Hydrochemical formation mechanism of mineral springs in Changbai Mountain (China)

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Abstract
Changbai Mountain area is an important mineral water storage and development area in China. The hydrochemical composition of mineral water is the decisive factor for mineral–water quality. Based on the hydrochemical data of 74 mineral water samples collected from 2018 to 2020, the characteristics and formation mechanism of the hydrochemical components of the mineral water were analyzed. The results show that the formation of single-type mineral springs (metasilicate mineral water) is controlled by rock weathering; compound mineral springs (metasilicate mineral water with high CO2 content) are the product of CO2-rich, weakly acidic, confined hot groundwater with high salinity, which are mixed with shallow groundwater as rising along the fracture. The volcanic geological process greatly influences the formation of the hydrochemical components of mineral springs on the North slope of Changbai Mountain. The mineral springs on the Longgang Mountain are greatly affected by human activities. The results of cluster analysis only consider that hydrochemical components are consistent with the classification of the areas which concentrated distributions of mineral as determined by hydrogeological and geomorphological studies. The results of this study are useful for understanding the distribution, hydrochemical characteristics, and formation mechanism of mineral springs in the Changbai Mountain area of China and provide the theoretical basis for the protection and development of mineral spring.

Keywords Hydrochemical formation mechanism · Cluster analysis · PCA · Metasilicic-acid mineral springs · The Changbai Mountain

Introduction
Mineral water is a kind of special groundwater formed under special hydrogeological conditions. This kind of water typically contains a certain amount of minerals and some trace elements. It is not polluted and it is a kind of precious water resource for human consumption. Mineral water generally has the characteristics of good oral feeling, fine water quality, and some minerals and trace elements needed by the human body, and hence, it is favored by people for consumption (Peh et al. 2010). The Changbai Mountain area in China is characterized by active geologic processes, and multiple stages of volcanic activities which have resulted in the formation of basalt, trachyte, pumice, and other magmatic rocks here. These rocks constitute the lava platform and volcanic cone of Changbai Mountain and Longgang Mountain (Meng-Meng Li et al. 2021a, b). The cracks and pores of volcanic rocks are very developed (Gao et al. 2011). The widely distributed volcanic eruptive clastic sediment favors the proliferation of forests and vegetation and promotes the recharge of groundwater by meteoric water. These factors have created unique conditions for the formation and enrichment of mineral water in the Changbai Mountain area (Bian et al. 2019). During groundwater flow, hydrogeochemical interactions occur between groundwater and the surrounding rock and soil, dissolving some mineral components and trace elements in the volcanic rocks and forming abundant

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and high-quality mineral water resources. Mineral springs, mainly of the metasilicic-acid (H₂SiO₃) type, are distributed radially around the main peak of the Changbai Mountain (Li et al. 2019). At present, this area is an important mineral water source in China and it is among the three major high-quality mineral water sources in the world, along with the Alps and Caucasus mountains.

Since mineral spring is directly discharged groundwater, the research methods used for groundwater can be applied to study mineral water. Research on the hydrochemical characteristics and formation mechanism of mineral spring mainly involves the use of graphic analysis, descriptive statistics, and multivariate statistical methods; in addition, hydrogen and oxygen isotopes and a variety of radioisotopes are used to analyze the formation time and cycles of cold spring and hot spring (Prtoljan et al. 2012; Haklidir 2013). The characteristics, origins, and influencing factors of the hydrochemical components of mineral springs are analyzed based on the statistical analysis of hydrochemical data and isotope method (Kopylova et al. 2011; Chelnokov et al. 2018). The dissolution rules of mineral components and the hydrogeochemical formation mechanism of mineral water were explored using TOUGHREACT and PHREEQC simulators and by water–rock interaction experiments (Choi et al. 2014; Yan et al. 2016). The conceptual model of the groundwater system was constructed by using the method of system science, and the groundwater system was characterized (Kulkarni et al. 2000; Dafny et al. 2006; Daniele et al. 2020). The origins, dynamics, and influencing factors of mineral spring flow were studied by wavelet analysis (Gao et al. 2016). Multi-factor statistical analysis and comprehensive evaluation methods were used to evaluate the quality of mineral water as daily life water (Leite et al. 2018). For mineral springs with industrial and medical care value, the distribution, content and function of special components, and development and utilization were studied (Vinograd 2004). Considering the development and utilization value of mineral water resources, the hydrochemical characteristics, types, distribution, development, and utilization modes of mineral springs were studied (Barut et al. 2004; Corral et al. 2014).

Research on mineral spring water in the Changbai Mountain area began in the 1980s. The initial research focused on the distribution characteristics of mineral water resources and the qualitative analysis of mineral spring formation. At present, studies on mineral water resources in Changbai Mountain mainly focus on their formation causes, runoff pathways, and the recharge relationship between mineral water and surface water (Yan et al. 2014). The main methods are a combination of isotopic, hydrochemical and numerical simulations (Zhang et al. 2017; Li et al. 2022). Most studies only analyze the genesis and hydrochemical characteristics of individual mineral. In contrast, mineral water in the Changbai Mountains densely distributed and inter-related in terms of water quantity, quality and dynamics. Although the mineral springs in the Changbai Mountain area distributed throughout the region, most of them are concentrated in certain small areas. Therefore, it is necessary to study spring clusters by considering them as a whole. In recent years, the development of mineral water resources has increased, and mineral water development has gained importance as a major industry. The mineral water in this area is developed by delimiting the protected areas where mineral springs are concentrated and establishing the mineral water industry base to realize the centralized development of mineral water in the protected areas. Within a given area with a high density of mineral springs, this kind of spring is interrelated in terms of water quantity, water quality, and dynamics, all kinds of the springs within the given area are a unified whole. Because of the continuous increase in development and utilization, problems such as reduction in water quantity and change in water quality have been observed in some kinds of the springs. This will also affect other mineral springs and underground aquifers within the same spring-concentrated area. The existing has studied the mineral springs in this area mostly, analyzing the genesis and hydrochemical characteristics of individual mineral springs. However, most springs in the area occur in clusters. Therefore, considering the integrity of the basin and groundwater system, the study area was divided into three areas with dense distribution of mineral springs, namely the Longgang Spring Group in Jingyu County (LG), the Western Slope of Changbai Mountain Spring Group in Fusong County (WS), and the Northern Slope of Changbai Mountain Spring Group in Antu County (NS). On the basis of hydrogeological survey and the sample collection, the hydrochemical data of mineral springs are analyzed by geostatistics, model construction, principal component analysis method and cluster analysis method. Determine the characteristics and formation mechanism of the hydrochemical components of the spring water in different regions. The objectives of this study are: (1) to classify the mineral springs, and reveal the hydrochemical characteristics; (2) to analyze the controlling factors of hydrochemical formation in different types of mineral water; (3) to describe the hydrochemical formation mechanism of single-type mineral spring and compound mineral springs. It provides a theoretical basis for protecting mineral water resources and ensuring sustainable development.

**Study area**

Changbai Mountain is the highest mountain range in Northeast Asia and is located at the border between China and Korea (Fig. 1). It is the largest volcanic mountain range in the humid areas of China. The volcanic geological process
is active, and the lava and pyroclastic rocks ejected form a giant volcanic cone and basalt platform with the Tianchi caldera as the center (Yan et al. 2018; Na et al. 2020). The total area of the basalt platform is 5500 km², and the average thickness is about 180 m. The climate of the study area is the East Asian monsoon climate with abundant rainfall and annual average precipitation of more than 600 mm. Songhua River, Yalu River, and Tumen River originate in this region. The forest cover is more than 90%. The interception and absorption of rainwater by vegetation promote the infiltration of rainwater. Groundwater is mainly supplied by atmospheric precipitation and converted to surface water and discharged into a low-lying valley (Wang et al. 2021). The main types of groundwater are basalt pore-fissure water, pore water in loose rock, karst-fissure water in carbonate rock, pore-fissure water in clastic rock, and fissure water in granite rock (Fig. 1).
Basalt has the characteristics of pore development, good connectivity, and high silica content and is an ideal medium for groundwater storage and migration. Material exchange occurs between basalt and groundwater, forming abundant metasilicic-acid-type mineral water resources. Mineral springs are concentrated in the low basaltic platform of Longgang Mountain and the basaltic plateau area of Changbai Mountain, forming a spring group. The low platform of the Longgang Mountain is located in the western part of the study area and has an altitude of 500–1200 m. In this platform, more than 170 craters were formed by the accumulation of basaltic trass (Fan et al. 2002). The Changbai Mountain basalt plateau is located under a giant volcanic cone, and around the volcanic cone, the altitude gradually decreases from a height of 1200 to 600 m (the valley) (Guo et al. 2014).

Data and methods

Sample collection and analytical method

A total of 74 mineral water samples were collected in 2018–2020 (Fig. 1). Considering the influence of season and precipitation, we sampled mineral water twice a year, respectively in July (wet season) and October (normal season), so as to analyze the changes of mineral water in different periods. These samples were collected from 8 springs in LG, 5 springs in WS and 8 springs in NS, respectively. We collected mineral water according to the flow direction and covered all types of springs in the study area. The sampling bottles were polyethylene bottles and glass bottles, which were washed with distilled water in advance. Before sampling, the sampling bottles were rinsed thoroughly with the spring water from the target sampling point thrice, and the pH value was measured on site. Subsequently, the water samples were sealed and labeled, stored at 4 °C, and sent to the Testing Science Experimental Center of Jilin University and Jilin Provincial Institute of Geological Sciences for water chemistry testing.

The mineral water parameters were tested according to “Methods for the examination of drinking natural mineral water” GB 8538-2016 (National Health Commission of the People’s Republic of China 2016). The test items include HCO₃⁻, Cl⁻, SO₄²⁻, Na⁺, K⁺, Ca²⁺, Mg²⁺, NO₃⁻, F⁻, TDS, TH, TFe, H₂SiO₃, etc. Among them, Na⁺ and K⁺ were determined by flame atomic absorption spectroscopy (FAAS), the detection limit is 1 ug L⁻¹ and the determination accuracy is 1%; Ca²⁺ and Mg²⁺ were determined by EDTA disodium salt titration, the detection limit is 0.02 mg L⁻¹ and the determination accuracy is 1%; SO₄²⁻ was determined by titration with Phenolphthalein and methyl orange, the detection limit is 1 mg L⁻¹ and the determination accuracy is 1%; Cl⁻ was titrated with silver nitrate, the detection limit is 1 mg L⁻¹ and the determination accuracy is 1%; NO₃⁻ and F⁻ were determined by ion chromatography, The detection limits were 0.01 mg L⁻¹ and the determination accuracy was less than 5%; Total dissolved solids (TDS) determination by electronic balance by dry weight method, the detection limit is 20 mg L⁻¹ and the determination accuracy was less than 5%; Total hardness (TH) by EDTA disodium titration, the detection limit is 10 mg L⁻¹ and the determination accuracy is 3%; Total hardness (TH) by EDTA disodium titration, the detection limit is 10 mg L⁻¹ and the determination accuracy is 5%; TFe was determined by inductively coupled plasma mass spectrometry, the detection limit is 0.05 mg L⁻¹ and the determination accuracy is 5%; TFe was determined by HG-AAS, the detection limit is 0.25 ug L⁻¹ and the determination accuracy is 5%. Other trace elements such as Sr²⁺, Li⁺ and Zn²⁺ were determined by FAAS, the detection limits were 0.01 mg L⁻¹, 0.05 mg L⁻¹, 0.05 mg L⁻¹ and the determination accuracy was less than 5%.

Data analysis

1. The hydrochemical data were analyzed by mathematical statistics. The Piper trigram can be plotted using the Grapher program. By distinguishing the types of mineral springs, the Gibbs diagram and the relationship graph of Ca²⁺/Na⁺ and Mg²⁺/Na⁺, Ca²⁺/Na⁺ and HCO₃⁻/Na⁺ (lithologic end element ratios diagram) were drawn to determine the dominant factors of hydrochemical formation. Based on the geophysical data and geological and hydrogeological conditions, the typical genetic diagram of a regional compound mineral spring was drawn. This diagram depicts the formation process of a series of compound mineral springs.

2. Open-source software R was used for statistical calculation and mapping and principal component analysis (PCA). The histogram of the contribution rate and loading diagram were drawn. The steps in PCA include calculating the mean value and standard deviation, calculating the covariance matrix, calculating the covariance eigenvalues and eigenvectors, arranging the eigenvalues in the increasing order, retaining several larger vectors, and finally, transforming the data into the new space constructed by the eigenvectors (Yang et al. 2010; Zhu et al. 2017; Celestino et al. 2019).

3. Cluster analysis is also called group analysis. The number of samples classified is unknown before analysis. According to the similarity degree, the samples are divided into several clusters, which are unaffected by other factors. Cluster analysis can be classified into
Q-Mode clustering for the research object itself and R-Mode clustering for variables and indicators of the research object. This study aims to classify spring water samples; therefore, SPSS software was used for Q-Mode clustering analysis of hydrochemical data (Katrine, Turner et al. 2014; Li et al. 2019). The Euclidean method regards all the variables of the sample as equally important. Since there is no difference in importance of each hydrochemical component, the Euclidean method for measuring the linear distance between two points is used to calculate the distance between different samples. The ward algorithm, which takes the square Euclidean distance as the distance between the two clusters, is used to calculate the distance between clusters. Because the order of magnitude of different indicators is different, it is necessary to standardize the whole range 0–1 to obtain the cluster dendrogram of mineral water chemistry (Cloutier et al. 2008; Rahbar et al. 2020).

Results

Classification of mineral springs

Water–rock interaction results in a type of dynamic, natural liquid mineral under the controls imposed by the rock medium and structure. This leads to a natural distribution of mineral water hydrochemistry. The mineral springs in the study area are concentrated in different hydrogeological units and basins. To analyze the similarity and differences in the chemical characteristics and hydrochemical origins of mineral water under the control of different hydrogeological and geomorphological conditions, digital elevation model (DEM) data were used. Using the hydrological analysis function of ArcGIS, the basin in the study area was extracted and divided. This process included the following steps: (1) creating non-depression DEM; (2) performing flow direction analysis and flow statistics; (3) defining the minimum surface runoff, river linking, and grid river vectorization. By comprehensive consideration of basin division and mineral spring enrichment, three areas with a dense distribution of mineral springs were selected—the West Slope of Changbai Mountain (WS), the North Slope of Changbai Mountain (NS), and the Longgang Mountain (LG). Each of these areas is located in different hydrogeological and watershed units. The three areas form 26.55% of the research area, and contain 61 sampling points, accounting for 82.43% of the total number of sampling points (Fig. 1). The mineral springs within any one area are regarded as a group for classification analysis.

Mineral spring water parameters include the contents of lithium, strontium, zinc, metasilicic, selenium, free carbon dioxide (CO₂), total dissolved solids (TDS), and the thresholds of the parameters are listed in Table 1. If only one parameter meets the standard of “Drinking natural mineral water” GB8537–2018 (National Health Commission of the People’s Republic of China 2018), the spring is a single-type mineral spring. If more than one parameter meets the standard, it is a compound mineral spring. Compound mineral springs are relatively rare in the study area, and their special components are listed in Table 2.

The special component contents of each mineral spring were analyzed. The results show that the H₂SiO₃, CO₂, and TDS of most of the compound mineral springs meet the quality standards of “Drinking natural mineral water” GB8537–2018, and the Li⁺ and Sr²⁺ contents of some mineral springs meet the standards. According to the types of mineral springs and the area with dense distributions of mineral springs, the mineral springs in the study area were classified into five categories. They are: (1) the spring group of west slope of Changbai Mountain (WS springs), (2) the spring group of north slope of Changbai Mountain (NS springs), (3) the spring group of Longgang Mountain (LG springs), (4) other regional mineral springs, and (5) compound mineral springs. Mineral springs of category 1–4 are all single metasilicic-acid type.

Based on the hydrochemical data of various types of mineral springs, the hydrochemical characteristics and genesis of different types and regions of mineral springs were analyzed and studied. Single metasilicic-acid mineral springs are widely distributed in the study area. All the single-type mineral springs in this study are of the metasilicic-acid type. The special component contents of compound mineral springs are quite different from that of single-type mineral springs. The average content of metasilicic-acid is 70.90 mg L⁻¹ of compound mineral springs, which is higher than 43.79 mg L⁻¹ of single-type mineral springs. The types and contents of special

| Types                  | Parameter            | Threshold               |
|------------------------|----------------------|-------------------------|
| Metasilicic            | H₂SiO₃/(mg L⁻¹)      | ≥ 25.0 (When the concentration is 25.0–30.0 mg L⁻¹, the water temperature should be above 25 °C) |
| Lithium                | Li/(mg L⁻¹)          | ≥ 0.20                  |
| Strontium              | Sr/(mg L⁻¹)          | ≥ 0.20 (When the concentration is 0.20–0.40 mg L⁻¹, the water temperature should be above) |
| Zinc                   | Zn/(mg L⁻¹)          | ≥ 0.20                  |
| Selenium               | Se/(mg L⁻¹)          | ≥ 0.01                  |
| Free carbon dioxide    | CO₂/(mg L⁻¹)         | ≥ 250                   |
| Total dissolved solids | TDS/(mg L⁻¹)         | ≥ 1000                  |
components of different compound mineral springs are also different (Table 2). Most compound mineral springs are far away from each other and have almost no hydraulic connection. The geological structure and stratigraphic conditions in this area is complex. The main reason for the difference of mineral spring special component is the difference of hydrogeological conditions such as formation lithology, groundwater circulation process and spring water discharge mode.

### Table 2

| No. | Sample name | H2SiO3 | CO2 | TDS | Li | Sr |
|-----|-------------|--------|-----|-----|----|----|
| I   | Yunhai      | 77.55  | –   | 1365.66 | –  | 0.33 |
| II  | Shixi       | 40.30  | 396.60 | 2646.72 | 0.64 | 1.48 |
| III | Xianren-1   | 40.00  | 308.00 | 2343.00 | 0.20 | 4.33 |
| IV  | Xianren-2   | 56.47  | 305.56 | 1402.40 | –  | –   |
| V   | Bailong     | 50.05  | 734.00 | 1371.00 | –  | 0.45 |
| VI  | Sanhao      | 96.20  | 658.11 | 2029.68 | –  | –   |
| VII | Yaoshui-1   | 98.80  | 1188.00 | 1672.00 | 0.19 | 0.44 |
| VIII| Yaoshui-2   | 107.83 | 1056  | 1672.00 | 0.21 | 0.52 |

### Hydrochemical types and component features

The minimum, maximum, average, and coefficient of variations (CV) of 14 hydrochemical parameters are shown in Table 3. The changes in the mean contents of different hydrochemical parameters of all types of mineral springs show good consistency. Except for F− and H2SiO3, the content of each component in the North slope springs is the lowest, and the TFe value of West slope springs is low. The

### Table 3

| Site       | K+  | Na+ | Ca2+ | Mg2+ | Cl− | SO42− | HCO3− | NO3− | TDS | pH | TH | TFe | H2SiO3 | F− |
|------------|-----|-----|------|------|-----|-------|-------|------|-----|----|----|-----|-------|----|
| WS (n = 16)|     |     |      |      |     |       |       |      |     |    |    |     |       |    |
| Min        | 1.67 | 3.08 | 5.39 | 1.08 | 0.81 | 0.00  | 29.22 | 1.00 | 84.39 | 6.50 | 22.15 | 0.01 | 36.38 | 0.17 |
| Max        | 15.19 | 66.25 | 24.39 | 7.25 | 8.92 | 300.78 | 8.08 | 484.93 | 7.85 | 199.00 | 0.38 | 84.64 | 0.79 |
| Md         | 3.88 | 12.97 | 11.88 | 5.11 | 2.49 | 4.13  | 88.34 | 4.84 | 166.72 | 7.25 | 53.04 | 0.08 | 45.04 | 0.41 |
| CV         | 1.01 | 1.19 | 0.44 | 0.93 | 0.66 | 0.50  | 0.85  | 0.39 | 0.66  | 0.06 | 0.81  | 1.21 | 0.30  | 0.47 |
| NS (n = 23)|     |     |      |      |     |       |       |      |     |    |    |     |       |    |
| Min        | 1.56 | 4.90 | 4.37 | 2.41 | 0.46 | 1.70  | 43.10 | 0.64 | 104.28 | 7.00 | 24.09 | 0.00 | 35.02 | 0.29 |
| Max        | 3.60 | 15.60 | 18.53 | 22.14 | 4.91 | 7.66  | 184.91 | 10.00 | 294.44 | 7.89 | 137.45 | 0.26 | 59.13 | 1.34 |
| Md         | 2.75 | 8.54 | 7.91 | 5.33 | 1.80 | 3.82  | 64.80 | 2.29 | 139.37 | 7.33 | 42.13 | 0.07 | 50.63 | 0.80 |
| CV         | 0.18 | 0.28 | 0.47 | 0.73 | 0.63 | 0.43  | 0.42  | 0.81 | 0.27  | 0.03 | 0.53  | 1.10 | 0.30  | 0.47 |
| LG (n = 16)|     |     |      |      |     |       |       |      |     |    |    |     |       |    |
| Min        | 2.42 | 5.76 | 10.98 | 5.96 | 1.76 | 4.26  | 70.12 | 1.87 | 142.50 | 6.67 | 62.06 | 0.01 | 30.46 | 0.07 |
| Max        | 4.71 | 12.35 | 25.07 | 12.69 | 7.37 | 16.63 | 125.01 | 16.20 | 218.70 | 8.05 | 97.73 | 0.32 | 41.80 | 0.26 |
| Md         | 3.79 | 9.43 | 15.81 | 9.20 | 3.29 | 7.83  | 104.42 | 4.84 | 180.24 | 7.58 | 78.74 | 0.11 | 36.49 | 0.16 |
| CV         | 0.17 | 0.20 | 0.22 | 0.30 | 0.46 | 0.44  | 0.42  | 0.81 | 0.27  | 0.03 | 0.53  | 1.10 | 0.12  | 0.32 |
| Others (n = 11)|     |     |      |      |     |       |       |      |     |    |    |     |       |    |
| Min        | 0.88 | 3.90 | 5.28 | 2.23 | 1.61 | 0.00  | 36.00 | 0.00 | 54.00 | 7.07 | 27.62 | 0.00 | 26.80 | 0.16 |
| Max        | 6.50 | 15.85 | 22.76 | 22.10 | 13.49 | 8.88  | 216.99 | 5.00 | 322.24 | 8.15 | 145.97 | 0.29 | 63.01 | 0.36 |
| Md         | 2.62 | 8.58 | 13.96 | 9.07 | 4.75 | 4.62  | 102.45 | 2.06 | 181.39 | 7.67 | 74.69 | 0.12 | 38.31 | 0.23 |
| CV         | 0.67 | 0.45 | 0.42 | 0.80 | 0.97 | 0.53  | 0.61  | 0.83 | 0.47  | 0.05 | 0.56  | 0.79 | 0.25  | 0.22 |

Compound mineral springs (n = 8)

| Min        | 6.53 | 43.75 | 98.08 | 48.77 | 5.89 | 0.00  | 678.13 | 0.00 | 1365.66 | 6.16 | 655.23 | 0.04 | 40.00 | 0.32 |
| Max        | 25.68 | 214.05 | 181.61 | 152.36 | 42.60 | 46.06 | 1706.40 | 0.54 | 2646.72 | 6.98 | 903.65 | 3.30 | 107.83 | 1.50 |
| Md         | 11.66 | 108.40 | 135.79 | 107.10 | 15.73 | 8.48  | 1239.34 | 0.12 | 1754.61 | 6.56 | 798.57 | 0.81 | 70.90 | 0.65 |
| CV         | 0.48 | 0.50 | 0.24 | 0.33 | 0.75 | 1.69  | 0.24  | 1.54 | 0.24  | 0.04 | 0.09  | 1.50 | 0.37  | 0.62 |

n is the number of samples, and the unit of each parameter (except pH value) is mg L−1.
The hydrochemical characteristics of single-type mineral springs and compound mineral springs are quite different. The pH value of compound mineral springs is lower than that of the single-type mineral springs, and the $F^-$ content is lower than that of the North slope springs; in comparison with the single-type mineral spring, the content of $SO_4^{2-}$, $K^+$ is 2–5 times, that of $H_2SiO_3$ is 1.5–1 time, the content of $NO_3^-$ is one order of magnitude lower, and the content of other hydrochemical parameters is 1–2 orders of magnitude higher. The coefficient of variation of LG springs is relatively small except for $NO_3^-$, and the $K^+$, $Na^+$, $Ca^{2+}$, $Mg^{2+}$, $Cl^-$, $HCO_3^-$, TDS, TFe, and $H_2SiO_3$ are the lowest among all types of mineral springs. In addition to $SO_4^{2-}$ and $NO_3^-$, the coefficient of variation of the WS springs is relatively large, and the coefficients of variation of $K^+$, $Na^+$, $Mg^{2+}$, $HCO_3^-$, TDS, and pH are the largest among all types of mineral springs. The coefficients of variation of $SO_4^{2-}$, $NO_3^-$, TFe, $H_2SiO_3$, and $F^-$ in compound mineral springs are the largest.

The Piper trilinear diagram of various mineral springs was drawn using Grapher, and their hydrochemical types were analyzed (Fig. 2). From the trilinear diagram, it can be seen that $Ca^{2+}$ accounts for the largest proportion of cations in the North slope springs, and cations of other types of mineral springs are distributed in the mixed area, and there is no dominant cation. The distribution of anions of all types of mineral springs is concentrated, and $HCO_3^-$ is the dominant anion. The hydrochemical types of all types of mineral springs are basically consistent, including the $HCO_3^--Ca$ type, $HCO_3^--Ca-Na$ type and $HCO_3^--Na$ type, among which $HCO_3^--Ca$ type accounts is the dominant type.

**Discussion**

There are many differences and similarities in the characteristics of the hydrochemical components of different categories of mineral springs. This result reflects the similarities and dissimilarities in the hydrochemical formation mechanisms of mineral water. The hydrochemical origins of various mineral springs are analyzed by using the Gibbs diagram and lithologic end element ratios diagram. The hydrochemical characteristics of single-type mineral springs and compound mineral springs are quite different. Different hydrogeochemical processes have various effects on the formation of single-type mineral water chemical components in different areas. There are obvious differences in their hydrochemical components. By using PCA, cluster analysis, and other hydrochemical analysis methods, the hydrochemical genesis of mineral springs in each category are explored with regional geological and hydrogeological conditions.
Controlling factors of hydrochemical formation

The Gibbs diagram is drawn based on the hydrochemical data of each mineral spring. According to the position of each spring in the Gibbs diagram, the main sources of the chemical components dissolved in water are analyzed, and the hydrochemical genesis of mineral springs are determined (Fig. 3). Most of the single-type mineral springs are located in the rock-weathering control area in the middle of the Gibbs diagram, and rock weathering is the main factor determining the chemical composition of this type of mineral water. Meteoric water with very low mineral content leaches the surface soil and infiltrates the underground rock stratum. The groundwater flows through basalts and pyroclastic rocks, and the mineral components in the weathered and broken rocks are transferred to groundwater, forming the metasilicic-acid mineral water. Compound mineral springs are located in the evaporation zone and the periphery of the model. Because the Changbai Mountain is a humid area, evaporation has little effect on the formation of mineral water. This type of mineral spring is not the product of evaporation and concentration, but it is affected by the special conditions during formation. The formation mechanism of hydrochemistry has changed, resulting in the atypical characteristics of hydrochemical components.

The lithologic end element ratios diagram (Fig. 4) for the mineral springs in the study area can be used to determine the rock types involved in water–rock interactions. It shows that the main mineral springs are located in the silicate rock area (Fig. 4a) or between the fields of silicate and carbonate rocks (Fig. 4b). It shows that the hydrochemical components of mineral springs in Changbai Mountain are mainly controlled by silicate rocks and influenced by carbonate rocks. The influence of carbonate rocks on the WS springs and the NS springs is less than that of LG springs.

PCA of single-type mineral springs

The hydrochemical characteristics of mineral springs are controlled by the dominant hydrogeochemistry reactions, but the formation process of mineral springs is often affected by many factors. Different hydrogeochemistry reactions will produce different hydrochemical characteristics and components (Yang et al. 2016; Ligavha-Mbelengwa and Gomo 2020). PCA can be used to evaluate the degree of influence of different hydrochemical actions in the process of mineral spring formation and the similarity and dissimilarity of each hydrochemical component source. PCA is a statistical analysis method that simplifies multiple indicators into a small number of comprehensive indicators. It uses a few variables to reflect the information of the original variables as much as possible to ensure that the loss of original information is small and the number of variables is as small as possible (Lourenço et al. 2010; Huang et al. 2013). Na⁺, K⁺, Ca²⁺, Mg²⁺, Cl⁻, SO₄²⁻ and HCO₃⁻ are the seven main components in groundwater; TDS and TH are hydrochemical comprehensive parameters that have an important impact on water quality; The contents of F⁻, NO₃⁻, TFe and H₂SiO₃ in some springs are relatively high. Based on this, the above 13 hydrochemical parameters were selected for PCA. First, standardize the data. Then Kaiser–Meyer–Olkin (KMO) and Bartlett’s test were performed to determine the suitability of the data for PCA. The results showed that KMO value is 0.666, and the Bartlett’s test reached significant level (P = 0.00 < 0.01), indicating that there is a correlation between variables, which meets the requirements of principal component analysis (Jolliffe and Cadima 2016; Lorenz et al. 2021). PCA can be used for data analysis. Using R, which integrates statistical analysis and graphic display, 13 hydrochemical parameters of single-type mineral spring in the study area were simplified and analyzed, and the histogram of factor contribution rate and factorial plane of samples in PCA was drawn.

The factor contribution rate histogram can be used to help determine the optimal principal component. The abscissa represents the main component, and the ordinate represents the contribution rate. The first several principal components with larger contribution rates are the main components that should be selected. According to contribution rate of each factor the number of selected principal components is determined. The histogram of contribution rate is shown in Fig. 5. The selected principal components can downgrade

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**Fig. 3** Gibbs diagram: TDS against Na/(Na+Ca) of different types of springs (WS springs, NS springs, LG springs, other springs and compound mineral springs)
the dimension of the data and provide more information.
The number of selected principal components is usually determined by the cumulative contribution rate of no less than 85%. The cumulative contribution rate of the first five principal components is 85.03%, which is greater than 85%.

The selected first five principal components can comprehensively reflect most of the information of hydrochemical, and meet the selection requirements of principal components with eigenvalues $> 1$. Aim to make the loadings of each PC more differentiation, which can be more conducive
to professional interpretation. Varimax normalized rotation is carried out to obtain the factor loading matrix of PCA (Haag and Westrich 2002; Jolliffe and Cadima 2016; Zhu et al. 2017). The eigenvalues, variance, cumulative variance and factor loading matrix of single-type mineral springs are shown in Table 4.

Table 4 Factor loadings matrix of the principal component analysis (PCA) with Varimax normalized rotation, values greater than 0.5 are marked in bold

| Variables | Factor loadings matrix |
|-----------|------------------------|
|           | PC1  | PC2  | PC3  | PC4  | PC5  |
| K⁺        | 0.850| 0.316| 0.066| 0.015| 0.074|
| Na⁺       | 0.851| 0.338| 0.062| −0.085| 0.134|
| Ca²⁺      | 0.705| −0.402| 0.284| 0.254| 0.150|
| Mg²⁺      | 0.854| −0.256| −0.044| 0.156| −0.050|
| Cl⁻       | 0.207| −0.161| −0.154| −0.056| 0.824|
| SO₄²⁻     | −0.068| −0.215| 0.332| 0.205| 0.643|
| HCO₃⁻     | 0.982| −0.071| −0.012| 0.135| 0.004|
| NO₃⁻      | 0.109| −0.105| 0.942| 0.004| 0.017|
| TDS       | 0.972| 0.050| 0.016| 0.156| 0.075|
| TH        | 0.922| −0.240| 0.066| 0.131| 0.119|
| TFe       | 0.223| 0.015| 0.017| 0.943| 0.070|
| H₂SiO₃⁻   | 0.231| 0.846| −0.175| 0.141| −0.155|
| F⁻        | −0.264| 0.746| −0.004| −0.128| −0.247|

Table 4 shows that the contribution rate of the first factor is 46.1%, and it mainly comprises HCO₃⁻, TDS, TH, Mg²⁺, Na⁺, K⁺ and Ca²⁺; it represent the process that mineral components in different types of rocks enter the groundwater, and various hydrochemical components of groundwater increase with the increase in the water–rock interaction time. Because of the different types of rock and soil, the flow length and the mineral contents in rock and soil, the factor loads of the main components in the first factor are relatively small. The contribution rate of the second factor is 17.6%, and it mainly comprises H₂SiO₃⁻ and F⁻ and represents the influence of volcanic activity and magmatic volatilization. In the volcanically active area, the release of F⁻ is closely related to deep magmatism and magma degassing. The solubility of silicate minerals increases with increase in temperature. In addition, the solubility of silicate minerals and the release capacity of F⁻ from rock and soil will also increase under weak alkaline conditions (Pachana et al. 2012; Sunkari et al. 2020). Hence, H₂SiO₃⁻ and F⁻ are the main components in the second factor. The contribution rate of the third factor is 7.8%, and it mainly comprises NO₃⁻. Human activities are the main source of nitrate, and hence, the third factor represents the impact of human activities (Singh et al. 2020). The contribution rate of the fourth factor is 7.1%, and it is mainly TFe, representing the high background value of iron in the rock matrix of Northeast China and the characteristics of pyroxene in trachyte and pantellerite that erupted in Holocene and are rich in Fe and poor in Mg (Li et al. 2004). The contribution rate of the fifth factor is

Fig. 5 Histogram of contribution rate of the first 13 principal components of hydrochemical principal component analysis
6.5%, and it mainly comprises Cl⁻ and SO₄²⁻, representing evaporation concentration.

PC1, PC2, and PC3 are the three factors that have the greatest impact on the chemical components of regional single-type mineral water. To analyze the influence degree of the three principal factors on springs located in different spring-concentrated areas, presenting the water samples on the PC1–PC2 and PC1–PC3 axes Factorial planes by using R (Fig. 6). Figure 6 show that there is no significant difference in the influence degree of weathering-leaching processes (the first factor) and volcanic geology (the second factor) on the hydrochemical components of the WS springs. Volcanic geology has a great influence on the formation of the hydrochemical components of the NS springs, but less influence on the LG springs. This is attributed to the different active degrees of regional volcanic geology. The massive volcanic eruption time of Longgang Mountain Volcanic Group is about 150,000 years ago. At present, the volcanic geological activity has weakened. The northern slope of Changbai Mountain has a developed fault structure, which comprises a ring structure and radial structure closely related to the giant volcano. The North slope springs are greatly affected by geological volcanic activity (Liu et al. 2009). The nitrate produced by human activities (the third factor) has a relatively large impact on LG springs, but a relatively small impact on the springs in the other regions—this is consistent with the fact that there are relatively more human activities in Longgang mountain area.

Cluster analysis of single-type mineral springs

The Q-Mode cluster analysis of mineral water chemistry is a method to classify different types of samples according to the similarities and differences in hydrochemical components (Lambrakis et al. 2004; Ghesquière et al. 2015). Only according to the chemical composition of mineral water, all single-type mineral springs are regarded as a single type for Q-Mode cluster analysis. In the analysis, the selection of hydrochemical parameters is consistent with the PCA. Q-Mode cluster analysis enabled the mineral springs to be divided into four categories, among these, cluster 12 and cluster 3–4 can be divided into two categories. On the whole, the content of hydrochemical components in cluster 3–4 is higher than that in cluster 1–2. The dendrogram of cluster analysis and the spatial distribution of mineral springs after clustering are shown in Fig. 7, and the average contents of hydrochemical components of mineral springs in different clusters are listed in Table 5.

The results of cluster analysis show that the majority of NS springs are in cluster 1; in the WS springs, cluster 2 accounts for the majority, and all the LG springs are in cluster 4, which shows that the hydrochemical characteristics within a spring cluster are similar, and different spring clusters show relatively large differences. Figure 7 and Table 5 show that the springs in cluster 1 are mainly located in the North slope springs area, along the second White River and third White River from south to north. It shows that the mineral springs in the upper reaches have a good hydraulic connection with those in the lower reaches, which are...
supplied with the groundwater of the basalt platform in the south. The volcanic geological process (the second factor in PCA) has a greater impact on this cluster of mineral springs. Most of the springs in cluster 2 are located in the West slope springs area. Most of these springs are supplied by basalt pore-fissure water with a flow rate of more than 10 Ls⁻¹ and are located near the river. The contents of most hydrochemical components are low, while those of $\text{SO}_4^{2-}$ and $\text{NO}_3^-$ are relatively high, indicating that they are closely related to surface water. The springs in cluster 3 are scattered and few, and most of the elements are present in relatively high contents. Some of the mineral springs are located in the bedrock area and the junction area between the basalt stratum and other types of strata, which are far from the supply area and concentrated areas of mineral springs. It shows that the flow path of this type of mineral spring is long, resulting in

Fig. 7 Dendrogram of Q-model hierarchical cluster analysis (HCA) including all water samples and spatial distribution of four types of mineral springs after cluster analysis
the inclusion of a large number of minerals from the rock and soil into groundwater. Most of the springs in cluster 4 are located in the LG springs area, and the distribution is relatively concentrated. Most of these springs are supplied by basalt pore-fissure water with a flow rate of more than 10 Ls⁻¹. The contents of H₂SiO₃ and F⁻ are low, whereas those of Cl⁻, SO₄²⁻, and NO₃⁻ are high. Human activities (the third factor in PCA) has a greater impact on this cluster. According to the topography, LG springs are located in Qinglong River valley (Fig. 1), which is surrounded by mountains. From the edge to the center of the valley, the amount of basalt pore-fissure water increased from less than 0.1 Ls⁻¹ to more than 10 Ls⁻¹. It shows that the mineral spring classified as cluster 4 in Longgang mountain area is the atmospheric precipitation of the mountains around the valley that infiltrates into the ground, flows in the basalt under the action of topography, and is exposed at the bottom of the valley.

The result of cluster analysis is consistent with the division of the concentrated distribution area of mineral springs. Because the hydrochemical composition is the only factor to be considered in cluster analysis, it shows that the hydrochemical composition of mineral water shows good consistency with the concentrated areas of mineral springs as determined by topography and landform. The division of the concentrated areas of mineral springs conforms to the basin conditions determined by topography and landform, as well as the hydrochemical characteristics of mineral springs.

### Formation of compound mineral springs

In the Gibbs map, some compound mineral springs are located at the periphery of the model, which shows that their formation is affected by special conditions. Tectonization and volcanic activity are strong in the study area. The ring structure and radial structure closely related to the volcanic activity are distributed around the main peak of the Changbai Mountain. There are many deep faults extending north–south, north–east, and north–west. Most of the groundwater in the deep fault is supplied by surface water and underground hot water, thereby affecting the mineral water quality (Chen et al. 2019). In the process of magmatism and metamorphism, CO₂ is released, lowering the pH value of groundwater. In addition, the temperature in the deep underground is higher. These factors increase the strength of the water–rock interaction. The groundwater circulation time in the deep fault is long, and a large number of minerals in the rock enter into the groundwater, increasing the content of each component and the TDS in the groundwater. The variation of mineral water temperature and atmospheric temperature in each month within a year is shown in Fig. 8. There are two lines each representing the springs in LG (gray), WS (green) and NS (red), one with a larger value and one with a smaller value, indicating the maximum and minimum temperatures of the springs, respectively. It can be found that the monthly variation of regional atmospheric temperature is large, and the monthly variation of mineral springs temperature is small. The temperature of single-type mineral springs is between 6 and 9 °C, and the temperature of compound mineral springs is higher than that of single-type mineral springs. The temperature difference between different compound mineral springs is also large. Through field observation, it can be seen that free CO₂ overflows occurs in the compound mineral spring, and travertine is attached to the spring mouth, and the rock surface has red iron oxide, and the pores are relatively developed (Fig. 9). The above analysis shows that the compound mineral spring is supplied by the underground hot water in the deep fault formed by geological volcanic activity, and the difference in hydrothermal cycle characteristics is the main reason for the temperature difference between different compound mineral springs.

According to the regional geological conditions, Xianren Spring, a high TDS compound mineral spring near the North–South fault zone, was selected for genetic analysis. The mineral spring is located in the Second White River Valley, and its location is marked in red in the NS region in Fig. 1. Because of the volcanic geological process of Changbai Mountain, north–south zonal geothermal reservoirs are buried underground. The geothermal reservoirs include shallow geothermal reservoirs in the fracture zone under the basalt caprock and a deep geothermal reservoir formed by the development of karst and structural fractures in the marble. Regional magma and magma chamber provide a stable
heat source for geothermal reservoirs. Based on measured data, previous studies and geothermal geological conditions, the genetic diagram of compound mineral springs is shown in Fig. 9 (Jin and Zhang 1994).

Figure 9 shows that there are local, intermediate, and regional groundwater flow systems in the region. Around the Changbai Mountain crater, radial and annular fractures are dense. Atmospheric precipitation and Tianchi lake water infiltrate into the rock fissures of high temperature and high pressure. Convection occurs under the action of temperature and pressure and rises along the marble fracture, forming a regional groundwater flow system from the main peak of Changbai mountain to the intersection of the fault structures in the Second White River valley. The regional groundwater flow system is mainly affected by volcanic geology. The groundwater in the upper part of the basalt platform is recharged by precipitation and the surface runoff of the giant volcanic cone in the upstream. Under the control of topography, the groundwater flows downstream and accumulates in the basalt fracture zone, forming an intermediate groundwater flow system from the upper part of the basalt platform to the discharge area of the lower valley. The intermediate groundwater flow system is mainly controlled by topography and geological structural conditions. Mineral springs are located in the valley with a low terrain. Atmospheric precipitation infiltrates into the basalt and gathers in the valley under the action of gravity. There are rivers and reservoirs in the upstream and downstream of mineral spring. The water levels of surface water and reservoir are higher than the groundwater level; hence, they supply groundwater and form a local groundwater flow system. The local groundwater flow system is mainly controlled by the terrain conditions. The regional and intermediate groundwater flow through the geothermal reservoirs and is heated; it then rises along the fracture connected with the mineral springs and mixes with the local groundwater flow with a lower temperature to form compound mineral springs. According to the above analysis, a part of the compound mineral springs in the study area is the product of dilution and subsequent mixing of the confined groundwater formed under the high-temperature and high-pressure conditions in the deep fault with the shallow groundwater as the confined groundwater rises along the fracture.

**Conclusions**

There are single metasilicic-acid mineral springs and compound mineral springs in Changbai Mountain area. Most of the single-type mineral springs are distributed in three areas. According to the types and distribution of mineral springs, they can be classified into five types: WS springs, NS springs, LG springs, other regional mineral springs, and compound mineral. The average values of hydrochemical parameters of all types of mineral springs show good consistency, and the hydrochemical type is mainly the HCO₃-Ca type. The content of metasilicic-acid in all mineral springs meets the standard of “Drinking natural mineral water.
(GB8537–2018)”, and the content of most hydrochemical parameters of compound mineral springs is 1–2 orders of magnitude higher than that of single-type mineral spring.

Using the general and special methods to explore the hydrochemical origin of mineral spring, it is concluded that the formation of single metasilicic-acid mineral spring is controlled by rock weathering, and that silicate rock is the main medium in the water–rock interaction. The hydrochemical formation mechanism of the compound mineral springs is different and shows the characteristics of high temperature and high CO₂. The formation of compound mineral springs is affected by the deep geothermal reservoirs of high temperature and high pressure and long-term groundwater circulation. It is the mixed product of the deep acidic hot water rising along the fault and mixed with the shallow groundwater.

PCA is used to judge the source of hydrochemical components and the influence of each factor on the hydrochemical components of mineral springs in different regions. The first five principal factors representing 85% contribution rate are extracted. The results show that the water–rock interaction of groundwater, regional volcanic geological process caused by volcanic activity and magmatic volatilization, human activities, high background value of iron in the rock, and evaporation concentration are the main reasons for the formation of the chemical characteristics of mineral water, with the contribution rates of the factors are 46.1, 17.6, 7.8, 7.1, and 6.5%, respectively. Volcanic geological process has a great influence on the formation of the chemical components of mineral water on the North slope spring, and the LG spring is greatly affected by human activities.

Using cluster analysis, the samples are classified into four categories: the mineral springs in cluster 1 are mainly located in the North slope springs area, and the mineral springs in cluster 1 on the North slope springs are supplied by the same groundwater aquifer. Most of the mineral springs on the West slope springs are in cluster 2, which is closely related to surface water. Some of the springs in cluster 3, which are scattered at the junction of different

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**Fig. 9** The genesis conceptual model of compound mineral spring, mineral water outlet and characteristics of mineral springs near rocks

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Legend
- Marbles of the Zhenzhumen Formation
- Quaternary basalt
- Fracture zone (Shallow geothermal reservoir)
- Deep fault and confined underground hot water

- Local groundwater flow system
- Intermediate groundwater flow system
- Regional groundwater flow system

- Meteoric water
- Compound mineral spring
- Deep hot fluid
- Shallow hot fluid

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strata. LG springs are all in cluster 4 and are distributed at the bottom of the valley, where enrichment and concentrated exposure of meteoric water is observed. The cluster analysis shows that the clustering results of only considering hydrochemical components are consistent with the division of the concentrated distribution area of mineral springs.

All the analysis results show that there are great differences between compound mineral springs and single-type mineral springs in the study area in terms of the hydrochemical characteristics and genesis. The compound mineral spring is a mixture of the confined groundwater formed in deep fractures and shallow cold groundwater, and the single-type mineral spring originates from meteoric water leaching the basalt. Mineral springs are distributed over the whole study area and concentrated in specific areas. Different hydrogeological conditions and human activities have different impacts on the mineral springs in variety regions, and there are obvious differences in their hydrochemical characteristics.

Author contributions JB: conceptualization, funding acquisition, project administration, resources, supervision. WS: data curation, investigation, methodology, writing—review & editing. JL: data curation, formal analysis, methodology, software, roles/writing—original draft. YL: investigation, validation. YM: investigation, visualization. YL: writing—review & editing. All authors read and approved the final manuscript.

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Declarations

Conflict of interest The authors have no competing interests to declare that are relevant to the content of this article.

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