

Evolution of hydrochemistry and isotopic characteristics in shallow alluvial aquifers of Poyang Lake Basin, Eastern China

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ABSTRACT

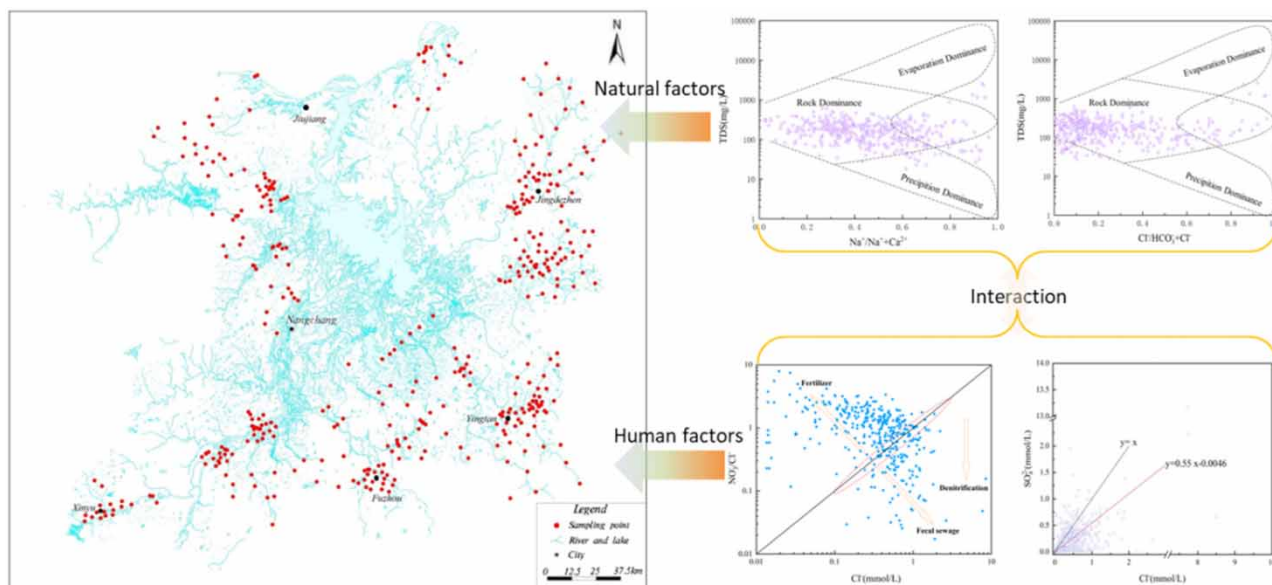
The hydrochemical characteristics of shallow groundwater and the factors affecting them were analyzed using descriptive statistical analysis, Piper trilinear charts, Gibbs charts, and ion ratios, to help improve the groundwater quality of the Poyang Lake Plain. The results showed that the groundwater was weakly acidic, with an average pH of 6.27. HCO_3^- was the main ion component, with high stability in the study area. The chemical ions in groundwater were mainly controlled by rock weathering, and Ca^{2+} and Mg^{2+} originated from the dissolution of carbonate, silicate, or evaporite. Cation exchange was an important driving factor in the formation of groundwater chemical components in the study area. The shallow groundwater and surface water in the region around Poyang Lake are replenished by precipitation, but both are more affected by evaporation. The large d-excess value indicated that the groundwater was greatly affected by water–rock interactions. The main source of NO_3^- is chemical fertilizer and NO_3^- levels were less affected by domestic sewage, which may be because the study area is a traditional agricultural area. Rock weathering, leaching, and human activities were the main driving factors for the evolution of the chemical characteristics of the groundwater in the study area. These results provide a technical reference and the basis for regional collaborative efforts to control groundwater and surface water pollution.

Key words: hydrochemistry, isotopic characteristics, Poyang Lake Basin, shallow groundwater, spatial analysis

HIGHLIGHTS

- The release of metal ions is accelerated in weakly acidic groundwater.
- Both natural and human activities influence the hydrochemical characteristics of groundwater.
- Nitrate comes from chemical fertilizer and domestic sewage in groundwater.
- The driving factors for the evolution of hydrochemical characteristics are rock weathering and human activities.

GRAPHICAL ABSTRACT



1. INTRODUCTION

Poyang Lake, located in the north of Jiangxi Province, China, is an important feeder for the Yangtze River and an important ecological economic zone (Shvartsev *et al.* 2016; Xu & Wang 2016). However, with rapid economic development, Poyang Lake has become polluted with nitrogen and phosphorus. The main sources of pollution in Poyang Lake are chemical fertilizers, livestock breeding activities, and urban sewage treatment plants, with pollutants entering the lake through surface runoff (Soldatova *et al.* 2017; Liao *et al.* 2018). However, groundwater is an important component of the water resources around Poyang Lake, and has a close hydraulic relationship with surface water. In fact, previous studies have shown that the surface water of the Poyang Lake basin (Ganjiang, Fuhe, Xiushui, Xinjiang) is composed of direct runoff, of which 23% is from precipitation and 77% is from the base flow of shallow groundwater (Hu *et al.* 2013; Liao *et al.* 2018). To understand the potential impact of groundwater on the water quality of Poyang Lake, it is necessary to study the changes in the chemical characteristics of groundwater due to the infiltration of surface water and atmospheric precipitation.

In recent years, researchers have analyzed the influence of the formation and evolution of groundwater on hydrochemical characteristics, which can effectively reveal the mechanisms involved in the interaction between groundwater and the environment and further identify the factors affecting groundwater quality (Gao *et al.* 2020; Solovey *et al.* 2021). Moreover, the types of surface water and groundwater hydrochemistry can be revealed using statistical diagrams from hydrochemistry analyses (Javadzadeh *et al.* 2020). The ratios of the main ions, Ca^{2+} and Mg^{2+} , and their combinations have been widely used to analyze the process of groundwater circulation and evolution, and to identify the factors influencing the chemical composition of groundwater and the degree of water-rock interaction (Xing *et al.* 2013; Xiao *et al.* 2017; Ramarosan *et al.* 2020). Stable hydrogen and oxygen isotopes (δD and $\delta^{18}\text{O}$) are ideal natural tracers for studying groundwater sources and evolution (Zhen *et al.* 2017; Li *et al.* 2019). Nitrate levels are generally low in groundwater, and their increase is also indicative of human activity (Wang *et al.* 2017).

In this study, we analyzed shallow groundwater hydrochemistry and the major ion ratios, hydrogen and oxygen isotopic composition, and hydrochemical characteristics of groundwater around the Poyang Lake area. Moreover, we investigated the regional groundwater recharge sources and the main water-rock interaction processes. The findings of this study will provide a greater understanding of nitrate pollution sources based on the regional groundwater evolution process, and provide support for regional water quality improvement programs.

2. MATERIALS AND METHODS

2.1. Study area

The region around Poyang Lake is located in the north of Jiangxi Province. It includes the cities Nanchang, Jingdezhen, and Yingtan, and 38 counties, including Jiujiang, Xinyu, Fuzhou, Yichun, Shangrao, and Ji'an (Figure 1). The total body of Poyang Lake covers an area of 51,200 km². The topography of the study area is a large asymmetric basin, with the Poyang Lake Plain as the base. The groundwater in the area can be divided into four types: loose rock pore water, clastic rock fissure pore water, carbonate-rock karst water, and bedrock fissure water. The depth of the shallow groundwater is 0.63–14.18 m, and the main sources of groundwater are atmospheric precipitation and river lateral recharge. The level of groundwater is affected by the topography. The groundwater in the whole region moves from east to west to the central region and from south to north according to the terrain and the runoff from hills and the low hill plain, with a general trend of migration to the central lake area.

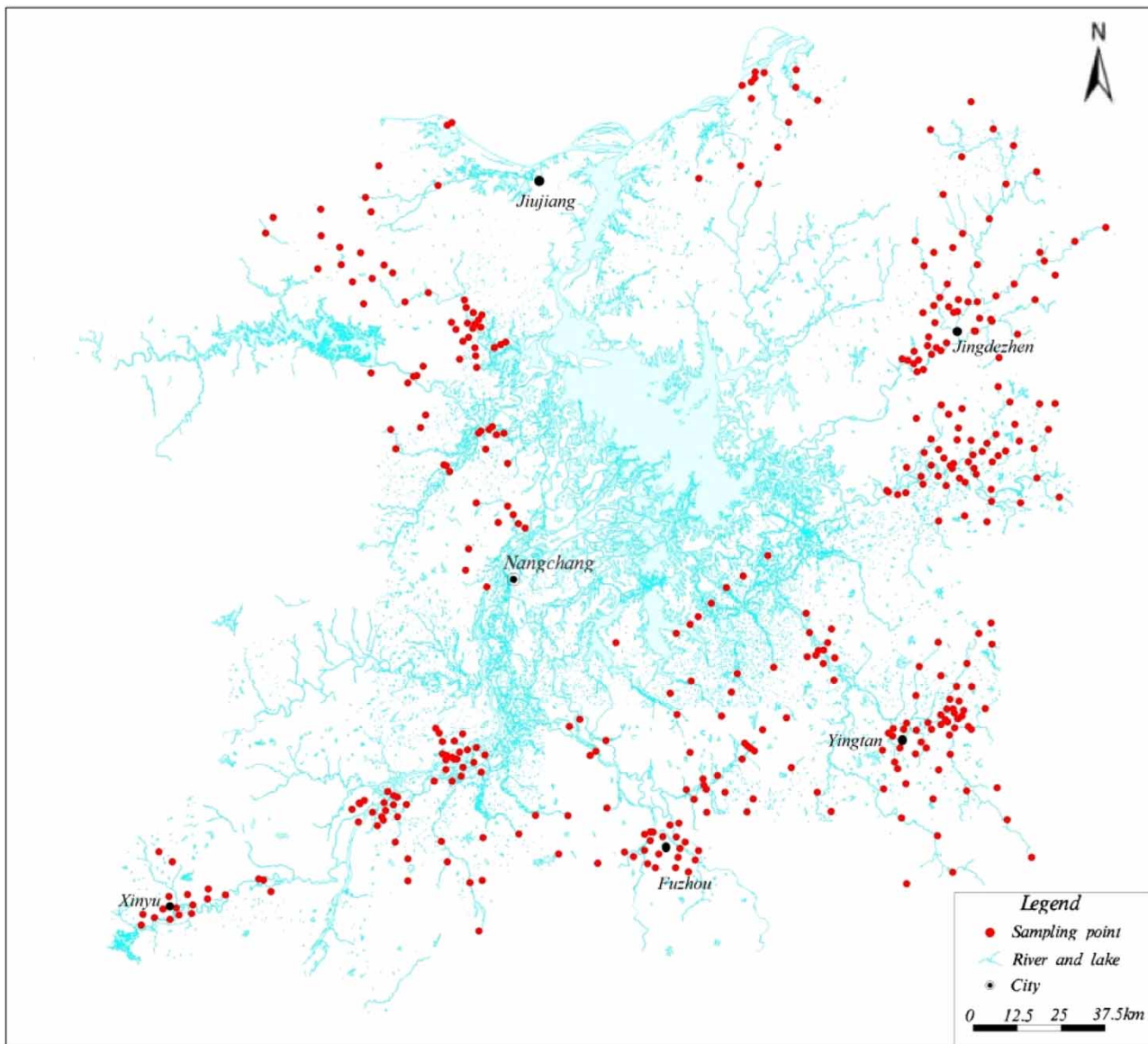


Figure 1 | Location of the study area and the sampling points of the shallow groundwater.

2.2. Sample collection and analysis

A total of 404 shallow groundwater sampling points were used to collect samples to test the hydrochemical indexes from the region around Poyang Lake from March 2018 to December 2019. A further 65 representative groundwater ($n = 37$) and surface water ($n = 28$) sampling points were also investigated. All the samples were collected at a depth of 0.5 m. The water was pumped to the site to test the stability of parameters before sampling, and then sampling was carried out. A 5 L plastic drum was filled with unaltered water samples to determine trinitrogen and inorganic salts. Before taking water samples, the sampling drum was washed with well water three times. Samples were tested in the laboratory within 1–7 days to ensure the authenticity and reliability of water quality testing data. Concentrations of Na^+ , K^+ , Ca^{2+} and Mg^{2+} were determined using inductively coupled plasma emission spectrometry. Total hardness (TH) was determined using the ethylenediamine di-sodium tetraacetic acid titration method. Total dissolved solids (TDS) was determined by the gravimetric method. Cl^- , SO_4^{2-} , NO_3^- and NO_2^- concentrations were measured by ion chromatography. NH_4^+ , CO_3^{2-} and HCO_3^- were analyzed with titrimetric methods. Stable isotopes of D and ^{18}O were determined using a gas isotope mass spectrometer. Isotope ratios are expressed in terms of deviation from Vienna Standard Average Ocean Water (VSMOW):

$$\delta(\text{‰}) = \frac{R_{\text{sample}} - R_{\text{vsmow}}}{R_{\text{vsmow}}} \times 1000$$

where δ is the isotopic deviation in ‰, R_{Sample} is the isotope ratio (D/H or $^{18}\text{O}/^{16}\text{O}$) in the sample, and R_{VSMOW} is the isotope ratio in VSMOW.

2.3. Chemical data assessment methods

A number of standard graphical and numerical techniques were used to assess the chemical data to determine the physical and chemical processes of groundwater in the study area. The Gibbs diagram and Piper diagram are widely used to estimate the origins of hydrochemical components in natural water (Gibbs 1970; Wang *et al.* 2017; Mu *et al.* 2021). The proportion of major components in groundwater reflects the hydrochemical processes that control groundwater chemistry (Marandi & Shand 2018; Yang *et al.* 2018). In the process of water evaporation, the dynamic fractionation coefficient of D is much larger than that of ^{18}O , resulting in deuterium excess (d-excess); d-excess is calculated based on the global meteoric water line (GMWL). When d-excess < 10 it represents the existence of secondary evaporation in the water (Craig 1961); d-excess can be calculated as follows:

$$d = \delta^{18}\text{O} - 8 \times \delta^2\text{H}$$

3. RESULTS AND DISCUSSION

3.1. Hydrochemistry of groundwater

Table 1 presents an overview of the statistical parameters established for the main ions in the phreatic aquifer of the urban agglomeration around Poyang Lake Basin. The results showed that the groundwater was weakly acidic, with a mean pH value of 6.27 ($n = 404$) and a minimum coefficient of variation of 0.76, indicating that the groundwater had a relatively stable pH. The order of anion and cation concentrations in groundwater was $\text{HCO}_3^- > \text{SO}_4^{2-} > \text{Cl}^- > \text{NO}_3^- > \text{NO}_2^-$ and $\text{Ca}^{2+} > \text{Na}^+ > \text{K}^+ > \text{Mg}^{2+} > \text{NH}_4^+$, respectively. HCO_3^- showed the largest mean concentration, but the coefficient of variation was relatively small. Therefore, HCO_3^- was the main ion in the groundwater and was present at high stability. The standard deviation of total dissolved solids (TDS) was higher than the standard deviation of the other components in the study area, indicating that the spatial distribution of TDS was significantly different.

The large coefficients of variation of NO_2^- , NH_4^+ , Na^+ , Cl^- , and SO_4^{2-} were mainly due to the large area from which measurements were taken, leading to large differences in ion concentration. This also indicates that these ions are easily affected by natural and anthropogenic activities. The maximum concentration of NO_3^- in the groundwater of the study area was 192.80 mg/L, with an average concentration of 23.77 mg/L, and the NO_3^- concentration exceeded groundwater quality standard Class III (20 mg/L; GB/T 14848 – 2017) in 44.44% of the sampling points. Moreover, the coefficient of variation was smaller for NO_3^- than for NH_4^+ and NO_2^- , indicating that the source of NO_3^- in the region was relatively stable, and it may be attributed to human activity.

Table 1 | Hydrochemical characteristics of the major ionic concentrations in the groundwater

Parameters	Min	Max	Mean	SD	CV
TH (mg/L)	1.70	507.90	101.48	78.42	0.77
TDS (mg/L)	18.30	2,557.00	197.10	208.48	1.06
pH	3.91	8.08	6.27	0.76	0.12
Ca ²⁺ (mg/L)	0.31	160.50	29.80	24.73	0.83
Mg ²⁺ (mg/L)	0.13	35.58	6.27	5.27	0.84
K ⁺ (mg/L)	0.24	84.65	7.59	10.80	1.42
Na ⁺ (mg/L)	0.99	764.20	17.82	55.75	3.13
Cl ⁻ (mg/L)	ND ^a	911.10	23.42	61.59	2.63
SO ₄ ²⁻ (mg/L)	ND ^a	1,265.00	30.57	76.39	2.50
HCO ₃ ⁻ (mg/L)	ND ^a	447.70	84.49	80.64	0.95
NO ₃ ⁻ (mg/L)	ND ^a	192.80	23.77	22.99	0.97
NO ₂ ⁻ (mg/L)	ND ^a	3.18	0.04	0.22	5.23
NH ₄ ⁺ (mg/L)	ND ^a	21.16	0.36	1.66	4.68

^aNone Detected.

TH: total hardness; TDS: total dissolved solids; SD: standard deviation; CV: coefficient of variation.

Piper trigrams can be used to analyze the content and evolution characteristics of the chemical composition of groundwater and further identify changes in the general chemical characteristics and control units in groundwater (Piper 1994; Zhang *et al.* 2017; Li *et al.* 2018; Adeyeye *et al.* 2021). As shown in Figure S1 (Supplementary Information), the groundwater in the study area had a variety of hydrochemical types. The main cations were Ca²⁺, Na⁺, and K⁺ and the main anions were SO₄²⁻ and HCO₃⁻. The high relative content of HCO₃⁻ indicated that the dissolution of carbonate rocks played a major role in controlling the hydrochemical characteristics of the groundwater. Strictly speaking, the Na⁺ and K⁺ content of the study area were significantly higher than the Ca²⁺ and Mg²⁺ content. Na⁺ and K⁺ mainly originated from the dissolution of potassium and sodium minerals in the aquifer. The sources of SO₄²⁻ in the groundwater environment were mainly sulfate minerals in sedimentary rocks, oxidized sulfide, meteoric atmospheric precipitation, and artificial input (chemical fertilizers) (Fang *et al.* 2021). In general, it is difficult to dissolve CaSO₄, which has low solubility, to produce SO₄²⁻. However, SO₄²⁻ content was higher than Cl⁻ content in the study area, indicating that sulfate in shallow groundwater was affected by meteoric precipitation and human activity.

3.2. Correlation analysis of main ions

Correlation analysis reflects the similarities and differences of the groundwater components, which is helpful in identifying the source consistency of each component. As shown in Figure 2, TDS was significantly correlated with the presence of ions in the groundwater ($r > 0.5$), and the strength of the correlation was in the order of Na⁺ > SO₄²⁻ > Cl⁻ > Ca²⁺ > Mg²⁺, indicating that Na⁺, SO₄²⁻, and Cl⁻ were the main ions contributing to TDS. There was a significant correlation between pH and Ca²⁺ and HCO₃⁻ levels, with correlation coefficients of 0.642 ($p < 0.01$) and 0.674 ($p < 0.01$), respectively. Moreover, the correlation coefficient between HCO₃⁻ and Ca²⁺ levels was 0.852 ($p < 0.01$). These results indicated that pH and Ca²⁺ and HCO₃⁻ levels were related to the weathering and dissolution of carbonate rocks. The correlations between NO₃⁻, NH₄⁺, NO₂⁻ and the other ions were weak, indicating that the source of nitrogen was different from the sources of other cations and anions. Groundwater nitrogen in the study area was mainly imported from external sources. These results indicated that the formation of the chemical characteristics of the groundwater around the Poyang Lake area was mainly affected by meteoric precipitation, water-rock interactions, and human activity.

3.3. Processes that influence groundwater hydrochemistry

3.3.1. Water-rock interactions

The shallow groundwater samples around Poyang Lake mainly lie in the water-rock interaction zone of the Gibbs diagram, with some points having a tendency to deviate towards meteoric precipitation (Figure 3). These results indicate that the

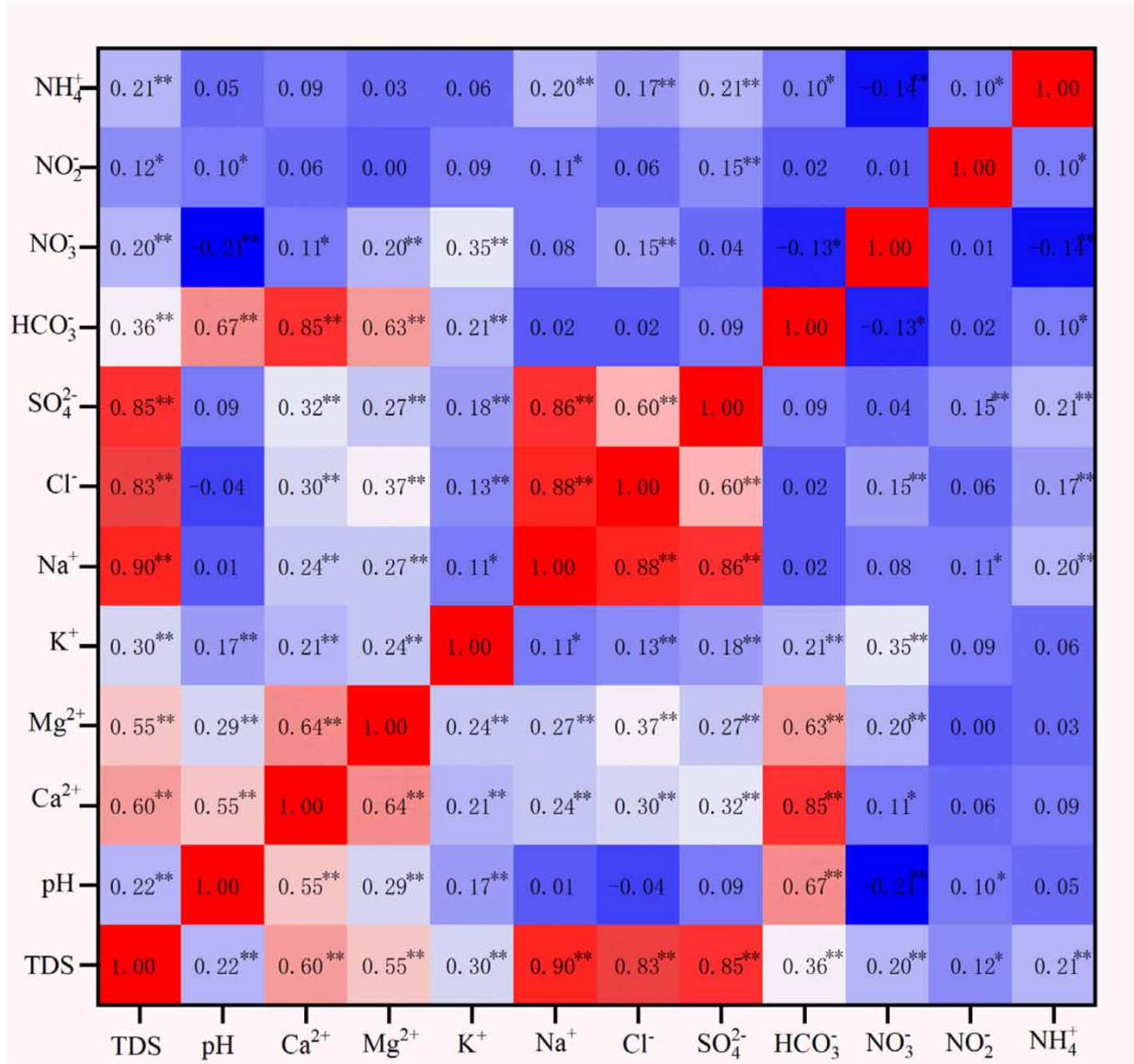


Figure 2 | Correlation heat map of the major ionic concentrations in the groundwater.

content of the major ions in the groundwater is mostly controlled by mineral dissolution in the aquifer matrix. In addition, meteoric precipitation had a certain effect on the ion content, but evaporation had no obvious effect. It can also be observed from Figure 3 that many samples fall outside the model block diagram, which may be due to the influence of human activity.

Changes in the ion ratio of soluble mineral components effectively indicates leaching in the aquifer (Figure 2, Supplementary Information). When the equivalent ratio of $(\text{Ca}^{2+} + \text{Mg}^{2+})$ to $(\text{SO}_4^{2-} + \text{HCO}_3^-)$ levels >1 , Ca^{2+} and Mg^{2+} mainly originate from the dissolution of carbonate in the groundwater, but when this ratio <1 , Ca^{2+} and Mg^{2+} mainly originate from the dissolution of silicate and evaporation (Qian *et al.* 2016; Guo *et al.* 2020; Liu *et al.* 2020). As shown in Figure S2 (Supplementary Information), these ratios were mainly located on both sides of the salt dissolution line (1:1). These results indicated that Ca^{2+} and Mg^{2+} content of the groundwater are mainly influenced by water-rock interactions and are mainly produced by the dissolution of carbonate and silicate and by evaporation (Zhu *et al.* 2021). More than 88% of the points for the relationship between $(\text{Ca}^{2+} + \text{Mg}^{2+})$ and HCO_3^- levels were distributed below the salt rock dissolution line (1:1 line), indicating that

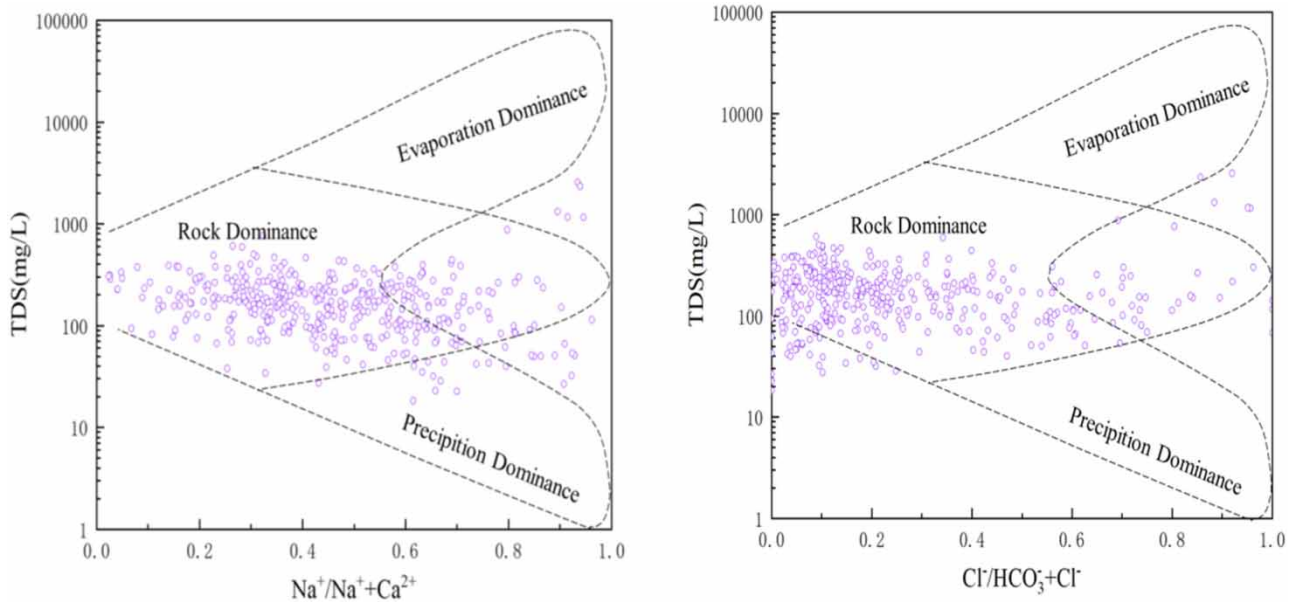


Figure 3 | Gibbs diagram of groundwater in the study area.

gypsum dissolution, but not carbonate mineral dissolution, generally occurs in the groundwater. Approximately 12% of the sample sites fell above the 1:1 line, which demonstrated that sodium (potassium) feldspar dissolution occurred in some aquifers in the study area. The Ca^{2+} formed after the dissolution of feldspar may affect the chemical balance of calcium bicarbonate in groundwater, and thus have a certain influence on the enrichment of HCO_3^- .

The correlation between $(\text{Na}^+ + \text{K}^+)$ and Cl^- levels reflects the dissolution of salt and feldspar minerals in the aquifer and the input of exogenous chloride ions (Sun *et al.* 2020). This ratio was distributed on both sides of the 1:1 line for most of the groundwater sampling sites in the study area. These results indicated that the sources of Na^+ and K^+ in the groundwater in the study area were different. In general, the dissolution of feldspar minerals was limited in most aquifers in the study area, indicating that positive cation exchange was the main reason for the relative enrichment of Na^+ and K^+ . The relative enrichment of Cl^- may have been greatly affected by exogenous sources in some of the groundwater sites in the study area.

3.3.2. Cation exchange

Cation exchange is an important factor affecting the chemical composition of groundwater. It has been found that the correlation between $(\text{Ca}^{2+} + \text{Mg}^{2+} + \text{HCO}_3^- + \text{SO}_4^{2-})$ and $(\text{Na}^+ + \text{Cl}^-)$ levels may be used to evaluate cation exchange between Ca^{2+} , Mg^{2+} , and Na^+ in aquifers (Wei *et al.* 2021). As shown in Figure 4, there was a significant negative correlation between $(\text{Ca}^{2+} + \text{Mg}^{2+} + \text{HCO}_3^- + \text{SO}_4^{2-})$ and $(\text{Na}^+ + \text{Cl}^-)$ levels in the groundwater ($r = 0.961$, $p < 0.01$). These results indicated that cation exchange was an important driving factor for the formation of the chemical components of groundwater in the study area.

3.3.3. Evaporation and concentration

The composition of the stable isotopes, δD and $\delta^{18}\text{O}$, in water is affected by meteorological processes, and their content and distribution characteristics are the basis for investigating and analyzing the source of groundwater recharge. The different positions of the δD and $\delta^{18}\text{O}$ data points of the water samples on the $\delta\text{D} - \delta^{18}\text{O}$ diagram reflect the different sources or formation processes of the water samples. If the data points fall on or near the meteoric water line, it indicates that the water sample originates from atmospheric water (Crawford *et al.* 2014). According to Craig (1961), there is a linear correlation between δD and $\delta^{18}\text{O}$ levels in global meteoric precipitation; therefore, the GMWL can be represented by $\delta\text{D} = 8\delta^{18}\text{O} + 10$. Chen *et al.* (2019) calculated the local meteoric water line (LMWL) in Lushan using the equation: $\delta\text{D} = 7.45\delta^{18}\text{O} + 8.36$. The slope and intercept of the LMWL were slightly smaller than those of the GMWL, which may be due to a certain degree of evaporation and may also indicate that water vapor circulation was more frequent in the study area than in previously studied

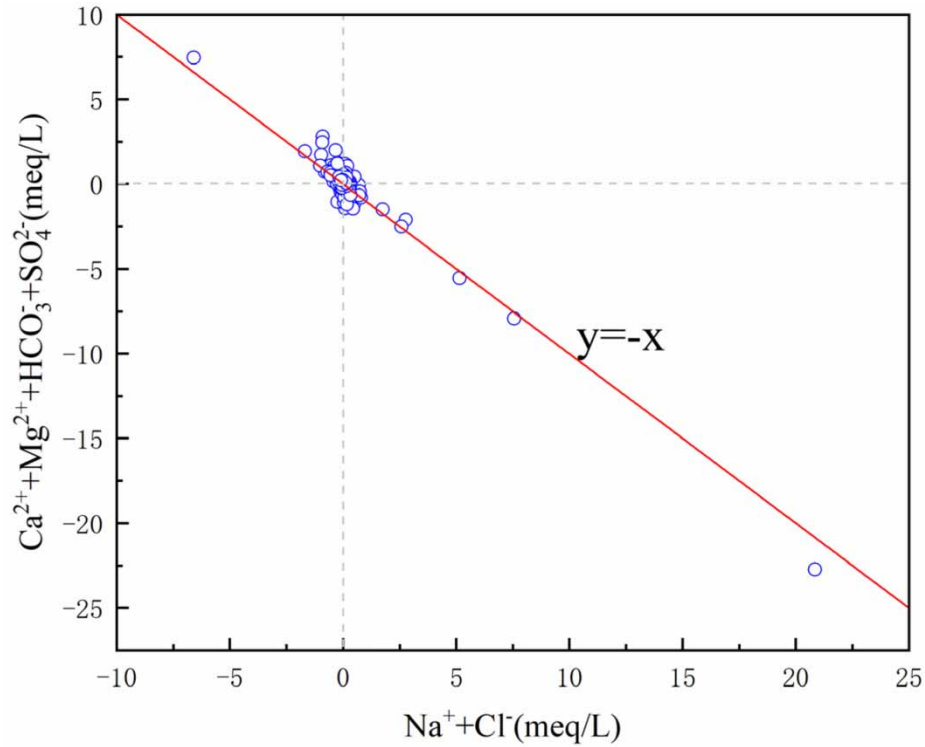


Figure 4 | Relationship between $(Ca^{2+} + Mg^{2+} + HCO_3^- + SO_4^{2-})$ and $(Na^+ + Cl^-)$ of groundwater.

regions. As shown in Figure 5(a), the slopes and intercepts of the evaporation lines of the surface water and groundwater were similar, and they were both smaller than those of the LMWL. The stable hydrogen and oxygen isotope compositions of the shallow groundwater and surface water were generally distributed on the upper right side of the LMWL, with a small number of samples from the region around Poyang Lake on the upper left side. These results indicated that surface water and

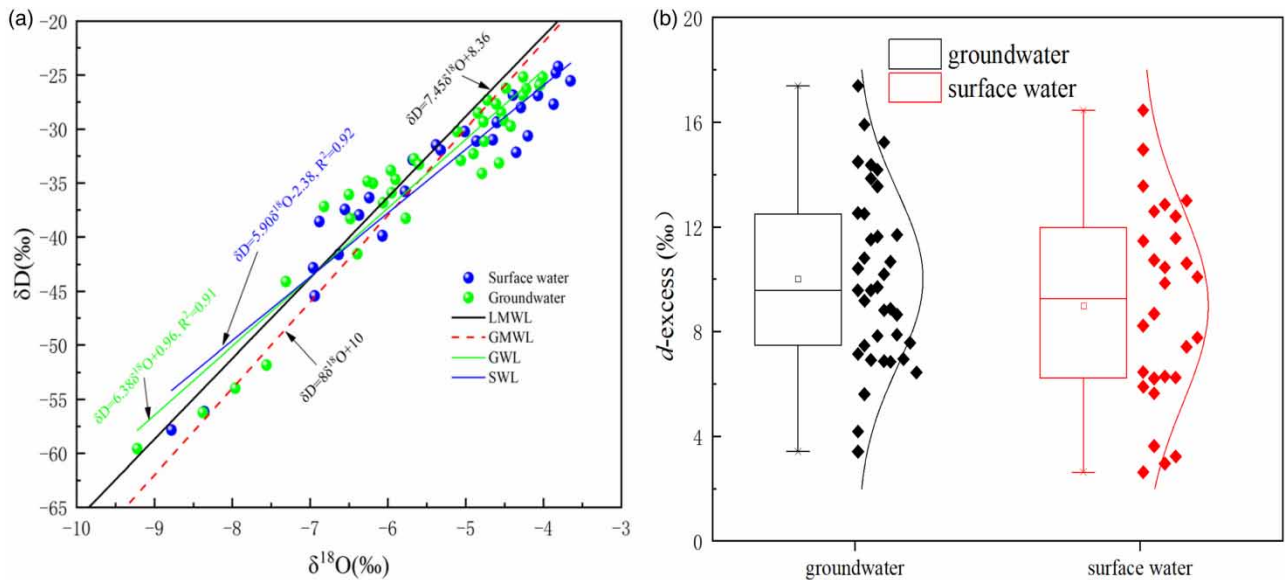


Figure 5 | Relationship between δD and $\delta^{18}O$ (a) and d -excess (b) analysis of groundwater and surface water.

groundwater interact with each other, are both recharged by precipitation, and are more susceptible to evaporation than to precipitation. The stable hydrogen and oxygen isotope levels in the surface water and shallow groundwater had a high degree of correlation, which further indicated a strong hydraulic relationship between groundwater and surface water.

The d-excess reflects the deviation of the hydrogen and oxygen isotopic compositions from the GMWL. The d-excess distribution of groundwater in the study area ranged from 3.42 to 17.39, with an average value of 10.01 (Figure 5(b)). The d-excess distribution of surface water ranged from 2.63 to 16.46, with an average value of 9.00. The d-excess value of groundwater was similar to that of the global average meteoric precipitation. Unexpectedly, the evaporation rate was lower for surface water than for groundwater. This may be because the sampling period was during the wet season, and surface water is easily affected by rainfall, whereas the groundwater was delayed due to evaporation in the dry season, which resulted in a large d-excess value. These results showed that groundwater is greatly affected by water-rock interactions.

3.4. Analysis of groundwater affected by human activity

Under natural conditions, the nitrate content of groundwater is usually low and constitutes a minor component. Therefore, the change in nitrate concentration in groundwater was used as an indicator of the effects of human activity. In addition, urban sewage contains SO_4^{2-} , which increases the concentration of SO_4^{2-} in the groundwater (Zhou *et al.* 2019).

The source of NO_3^- in groundwater was identified based on the ratio of $(\text{NO}_3^-/\text{Cl}^-)$ to Cl^- . When the Cl^- content was high and the $(\text{NO}_3^-/\text{Cl}^-)$ content ratio was low, the main source of NO_3^- in groundwater was manure and domestic sewage, but when the $(\text{NO}_3^-/\text{Cl}^-)$ content ratio was high and the Cl^- content was low, the main source was chemical fertilizer (Egbi *et al.* 2020; Zhang *et al.* 2020a, 2020b). As shown in Figure 6(a), the study area was a traditional agricultural area, and the sampling points fell within the range of a high $\text{NO}_3^-/\text{Cl}^-$ content ratio and low Cl^- content. The results showed that the main source of NO_3^- was chemical fertilizer. Some sampling points were distributed near the 1:1 line, indicating that NO_3^- mainly originated from chemical fertilizer, manure, and domestic sewage. Thus, these sampling points may be located in an urban-rural transitional area. Similarly, previous studies have found that domestic sewage and fertilizers are the principal sources of nitrogenous compounds (Soldatova *et al.* 2017).

The $\text{SO}_4^{2-}/\text{Cl}^-$ content ratios in most groundwater samples were less than 1 (Figure 6(b)), and the results showed that the SO_4^{2-} content was less affected by urban domestic sewage, which further indicated that the NO_3^- content of the groundwater was also less affected by domestic sewage and more affected by chemical fertilizers.

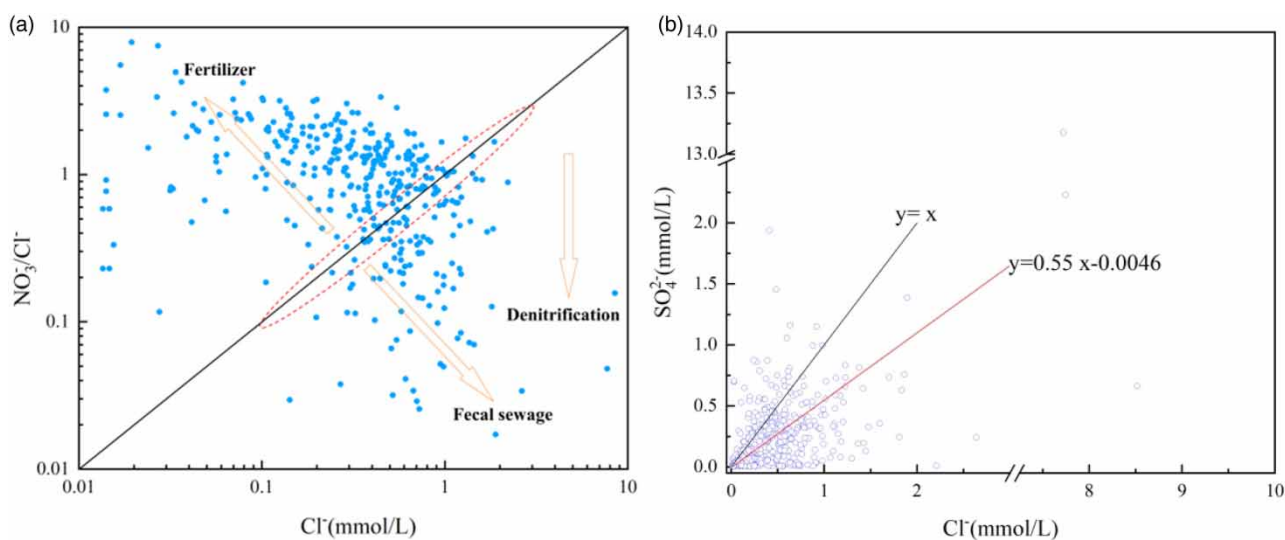


Figure 6 | Relationship of (a) Cl^- and $\text{NO}_3^-/\text{Cl}^-$ and (b) Cl^- and SO_4^{2-} in the groundwater.

4. CONCLUSIONS

The shallow groundwater around Poyang Lake was mainly derived from meteoric water. The average pH value was 6.27. The main cations were Ca^{2+} , Na^+ and K^+ in groundwater, and the main anions were SO_4^{2-} and HCO_3^- . There were significant differences in NO_2^- , NH_4^+ , Na^+ , Cl^- , and SO_4^{2-} , which demonstrates that these ions were susceptible to natural and human activity.

The hydrochemical characteristics of shallow groundwater were influenced by both natural factors and human factors. Natural factors include rock weathering, dissolution of dissolved minerals such as carbonate silicate or evaporite, and ion exchange. Human factors such as domestic sewage and chemical fertilizer lead to the difference of nitrate in groundwater in this region. In addition, precipitation also had a certain degree of influence on it, but the influence of evaporation was not obvious.

NO_3^- mainly comes from chemical fertilizers, feces and domestic sewage in the shallow groundwater around Poyang Lake. However, the value of $\text{SO}_4^{2-}/\text{Cl}^-$ in most groundwater sampling points was less than 1 in the study area. This result indicated that SO_4^{2-} was less affected by urban domestic sewage. Therefore, NO_3^- was more susceptible to the influence of chemical fertilizers in the groundwater.

CREDIT AUTHORSHIP CONTRIBUTION STATEMENT

Xiaodong Chu: Conceptualization, Investigation, Formal analysis, Software, Writing – original draft, Visualization, Writing – review & editing. Hao Wang: Supervision, Writing – review & editing. Liangzhong Li: Data curation, Writing – original draft, Funding acquisition. Zhifei Ma: Data curation, Writing – original draft. Daishe Wu: Validation, Data curation, Writing – original draft, Writing – review & editing. Ting Chen: Conceptualization, Funding acquisition.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict of interest.

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