	Ion chromatography
SO_4	Fp	Ion chromatography
Al	Fa	Atomic absorption spectrometry
Fe	Fa	Atomic absorption spectrometry
B	Fa	Modification of the curcumin
pH	Ru	Ion selective electrode
$\delta^{18}O$, δD	Ru	Mass spectrometry
$\delta^{13}C$, ^{14}C	Gas	MAT-252 mass spectrometer
^3He,^4He	Gas	VG5400 Mass spectrometry

Ru - Raw, untreated; Rd - Raw, diluted; Fu - Filtered, untreated spectroscopy; Fp - Filtered, precipitated; Fa - Filtered, (0.2 μ, 0.45 μm) acidified.

The oxygenic and hydrogen isotopic composition of groundwater was analyzed and the data is represented as δ-values in per mil. Oxygen was extracted from the water by equilibrating 5 ml of degassed water with small amount of CO_2 gas in a sealed tube for 3 hours, in a shaking water bath at 20.0°C (Epstein and Mayeda, 1953). The measurements are subsequently corrected for the CO_2 gas which is also used as the secondary reference standard. In Iceland hydrogen isotope analysis based on the zinc reduction method (Coleman et al., 1982) was used until 1998 but later by the H_2-water equilibration method using a Pt-catalyst (Horita, 1988). The samples were analyzed on the Finnegan MAT252 at the National Lad., Nanjing University. The accuracy of the measurements is better than 0.2 ‰ for $\delta^{18}O$ and 3‰ for δ^2H.

The sample preparation for carbon isotopes was carried out in accordance with McNichol et al. (1994), i.e. the water samples (1liter) were acidified in a vacuum system and CO_2 extracted directly by nitrogen flow through the water sample. The released CO_2 from the Hengjing and Gangxinan areas was measured only for $\delta^{13}C$ at the Finnegan MAT 252 mass spectrometer at the Lanzhou Institute of Geology, Chinese Academy of Sciences. The accuracy of the measurement is better than 0.1‰ (PDB).

Helium isotope data is available for the Hengjing and Gangxi'nan areas in China (Zhou and Zhang, 2001). In China, sample collection took place over two periods (1997, 2003) following closely the standard sampling techniques as described by Hilton (1996). All samples were collected in AR-glass flasks with prolonged flushing of all connecting tubing carried out to ensure minimal atmospheric contamination. The helium isotope ratio of the volatile for the Chinese samples was analyzed on the VG5400 at the Lanzhou Institute of Geology, Chinese Academy of Sciences, China. The accuracy of $^4He/^3He$ measurement is better than 0.3×10^{-6}%.

4. CHEMICAL AND ISOTOPIC CHARACTERISTICS OF THE THERMAL FLUIDS

4.1 Chemical Composition of Waters

In the Hengjing hot spring area, the chemical compositions of water discharges are from cold and hot springs, according to measured temperature. Sampling positions are shown in Figure 1. An initial classification of the waters is carried out on the basis of relative contents of the three major anions, Cl, SO_4 and HCO_3. None of the waters plot in the fields marked “mature water” or “volcanic water” in the diagram. All the thermal waters plot close to the HCO_3 corner and contain HCO_3 as the predominant anion. This would suggest that the thermal waters are of meteoric origin and that absorption of CO_2 diffusing from deeper levels into peripheral groundwater has occurred. The proportions of HCO_3 of the thermal waters are the same as of cold springs, but the thermal waters contain higher SO_4/Cl ratios, probably due to water-rock interaction that increases the SO_4 concentration in the thermal fluids. Clearly, thermal waters have significantly different chemical properties from the cold waters due to water-rock interaction. It is however suggested that all waters in this area are of meteoric origin (Sun and Zhang, 2005).

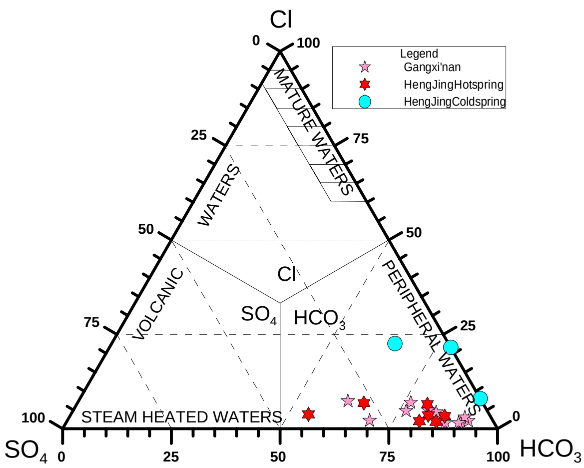

Figure 1: Classification of waters from the hot spring areas on the basis of relative Cl, SO4 and HCO3 content, in mg/l.

All of the chemical compositions of water discharged are from hot springs in the Gangxinan hot spring area, together with measured discharge temperature and isotopes. An initial classification of the waters on the basis of relative contents of Cl-SO_4-HCO_3 is also shown in Figure 1. All the samples are located in the HCO_3 corner in this figure. This indicates that the thermal waters can be classified as HCO_3 type water and all of the waters are highly immature.

The main reasons for the chemical difference are that the geology of the areas is different, where most of the rock in Hengjing is carbonic rock, but in Gangxinan the rock matrix is not carbonic. Thus water rock interaction will lead to different water types during the thermal water up flow and mixing with the cold water.

In Hengjing hot spring area, all water samples are plotted in the triangular diagram as shown in Figure 2. Points close to the √ Mg corner usually suggest a high proportion of relatively cold groundwater, not "immature". As shown in the diagram the positions of all the data from Hengjing are classified into two groups: the hot spring group and the cold spring group including sample HJ07, HJ08 and HJ10. Most hot springs of the first group are located in the partially equilibrated and mixed water area or close to this area. The cold water is, however, very immature and is located in the immature waters zone. None of the data points suggests full equilibrium. This has been indicated that the Hengjing waters are not fully mature and the geo-indicators can therefore only be used with limits. In this case, more detailed information is needed on possible processes that have affected the waters. This can be obtained by using a square diagram based on the relative Na, K, Mg and Ca content (Giggenbach, 1988), as shown in Figure 3. There it can be seen that all the water points can be classified into three groups. Firstly the mature group is close to the full equilibrium line including HJ05, HJ012 and HJ06, secondly the less mature water group is located in the centre of the diagram including HJ11, HJ09 and HJ02, and thirdly the cold water group is located at the Mg and K corner, including HJ07 HJ08 and HJ10. As the different symbols representing different water temperature Figure 3 demonstrate that with increasing temperature the maturity of the waters increases. The occurrence of mixing of thermal waters with surface waters has also been proposed by Sun and Zhang (2005). Figure 3 shows the mixing lines through the hot waters emanating from the cold waters and extrapolation until they intersect the full equilibrium line. The temperatures that can be read off as indicated by these intersect points suggest that deep water-rock equilibration temperatures are ranging from 130°C to 180°C given an average value of 155°C.

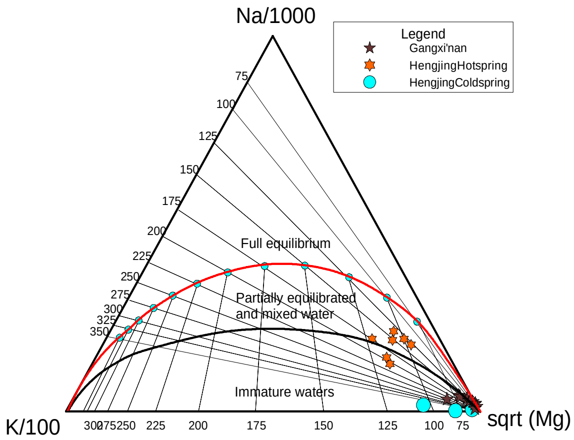

Figure 2: Na-K-Mg equilibrium diagram for Arnórsson (2000) and the fluids of Hengjing Hot Spring Area.

From Guanxinan hot spring area, the data of the hot springs are also shown on the Na-K-Mg ternary diagram in Figure 2. All the hot spring waters plot in the √Mg corner, suggesting that these waters have not attained equilibrium and are immature water. This indicates that they have possibly mixed with cold water during the up flow. For such waters, the application of solute geothermometers to estimate discharge temperature is not reliable.

The Na-K-Mg ternary diagram provides information about the geothermal water affected by mixing with cold groundwater. Figure 2 shows that all the waters from the Hengjing and Guanxinan areas are partially equilibrated or mixed waters and some of the cold water is immature. Also many of the waters from the Western fjords are partially equilibrated and mixed waters, but some are fully equilibrated. The unequilibrated waters are generally interpreted to have been affected by mixing with cold water. This is in agreement with the oxygen and hydrogen isotope study. The temperature of the geothermal reservoir in the Hengjing area obtained by Ca-Na-K-Mg diagram ranges from 130℃ to 180℃. More information could be achieved by other methods such as quartz geothermometer and silica-enthalpy diagram (Sun and Zhang, 2005). The temperature of the geothermal reservoir in the Guanxinan area, however, cannot be obtained by cation geothermometers because all the waters are very immature waters.

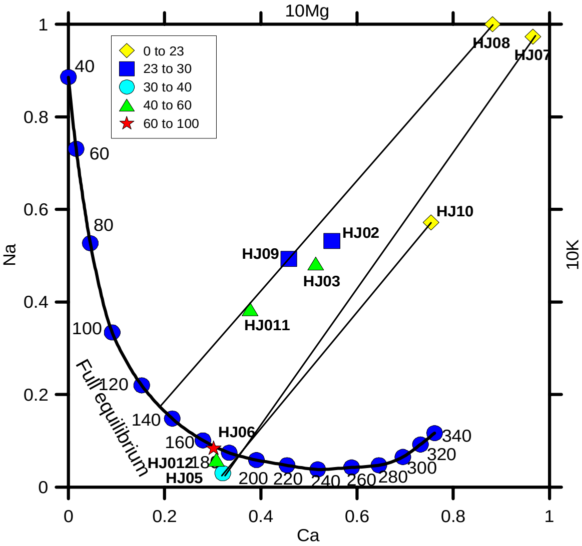

Figure 3: Evaluation of Na-K-Mg-Ca equilibration temperatures by use of values of 10cK/(10cK+cNa) versus 10cMg/(10cMg+cCa), ci in mg/kg, where the waters temperature ranging from 0-23°C are represented by the diamond, from 23-30°C by the square, from 30-40°C by open circles, from 40-60°C by triangles and above 60°C by stars.

4.2 Stable Isotopes of O and H

In the Hengjing Hot Spring Area, fourteen samples including four cold springs with the water temperature varying from 18°C to 22°C, and ten hot springs from 25°C to 73°C, were plotted in Figure 4. In the plot, the isotopic composition of the cold waters lies in the range of -39‰ to -55‰ and -5.6‰ to -7.4‰ for δ^2H and δ^{18}O, respectively. The δD-δ^{18}O relationship of the cold water is shown in Figure 4 defining the local meteoric water line as δD= 8.33δ^{18}O + 8.52, with the correlation coefficient of 0.97. Compared to the global meteoric water line defined by Craig (1961), the local meteoric line has similar slope but the intercept is about 1.5 lower. The δ^2H values of the geothermal waters in this area range from -47‰ to -57‰, i.e. are more negative than those of the cold springs. The δ^{18}O values of the thermal waters (-5.9‰to -7.2) are, however, similar to those of the cold waters. This has been considered to indicate that the origin of the geothermal water is local precipitation from higher latitudes than the cold water. The slight oxygen shift, in the range of 0.2‰ to 1.0‰, has been explained by interaction between groundwater and the bedrock for the geothermal water. However, the oxygen shift is not obvious, especially when the accuracy of the measurements (0.2‰ for oxygen and 3‰ for hydrogen) is taken into consideration. Lack of oxygen shift is taken to indicate low reservoir temperatures in agreement with Sun and Li (2001), who concluded that the Hengjing Area belongs to a deep circulation type and low enthalpy geothermal resources. In the central part of the Jiangxi province, the correlation lines between altitude and isotopic composition of local precipitation were defined as (Sun and Li, 2001):

$$\delta D = -25.11 - 0.047H \tag{1}$$

$$\delta^{18}O = -4.82 - 0.0032H \tag{2}$$

Where, δD and δ^{18}O values are expressed in per mill versus the SMOW, H denotes altitude in meters. According the isotopic contents in these thermal waters, the recharge altitudes of the thermal waters are in the range of 350 m to 700 m (Sun and Li, 2001)

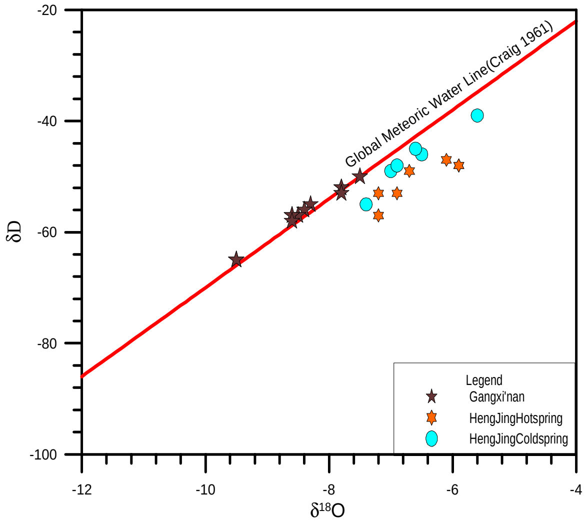

FIGURE 4: δ^{18}O and δD relationship of cold and hot spring waters in the Hengjing hot spring area. Both the global meteoric water line (Craig, 1961) and the local meteoric water line are shown for comparison.

Figure 4 also shows the δ^{18}O and δD values of thermal waters in the Gangxinan Area. The figure demonstrates a considerable range both in δ^{18}O (-9.5‰ to -7.5‰) and in δD (-65‰ to -50‰). Lacking isotopic composition of cold water in this area, both the global meteoric water line (GMWL) is shown for comparison in Figure 4. The best fit through the thermal water data points is δD = 7.36δ^{18}O + 5.49, very similar to the GMWL. This indicates that the thermal water is of meteoric origin and temperatures of geothermal reservoir are low. Using equations (1) and (2), the altitudes of the recharge areas are estimated to be in the range of 530 m to 1460 m.

All data from the three hot spring areas are plotted in the δD and δ^{18}O diagram shown in Figure 4. δD of the thermal waters from all two place is in the range of -72‰ to -39‰ and δ^{18}O ranges from -9.2‰ to -5.8‰. The water samples follow the global meteoric water line (GMWL). This demonstrates that the thermal waters are of meteoric origin and also indicates that the reservoir temperatures in these areas is relatively low as no oxygen shift is observed, apart from the warm waters in the Hengjing area, which is explained by high CO_2 concentration of the bedrock. This is also in good agreement with other geochemical data that have characterized these areas as low temperature reservoirs. However, there are some differences between the areas. As pointed out above, the thermal waters from the Hengjing area show slight oxygen shift. One possible reason is the high concentration of CO_2 (96.7~99.8%), which can provide more protons for water-rock interaction to cause oxygen isotopic exchange. Even in the same geographical area, the isotope concentration can range considerably. This is taken to suggest that these hot springs have different recharge regimes.

4.3 Helium and Carbon Isotopes

Helium isotopes are useful to study the origin of geothermal gases. From the Hengjing hot spring area, helium isotope results were obtained for 4 samples, as shown in Figure 5, where are represented by circles, ranging from 1.90×10^{-6} to 2.95×10^{-6}. These R/Ra ratios which are in the range of 1.36 and 2.11 are approximately one quarter of the MORB (mid-ocean ridge basalt) average level value. Since the $^{3}He/^{4}He$ values for the hot springs are higher than 1 (1Ra), mantle degassing must be occurring. Thus the helium isotopes have been suggested to indicate that the Hengjing area is located in a region of degassing of mantle-derived volatiles.

Carbon isotopes $\delta^{13}C$ are also very useful in indicating the source of the gasses (mantle, crust, subducted sediments or atmosphere). Only four carbon isotope samples were analyzed. $\delta^{13}C_{DBP}$‰ lies in the range of -6‰ to -4‰, i.e. somewhat heavier than the air (-7‰). In China, the $\delta^{13}C$ (CO_2) values associated with inorganic carbon are normally greater (heavier) than -8‰ and mainly fall into the range of -8‰ to +3‰, whereas values associated with organic carbon are normally in the range of -8%~-26%. Values between -10% and -8% are considered to be associated with a mixture of inorganic and organic carbon. $\delta^{13}C$ (CO_2) values associated with magmatic mantle origin carbon normally lie between -8‰ and -4‰ and the values associated with $CaCO_3$ metamorphous carbon range from -3‰ to +3‰ (Dai, 1994). In Figure 5 these typical values for different geological settings are shown together with the data from the Hengjing Area. All the data groups together in the mantle or magmatic source zone, where -8‰<$\delta^{13}C_{DBP}$‰<-4‰ and 1<R/Ra<MORB(8). This is in agreement with the geological observation that carbonate rocks are not found in the area. Thus it is suggested that the gas from the hot springs is of inorganic origin and possibly partly of mantle origin.

From the Gangxinan hot spring area, nine samples from five hot springs were analyzed for helium (^{3}He and ^{4}He) and carbon isotopes ($\delta^{13}C$), which are represented by rectangles as shown in Figure 5. The $\delta^{13}C$ from the hot springs are in the range of -12.6‰ to -23.7‰, i.e. considerably lower (more negative) than the atmosphere (-7‰). $\delta^{13}C$ values associated with organic carbon have been found to be in the range of -8‰ to -39‰ and most values lie in the range of -12 ‰ to -17‰, whereas inorganic carbon falls in the range of -8 ‰ to +7‰. Accordingly CO_2 from the hot springs originates most likely from organic (sedimentary) carbon or from some mixture of organic carbon with either atmospheric carbon or with inorganic carbon. The measured $^{3}He/^{4}He$ ratios(R) from the sample range from 1.90×10^{-6} to 2.95×10^{-6} and R/Ra ratios range from 0.05 to 0.13. All of the R/Ra values are smaller than 1, and most of them are less than 0.1. This indicates that the helium in the thermal springs originates from crust radiogenic production. All the data were plotted in the diagram showing the relationship between helium isotope ratio (R/Ra) and $\delta^{13}C_{DBP}$‰. It can clearly be seen that all data points fall into the organic CO_2 zone and lie between a typical radiogenic helium value (R/Ra= 0.05) and atmospheric helium value (R/Ra= 1). Furthermore, there is a tendency for the lighter carbon isotope values to be associated with lower R/Ra ratios indicating a possible common provenance of radiogenic helium and light carbon. Therefore, the thermal gases can be considered to be of "crustal" radiogenic origin possibly mixed with atmosphere.

As all He isotope data considered in this study and the $\delta^{13}C$ values of gas samples are compiled in Figure 5, there are some clear differences that have been seen in the data from these two areas. This can be explained by the following. Firstly, the Hengjing hot spring area is situated ~800 to 1000 km away from the convergent boundary between the Philippine plate and the Eurasian plate. The helium isotope values (R/Ra) in this region range between 1 and 2, whereas the $\delta^{13}C$ values fall into the MORB $\delta^{13}C$ range (-5‰ ± 1). It has been suggested that partial mantle-derived input dominates the helium systematic. The Gangxinan hot spring area is even further away from the convergent boundary, or about 1000-1200 km. The isotope (R/Ra) values are in the range of 0.01 to 0.2, which are much lower than in the other area, and most of them fall into the typical radiogenic production value (0.01~0.1). The $\delta^{13}C$ values are very light and lie in the range of -24‰ to -12‰.

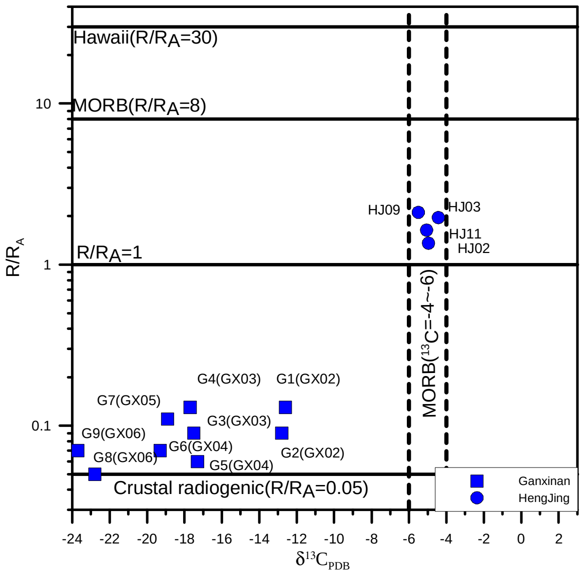

FIGURE 5: Plot of carbon isotope ^{13}C concentration vs. helium ratio.

5. CONCLUSIONS

The main conclusions and recommendations can be summarized as follows:

(1) The thermal waters from the Hengjing and Gangxi'nan areas are HCO_3 dominated type waters.

(2) According to the Na-K-Mg ternary diagram, the thermal waters from the Hengjing area are partially equilibriated or mixed waters. The Na-K-Mg-Ca ternary diagram indicates that the temperature of deep thermal water reservoirs in the Hengjing area range from 130 to 180°C. While the thermal waters from Gangxinan are immature and cannot be used for reservoir temperature estimates.

(3) The thermal waters are considered to be of meteoric origin according to the $\delta^{18}O$ and $\delta^{2}H$ relationship. In the Hengjing and Gangxinan areas, thermal waters are considered to be of local precipitation origins. The recharge altitudes are around 350 m~800 m for the hot springs in the Hengjing and 530 m~1460 m for those in the Gangxinan area.

(4) Helium isotopes $^{3}He/^{4}He$ and $\delta^{13}C$ concentration of gas escaping from the hydrothermal waters indicate different origins for the three studied areas. In the Hengjing Area, helium and light carbon isotopes show that they could partly be derived from the mantle.

Helium in Gangxinan is derived from radiogenic production of the upper most crust by addition of much ^{13}C depleted carbon.

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