Recognition of Significant Multi-Element Geochemical Signatures of Lower Soil on Hainan Island, China: Implications for Thermal Mineral Water Exploration

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Abstract: As an important geothermal resource, thermal mineral water has high resource efficiency and thermal energy efficiency. The aim of this study was to delineate prospective areas of thermal mineral water based on potential thermal mineral water sites and faults. Linear regression was used to process the temperature of 22 known thermal mineral water sites as dependent variables, and 54 indices of the lower soil of multipurpose regional geochemical surveys as independent variables, in the area of intermediate-acid intrusive rocks and sediment degeneration rocks on Hainan Island. Published data were quoted from the National Multi-Purpose Regional Geochemical Survey (Hainan Province, China). According to the regressive modelling of 2197 lower soil samples, 547 potential thermal mineral water sites were delineated after considering 4 factors—geological background, regional structure, interval of dependent variable’s predictive temperature, and boundary of independent variable’s contents—which were compared against 22 known thermal mineral water sites to choose the 2197 lower soil samples, based on the choice of prospective sites of thermal mineral water on Hainan Island. The results showed that the proportion of A1-level sites that were >70 °C constituted 11% of all A1-level prospective sites, reflecting the superiority of east–west or north–east directional regional faults in controlling the distribution of thermal mineral water. This study shows the indications of the multipurpose regional geochemical survey with regards to thermal mineral water, which is one of the most important tourist resources of Hainan Island.

Keywords: multipurpose regional geochemical survey; thermal mineral water; indication; Hainan Island

1. Introduction

Geothermal resources are buried underground, and are controlled by conditions such as geological structure, stratum lithology, and water–rock interaction [1–5]. Their distribution is closely related to plate tectonics, and plate tectonic movement has a controlling effect on global geotropic activities. Among the geothermal resources, the groundwater that absorbs geothermal heat in the porous or fractured rock layer and has a temperature of 25 °C or higher is called thermal mineral water, which is a hydrothermal geothermal resource. Understanding the geochemical evolution process and geochemical characteristics of geothermal fluids is of great significance to the sustainable development of geothermal resources [6,7]. China’s medium- and low-temperature geothermal resources are widely distributed in the continental crustal uplift and crustal subsidence areas within the plate. The fault zones formed in different geological periods are widely distributed in the crustal uplift. Among the crustal uplift areas, the geotropics along the southeast coast are the most densely distributed areas of hot springs. Hainan Island is very rich in thermal mineral water resources, and 32 rift-type belt distributions of thermal mineral water have been
identified to date, distributed in the central part of Hainan Island and strictly controlled by the relevant faults.

Thermal mineral water is widely used for power generation, heating, aquaculture, textile printing and dyeing, etc. [8–13]. Meanwhile, geothermal water has higher medical and healthcare value when the content of some trace elements in hot spring water reaches the water quality standard of medical mineral water [14–19]. Although Hainan Province is rich in thermal mineral water resources, their level of development and utilization is not high, due to the constraints of many factors. Therefore, under the premise of rational planning of resource development, finding effective methods of searching for minerals will be of great help to improve the utilization rate of thermal mineral water [20].

Studies have shown that the trace element content of soils around thermal mineral water tends to increase or decrease to varying degrees [21]. Among them, elements that are more than 10 times more abundant than the crustal Clarke numbers include Hg, As, Sb, Bi, W, Mn, etc.; elements that are several times more abundant include Pb, Zn, Ni, Li, Rb, Be, B, etc.; and elements that are slightly less abundant include Cu, Mo, Sn, etc. Therefore, the search for thermal mineral water based on soil element geochemistry can be an effective technical method. The use of multipurpose regional geochemical surveys to obtain elemental index data is a very effective method [22]. This work aimed to search and forecast the island-wide thermal mineral water distribution based on the discovered thermal mineral water and the deep-soil geochemical data of a multipurpose regional geochemical survey.

2. Overview of the Study Area

Hainan Island is located in the northwest of the South China Sea, with a diverse and tropical ecosystem (Figure 1); it is a dome-shaped island composed of mountains, hills, and terraces—high in the middle, and low on all sides. The central part is mountainous, with the Wuzhishan Mountain as the main peak, while the geological structure in the north is similar to that of the Leizhou Peninsula, which is a low-platform land. Hainan Island belongs to the southern extension of the second uplift zone of the Neocathaysian system. The regional tectonics are relatively developed, with four main groups: east–west, north–east, north–west, and north–south. Fault zones are more widely distributed on Hainan Island, and this geological structure provides favorable conditions for heat rising along the fault zones at depth, and for water infiltrating, flowing, pooling, and storing along the fault zones. Due to the development of faults, different rock layers are in contact with one another, causing the thermal mineral water to come into contact with different strata and absorb various chemical elements. The east–west rupture is the seismic control structure of Hainan Island. At the same time, the north–east fault zone was formed before the Cretaceous period, and exerts very obvious control over the seismic, geothermal, hydrogeological, and mineral resources. The combination and interaction of these tectonic zones is an important factor affecting the formation and outcrop of emergence of thermal mineral water on Hainan Island [23].

Hainan Island is located over two geotectonic units; with the east–west Jinjiao–Lingshui fault zone in the south of Hainan Island being the boundary, the Sanya area to the south of the fault zone belongs to the Sanya platform edge depression zone of the South China Sea platform, while the north of the fault zone belongs to the South China Fold System. As the main part of Hainan Island, the South China Fold System is divided into the Wuzhishan Mountain Fold Belt to the south of the Wangwu–Wenjiao fault, and the Leiqiong depression to the north.
Figure 1. Geographical location of Hainan Island and distribution of lower soil samples used for prediction of thermal mineral water sites, along with known sites. 1: Geotectonic boundary; 2: Quaternary sediments; 3: Cretaceous sedimentary rocks; 4: Mesoproterozoic–Paleozoic sedimentary–metamorphic rocks; 5: Cenozoic basic volcanic rocks; 6: Paleozoic–Mesozoic intermediate-acid intrusive rocks; 7: known thermal mineral water sites (S: natural spring); 8: known thermal mineral water sites (B: borehole); 9: bedded thermal mineral water in northern Hainan Island; 10: distribution of lower soil samples used for prediction.

Sedimentary rocks exposed on the surface of Hainan Island originate from all eras, except for the Devonian and Jurassic periods, starting from the oldest Mesoproterozoic strata to the Cenozoic erathem. Among them, the Mesoproterozoic to Paleozoic strata have undergone different degrees of metamorphism and are distributed in an island-like manner in the middle of Hainan Island within intermediate-acid intrusive rocks; the Mesozoic strata consist mainly of Cretaceous red beds distributed in areas such as Baisha, Leiming, and Yangjiang; the Cenozoic strata consist mainly of Quaternary rocks distributed around the island and along the Wangwu–Wenjiao fault. Intrusive rocks cover the most extensive area of the island, distributed mainly in the central area of Hainan Island, and consisting mostly of intermediate-acid rocks and acidic rocks, more than 60% of the lithology of which is monzogranite. Volcanic rocks are mainly distributed in the northern part of Hainan Island, and are Cenozoic Neogene–Quaternary mafic volcanic rocks.

The 32 rift-type belt distributions of thermal mineral water found on Hainan Island are mainly located in the central part of the island, in the area of intermediate-acid intrusive rocks and sedimentary–metamorphic rocks, with small amounts exposed in the Quaternary and Cretaceous red bed areas. In the northern part of Hainan Island, there are porous layers of thermal mineral water, 29 of which have been exploited—mainly in the central area of Haikou.

3. Material and Methods
3.1. Sampling and Analysis

Selection of soil samples: All sampling data were obtained from the multipurpose regional geochemical survey project of Hainan Island conducted in 2004–2011. The method of collecting samples was geochemical measurement with two-layer grid sampling. Surface samples at depths of 0–20 cm were taken, with a density of 1 sample per 4 km². The deep soil samples (150–200 cm) were generally collected at the center of each 16 km² grid. Deep soils are more profoundly influenced by subsurface thermal mineral water than surface soils, and at the same time are less affected by the secondary effects of the surface environment. Therefore, deep soil samples from the multipurpose regional geochemical
survey of Hainan Island were selected as carriers for indicating subsurface thermal mineral water. The total number of samples was 2197, and the analytical test items included 54 elemental indicators, such as Hg content, pH, and SiO$_2$ content. Results for some of these elements of solid samples are shown in Table 1. The analysis methods were the same as those of Li Min et al. [24].

Table 1. Results of the indicators in the solid samples (unit: mg/kg).

|        | Hg  | As  | Pb  | Zn  | Cu  | Mn  | Th  | Be  | Ce  | Sn  |
|--------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| Mean   | 0.024 | 9.79 | 33.9 | 46.1 | 10.68 | 362 | 23.0 | 2.28 | 91.5 | 4.8 |
| Min    | 0.001 | 0.08 | 5.8  | 3.4  | 0.24 | 37  | 0.1  | 0.18 | 8.1  | 0.6 |
| Max    | 0.167 | 309.4 | 143.3 | 294  | 97.45 | 4291 | 122.5 | 9.6  | 706.6 | 35.9 |

3.2. Methods

Selection of known thermal mineral water outcrops and their evaluation indicators: The thermal mineral water temperature results obtained by the two collection methods are shown in Table 2. A total of 9 of the 32 identified fissure-type zones of thermal mineral water were found in the Quaternary (7) or Cretaceous strata (2), while the others were found in intermediate-acid intrusive rocks (17) or sedimentary–metamorphic rocks (6). Considering that sources of soil materials in the Quaternary and Cretaceous strata have a longer material migration distance, only the remaining 23 thermal mineral water sites were included in the analytical study. The thermal mineral water sites B2 and B1 are shown in Figure 1; among them, site B2 thermal mineral water was not used because it was too close to site B1, meaning that we could not distinguish it from site B1 in terms of precision of soil sampling (one sample per 16 km$^2$). The measured temperatures of thermal mineral water at 22 locations were used as the calibration indices for thermal mineral water, and their corresponding elemental contents were calculated based on the average of four surrounding deep soil samples (with distance as the calculation weight). However, these four samples needed to have the same geological background as the location of the thermal mineral water; otherwise, only 1–3 samples of the same geological background were taken for calculation.

Table 2. Actual measured temperature of thermal mineral water.

| Borehole | No. | B1   | B2   | B3   | B5   | B6   | B11  | B12  | B17  | B18  | B21  | B23  | B24  | B27  | B28  | B29  | B32  |
|----------|-----|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|
| T (°C)   |     | 83   | 59   | 70   | 73.5 | 38   | 47   | 78   | 58   | 75   | 63   | 84   | 54   | 49   | 78   | 47   | 63   |
| Natural Spring | No. | S4   | S7   | S8   | S9   | S10  | S13  | S14  | S15  | S16  | S19  | S20  | S22  | S25  | S26  | S30  | S31  |
| T (°C)   |     | 39.5 | 41   | 40   | 51   | 51.5 | 49   | 38   | 34.5 | 48   | 34   | 41   | 35   | 77   | 45   | 72   | 68   |

Linear regression analysis was performed with the thermal mineral water temperature as the dependent variable, and with the 54 elemental indicators of the deep soil corresponding to the thermal mineral water at 22 locations as the independent variables; the regression method used was stepwise regression. The predicted temperatures of all deep soil samples were calculated based on the regression model developed.

The prospective sites for thermal mineral water prospecting were delineated based on the prediction intervals of the dependent variables and the content intervals of the independent variables of the 22 regression samples, as well as the geological backgrounds they represented and the regional fault of the location.

Data processing and mapping were conducted with the MAPGIS 6.5 (Zondy Cyber, China, Wuhan) or SPSS 11.0 software.
4. Results and Discussion

4.1. Regression Analysis Results

Table 3 shows the regression analysis coefficients for the 22 regression analysis samples.

Table 3. Liner regressive coefficients.

| Model | Unstandardized Coefficients | Standardized Coefficients | t | Sig. | 95% Confidence Interval for B | Tolerance | VIF |
|-------|-----------------------------|---------------------------|---|-----|-------------------------------|-----------|-----|
|       |                             |                           |   |     | Lower Boundary | Upper Boundary |       |     |
|       |                             |                           |   |     |                 |                   |       |     |
| 1     | Constant 16.9012            | 1.1343                    | 0.2701 | −14.1796 | 47.9820 | 1         | 1 |
|       | Th 1.8995                  | 0.5014                    | 2.5916 | 0.0174 | 0.3706 | 3.4284 | 1 |
| 2     | Constant 33.6614            | 2.3327                    | 0.0308 | 3.4590 | 63.8639 | 0.8337   | 1.1995 |
|       | Th 2.6679                  | 0.7042                    | 3.8055 | 0.0012 | 1.2005 | 4.1352 | 0.8337 | 1.1995 |
|       | Be −13.2723                | −0.4973                   | −2.6874 | 0.0146 | −23.6093 | −2.9353 | 0.8337 | 1.1995 |
| 3     | Constant 11.9989            | 0.7599                    | 0.4571 | −21.1728 | 45.1706 | 1         | 1 |
|       | Th 2.3603                  | 0.6230                    | 3.6832 | 0.0017 | 1.0140 | 3.7066 | 0.8000 | 1.2501 |
|       | Be −12.6880                | −0.4754                   | −2.8648 | 0.0103 | −21.9928 | −3.8331 | 0.8311 | 1.2032 |
|       | Ce 0.3154                  | 0.3688                    | 2.3871 | 0.0282 | 0.0378 | 0.5930 | 0.9587 | 1.0431 |
| 4     | Constant 10.3082            | 0.7178                    | 0.4826 | −19.9907 | 40.6072 | 1         | 1 |
|       | Th 1.9977                  | 0.5273                    | 3.3020 | 0.0042 | 0.7213 | 3.2741 | 0.7404 | 1.3507 |
|       | Be −14.8234                | −0.5554                   | −3.5819 | 0.0023 | −23.5548 | −6.9020 | 0.7852 | 1.2735 |
|       | Ce 0.2898                  | 0.3389                    | 2.4035 | 0.0279 | 0.0354 | 0.5442 | 0.9497 | 1.0530 |
|       | Sn 3.6943                  | 0.3391                    | 2.1957 | 0.0423 | 0.1444 | 7.2442 | 0.7915 | 1.2634 |

Note: VIF: variance inflation factor; Sig.: significance level.

The basic idea of stepwise regression analysis is that the selected variable can be eliminated when it becomes unimportant after the introduction of new variables, and the eliminated variable can be reselected into the equation when it becomes important after the introduction of new variables. This can avoid multicollinearity and obtain a relatively simple model with fewer variables, in which only a few variables with significant effects are usually retained, thus having a better simulation prediction effect [25].

In the fourth step of the stepwise regression analysis (model 4), only 4 of the 54 elemental indicators—namely, Th, Be, Ce, and Sn—entered the regression equation. From the standardized coefficients (without the effects of elemental content magnitude), the elements Th and Be affected the temperature of thermal mineral water most significantly, but Be had a negative effect. Since Be is mainly enriched in pegmatite and during pneumatolysis of the late stage of intermediate-acid magma intrusion [26,27], in terms of time, the temperature of this phase is obviously lower than that of the early intrusion phase, while in terms of space, it occurs mostly at structurally open tectonic sites, where the temperature will be further decreased. Therefore, the entry of Be into the regression equation indicates the locations of convergent outcrops of thermal mineral water, i.e., tectonic sites with high Be levels and cooling. As mentioned above, the thermal mineral water sites in the rift belt on Hainan Island are strictly controlled by the respective fracture tectonics. In addition, the acidic intrusive rocks in which Th, Ce, and Sn occur are the heat sources for the thermal mineral water. Thus, the combination of the elements is a common manifestation of the thermal source and the propagation sites of the thermal mineral waters, with great geological significance.

The significance levels of the coefficients of the independent variables in model 4 were all less than 0.05, indicating that the four independent variables were significantly indicative of the thermal mineral water temperature; the corresponding index tolerance was
relatively large, and the variance inflation factor was relatively small (< 10), thus allowing the rejection of the hypothesis of covariance between them.

The regression Equation (1) was obtained from the unstandardized coefficients as follows:

\[
\text{Thermal mineral water temperature (°C)} = 10.3082 + 1.9977 \times W_{Th} + (-14.8234) \times W_{Be} + 0.2898 \times W_{Ce} + 3.6943 \times W_{Sn} (1)
\]

where \( W \) is the original unstandardized content value of each element.

In fact, a reasonable linear regression analysis requires an existing linear correlation between the dependent and independent variables. The correlation analysis showed that the correlation coefficients between the thermal mineral water temperature and the elemental content of Th, Be, Ce, and Sn at 22 locations were 0.5014, −0.2101, 0.4764, and 0.4024, respectively, which were the largest or smallest relative to the other elements. In addition, the correlation coefficient between the predicted and measured temperatures from the regression analysis was 0.8240 at a 0.01 significance level, making it highly significant. Therefore, the linear regression model was considered reasonable, and could be used for thermal mineral water temperature prediction calculations based on corresponding deep soil samples.

4.2. Calculation of Predicted Values of Thermal Mineral Water Temperature and Sample Selection of Deep Soils

Based on the above regression equation, the predicted thermal mineral water temperature was calculated for all deep soil samples. Overall, 2197 samples had a predicted maximum value of 403.95 °C and a minimum value of −106.54 °C, with a mean value of 59.20 and a standard deviation of 39.97 °C. Naturally, this was a purely mathematical calculation, and trade-offs were necessarily made while considering many factors, such as geological background.

The geological backgrounds of the 22 regression samples were all intermediate-acid intrusive rocks or sedimentary–metamorphic rocks; hence, the same geological backgrounds were required for deep soils, meaning that only 1257 samples were suitable. The content intervals of the four independent variables of the regression samples were 12.05–29.55 for Th, 1.17–3.77 for Be, 47.00–122.85 for Ce, and 1.90–7.89 for Sn (all units in mg/kg), which were also used as the qualifying conditions for deep soil sample selection, and only 555 samples remained out of 1257 samples. The range of regression prediction values of regression analysis samples (25.20–87.83 mg/kg) was also used as one of the qualifying factors, so that only 547 out of 555 samples remained. Under the three mutually inclusive conditions, the 547 samples were the most equivalent predicted samples to the regression analysis samples; this was the fundamental basis for delineating the prospective thermal mineral water areas, and its spatial distribution is shown in Figure 1.

4.3. Thermal Mineral Water Prospecting Distant Site Trapping

As mentioned earlier, the rift-type belt distribution of thermal mineral water in Hainan Island is strictly controlled by the relevant faults, and almost all of them are east–west and north–east directional. Therefore, in addition to the above-mentioned deep soil prediction sample points as the fundamental basis for delineating prospective areas of thermal mineral water, faults are also one of the essential bases.

The regional faults within the control area of the predicted sample point were the necessary elements for classifying the prospective sites (areas). The main fault zones on Hainan Island are listed in Figure 2a. The sample sites with east–west or north–east directional fracture structures passing through them, intersected by other directional faults, were considered to be A1-level prospecting sites; those with only east–west or north–east directional faults passing through them were considered to be A2-level prospecting sites; those with only other directional faults passing through them were considered to be A3–
level prospecting sites; and those without faults passing through them were considered to be B-level prospecting sites.

Figure 2. Distribution of major fault zones and prospecting sites of thermal mineral water on Hainan Island (a). Sketch of the main faults on Hainan Island (b). Prospective sites of thermal mineral water on Hainan Island. ① Wangwu–Wenjiao fault zone; ②: Changjiang–Qionghai fault zone; ③: Jianfeng–Diaoluo fault zone; ④: Jiusuo–Lingshui fault zone; ⑤: Guangcun–Puqian fault zone; ⑥: Gezhen fault zone; ⑦: Baisha fault zone; ⑧: Yacheng–Gangbei fault zone; ⑨: Qiongshan–Shuhe fault zone; ⑩: Puqian–Beiao fault zone. 1: A1-level prospecting site; 2: A2-level prospecting site; 3: A3-level prospecting site; 4: B-level prospecting site; 5: predicted temperature 25–40 °C; 6: predicted temperature 40–50 °C; 7: predicted temperature 50–60 °C; 8: predicted temperature 60–70 °C; 9: predicted temperature >70 °C.
The prospective thermal mineral water sites in the rift-type belt distribution on Hainan Island are shown in Figure 2b. Of the 547 prospective sites, 91 were at A1-level, 90 were A2, 185 were A3, and 181 were B. The distribution of prospective sites by temperature interval is shown in Table 4; among them, 14 of the A1-level prospecting sites had predicted temperatures of 25–40 °C, 27 of 40–50 °C, 25 of 50–60 °C, 16 of 60–70 °C, and 10 of >70 °C; 11 of the A2-level prospecting sites had predicted temperatures of 25–40 °C, 29 of 40–50 °C, 30 of 50–60 °C, 30 of 60–70 °C, 16 of 60–70 °C, and four of >70 °C; 29 of the A3-level prospective sites had predicted temperatures of 25–40 °C, 63 of 40–50 °C, 43 of 50–60 °C, 33 of 60–70 °C, and 17 of >70 °C.

Table 4. Distribution of the number of prospective sites in each temperature interval.

| Number of Prospective Sites | 25–40 °C | 40–50 °C | 50–60 °C | 60–70 °C | >70 °C |
|-----------------------------|---------|---------|---------|---------|--------|
| A1                          | 14      | 27      | 25      | 16      | 10     |
| A2                          | 11      | 29      | 30      | 16      | 4      |
| A3                          | 29      | 63      | 43      | 33      | 17     |

Note: A1, A2, and A3 are the levels of prospective sites classified by regional fault structure.

5. Conclusions

A linear regression analysis was performed based on the temperature of known thermal mineral water (the dependent variable) and the content of deep soil elemental indicators from a multipurpose regional geochemical survey of Hainan Island (the independent variable); among the 54 basic indicators, 4 indicators—Th, Be, Ce, and Sn—entered the regression equation. The regression model has significant geological significance for thermal mineral water temperature prediction.

Under the constraints of requirements such as the geological background represented by the regression analysis samples, the independent variable content interval, the prediction interval of dependent variables, and the regional faults, 547 prospective thermal mineral water sites were delineated, of which 16.6% were at A1-level. The proportion of A1-level sites that were >70 °C constituted 11% of all A1-level prospective sites, which was greater than the proportion of the A2-level (4%) and A3-level (9%) sites, reflecting the superiority of east–west and north–east directional regional faults in controlling the distribution of thermal mineral water.

The information cited in this study is of low precision, limited completeness, and limited accuracy. Each sample represents an area of 16 km², so higher precision chemical prospecting and other work are required in order to search for thermal mineral water within the predicted prospective areas. The temperature acquisition methods used in drilled wells and natural outcrops are different, with the latter being less accurate. The thickness of the overlying strata from the elevation of the thermal mineral water outcrop to the surface at the wellhead—as one of the evaluation indicators of thermal mineral water exploration—was also an important indicator for this study; however, this indicator was also not considered, because drilling evaluation work has not been carried out at some of the thermal mineral water outcrops, and the thickness of the cover is unknown. The focus of further research should be to enhance the exploration and evaluation of known thermal mineral water outcrops, and to accurately measure the evaluation indicators in order to provide a scientific basis for understanding of the unknown factors.

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