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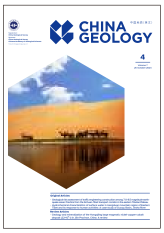

Hydrochemical characteristics of surface water in Hengduan mountain region of Eastern Tibet and its response to human activities: A case study of Duoqu Basin, Jinsha River

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ABSTRACT

The analysis of hydrochemical characteristics and influencing factors of surface river on plateau is helpful to study water hydrological cycle and environmental evolution, which can scientifically guide rational development and utilization of water resources and planning of ecological environment protection. With the expansion and diversification of human activities, the quality of surface rivers will be more directly affected. Therefore, it is of great significance to pay attention to the hydrochemical characteristics of plateau surface rivers and the influence of human activities on their circulation and evolution. In this study, surface water in the Duoqu basin of Jinsha River located in Hengduan mountain region of Eastern Tibet was selected as the representative case.Twenty-three groups of surface water samples were collected to analyze the hydrochemical characteristics and ion sources based on correlation analysis, piper trigram, gibbs model, hydrogen and oxygen isotopic techniques.The results suggest the following: (1) The pH showed slight alkalinity with the value ranged from 7.25 to 8.62. Ca^{2+}, Mg^{2+} and HCO_3^- were the main cations and anions. HCO_3-Ca and HCO_3-Ca·Mg were the primary hydrochemical types for the surface water of Duoqu River. The correlation analysis showed that TDS had the most significant correlation with Ca^{2+}, Mg^{2+} and HCO_3^-. Analysis on hydrogen and oxygen isotopes indicated that the surface rivers were mainly recharged by atmospheric precipitation and glacial melt water in this study area. (2) The surface water had a certain reverse cation alternating adsorption, and surface water ions were mainly derived from rock weathering, mainly controlled by weathering and dissolution of carbonates, and secondly by silicates and sodium rocks. (3) The influence of human activities was weak, while the development of cinnabar minerals had a certain impact on the hydrochemistry characteristics, which was the main factor for causing the increase of SO_4^{2-}. The densely populated county towns and temples with frequent incense burning activities may cause some anomalies of surface water quality. At present, the Duoqu River watershed had gone through a certain influence of mineral exploitation, so the hydrological cycle and river eco-environment at watershed scale will still bound to be change.The results could provide basic support for better understanding water balance evolution as well as the ecological protection of Duoqu River watershed.

1. Introduction

Surface water of plateau rivers is an important part of global hydrogeochemical cycle, an important channel for material and energy transfer at plateau, and plays an important indicator and regulation role in ecosystem balance and climate change (Li CC et al., 2021; Shi D et al., 2021; Tan H et al., 2021; Fu CC et al., 2023). River hydrochemical characteristics are the results of long-term interaction between water body and rock, soil, sediment, suspended matter, microorganisms (He XJ et al., 2019; Li XY et al., 2020). The analysis of chemical characteristics and influencing factors of surface river on plateau is helpful to study water geochemical process, hydrological cycle and environmental evolution, which can scientifically guide rational development and utilization of water resources and planning of ecological environment protection (Wang MG et al., 2022; Zhang T et

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al., 2018; Tang XW and Wu JK, 2014). Human activities such as mining development, animal husbandry and urban life affect the water cycle of regional surface rivers and the ecological environment dependent on surface rivers to some extent. Meanwhile, with the expansion and diversification of human activities, the circulation and evolution of surface rivers will be more directly affected. These changes of external conditions make the circulation and evolution of plateau surface water more complicated (Li WP et al., 2021; Wang MG et al., 2023). Therefore, it is of great significance to pay attention to the hydrochemical characteristics of plateau surface rivers and the influence of human activities on their circulation and evolution, which can promote water resource management and water quality security assurance.

There are many researches on surface river hydrochemistry in Qinghai-Tibet Plateau area. However, hydrochemistry mainly focuses on obtaining average information at large watershed scale (Li Z et al., 2022; Liu CY et al., 2023). For example, Yu TT et al. explored Characteristics of oxygen and hydrogen isotope distribution of surface runoff in the Lhasa River basin, and proposed that precipitation was the main recharge source and that the Lhasa River has experienced strong evaporation in the Lhasa River basin (Yu TT et al., 2010); Huang X et al. found that Ca^{2+} and HCO_3^- was the dominant ion and the concentration of dissolved metal ions was small in the study of Yarlung Zangbo River main stream river water (Huang X et al., 2011); Song HW and Yan YP et al. studied the isotopic characteristics of surface water and groundwater in the middle reaches of the Yarlung Zangbo River and their indicative functions (Song HW et al., 2021; Yan YP et al., 2022). Jiang L et al. studied the water chemical components and chemical weathering in the whole basin of the Yarlung Zangbo River and obtained the water body types in the whole basin (Jiang LG et al., 2015b).

In recent years, researchers have paid more and more attention to the study of water chemistry in Jinsha River. For example, Zhang LL et al. explored major ion compositions for water samples from Jinshajiang, Lancangjiang, and Nujiang drainage basins of China, and determined natural chemical weathering rates on the eastern Himalayan and Qinghai-Tibet Plateau (Zhang LL et al., 2016); Li J et al. analyzed the spatial variations of δ^2H and δ^{18}O in river and lake waters during flooding season, and revealed the factors underlying their variations along the middle and lower reaches of the Yangtze River (Li J et al., 2020); Wang SY et al. illuminated that the characteristics and influencing factors of stable isotopes in groundwater in the Permafrost Region of the Source Region of the Yangtze River, and further discussed the groundwater recharge sources (Wang SY et al., 2020); Wang MG et al. studied hydrochemical characteristics and material source of Requ River in eastern Tibet, and discussed the influence of urban life,farming and animal husbandry activities on the water chemistry of surface water and the primary ion sources (Wang MG et al., 2023). However, these studies have only focused on the main stream and some downstream tributaries of Jinsha River. As for systematic study on the main tributaries of Hengduan Mountain area of Jinsha River is seldom reported.

Based on the above factors, this paper takes the Duoqu River Basin of Jinsha River as the research object. The Duoqu River basin is located in the upper section of the Hengduan Mountains and belongs to the first class tributary of the Jinsha River, and hydrochemistry mainly focuses on obtaining average information at small watershed scale. The team collected 23 surface water samples, combined with piper trigrapH, gibbs model (Yi P et al., 2018) and isotopic techniques, identified the hydrochemical and isotopic characteristics as well as the hydrochemical evolution and circulation characteristics, and discussed the impacts of human activities such as mining development, animal husbandry and urban life on water quality. The research results could support the scientific development and rational utilization of surface water resources in Duoqu River Basin, and also provide reference significance for the study of surface water circulation and water resources sustainable utilization in other tributaries of Jinsha River.

2. Study area

Duoqu River is a primary tributary of the Jinsha River, mainly flowing through the eastern part of Jiangda County in eastern Tibet, flowing from northwest to southeast, and finally flowing into the Jinsha River in Boluo Township, with a total length of 12.65 km. The Duoqu River basin is located in the upper section of the Hengduan Mountains, belonging to the junction of the Pan-Chuaxia continent and the Gondwana continent in terms of tectonic units. It is formed by welding the four continental blocks (South Qiangtang-Zuogong, Gangdise-Nianqing Tanggula, Dege-Zhongdian, Changdu-Simao) and the suture zones between them (Lancang River, Jinsha River, Bangong Lake-Nujiang River). After the long-term depositional-tectonic evolution of the Nujiang Tethys Ocean, a complex geological structure pattern was formed.

The terrain in the Duoqu River basin fluctuates greatly, with the highest altitude of 5153 m and the lowest altitude of 2907 m. The relative elevation difference is about 2246 m, the average slope is about 47.2 ‰, the main river channel is about 12–55 m wide, and the average annual runoff of the river is about 800 million cubic meters. The overall topography is high in the north and low in the south, and the geomorp Hology of the basin develops slowly. The Duoqu River Basin is located in the plateau cold temperate semi humid climate zone. Rainfall in the basin is unevenly distributed in time, with frequent rainfall in summer and autumn and large water flow, which is prone to flood disasters. There is little rain in winter and spring, and the water flow is stable and small (Jia LR, 2017). The climate of the study area is mainly the plateau temperate subhumid mountain climate, because the mountain is high and the valley is deep, the climate has obvious vertical change. The annual sunshine duration is 1500–2700 hours, the average annual temperature is 2–10°C.

The main strata exposed from south to north in the study area belongs to Changdu-Simao strata area and West Jinwulan-Jinshajiang strata area, the main exposed strata are mesoproterozoic, paleozoic permian devonian and mesozoic triassic. The distribution of strata and sampling points in the study area is shown in Fig. 1. Gabbro, basalt, diorite, granite, limestone, dolomite, and volcanic rocks are the main rock types.The study area is mainly distributed in the northeast of Jiangda County. Animal husbandry is the main industry in the area, supplemented by agriculture and industrial foundation. Livestock mainly include sheep and pork. Mineral resources distributed in the area are mainly gold and cinnabar ores, followed by a small amount of copper and lead ores.

3. Materials and methods

3.1. Sample collection

In order to comprehensively consider the chemical evolution of surface water in the Duoqu River Basin, 23 groups of samples collected were relatively evenly distributed to ensure that the sampling points were distributed in different geological and geomorphological units, hydrogeological conditions and other background conditions, which can ensure that each sample point was representative.The water sample collection was generally 20 cm below the water surface, and three samples were collected from each sampling point using a 250 mL polyvinyl chloride sampling bottle. All samples passed through 0.45 μm membrane filtration, the water sample was added with 5 mL 1% nitric acid protection solution to test cations, so that the pH value of the water sample was less than 2, the remaining 2 vials were not acidified to test anions. 15 groups of water samples were collected at least 20 cm below the water surface using a Plexiglas container for δD, δ^{18}O isotopes analysis, 1500 mL of sample was collected at each sampling point.The air in the bottle was exhausted before sampling to avoid further isotope fractionation, all samples pass through 0.45 μm membrane filtration.

3.2. Sample analysis method

The chemical index of water samples were analyzed by the Hebei Geological Testing Center. The concentrations of Ca^{2+}, K^{+}, Mg^{2+} and Na^{+} were determined by flame atomic absorption spectromete (contrAA300, Jena, Germany). CO_3^{2-} and HCO_3^{-} were determined by hydrochloric acid titration analysis. Cl^{-}, SO_4^{2-}, F^{-} and NO_3^{-} were determined by ion chromatograph (883, Vantone, Switzerland). PH was measured by insitu multi-parameter water quality analyzer. The of total soluble solids (TDS) were determined by gravimetric method. The precision of the test method was expressed as the relative standard deviation (*RSD*), and the precision was expressed as the relative deviation (*RD*). The precision and accuracy of main anions and cations were shown in Table 1.The δD and δ^{18}O isotopes of the 15 groups of samples were analyzed by the liquid water isotope analyzer (L2130-i) with the test accuracy of 0.12 ‰ and 0.02‰, respectively.

4. Results and discussion

4.1. Hydrochemical characteristics of surface water

4.1.1. Characteristcs of major ions

23 groups of hydrochemical samples were collected in the study area. Table 2 showed the statistics of main ion contents in surface water. The results showed that Mg^{2+} and Ca^{2+} were

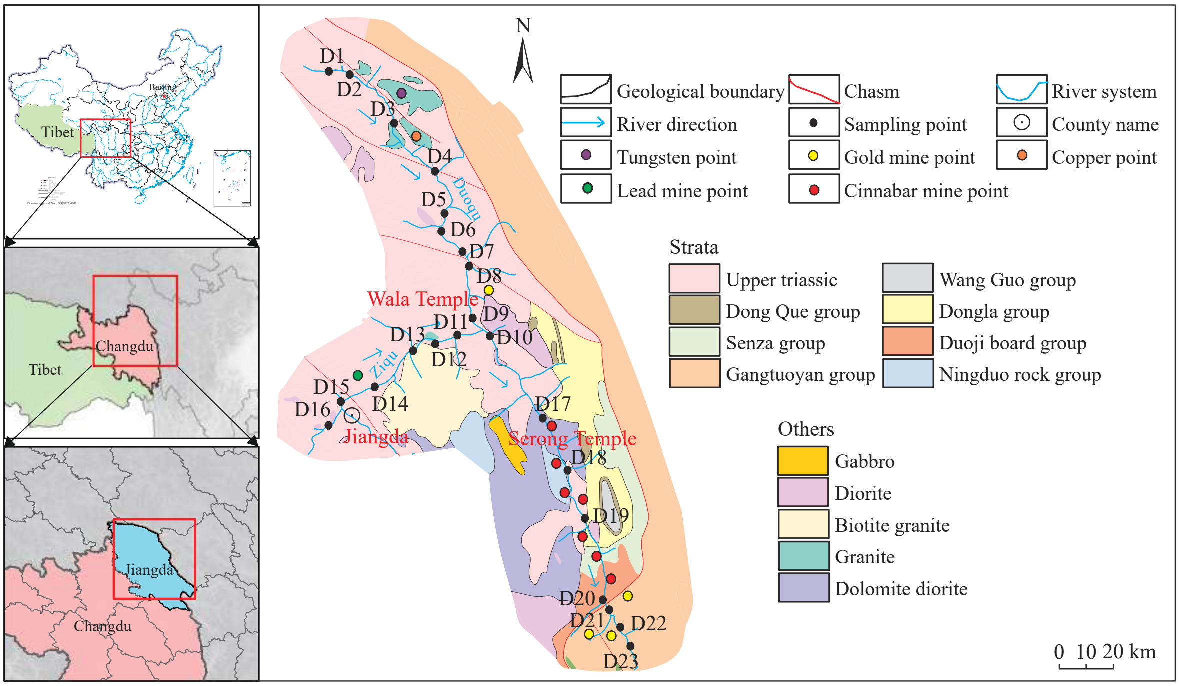


Fig. 1. Distribution diagram of strata and sampling points in Duoqu River Basin.

the main cations in the surface water of Duoqu River Basin, and the contents of K^+ and Na^+ were low. Na^+ ranged 1.07–6.60 mg/L with the mean value of 2.16 mg/L, while K^+ value was from 0.57 mg/L to 1.85 mg/L with the mean value of 0.98 mg/L. The main anion was HCO_3^- and its content ranged 43.65–196.44 mg/L with the mean value of 131.01 mg/L. The surface water components were relatively stable in Duoqu River Basin, the TDS values ranged 49.22–205.74 mg/L with the mean value of 135.26 mg/L, higher than the average value of world rivers (115 mg/L) (Zhang Y et al., 2015). The pH values were from 7.25 to 8.62, showed neutral to slightly alkaline. This result was similar to the study conclusion of Tao Zhang (Zhang T et al., 2020) in the hydrochemical characteristics and controlling factors of surface water in Ranwu Lake Basin. It reflected that the surface runoff in the plateau area was affected by the actions of glacial melt water and atmospheric precipitation, and the water body presented a low salinity and slightly alkaline state.

Fig. 2 showed the proportion of equivalent concentrations of main ions in surface water in the survey area. The relation of cation equivalent concentration was $Ca^{2+} > Mg^{2+} > Na^+ > K^+$. Ca^{2+} equivalent concentration accounted for 49.07%–87.06% of the total cations, with an average of 79.82%. Mg^{2+} equivalent concentration accounted for 10.45%–43.67% of the total cations, with an average of 15.10%. Ca^{2+} and Mg^{2+} accounted for about 94.92% of total cations. The anions in surface water were mainly HCO_3^-, and the anion equivalent concentration was in the order of $HCO_3^- > SO_4^{2-} > CO_3^{2-} > NO_3^- > Cl^- > F^-$. HCO_3^- accounted for 65.12%–86.64% of total anions, with an average of 81.61%. SO_4^{2-} accounted for 2.77% to 19.30% of the total anions, with an average of 9.13%. HCO_3^- and SO_4^{2-} accounted for about 90.74% of the total anions. In summary, the main cations in the study area were Ca^{2+} and Mg^{2+}, and the main anions were HCO_3^-.

4.1.2. Correlation analysis of chemical indexes

Correlation analysis is commonly used to research the ion sources in the hydrochemistry evolution. Components from the same source are highly correlated while those from different sources are poorly correlated (Dong WH et al., 2017). The correlation between chemical parameters in the Duoqu River Basin was shown in Fig. 3. TDS was strongly correlated with Mg^{2+},Ca^{2+}, HCO_3^- and SO_4^{2-}, indicating that Mg^{2+}, Ca^{2+}, HCO_3^- and SO_4^{2-} were the main sources of TDS for surface water in Duoqu River, among which TDS had the most significant correlation with Mg^{2+}, $Ca^{2+}$and HCO_3^-. The correlation coefficients were 0.842, 0.671 and 0.954, respectively, indicating that TDS mainly came from Ca^{2+}, Mg^{2+} and HCO_3^- in surface water. HCO_3^- had a significant correlation with Ca^{2+} and Mg^{2+}, indicating that they had common material sources, mainly came from weathering and dissolution of carbonate rocks such as calcite and dolomite. Cl^-, F^- had significant correlation with Na^+, K^+, and the main mineral components in the study area were quartz, feldspar, calcite, kaolin, mica. It indicated that they also had a common material source, mainly from the weathering and dissolution of sodium and potassium silicate rock minerals such as albite and potassium feldspar, as well as some from atmospheric precipitation and evaporation salt dissolution.

Table 1. Precision and accuracy of main anions and cations.

Index	*RSD*/%	*RD*/%	Index	*RSD*/%	*RD*/%
Ca^{2+}	3.33	0–0.23	SO_4^{2-}	2.17	0–0.75
K^+	6.35	0–0.65	Cl^-	2.14	0–0.93
Mg^{2+}	4.46	0–1.16	CO_3^{2-}	3.36	0
Na^+	2.67	0.46–3.34	NO_3^-	1.84	0–2.84
F^-	1.42	0–3.65	HCO_3^-	2.87	0

4.1.3. Hydrochemical type

The hydrochemical evolution and hydrochemical types are usually evaluated by using the microgram equivalent percentage (meq%) points of the major anions and cations in the piper ternary diagram (Li CZ et al., 2019; Cui BL and Li XY, 2015; Wang WH et al., 2018; Qi HH et al., 2019). The relative contents of various ions can be seen in the three-line diagram (Fig. 4). On the cation diagram, most of the water sample points were located at the lower left corner near the distribution of Ca^{2+} and Mg^{2+} axes, indicating that the main cations of surface water were Ca^{2+}, followed by Mg^{2+}, which mainly came from the weathering and dissolution of rocks.On the anionic diagram, all water sample points were near the the lower left corner of CO_3^{2-}+HCO_3^- region, indicating that the anions were dominated by HCO_3^-. According to Shukalev classification method, the hydrochemical types of surface water in this area were mainly HCO_3-Ca and HCO_3-Ca·Mg, indicating that the area was mainly controlled by weathering and dissolution of silicate rocks, carbonate rocks and evaporative rocks.

4.1.4. Hydrogen and oxygen isotope characteristics

The stable isotope characteristics of hydrogen and oxygen are one of the important indicators reflecting water quality changes. Fifteen groups of water samples collected in the

Table 2. Statistics of major ions in Duoqu River Basin (mg/L).

Projects	HCO_3^-	CO_3^{2-}	NO_3^-	Cl^-	SO_4^{2-}	F^-	Mg^{2+}	Na^+	K^+	Ca^{2+}	TDS	pH
Minimum	43.65	5.00	0.02	0.36	2.67	0.05	1.75	1.07	0.57	12.34	49.22	7.25
Maximum	196.44	8.70	1.84	1.84	38.19	0.20	21.10	6.60	1.85	51.30	205.74	8.62
Median	130.00	5.00	1.08	0.68	10.00	0.08	3.82	1.77	0.91	44.30	138.64	8.50
Mean	131.01	5.69	0.98	0.73	11.53	0.09	4.87	2.16	0.98	41.03	135.26	8.18
Standard deviation	30.33	1.21	0.45	0.33	6.75	0.03	3.65	1.47	0.32	9.51	31.15	0.45
Variance	920.20	1.47	0.20	0.11	45.56	0.00	13.31	2.17	0.10	90.42	970.17	0.20

study area had been tested for hydrogen and oxygen isotopes. The sampling information and test results were shown in Table 3. The results showed that the hydrogen and oxygen isotopes in the study area had little change. The $\delta^{18}O$ value ranged from −16.85‰ to −16.23‰, with an average value of −16.48‰, the δD value ranged from −127.58‰ to −122.78‰, with an average value of −124.50‰.

Generally speaking, rivers are mainly supplied by atmospheric precipitation. Therefore, studying the relationship between hydrogen and oxygen isotopes of rivers and atmospheric precipitation isotopes can better understand the recharge of rivers by atmospheric precipitation, and further trace the hydrological cycle processes.

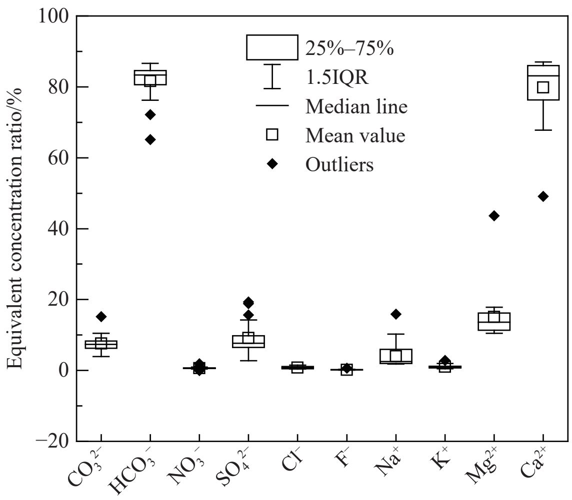


Fig. 2. Proportion of equivalent concentration of main anions.

There was a significant linear relationship between δD and $\delta^{18}O$ in the Duoqu River, and the fitting equation was shown in equation (1).

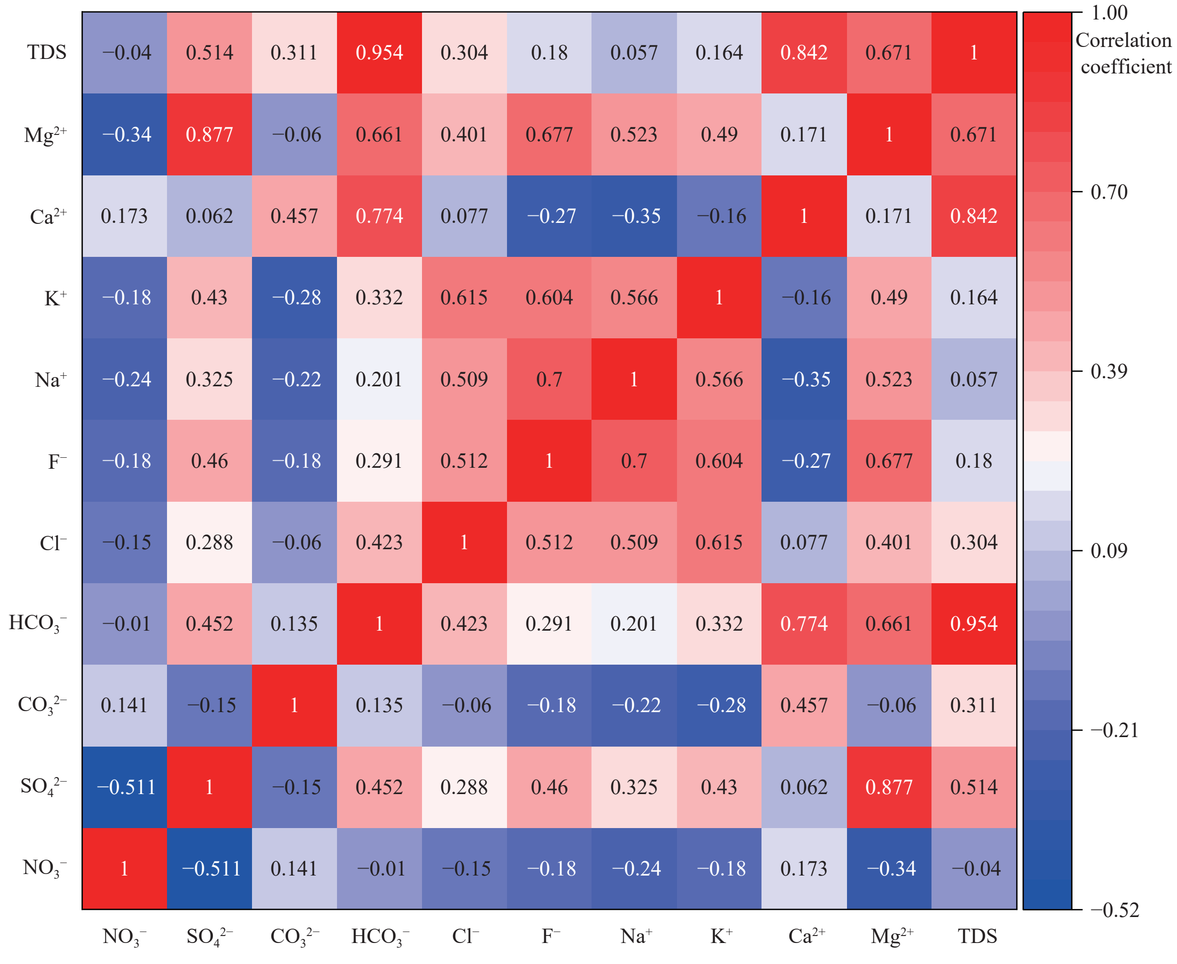
$$\text{The study area}: \delta D = 7.30\delta^{18}O - 4.25 (R^2 = 0.789) \tag{1}$$

Compared with the global atmospheric precipitation line (Craig H,1961) [Equation (2)] and the Eastern Tibetan Plateau atmospheric precipitation line [EMWL, Equation (3)] (Yu JS et al., 1980), the slope and intercept of the fitting curves for δD and $\delta^{18}O$ in this study area were smaller than the global and local atmospheric precipitation lines.Which indicated that the hydrogen and oxygen isotopes of the river were affected by various types of recharge sources in addition to different degrees of evaporation (Song MY et al., 2020).

$$\text{GMWL}: \delta D = 8\delta^{18}O + 10 \tag{2}$$

$$\text{EMWL}: \delta D = 8.2\delta^{18}O + 19 (R^2 = 0.98) \tag{3}$$

Fig. 5 showed the relationship between the fitting curves



	NO_3^-	SO_4^{2-}	CO_3^{2-}	HCO_3^-	Cl^-	F^-	Na^+	K^+	Ca^{2+}	Mg^{2+}	TDS
TDS	−0.04	0.514	0.311	0.954	0.304	0.18	0.057	0.164	0.842	0.671	1
Mg^{2+}	−0.34	0.877	−0.06	0.661	0.401	0.677	0.523	0.49	0.171	1	0.671
Ca^{2+}	0.173	0.062	0.457	0.774	0.077	−0.27	−0.35	−0.16	1	0.171	0.842
K^+	−0.18	0.43	−0.28	0.332	0.615	0.604	0.566	1	−0.16	0.49	0.164
Na^+	−0.24	0.325	−0.22	0.201	0.509	0.7	1	0.566	−0.35	0.523	0.057
F^-	−0.18	0.46	−0.18	0.291	0.512	1	0.7	0.604	−0.27	0.677	0.18
Cl^-	−0.15	0.288	−0.06	0.423	1	0.512	0.509	0.615	0.077	0.401	0.304
HCO_3^-	−0.01	0.452	0.135	1	0.423	0.291	0.201	0.332	0.774	0.661	0.954
CO_3^{2-}	0.141	−0.15	1	0.135	−0.06	−0.18	−0.22	−0.28	0.457	−0.06	0.311
SO_4^{2-}	−0.511	1	−0.15	0.452	0.288	0.46	0.325	0.43	0.062	0.877	0.514
NO_3^-	1	−0.511	0.141	−0.01	−0.15	−0.18	−0.24	−0.18	0.173	−0.34	−0.04

Fig. 3. Correlation between chemical parameters in the Duoqu River Basin.

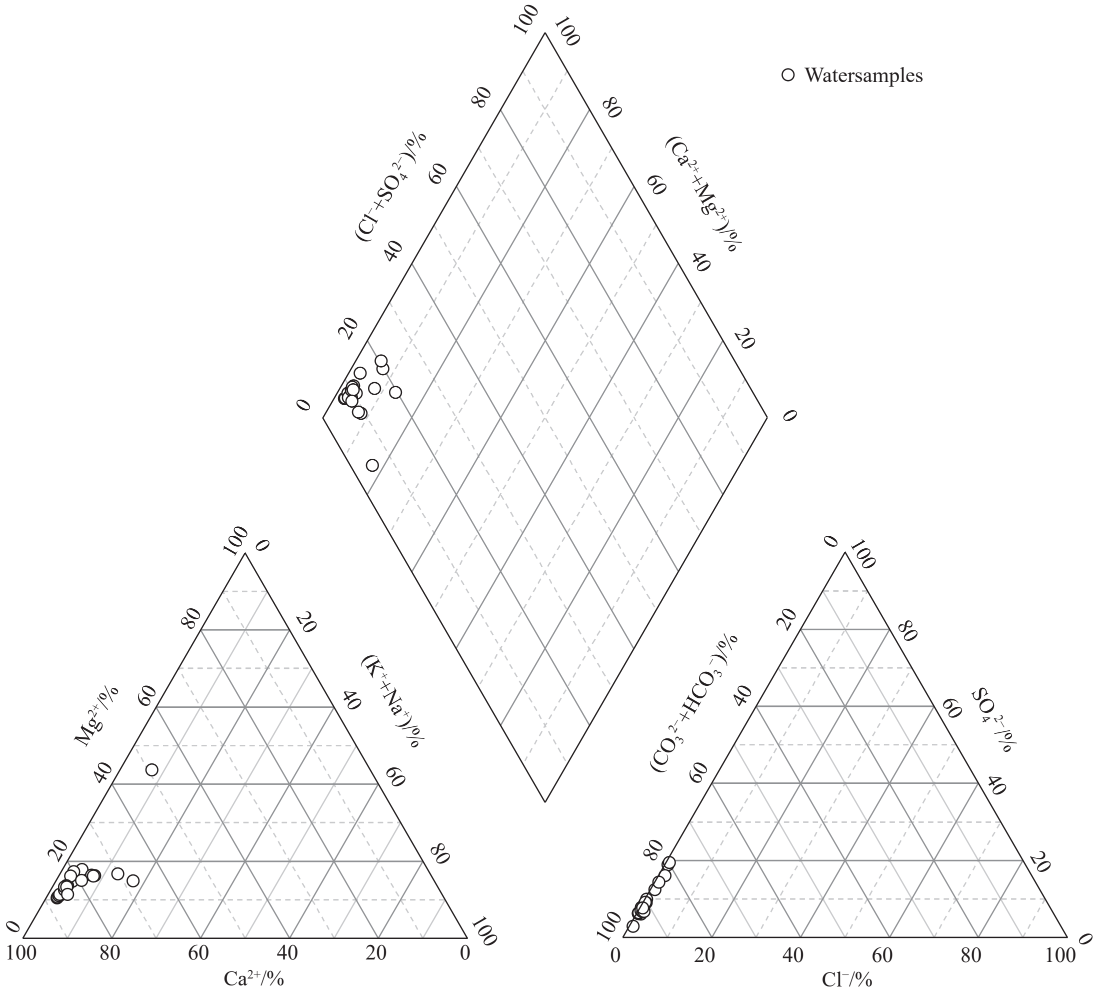


Fig. 4. Piper three-line diagram of main ions in the Duoqu River Basin.

Table 3. Hydrogen and oxygen isotope data of Duoqu River.

No	longitude	latitude	altitude	$\delta^{18}O$/‰	δD/‰	water type
D1	98.17135	31.86067	3814	−16.25	−122.78	river
D3	98.22358	31.84902	3740	−16.29	−123.26	river
D4	98.28755	31.80287	3607	−16.34	−123.34	river
D5	98.32975	31.75179	3506	−16.29	−123.18	river
D7	98.34507	31.69933	3428	−16.23	−122.90	river
D8	98.37651	31.65790	3384	−16.31	−123.30	river
D9	98.37646	31.65650	3327	−16.60	−122.78	spring
D10	98.37770	31.60161	3252	−16.25	−123.12	river
D11	98.39778	31.58534	3242	−16.49	−124.96	river
D12	98.36089	31.58577	3270	−16.70	−126.83	river
D18	98.46326	31.49951	3148	−16.85	−127.58	river
D19	98.48698	31.45318	3119	−16.76	−126.92	river
D20	98.50608	31.40601	3082	−16.74	−126.78	river
D21	98.53058	31.31999	2997	−16.57	−124.68	river
D24	98.55961	31.27428	2974	−16.54	−125.00	river
min				−16.85	−127.58	
max				−16.23	−122.78	
mean				−16.48	−124.50	
std				0.22	1.75	

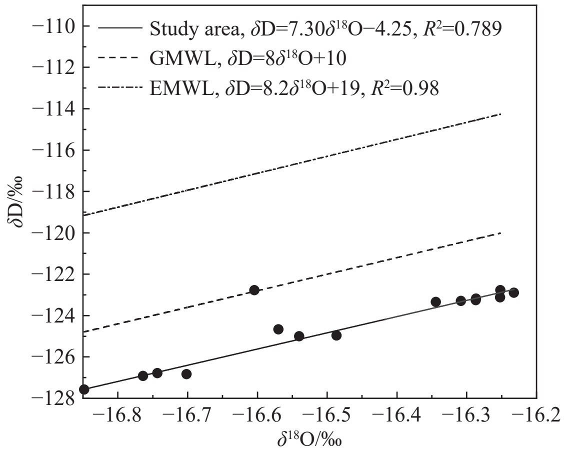


Fig. 5. Relations of δD and $\delta^{18}O$ isotopes.

of δD and $\delta^{18}O$ in the Duoqu River and the atmospheric precipitation lines of GMWL and EMWL. It could be seen from the figure that except for the sampling point D9, all the other points were located at the lower right side of the global and eastern Qinghai-Tibet Plateau atmospheric precipitation line, indicating that the surface water in this area had undergone considerable evaporation in the formation process compared with the average level of global and eastern Qinghai-Tibet Plateau precipitation. In addition, which indicated that the recharge source of the Duoqu River was not only the atmospheric precipitation, but also a certain proportion of glacial meltwater. The hydrogen and oxygen isotope composition of river samples was similar, compared with that, the δD value of spring water at sampling point D9 was significantly higher. It was analyzed that spring water recharge source was mainly groundwater, and its hydrogen and oxygen isotope composition was quite different from that of surface water (Gao JQ et al., 2021).

4.2. Major ions control factors

4.2.1. Cation alternating adsorption

Cation alternating adsorption means that under certain conditions, particles absorb some cations in water and convert some of the cations they previously adsorbed into water (Wang XX et al., 2014).[(HCO_3^-+ SO_4^{2-}) − (Ca^{2+} − Mg^{2+})] (meq) and (Na^+− Cl^-)(meq) diagrams can determine whether cation exchange takes place(Xiao J et al., 2015). If cation exchange is an important process controlling the ionic composition of the samples, the relationship between these parameters should be linear with a slope of 1∶1.The direction and strength of cation exchange in groundwater can be determined by the chlor-alkali index. According to Equations (4) and (5), when both CAI1 and CAI2 are positive, K^+ and Na^+ exchange with Mg^{2+} and Ca^{2+} in the surrounding rocks; When both CAI1 and CAI2 are negative, reverse exchange occurs, that is, $Mg^{2+}$and Ca^{2+} exchange with K^+ and Na^+ in surrounding rocks (Peng C et al., 2021).

$$\mathrm{CAI1} = \frac{Cl^- - (Na^+ + K^+)}{Cl^-} \qquad (4)$$

$$\mathrm{CAI2} = \frac{Cl^- - (Na^+ + K^+)}{HCO_3^- + SO_4^{2-} + NO_3^- + Cl^-} \qquad (5)$$

As shown in Fig. 6(a) and Fig. 6(b), most water samples in Duodu River were located near the 1∶1 line.The CAI1 value was between −9.97 and −0.90, with a mean value of −3.53, the CAI2 value was between −0.0064 and −0.063, with a mean value of −0.018. CAI1 and CAI2 in water body were both negative values, which indicated that the surface water of Duodu River had undergone some reverse cation alternating adsorption during the hydrogeochemical process, That is, Ca^{2+} and Mg^{2+} dissolved from carbonate rocks in the surface water had exchanged with Na^+ and K^+ in the surrounding rocks.The adsorption of Ca^{2+} and Mg^{2+} and the release of K^+ and Na^+ will inevitably cause the change of hydrochemical components in the surface water.

4.2.2. Water-rock interaction

Gibbs diagrams are often used to deduce hydrogeochemical processes in natural water (Li ZJ et al., 2019; Jasrotia AS et al., 2019). The Gibbs diagram is divided into three terminal elements, namely rock weathering, evaporation-crystallization and atmospheric precipitation. The control area of atmospheric precipitation is located at the lower right corner of Gibbs diagram, which has a lower total dissolved solid concentration (<10 mg/L) and a higher Ratio of $Cl^-/(Cl^-+HCO_3^-)$ and $Na^+/(Na^++Ca^{2+})$, generally ranging from 0.5 to 1. The middle left of the Gibbs diagram is the rock weathering zone, and the TDS values are generally in the range of 70–300 mg/L, and the ratios of $Cl^-/(Cl^-+HCO_3^-)$ and $Na^+/(Na^++Ca^{2+})$ are generally less than 0.5. The upper right corner of Gibbs diagram is the evaporative crystallization control zone, which has a high TDS (>300 mg/L) and a high ratio of $Cl^-/(Cl^-+HCO_3^-)$ and $Na^+/(Na^++Ca^{2+})$ (0.5–1).When the water sample points of the Duoqu River basin were put into Gibbs diagram, the water samples in this area had lower TDS and lower ratios of $Cl^-/(Cl^-+HCO_3^-)$ and $Na^+/(Na^++Ca^{2+})$.The water sample points all fell in the control area of rock weathering (Fig. 7), which indicated that rock weathering was the main ion source of the Duoqu River basin.

Different ions come from weathering dissolution of different rocks. HCO_3^-mainly comes from weathering and dissolution of silicate rock and carbonate rock, SO_4^{2-} and Cl^- mainly comes from the dissolution of evaporate rocks, weathering and dissolution of silicate and evaporative rocks can release K^+ and Na^+. Mg^{2+} and Ca^{2+} are mainly derived from weathering of carbonate rocks, silicate rocks and evaporative rocks. Mixed graph are commonly used to reveal

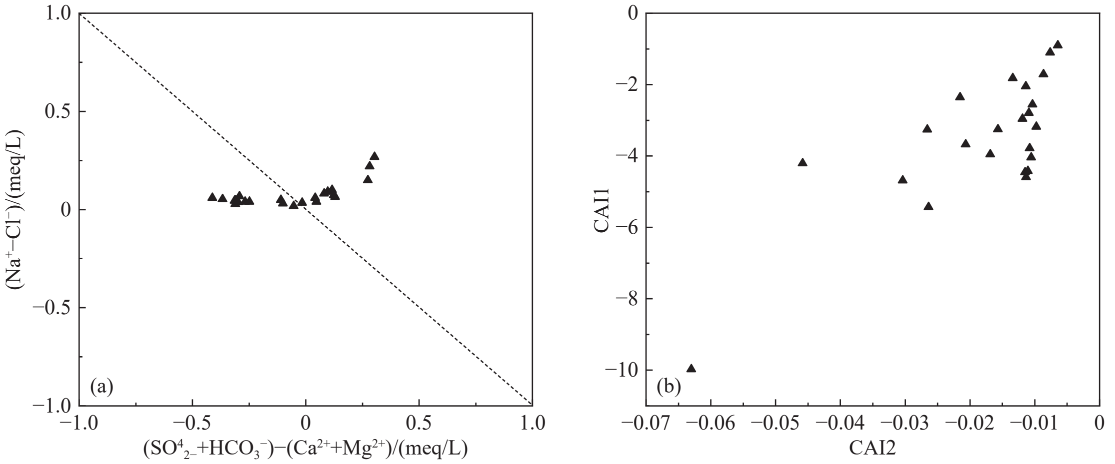


Fig. 6. Ion exchange (a) for the plot of (HCO_3^- + SO_4^{2-}) − (Ca^{2+}+ Mg^{2+}) vs. Na^+− Cl^-, and (b)for the Chlor-alkali index plot.

the source of ions generated by chemical weathering in watersheds. Since Ca^{2+}/Na^{+}, $Mg^{2+}/Na^{+}$and HCO_3^{-}/Na^{+} are not affected by flow rate, dilution and evaporation (Jiang LG et al., 2015a; Thomas J et al., 2015), the relationship between Mg^{2+}/Na^{+} and Ca^{2+}/Na^{+}, HCO_3^{-}/Na^{+} and Ca^{2+}/Na^{+} can reveal the hydrochemical origin, and it can also reveal which mineral dissolution is the main ion source. The ion ratios of Mg^{2+}/Na^{+}, Ca^{2+}/Na^{+} and HCO_3^{-}/Na^{+} are used to study the reactions between water and different rocks. The controlled end-member ratios of carbonate weathering are close to 50, 10 and 120, respectively. The ratios of the control end members of silicate rock are close to 0.35±0.15, 0.24±0.12 and 2±1 (Xiao J et al., 2012; Zhu BQ et al., 2011; Fan BL et al., 2014). Fig. 8 showed that the sample points of Duoqu River were located between the silicates and carbonates, and tended to be controlled by carbonates, which indicated that the surface water of Duoqu River basin was mainly controlled by weathering and dissolution of carbonates, and secondly by contribution from weathering of silicates, this corresponded to the formation lithology in the investigation area,which mainly contained carbonate rocks with calcite as the main component, and volcanic rocks with silicate as the main component.

The relationship between the calcium sodium ratio and TDS in surface water and rainwater can be used to determine the main factors controlling the evolution of surface water (Wu Y et al., 1996). The average contents of TDS, $Ca^{2+}$and Na^{+} in rainwater in this study area were 15.20 mg/L, 4.10

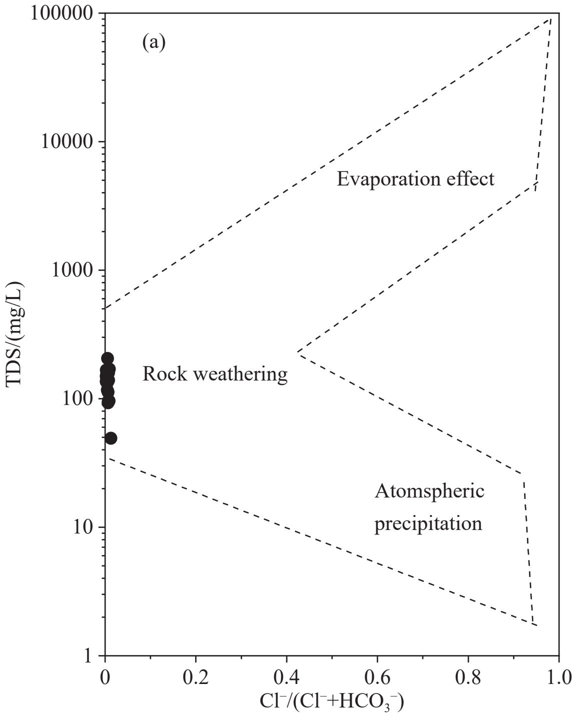


Fig. 7. Gibbs model of surface water in the Duoqu River Basin.

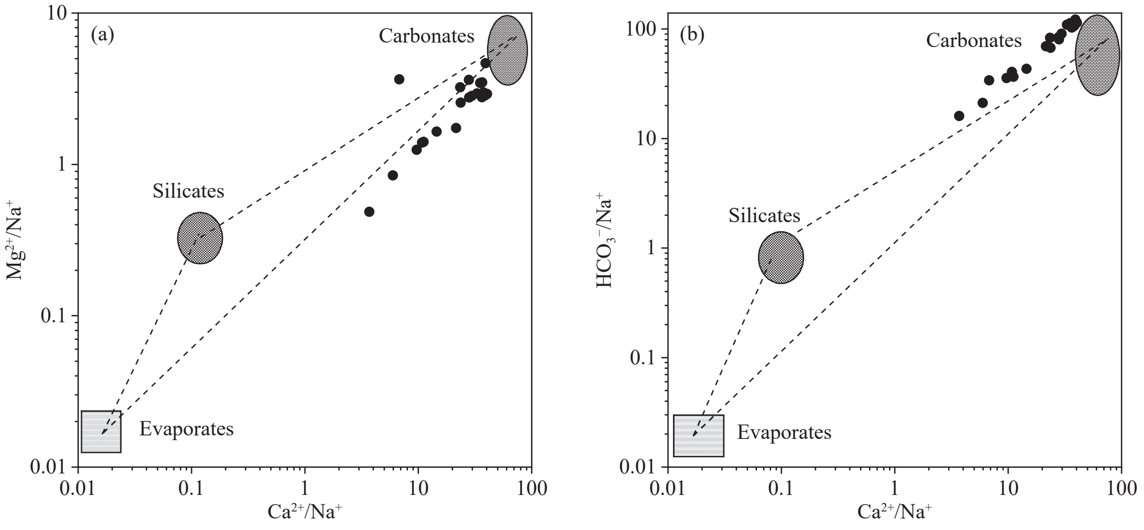


Fig. 8. Ratios of Ca^{2+}/Na^{+} to Mg^{2+}/Na^{+} and HCO_3^{-}/Na^{+}.

mg/L and 0.10 mg/L. The relationship between lg(Ca/Na) and lg(TDS) was plotted with the contents of ions in atmospheric precipitation as reference values (Fig. 9). The results indicated that the surface water points all fell in the area controlled by the weathering of sodium- bearing rocks, which revealed that the surface water in the Duoqu River was mainly controlled by rock weathering. The conclusions were consistent with the results of Gibbs model analysis.

4.3. Effect of human activities

Human activities have a very important impact on the evolution of hydrochemistry. Cl^- and $\mathrm{NO_3}^-$ are relatively stable conservative ions with little influence from water rock interaction. However, human activities such as fertilization, sewage and the discharge of poultry and livestock manure produce more Cl^- and $\mathrm{NO_3}^-$, which can change the hydrochemical composition of surface water and form higher equivalent concentration ratios of $\gamma(\mathrm{Cl}^-)/\gamma(\mathrm{Na}^+)$and $\gamma(\mathrm{NO_3}^-)/\gamma(\mathrm{Na}^+)$ (Li CC et al., 2019). Therefore, the ratio of $\gamma(\mathrm{Cl}^-)/\gamma(\mathrm{Na}^+)$ and $\gamma(\mathrm{NO_3}^-)/\gamma(\mathrm{Na}^+)$ can be used to study the impact of human activities on surface water. As can be seen from Fig. 10, the ratios of $\gamma(\mathrm{Cl}^-)/\gamma(\mathrm{Na}^+)$ and $\gamma(\mathrm{NO_3}^-)/\gamma(\mathrm{Na}^+)$ in the surface water were relatively low, indicating that the surface water quality in the study area was less affected by human activities such as agricultural, animal husbandry activities and residents' lives, which was mainly due to the hydrochemical evolution under natural conditions.

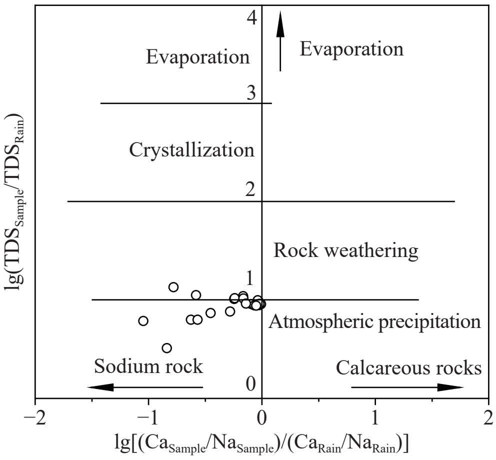


Fig. 9. Ca-Na-TDS relationship in the Duoqu River basin.

$\mathrm{SO_4}^{2-}$ usually comes from gypsum dissolution, sulfide oxidation, agricultural fertilization. The ratio relationship between $\gamma(\mathrm{SO_4}^{2-})/\gamma(\mathrm{Na}^+)$ and $\gamma(\mathrm{NO_3}^-)/\gamma(\mathrm{Ca}^{2+})$ can indicate the source of $\mathrm{SO_4}^{2-}$ and $\mathrm{NO_3}^-$ in surface water (Sun HY et al., 2020). As shown in Fig. 11, the ratio of $\gamma(\mathrm{NO_3}^-)/\gamma(\mathrm{Ca}^{2+})$ in the study area was relatively low, which was similar to the ratio characteristics of $\gamma(\mathrm{Cl}^-)/\gamma(\mathrm{Na}^+)$and $\gamma(\mathrm{NO_3}^-)/\gamma(\mathrm{Na}^+)$, indicating that agricultura activities and domestic sewage had a small impact on their water quality. Among the 24 surface water points in the Duoqu River, there were 4 water points with higher values of $\gamma(\mathrm{SO_4}^{2-})/\gamma(\mathrm{Na}^+)$, and the values of $\gamma(\mathrm{SO_4}^{2-})/\gamma(\mathrm{Na}^+)$ were greater than 4, which were respectively D15 (4.36), D18 (7.03), D19 (4.34), and D20 (4.54). The ratios of other points were lower (0.19 to 3.79, with an average of 2.65). Except for D15 point, D18, D19, and D20 were all located in the downstream of the Duoqu River basin. According to Fig.1, there were cinnabar minerals on both sides of D18, D19, D20. The main component of cinnabar minerals is mercury sulfide (HgS), which can be weathered and dissolved to release $\mathrm{SO_4}^{2-}$ions, resulting in a high ratio of $\gamma(\mathrm{SO_4}^{2-})/\gamma(\mathrm{Na}^+)$ at nearby water points. The ratio of $\gamma(\mathrm{SO_4}^{2-})/\gamma(\mathrm{Na}^+)$ at Point D18 was particularly high, which may be due to the fact that this point was not only affected by the cinnabar minerals, but also it was located downstream of the Serong Temple, a famous temple in eastern Tibet, incense burning activities in temples could result in abnormal water quality.

The D15 point was located on the tributary of Ziqu River, and the changes of major ions in the direction where Ziqu River flowed into Duoqu River were analyzed (Fig. 12). The results showed that the content of major ions fluctuated greatly along the flow direction of surface water in Ziqu River, and generally showed a slow upward trend. It is worth noting that there were two sudden increasing points along the path, D15 and D11. Compared with the upstream D16, the main ions content of D15 increased significantly, Ca^{2+}

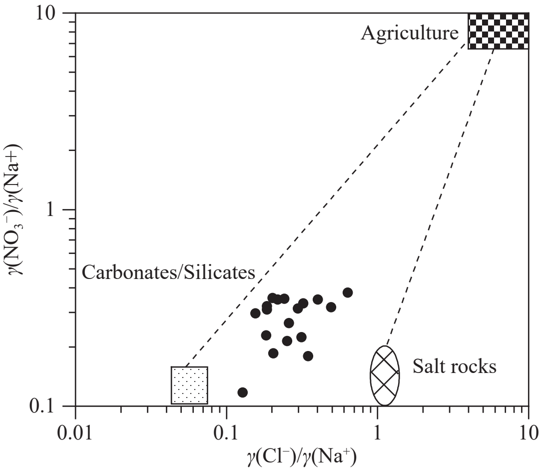


Fig. 10. Variation of $\gamma(\mathrm{NO_3}^-)/\gamma(\mathrm{Na}^+)$ with $\gamma(\mathrm{Cl}^-)/\gamma(\mathrm{Na}^+)$.

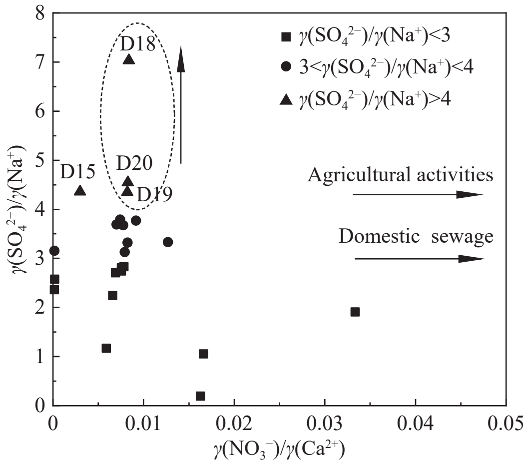


Fig. 11. Variation of $\gamma(\mathrm{SO_4}^{2-})/\gamma(\mathrm{Na}^+)$ with $\gamma(\mathrm{NO_3}^-)/\gamma(\mathrm{Ca}^{2+})$.

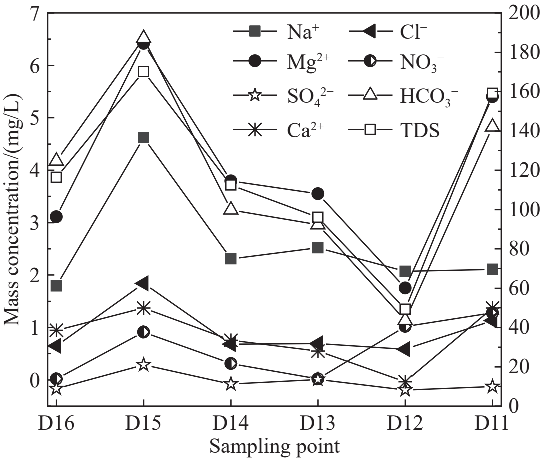


Fig. 12. Spatial variation of major ion concentrations along the Ziqu River. The left axis represents solid, while the right axis represents hollow.

increased by 29.30% (from 38.56 mg/L to 49.86 mg/L), Na^+ increased by 158.1% (from 1.79 mg/L to 4.62 mg/L), SO_4^{2-} increased by138.10% (from 8.82 mg/L to 21.00 mg/L) and HCO_3^- increased by 50.00% (from 124.72 mg/L to 187.09 mg/L). Compared with the upstream D16, the main ions content of D11 increased significantly, Ca^{2+} increased by 29.41% (from 38.56 mg/L to 49.90 mg/L), Na^+ increased by 17.88% (from 1.79 mg/L to 2.11 mg/L), SO_4^{2-} increased by 11.90% (from 8.82 mg/L to 9.87 mg/L) and HCO_3^- increased by 13.86% (from 124.72 mg/L to 142.00 mg/L). D15 was located in Jiangda County, while D11 was located near Wala Temple (Fig. 1), and the concentration of major ions at D15 and D11 increased suddenly, which could be attributed to the densely populated county and temple. A large number of visitors and frequent human activities would cause abnormal water quality.

In summary, the ratio characteristics of the $\gamma(Cl^-)/\ \gamma(Na^+)$, $\gamma(NO_3^-)/\ \gamma(Na^+)$ and $\gamma(NO_3^-)/\ \gamma(Ca^{2+})$ indicated that the surface water quality in the study area was weakly affected by human activities such as agricultural and domestic sewage. The ratio characteristics of $\gamma(SO_4^{2-})/\ \gamma(Na^+)$ indicated that the development of cinnabar minerals had a certain impact on the hydrochemistry of surface water in the study area, which was the main factor for causing the increase of SO_4^{2-} in nearby surface water points. The changes of major ions along the Ziqu River indicate that densely populated county towns and temples with frequent human activities may cause some anomalies of surface water quality nearby.

5. Conclusions

The key findings of this study are:

(i) The surface water of Duoqu River Basin was weak alkaline with a pH of 7.25–8.62 and a high soluble total solids (49.22–205.74 mg/L). The main cations were Mg^{2+} and Ca^{2+}, and the equivalent concentrations of Mg^{2+} and Ca^{2+} accounted for 94.92% of the total cations. The anion was mainly HCO_3^- with the equivalent concentration of HCO_3^- accounting for 65.12%–86.64% of the total anion. The correlation analysis showed that TDS had the most significant correlation with Mg^{2+}, Ca^{2+}, HCO_3^-, with correlation coefficients of 0.842, 0.671 and 0.954, respectively. HCO_3-Ca and HCO_3-Ca·Mg were the main hydrochemical types in the basin.

(ii) The hydrogen and oxygen stable isotopes comprehensive analysis showed that the Duoqu River was mainly recharged by atmospheric precipitation and glacial melt water. The difference in hydrogen and oxygen isotope composition of surface rivers was mainly caused by different recharge sources and evaporation degree.

(iii) The surface water of the Duoqu River had undergone a certain reverse cation alternating adsorption in the hydrogeochemical process, that was,the Ca^{2+} and Mg^{2+} dissolved in the surface water carbonate rocks exchanged with K^+ and Na^+ in the surrounding rocks.

(iv) Analysis on water-rock interaction revealed that surface water ions in the Duoqu River basin were mainly derived from rock weathering, mainly controlled by weathering and dissolution of carbonates, and secondly by silicates and sodium rocks.

(v) The influence of human activities on water chemical composition was weak, which was mainly influenced by water chemical evolution in natural state. While the development of cinnabar minerals had a certain impact on the hydrochemistry of surface water, which was the main factor for causing the increase of SO_4^{2-} in nearby surface water points. The densely populated county towns and temples with frequent anthropogenic activities may cause some anomalies of surface water quality nearby.

CRediT authorship contribution statement

Jing-jie Li, Sheng Lian and Ming-guo Wang conceived of the presented idea , performed the data analysis and results interpretation, as well as took the lead in writing the manuscript. Ming-guo Wang carried out the field investigation and water sampling. Sheng Lian drew and modified diagram. Huai-sheng Zhang and Tao Yang revised the manuscript. All authors discussed the results and contributed to the final manuscript.

Declaration of competing interest

The authors declare no conflicts of interest.

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