



## Geochemical Study of the Groundwater from Coal Bearing Aquifer in Zhuxianzhuang Coal Mine, Northern Anhui Province, China

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### ABSTRACT

Major ion concentrations were measured in 22 groundwater samples collected from a coal bearing aquifer in Zhuxianzhuang coal mine, northern Anhui Province, and the ion source and geochemical characteristics were discussed by the methods such as traditional graphic and multivariate statistic analysis. The results showed the presence of three water types, i.e.,  $\text{HCO}_3\text{-Na}$ ,  $\text{Cl-Na}$  and  $\text{SO}_4\text{-Na}$ . The surrounding rock weathering and evaporation, both controlled the chemical characteristics of groundwater, evidenced by the Gibb's diagrams. In detail, the features of ion ratios such as  $\text{Ca/Na}$ ,  $\text{HCO}_3\text{/Na}$  and  $\text{Mg/Na}$  show that the evaporate dissolution and silicate weathering were the dominant water-rock interactions. Multivariate analysis, such as correlation analysis, cluster analysis and FA analysis were also conducted to reveal the ion source in groundwater, and the  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  were mainly released from the dissolution of gypsum and/or magnesium and/or anorthite; while the  $\text{Na}^+$  is influenced by the weathering of feldspar and halite, with the cation exchange in a certain extent. In contrast, the  $\text{Cl}^-$ ,  $\text{HCO}_3^-$  and  $\text{SO}_4^{2-}$  are also controlled by the halite dissolution, silicate mineral weathering and sulphate mineral dissolution, respectively. In addition, the  $\text{SO}_4^{2-}$  in groundwater could be influenced by the oxidation of pyrite, which is abundant in the coal bearing aquifers.

### INTRODUCTION

Quality and quantity of groundwater are essential components for the life and environment, that play a key role in determining the sustainable development of agriculture and industry (Hidalgo et al. 2001, Rubasinghe et al. 2015). With the rapid development of economics in recent decades, an irrefutable fact is that the quality of groundwater is deteriorating, and a considerable consumption of contaminated water resources has an important repercussion on the environment and human health (Emmanuel et al. 2009, Qin et al. 2012, Wang et al. 2013).

In China, groundwater system in coal mining-field is not only the major source of water supply, it also influences the safety production during the period of mining coal. Out of 4.2 billion  $\text{m}^3$  mine water discharged by coal mine every year, only 26.2% was utilized as mining area production, afforesting, dust-proof, agricultural irrigation, as well as domestic water (He et al. 2008). From 2006 to 2010, 26 accidents of fatal water hazard, all caused 10 professionals or over dead, and the total casualty of these 5 years is 256 (Wu et al. 2013). Therefore, with the purpose of preventing and controlling water disaster and the mining depth increasing, much attention has been made to focus on the water source discrimination, using the hydrochemistry characteristics of the deep-buried groundwater (Gui et al. 2011).

In this paper, major ion concentration of the coal bearing aquifer in Zhuxianzhuang coal mine, northern Anhui province, China has been studied for revealing the ion source of the composition and their hydro-geochemical characteristics. The results will be utilized to establish basic data for understanding the evolution of the groundwater, and provide some useful identification information to distinguish the source of inrush water.

### MATERIALS AND METHODS

The Zhuxianzhuang coal mining region is located in the center of Huaibei plain, 13 km southeast of Suzhou city, northern Anhui Province, China (Fig. 1). The region has a moderate climate with an annual temperature of  $14.3^\circ\text{C}$ , the average annual precipitation and evaporation is 940.55 mm and 1553 mm, respectively. The length of the study site from south to north is 9 km, and the width from west to east is 1.5-5.8 km, with an approximate area extent of  $21.5 \text{ km}^2$ . The study area is located between longitudes  $117^\circ05'37.6'' \text{ E} \sim 117^\circ09'23'' \text{ E}$  and latitudes  $33^\circ33'31.4'' \text{ N} \sim 33^\circ39'37.6'' \text{ N}$ , respectively. The mining depth ranges from 250 to 800 m, and the coal output is about 2.2 million tons per year.

According to different composition, lithology and formation age, the groundwater system of the study area is divided into five main aquifers from shallow to deep (Chen

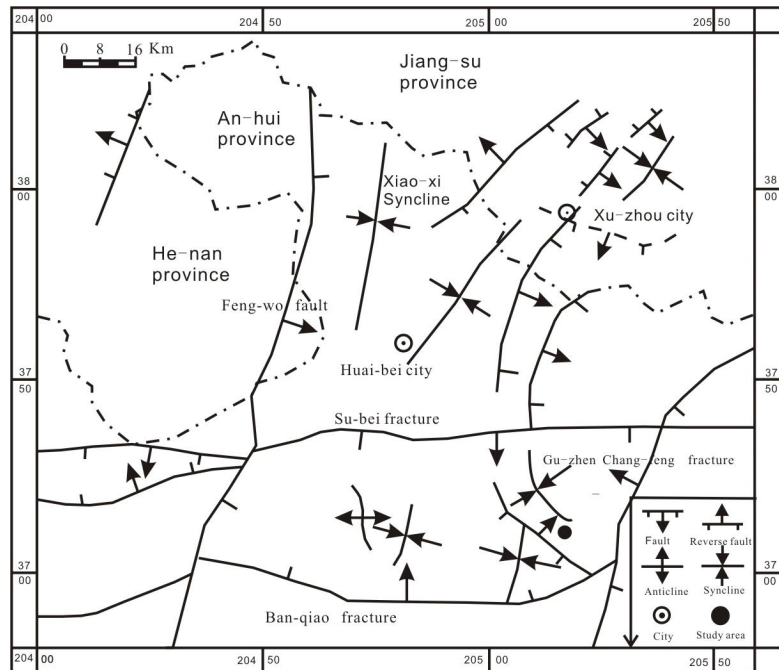


Fig. 1: Location and simple geological map of the study area.

et al. 2013). The Quaternary aquifer contains conglomerate, sandy clay and mudstone. The Jurassic aquifer is featured by limestone fragment and mud, with the grain diameter of 2-7 cm. The Coal bearing aquifer is constituted of mudstone, sand and siltstone. Limestone aquifer is featured by two types, one type is formed of interbedded thin limestone and mudstone or sandstone, which belong to Taiyuan formation; the other type is constituted of thick Ordovician limestone, with proved thickness of more than 500 m.

A total of 22 groundwater samples were analysed for major ions, which were collected from boreholes and downhole outflow points of the coal bearing aquifer. Water samples were filtered through 0.45 mm pore-size membrane and stored in polyethylene bottles, which had been cleaned in the laboratory. Before the collection of samples, bottles were further rinsed with the water to be sampled. Major ions were measured in the Anhui No.3 Coal Geological Bureau Province exploration team.  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  were analysed by EDTA titration,  $\text{Na}^+$  by atomic absorption spectrometry,  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$  by ion chromatography, and alkalinity by acid-based titration.

Piper diagram was established using Aq.Qa software (Fig. 2), and statistical analysis was carried out by the SPSS and MYSTAT software (version 11).

## RESULTS AND DISCUSSION

**Hydrochemical characteristics of major ions:** To reveal

the hydrochemical characteristics of groundwater, the major ion concentrations of the coal bearing aquifer are presented in Table 1, along with the pH and TDS. Generally, the pH values ranged from 7.2 to 8.7, with a mean value of 7.95, reflecting that all the groundwater samples were naturally alkaline. The concentration of the total dissolved solids (TDS) varied from 1209.56 to 1880.97 mg/L, with an average value of 1634.52 mg/L. Referring to the drinking water standards from the World Health Organization (WHO), the pH values of almost all samples meet the criteria of 6.5-8.5. However, the TDS values of all groundwater samples are higher than the permissible limit of WHO, which are not suitable for drinking, but can be used for agricultural irrigation.

The cationic concentrations (Na, Ca and Mg) range from 246.4 to 574.15 mg/L, 3.91 to 214.83 mg/L, 2.57 to 95.72 mg/L, with an average value of 370.67 mg/L, 87.68 mg/L, 44.32 mg/L, respectively. The anionic concentrations ( $\text{HCO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$ ) ranged from 156.03 to 920.83 mg/L, 2.88 to 581.18 mg/L, 246.39 to 373.57 mg/L, with an average value of 500.68 mg/L, 299.16 mg/L, 331.10 mg/L, respectively. The groundwater samples could be recognized as three water types, i.e. the  $\text{Cl-Na}$ ,  $\text{HCO}_3\text{-Na}$  and  $\text{HCO}_3\text{-Na}$  types and they have 4, 8 and 10 samples, respectively. The different water types could reflect the diverse hydrochemical characteristics controlled by the different water-rock interaction. Further analysis showed that ionic

Table 1: Major ion properties of groundwater in the study area (The units are in mg/L except pH) .

ID	Ca <sup>2+</sup>	Mg <sup>2+</sup>	Na <sup>+</sup>	HCO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	pH	TDS	Water type
ZXZ1	12.44	8.15	574.15	919.37	2.88	363.22	8.2	1880.97	HCO <sub>3</sub> <sup>-</sup> -Na
ZXZ2	12.16	2.74	383.25	476.44	4.53	329.64	8.3	1209.56	Cl <sup>-</sup> -Na
ZXZ3	172.95	16.44	330.07	455.08	304.55	373.57	7.4	1652.8	Cl <sup>-</sup> -Na
ZXZ4	89.56	35.02	275.63	580.96	30.87	325.53	7.2	1338.31	HCO <sub>3</sub> <sup>-</sup> -Na
ZXZ5	162.68	78.25	279.89	322.42	579.94	327.48	7.9	1751.85	SO <sub>4</sub> <sup>2-</sup> -Na
ZXZ6	80.85	95.72	369.61	438.91	516.15	356.49	7.7	1858.03	SO <sub>4</sub> <sup>2-</sup> -Na
ZXZ7	203.11	76.28	272.78	408.03	581.18	336.81	7.3	1878.54	SO <sub>4</sub> <sup>2-</sup> -Na
ZXZ8	192.26	81.67	263.72	395.35	570.48	333.91	7.7	1837.39	SO <sub>4</sub> <sup>2-</sup> -Na
ZXZ9	75.43	48.93	383.22	156.03	551.54	354.36	7.9	1572.32	SO <sub>4</sub> <sup>2-</sup> -Na
ZXZ10	214.83	81.99	246.4	387.41	579.12	346.41	7.3	1856.16	SO <sub>4</sub> <sup>2-</sup> -Ca
ZXZ11	95.53	63.68	352.08	341.34	520.26	319.01	7.9	1693.9	SO <sub>4</sub> <sup>2-</sup> -Na
ZXZ12	138.1	75.99	315.19	308.88	556.07	322.94	8.3	1717.47	SO <sub>4</sub> <sup>2-</sup> -Na
ZXZ13	112.87	68.78	289.23	264.4	519.86	308.44	8	1564.59	SO <sub>4</sub> <sup>2-</sup> -Na
ZXZ14	155.79	73.88	306.64	380.94	515.32	363.22	7.8	1795.97	SO <sub>4</sub> <sup>2-</sup> -Na
ZXZ15	14.77	19.	436.52	702.9	5.76	311.14	8.4	1491.19	HCO <sub>3</sub> <sup>-</sup> -Na
ZXZ16	19.92	30.8	370.16	413.79	86.02	367.16	8	1289.1	Cl <sup>-</sup> -Na
ZXZ17	47.9	32.89	440.84	920.83	57.62	272.06	7.8	1772.59	HCO <sub>3</sub> <sup>-</sup> -Na
ZXZ18	10.02	5.74	521.71	702.95	95.9	339.05	8.3	1677.01	HCO <sub>3</sub> <sup>-</sup> -Na
ZXZ19	3.91	2.57	511.96	744.44	133.36	246.39	8.5	1644.11	HCO <sub>3</sub> <sup>-</sup> -Na
ZXZ20	70.12	43.17	394.96	612.64	298	284.5	7.85	1703.43	HCO <sub>3</sub> <sup>-</sup> -Na
ZXZ21	30.54	26.2	426.24	636.72	32.1	334.15	8.36	1486.67	HCO <sub>3</sub> <sup>-</sup> -Na
ZXZ22	13.21	7.11	410.46	445.22	39.93	368.65	8.7	1287.52	Cl <sup>-</sup> -Na

concentrations of alkali metal (Na) exceed the alkaline earth metal (Ca and Mg), and strong acids (SO<sub>4</sub> and Cl) exceed the weak acids (HCO<sub>3</sub>).

**Mechanism of main ion source:** To determine the main ion source of the coal-bearing groundwater, such as precipitation dominance, rock dominance and evaporation dominance, Gibbs diagram was used for assessing the chemical controlling mechanism (Gibbs 1970). Gibbs ratios of Na/(Na+Ca) and Cl/(Cl+HCO<sub>3</sub>) as a function of TDS were plotted. From the Gibbs diagram (Fig. 3), it can be revealed that the ratios roughly varied in a certain range of rock dominance and evaporation dominance. This phenomenon indicates that chemical weathering and evaporation, related to the dissolution of evaporated and hydrological discharge, are the remarkable factors during the evolution of the chemical composition of the groundwater in the study area.

Current chemical constituents were affected by circumstances, such as formation lithology, recharge, discharge and water-rock interaction. Generally, three normal processes of the generation of solutes in groundwater contains: evaporation dissolution, carbonate dissolution and silicate weathering (Sun et al. 2013). As is shown in Fig. 4, Ca/Na, HCO<sub>3</sub>/Na and Mg/Na ratios range from 0.009 to 1.003, 0.154 to 0.795 and 0.010 to 0.638, with average values of 0.332, 0.506 and 0.269, respectively. This result shows that the evaporate dissolution and silicate weathering were the dominant hydrochemical interactions.

If the dissolution of halite is under the pure chemical reaction, the ratios of Na against Cl can be distributed along 1:1 line. However, Na-Cl relationship of all the samples deviated from the 1:1 equiline could explain two reaction(s): 1) Those that fall below the 1:1 line resulted from the effect of silicate weathering (Na/Cl was more than 1); 2) Those that above the equiline are related to carbonate dissolution

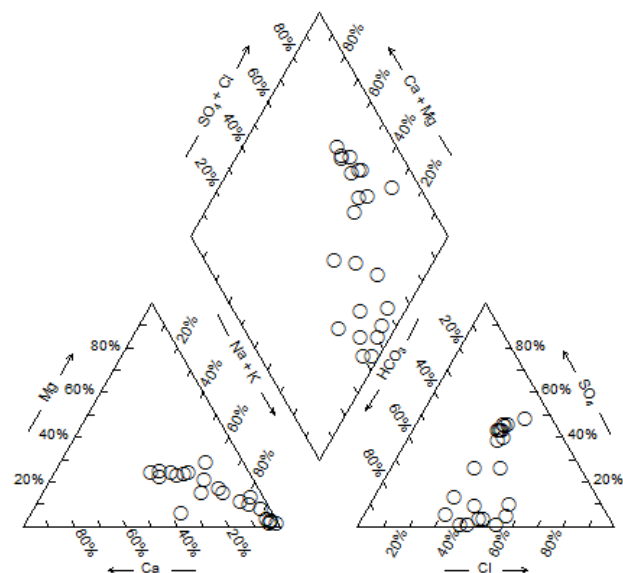


Fig. 2: Piper plot of groundwater hydrochemical data in the study area.

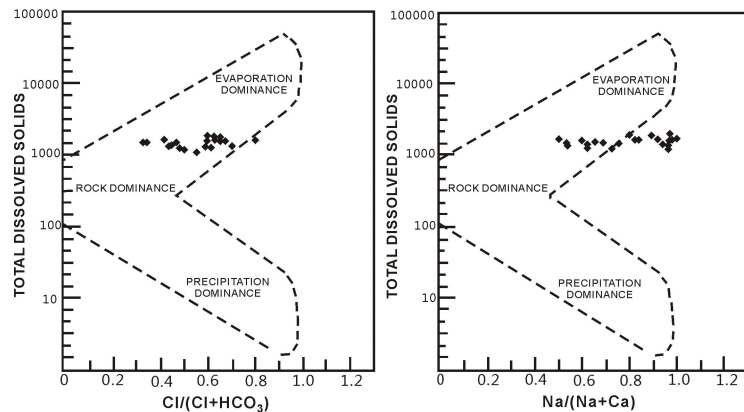
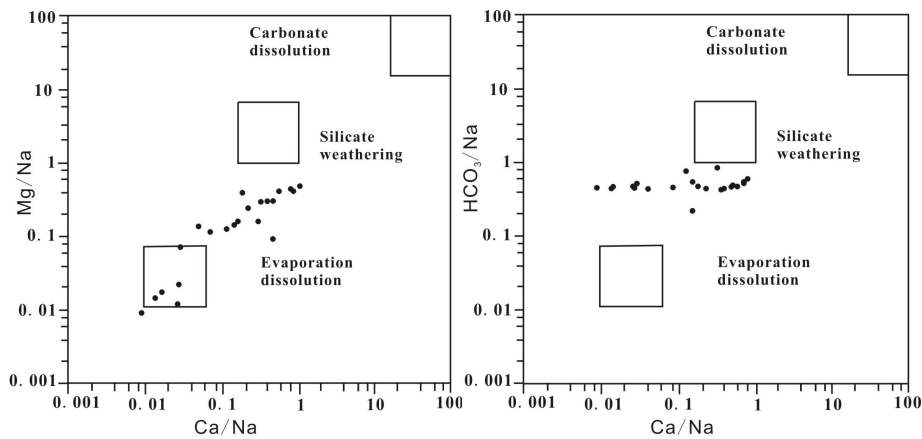


Fig. 3: Gibbs diagram.

Fig. 4: Scatter plots of  $\text{Na}^+$  normalized  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  and  $\text{HCO}_3^-$  of groundwater samples in the study area.

( $\text{Na}/\text{Cl}$  was less than 1). Furthermore, high Na concentration can be derived from human activities, however, all the samples of the coal bearing aquifer were collected from a depth > 300 m, so anthropogenic sources of  $\text{Na}^+$  cannot be a right way to explain the high concentration of Na. In Fig. 5, all of the points fall below the equiline, signifying the high sodium concentration (Meybeck 1987). It is obvious from the plot that the weathering of silicate provides the main inputs of Na, such as feldspar.

Moreover, another process of sodium concentration, occurred along the flow path, could be ion exchange (Kurdi et al. 2014). According to chloro-alkaline indices,  $(\text{Cl}-\text{Na})/\text{Cl}$  (CAI-I) and  $(\text{Cl}-\text{Na})/(\text{SO}_4+\text{HCO}_3)$  (CAI-II) can be used to estimate the ion exchange between the groundwater and its host rocks. If the indices are positive, the cation exchange occurs between Ca and/or Mg in the host rock and Na in water. However, if the indices are negative, it presents a reverse cation exchange. From the Fig. 6, all of the samples calculated are negative values, indicating Na in host rocks and Mg and/or Ca in water generate the cation exchange, such as albite.

A plot of  $(\text{HCO}_3+\text{SO}_4)$  against  $(\text{Ca}+\text{Mg})$  was used to assess the effect of carbonate dissolution and silicate weathering. In Fig. 7, all the points fall below the equiline, and it can be supported that 22 samples have higher  $\text{HCO}_3+\text{SO}_4$  concentration relative to  $\text{Ca}+\text{Mg}$ , which suggests an extra source of  $\text{HCO}_3^-$  balanced by  $\text{Na}^+$ , supporting the weathering of silicate minerals.

Ca, Mg and  $\text{SO}_4$  ions in groundwater can be provided by the dissolution of gypsum and the weathering of an  $\text{Mg}^{2+}$  sulphate mineral (Jalali 2009), respectively. In Fig. 8, a plot of  $\text{Ca}+\text{Mg}$  against  $\text{SO}_4$  show that some of the groundwater samples are around the 1:1 line, but most of the points fall below the equiline, indicating that besides the dissolution of gypsum and/or the weathering of an  $\text{Mg}^{2+}$  sulphate mineral. Moreover, the oxidation of pyrite could afford the source of  $\text{SO}_4$ , and the dissolution of anorthite can provide another Ca ion source in groundwater.

**Multivariate statistic analysis:** In order to reveal the relation between the parameters, the correlation analysis was obtained in this study, the results are presented in Table 2. It

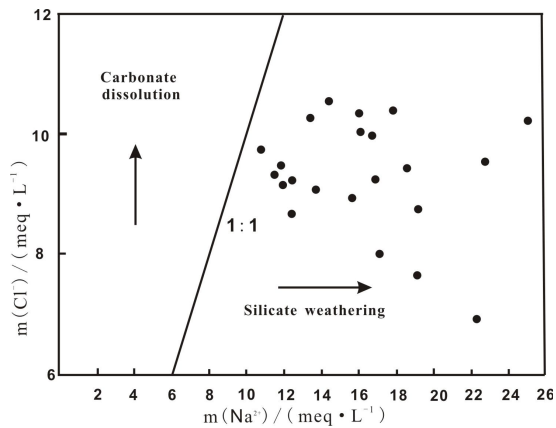


Fig. 5: Scatter plots of Na<sup>+</sup> and Cl<sup>-</sup> of groundwater samples in the study area.

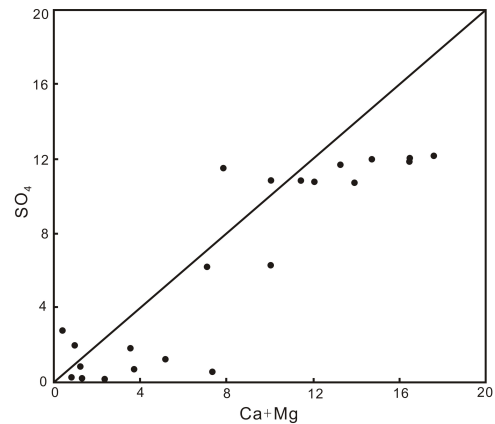


Fig. 8: (Ca+Mg)-SO<sub>4</sub> diagram.

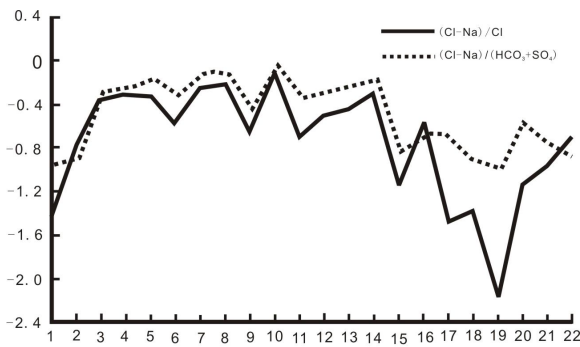


Fig. 6: Variation of (Cl-Na)/Cl and (Cl-Na)/(SO<sub>4</sub>+HCO<sub>3</sub>).

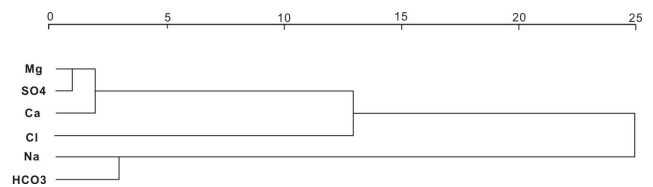


Fig. 9: R mode cluster analysis.

Table 2: Correlation matrix of major ions.

Parameters	Na <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	Cl	SO <sub>4</sub>	HCO <sub>3</sub>
Na <sup>+</sup>	1.00					
Ca <sup>2+</sup>	-0.84	1.00				
Mg <sup>2+</sup>	-0.74	0.77	1.00			
Cl	-0.20	0.21	0.11	1.00		
SO <sub>4</sub>	-0.69	0.82	0.89	0.13	1.00	
HCO <sub>3</sub>	0.74	-0.56	-0.61	-0.39	-0.75	1.00

Table 3: Principal component analytical results of coal bearing aquifer.

Component	Before Rotation		After Rotation	
	FC1	FC2	VF1	VF2
Na <sup>+</sup>	-0.90	0.04	-0.88	-0.18
Ca <sup>2+</sup>	0.90	-0.11	0.90	0.11
Mg <sup>2+</sup>	0.90	-0.22	0.92	0
Cl <sup>-</sup>	0.30	0.93	0.07	0.98
SO <sub>4</sub> <sup>2-</sup>	0.92	-0.17	0.94	0.05
HCO <sub>3</sub> <sup>-</sup>	-0.83	-0.26	-0.75	-0.45
Eigen values	4.04	1.02	3.87	1.20
Var/%	67.34	17.06	64.46	19.93
Cum/%	67.34	84.40	64.46	84.40

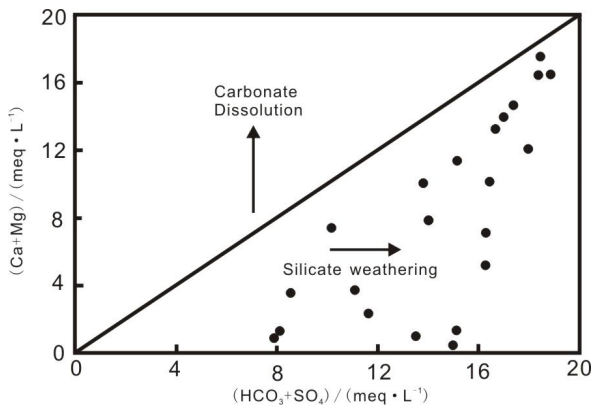


Fig. 7: ( HCO<sub>3</sub>+SO<sub>4</sub> ) - ( Ca+Mg) diagram.

could be concluded that there exists a good correlation between Na and HCO<sub>3</sub>, with a correlation matrix 0.74, which indicates the weathering of silicate mineral in the hydrochemical process. The positive correlation also existed between Ca and Mg, Ca and SO<sub>4</sub>, and Mg and SO<sub>4</sub>, with the correlation matrix 0.77, 0.82 and 0.89, respectively. The results suggest that the dissolution of sulphate mineral, such as magnesium sulphate and gypsum, could be existed in the process of water-rock interaction.

Cluster analysis has an advantage in identifying the relationship among massive samples (Kamble et al. 2011), including Q mode cluster analysis (sample cluster) and R mode cluster analysis (parameter cluster). The results shown in Fig. 9 were consistent with correlation analysis, which can explain that the main ions can be classified in two

groups. Group one containing Mg, SO<sub>4</sub>, Ca and Cl, reflects that the main sources of these ions are the dissolution of sulfide and chloride. Group two including Na and HCO<sub>3</sub><sup>-</sup>, indicates that the main sources of these two ions are from the silicate weathering.

FA analysis was considered as a popular method of mastering the principal contradiction, aiming at simplifying multivariable information, and it was used to analyse the source of aquifer, contaminant, etc. Two factors have been obtained with eigen value higher than one after varimax rotation of groundwater (Table 3), and the cumulative variance is about 84.4%. The parameters of Ca, Mg and SO<sub>4</sub>, with a high value in VF1 (account for 64.46%), are 0.90, 0.92 and 0.94, respectively, which could reveal that the factor could be related to the dissolution of gypsum. VF2 accounts for 19.93% of the total variance, with a high loading for Cl, indicating the dissolution of chlorine salt mineral, such as halite.

## CONCLUSIONS

The concentrations of major ions in groundwater samples collected from coal bearing aquifer in Zhuxianzhuang coal mine, northern Anhui Province has been analysed, combined with the discussion about the ion source and hydrochemistry characteristics, the following conclusions can be obtained:

1. The groundwater samples in this study can be subdivided into Cl-Na, HCO<sub>3</sub><sup>-</sup>-Na and HCO<sub>3</sub><sup>-</sup>-Na. And alkali metal (Na) exceed alkaline earth metals (Ca and Mg), and strong acids (SO<sub>4</sub> and Cl) exceed the weak acids.
2. Gibb's diagrams indicate that chemical weathering and evaporation are the main factors in controlling the hydrochemistry of groundwater. Especially, the evaporation dissolution and silicate weathering are the dominant factors in the interaction between surrounding rock and groundwater, manifested from the ion ratios such as Ca/Na, HCO<sub>3</sub><sup>-</sup>/Na and Mg/Na.
3. Ca and Mg are mainly sourced from the dissolution of gypsum and/or magnesium and/or anorthite, while Na is releasing from the halite and feldspar, with the cation exchange existing in a certain degree. Whereas, the Cl<sup>-</sup>, HCO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> are controlled by the halite dissolution, silicate mineral weathering and gypsum dissolution, respectively. In addition, the SO<sub>4</sub><sup>2-</sup> in groundwater is also influenced by the oxidation of pyrite, which is abundant in the coal bearing aquifers.
4. From multivariate statistic analysis, it is clear that the main composition of the coal bearing aquifer is controlled by the dissolution of gypsum, sulfide, chloride and silicate mineral weathering.

## ACKNOWLEDGEMENTS

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