

## Source identification of inrush water based on groundwater hydrochemistry and statistical analysis

Linhua Sun<sup>a,b,\*</sup>, Song Chen<sup>a,b</sup> and Herong Gui<sup>a,b</sup>

<sup>a</sup>School of Resources and Civil Engineering, Suzhou University, Anhui 234000, China

<sup>b</sup>National Engineering Research Center of Coal Mine Water Hazard Controlling, Anhui 234000, China

\* Corresponding author. E-mail: sunlinh@126.com

### Abstract

Water source identification is important for water hazard controlling in coal mines. In this study, major ion concentrations of the groundwater collected from four representative aquifer systems in the Baishan coal mine, northern Anhui Province, China, have been analysed by a series of statistical methods. The results indicate that the major ion concentrations of the groundwater from different aquifer system are different with each other, and provided the possibility of water source identification based on hydrochemistry. Factor analysis indicates that these differences are controlled by different types of water rock interactions. The analysis based on US Environmental Protection Agency (EPA) Unmix model identified three sources (weathering of silicate minerals, dissolution of carbonate and evaporate minerals) responsible for the hydrochemical variations of the groundwater. Also, it shows that their contributions for the groundwater in different aquifer systems vary considerably. Based on these variations and on step by step analysis, the source aquifer system for the groundwater samples with unknown source has been determined and, similar to the result obtained by the cluster and discriminant analysis. Therefore, EPA Unmix model can be applied for water source identification in coal mine, as it can provide information about water rock interaction and water source identification simultaneously.

**Key words:** coal mine, groundwater, source of inrush water, water-rock interaction

### INTRODUCTION

Among the five most dangerous disasters (including water, fire, gas, dust and roof) in coal mine, water inrush has brought to human with highest loss, not only the loss of property, but most importantly, the death of people. In recent years, more than 100 accidents related to water inrush have occurred per year (Wu *et al.* 2013). During the last twenty years, more than 220 coal mines have been flooded, more than 8,000 persons died and more than 30 billion Yuan (¥ RMB) have been lost in the accidents. Previous studies revealed that a large number of factors can affect the occurrence of water inrush: the geographical characteristics of the coal mines, the conditions of structure and hydrogeology, as well as the status of coal mining (Xu & Bu 2015).

In recent years, two kinds of methods have been applied for the controlling of water inrush: one is passive prevention, which focused on finding out the source of water after the water inrush; another one is positive prevention, which focused on forecasting the possibility of water inrush based on the comprehensive understanding of the hydrogeological conditions of the coal mine (Wu *et al.* 2013). Before or after water inrush, some of the information is contained in the groundwater characteristics, including the quantity, temperature and hydrochemistry etc. They have been used for identifying the source of inrush water, which is important for either management after water inrush or, prevention before water inrush. Based on the information, a large number of studies have been brought to

light, and a series of methods have been reported, including quality-level-temperature method, hydrochemical method, mathematical method, GIS method and others (Jiang & Liang 2006; Sun *et al.* 2007; Chen *et al.* 2009; Zhang *et al.* 2009; Zhou *et al.* 2010; Huang & Chen 2011).

However, there are still some problems that have not been well documented, which are considered to be important in the future. Especially, the mechanism of water-rock interaction in the groundwater system has not been well understood, which is considered to be the basis for applying of the hydrochemistry of groundwater for water source identification (Sun & Gui 2012). If the information about water rock interaction (source of chemical constituents) and statistical analysis can be used together, it might be a better choice for solving the problem related to the source identification of inrush water in coal mine.

Therefore, in this study, a total of twenty-five groundwater samples have been collected from four representative aquifer systems in the Baishan coal mine, northern Anhui Province, China. Their major ion concentrations have been analyzed by the EPA Unmix model for identifying the source of water, which has previously been used for identifying the source of major ions in groundwater (Sun & Gui 2015a) and the wall rock compositions of the aquifer system (Sun 2015). The study can provide information for understanding the water rock interactions in the groundwater system as well as water source identification for water hazard controlling in the coal mine.

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## MATERIALS AND METHODS

### Hydrogeological background

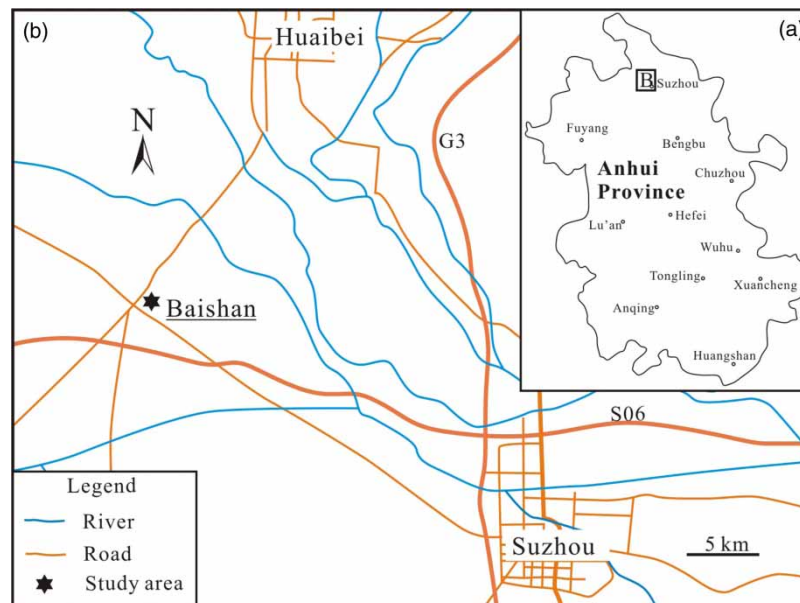
The coal field in northern Anhui Province is an important energy base in China. There are more than thirty coal mines located in the area and more than 100 million tons of coals have been exploited per year. The Baishan coal mine is located northwest to the Suzhou city and southwest to the Huaibei city, the distances are 35 and 15 km, respectively. Previous investigations revealed that the aquifer in the coal mine can be subdivided into four systems from shallow to deep: loose layer (LA), coal bearing sandstone (CA), Taiyuan Formation limestone (TA) and Ordovician limestone (OA). Their characteristics are as follows:

The LA is mainly composed of orange-deep yellow mudstone, sandstone and conglomerate (mostly composed of calcite), with thickness between 20 and 57 m. The depth of the aquifer system is 220–280 m below the surface. The CA is mainly composed of mudstone, siltstone, sandstone and coal seams, and the plagioclase is the main mineral. The thickness of the aquifer system varies significantly, from 37 to 128 m, and the depth is 280–400 m below the surface. In what concern TA and OA, they are mainly composed of thick layer of light gray limestone, the minerals are mainly calcite and dolomite, the thickness of these two aquifer systems are small than 136 m and depth between 530 and 680 m.

Previous investigations suggested that the surface water and the water in the CA are the direct threaten for the safety of coal mining in the study area. However, the water from the LA and TA can recharge the CA through the fault system or other channels. Therefore, during the management of water inrush in the coal mine, the water source identification related to the LA, CA and TA should be paid attention to.

### Sampling and analysis

A total of twenty-five groundwater samples have been collected from four representative aquifer systems in the Baishan coal mine (Figure 1), five from the LA, CA and OA, respectively, and four from the TA through drainage holes with known source in the shafts. There are other six samples, which were collected from the drilling hole in the coal seam floor and their source aquifer have not been



**Figure 1** | Location of the Baishan coal mine.

determined (maybe TA or OA according to their depth). Concentrations of eight major ions ( $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{HCO}_3^-$  and  $\text{CO}_3^{2-}$ ) have been analyzed, and because of the low concentrations of  $\text{K}^+$ ,  $\text{Na}^+$  and  $\text{K}^+$  were merged and presented as ( $\text{Na}^+ + \text{K}^+$ ). The analytical methods are as follows:  $\text{Na}^+ + \text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  were analyzed by ion chromatography, whereas  $\text{HCO}_3^-$  and  $\text{CO}_3^{2-}$  were analyzed by acid–base titration in the Engineering and Technological Research Center of Coal Exploration, Anhui Province, China.

All of the data were firstly processed by Microsoft Excel for calculating the mean concentrations of the groundwater samples from each aquifer systems. Then, all of the data were analyzed by factor analysis (Mystat 12) for identifying the main sources of the major ions, method often used in environmental studies (Maiz *et al.* 2000; Liu *et al.* 2003; Shukla *et al.* 2006). Then, Q-mode cluster and discriminant analyses (Mystat 12) have been applied in order to allow the identification of the relationships between samples, and then for determination of the source aquifer of the unknown samples based on the relationships between samples (Panda *et al.* 2006; Shi & Xu 2010). Finally, the Unmix model has been applied for quantifying the source of major ions and to obtain the contributions of each source. Then the source aquifer of the unknown samples was determined according to the contribution ratios, because samples from similar source aquifer should have similar contributions from same host rocks.

## RESULTS AND DISCUSSIONS

### Concentrations of major ions

Range and mean concentrations of major ions in the groundwater samples from different aquifer systems are synthesized in Table 1. As can be seen from the table, the chemical compositions of the groundwater from the four aquifer systems vary considerably: the groundwater samples from the CA possess the highest mean concentrations of  $\text{Na}^+ + \text{K}^+$  and  $\text{HCO}_3^-$  relative to other aquifer systems, whereas the groundwater samples from the TA and OA have relatively higher  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Cl}^-$ , and  $\text{SO}_4^{2-}$  concentrations, especially the OA. These differences suggest that the types and degrees of water rock interactions in these four aquifer systems are different. Moreover, the water samples with unknown sources are similar to the groundwater samples from the TA and OA, but different with those from the LA and CA.

**Table 1** | Range and mean concentrations (mg/l) of major ions in the groundwater samples from different aquifer systems

<b>Aquifer</b>	<b>Na<sup>+</sup> + K<sup>+</sup></b>	<b>Ca<sup>2+</sup></b>	<b>Mg<sup>2+</sup></b>	<b>Cl<sup>-</sup></b>	<b>SO<sub>4</sub><sup>2-</sup></b>	<b>HCO<sub>3</sub><sup>-</sup></b>	<b>CO<sub>3</sub><sup>2-</sup></b>
LA ( <i>n</i> = 5)	99.7–245 (208)	73.6–105 (96.4)	51.4–64.8 (60.2)	97.0–155 (141)	204–368 (329)	524–639 (559)	0–0 (0)
CA ( <i>n</i> = 5)	480–504 (494)	31.0–44.3 (37.6)	26.8–30.7 (28.7)	144–154 (147)	495–513 (505)	565–701 (642)	0–41.1 (15.5)
TA ( <i>n</i> = 4)	171–245 (197)	124–162 (137)	41.8–144 (74.9)	90.0–210 (136)	407–805 (569)	325–388 (359)	0–0 (0)
OA ( <i>n</i> = 5)	123–181 (158)	137–178 (168)	128–156 (147)	141–178 (168)	691–895 (843)	247–349 (300)	0–29.5 (5.90)
UN ( <i>n</i> = 6)	177–276 (212)	113–189 (166)	74.9–141 (120)	163–206 (192)	672–785 (760)	321–450 (397)	0–0 (0)

Note: LA, CA, TA and OA are the loose layer, coal bearing sandstone, Taiyuan limestone and Ordovician limestone aquifer systems, respectively.

## Factor analysis

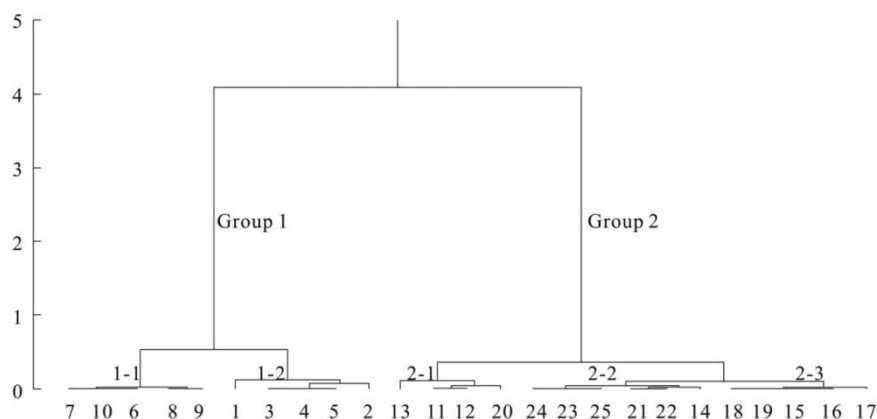
Two factors have been obtained based on the factor analysis with Eigen value higher than one after varimax rotation (Table 2). As can be seen from the table, the first factor, which accounts for 72.1% of the total variance explanation, has high positive loadings of  $\text{Na}^+ + \text{K}^+$  and  $\text{HCO}_3^-$ , whereas factor with 18.6% of the total variance explanation, has high positive loadings of  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$ . Based on the investigations and previous studies (Sun & Gui 2012; Wu *et al.* 2013), these two factors can be explained to be the weathering of silicate minerals (Factor 1) and dissolution of carbonates, sulfates and chlorides (Factor 2). This consideration is consistent with the characteristics of major ion concentrations of the groundwater from each aquifer (Table 1). The groundwater samples from the CA (sandstone dominant) show high  $\text{Na}^+ + \text{K}^+$  and  $\text{HCO}_3^-$  concentrations, whereas the groundwater samples from the TA and OA with high  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$  might be an indication of high contents of evaporate minerals.

## Cluster analysis

Hierarchical Q-mode cluster analysis has been applied to the data, and the 'Ward' linkage and the 'Pearson' distance have been chosen for calculation. The results are shown in Figure 2 as a dendrogram. As can be seen from the figure, two main groups and five subgroups can be identified: the Group 1 can be subdivided into two subgroups, including samples 1–5 (Group 1-1) and 6–10 (Group 1-2), which are consistent with the samples collected from the LA and CA, respectively.

**Table 2** | Results of factor analysis

Species	Factor 1	Factor 2
$\text{Na}^+ + \text{K}^+$	0.98	-0.05
$\text{Ca}^{2+}$	-0.75	0.46
$\text{Mg}^{2+}$	-0.57	0.65
$\text{Cl}^-$	0.10	0.97
$\text{SO}_4^{2-}$	-0.15	0.69
$\text{HCO}_3^-$	0.60	-0.12
Eigen value	4.32	1.12
Variance explained	72.1%	18.6%



**Figure 2** | Results of Q-mode cluster analysis. Samples 1–5, 6–10, 11–14 and 15–19 are collected from the LA, CA, TA and OA, whereas samples 20–25 are with unknown sources.

The Group 2 can be subdivided into three subgroups, including Group 2-1 (samples 11–13, 20), Group 2-2 (samples 14, 21–25) and Group 2-3 (samples 15–19). It can be found that all of the samples collected from the OA are classified to be Group 2-3, and the samples collected from the TA and unknown source aquifer system are classified to be Group 2-1 and 2-2. Such a classification indicates that the samples with unknown source are chemically similar to the samples collected from the OA and TA, especially the TA, as already observed based on descriptive statistics.

### Discriminant analysis

Discriminant analysis provides linear or quadratic functions of the variables that ‘best’ separate cases into two or more predefined groups. In this study, the groundwater samples collected from the LA, CA, TA and OA are signed to be 1, 2, 3, and 4, respectively, whereas the samples with unknown source are signed to be ‘blank’. The classical method of Mystat (version 12) has been chosen for calculation and the results are shown in Table 3.

As can be seen from Table 3, the discrimination of all of the samples collected from the known aquifer systems (LA, CA, TA and OA) is corrected to be 100%: samples 1–5, 6–10, 11–14 and 15–19 have the lowest distance value to group 1 (LA), 2 (CA), 3 (TA) and 4 (OA), respectively, which suggests that the efficiency of the discrimination is valid. Based on the discrimination model established by the

**Table 3** | Results of discriminant analysis

ID	Source	Predicted	Distance 1	Distance 2	Distance 3	Distance 4
S1	LA	1	10	309	368	1,014
S2	LA	1	11	336	380	1,050
S3	LA	1	2	310	403	1,103
S4	LA	1	3	324	424	1,138
S5	LA	1	1	293	374	1,050
S6	CA	2	301	1	288	757
S7	CA	2	295	3	290	768
S8	CA	2	347	3	336	803
S9	CA	2	311	3	358	866
S10	CA	2	303	1	299	776
S11	TA	3	458	362	5	153
S12	TA	3	404	244	8	194
S13	TA	3	329	331	8	236
S14	TA	3	379	344	11	181
S15	OA	4	978	724	156	11
S16	OA	4	1,095	828	195	1
S17	OA	4	1,178	881	230	4
S18	OA	4	1,065	786	182	2
S19	OA	4	1,031	759	170	2
S20	UN	3	548	253	45	169
S21	UN	3	389	324	6	184
S22	UN	3	343	311	7	209
S23	UN	3	308	383	31	256
S24	UN	3	207	353	69	400
S25	UN	3	328	396	30	249

Note: Distance 1–4 are distances of each samples to each cluster.

known samples, all of the groundwater samples with unknown source are classified to be 3 (TA), implying that the TA is the source aquifer system for them, similar to the result obtained by cluster analysis that the unknown source samples are clustered together with the TA samples.

### Unmix model analysis

Based on the calculation of UNMIX model, three sources have been identified and the results are listed in Tables 4 and 5, and shown in Figures 3 and 4. These three sources have Min Rsq = 0.99 and Min Sig/Noise = 2.53, higher than the minimum requirement of the model (Min Rsq > 0.8 and Min Sig/Noise > 2). It can also be obtained from the relationships between predicted and observed values of the major ions, that the  $r^2$  values for  $\text{Na}^+ + \text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$  and  $\text{HCO}_3^-$  are 1.00, 0.95, 0.97, 0.83, 0.98 and 0.98, respectively (Figure 3), suggesting that the modeling is efficient (Ai *et al.* 2014).

**Table 4** | Source compositions ( $S_n$  in mg/l)

Species	Source 1	Source 2	Source 3
$\text{Na}^+ + \text{K}^+$	168	30.3	56.2
$\text{Ca}^{2+}$	12.4	33.3	76.1
$\text{Mg}^{2+}$	8.64	18.1	61.7
$\text{Cl}^-$	49.1	35.4	75.2
$\text{SO}_4^{2-}$	169	83.1	355
$\text{HCO}_3^-$	209	137	108

The relationship between the hydrochemistry of the groundwater and the sources are as follows:

$$C_m = \sum X_1 \times S_1 + X_2 \times S_2 \dots \dots X_n \times S_n$$

$C_m$  is the measured concentration of groundwater;  $S_n$  is the source concentration (Table 4);  $X_n$  is the portion of source  $n$  (Table 5), and  $(X_n \times S_n) / \sum (X_n \times S_n)$  is the contribution of source  $n$  for the  $C_m$  (Figure 4).

Weathering of silicate minerals, dissolution of carbonate and evaporate minerals are the main processes controlling the groundwater chemistry in the coal mining related strata (Sun 2015; Sun & Gui 2015a). Therefore, the Source 1 dominated by  $\text{Na}^+ + \text{K}^+$ , and  $\text{HCO}_3^-$  should be explained to be the weathering of silicate minerals, and the Source 3 dominated by  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Cl}^-$ , and  $\text{SO}_4^{2-}$  can be explained to be the dissolution of evaporate minerals (sulfates and chlorides).

Such considerations can be demonstrated by the contributions of these sources (Figure 4), as the samples from the CA have highest contributions from the Source 1, consistent with the truth that this aquifer system is rich in plagioclase sandstone. Samples from the OA have highest contributions from the Source 3, which is also consistent with the previous investigations that the rocks in the Ordovician strata are mainly composed of calcite, dolomite and gypsum.

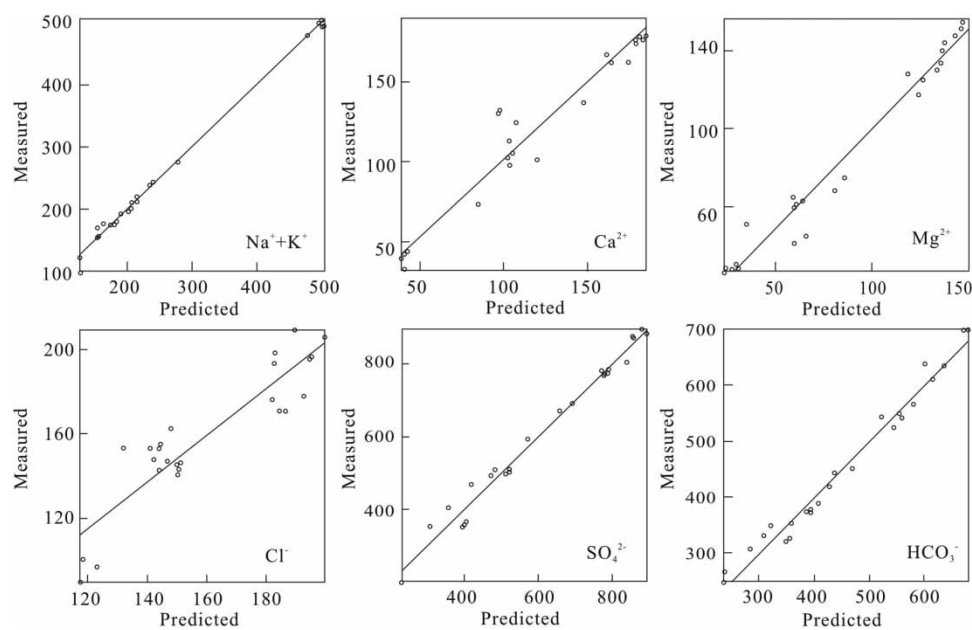
The Source 2 should be considered to be the carbonate source, because it cannot be explained to be silicate and /or evaporate sources relative to the Source 1 and 3, since the weathering of silicate will generate high  $\text{Na}^+ + \text{K}^+$  and  $\text{HCO}_3^-$  simultaneously (Source 1), whereas the high  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  concentrations can be generated by dissolution of evaporate minerals (Source 3) rather than carbonate minerals.

However, this consideration is also ambiguous because: (1) the Source 2 shows only medium contributions of  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{HCO}_3^-$ , which is not consistent with the characteristics generated by

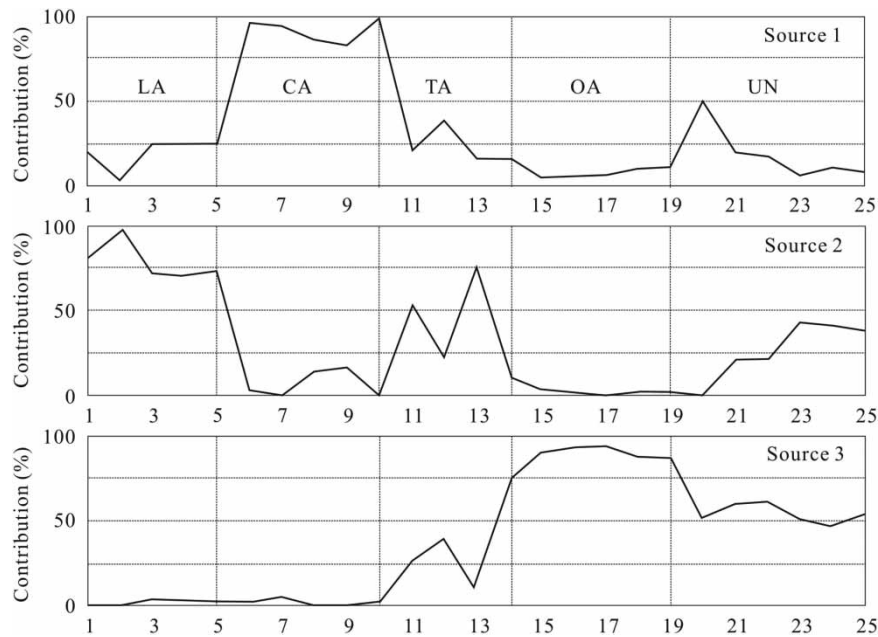
**Table 5** | Portions of the sources ( $X_n$ )

ID	Source aquifer	Source 1	Source 2	Source 3
S1	LA	0.831	3.609	-0.605
S2	LA	0.104	3.707	-0.058
S3	LA	0.907	2.508	0.103
S4	LA	0.928	2.611	0.087
S5	LA	0.909	2.610	0.067
S6	CA	2.955	0.089	0.039
S7	CA	2.836	-0.229	0.169
S8	CA	2.971	0.498	-0.170
S9	CA	2.957	0.606	-0.221
S10	CA	2.951	-0.047	0.047
S11	TA	0.520	1.326	0.616
S12	TA	0.990	0.576	1.001
S13	TA	0.447	2.067	0.299
S14	TA	0.443	0.302	2.090
S15	OA	0.106	0.109	1.878
S16	OA	0.131	0.046	2.345
S17	OA	0.172	-0.558	2.570
S18	OA	0.267	0.062	2.275
S19	OA	0.300	0.061	2.333
S20	UN	1.283	-0.484	1.360
S21	UN	0.577	0.619	1.757
S22	UN	0.505	0.636	1.802
S23	UN	0.219	1.481	1.751
S24	UN	0.398	1.511	1.678
S25	UN	0.293	1.284	1.788

Note:  $X_n$  (in this table)  $\times S_n$  (in Tables 4) is the contribution of source  $n$  in mg/l.

**Figure 3** | Predicted versus measured concentrations (mg/l).





**Figure 4** | Variations of source contributions  $((X_n \times S_n) / \sum(X_n \times S_n))$ . Samples 1–5, 6–10, 11–14 and 15–19 are collected from the LA, CA, TA and OA, whereas samples 20–25 are with unknown sources. Take sample 1 as an example, its chemical composition is contributed by 20% of source 1 and 80% of source 2.

the dissolution of carbonates; (2) it is not consistent with the truth that the TA and OA have more carbonate minerals than the other two aquifer systems (LA and CA), whereas the groundwater samples collected from the LA possess the highest contributions from this source.

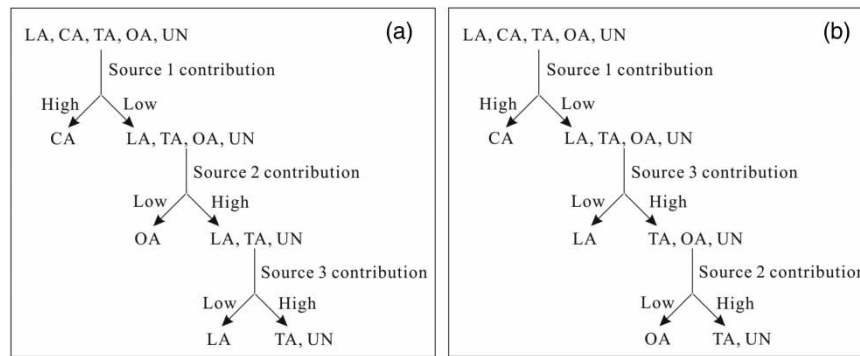
In consideration with the hydrogeological conditions of the study area, the high contributions of the Source 2 (dissolution of carbonate minerals) for the LA samples may be related to the high contents of carbonate conglomerate in the strata, whereas the low contributions of this source for the TA and OA samples might be replaced by the dissolution of evaporate minerals, because evaporate minerals have much higher solubility than the carbonates in similar conditions.

#### Implications for water source identification

As mentioned in the introduction, several methods have been applied for water source identification in coal mines (Jiang & Liang 2006; Sun *et al.* 2007; Chen *et al.* 2009; Zhang *et al.* 2009; Zhou *et al.* 2010; Huang & Chen 2011). However, hydrochemical analysis is the most popular and reliable method, because water flow over the rocks can inherit the chemical signature of the rocks, and the rocks in the different aquifer systems are different. In this study, the main rocks in the LA, CA, TA and OA are different with each other, which therefore, provided the possibility of water source identification based on water hydrochemistry.

The concentrations of major ions in the groundwater samples firstly revealed that they have undergone different types and degrees of water rock interactions in their aquifer systems. Factor and cluster analysis confirmed that the groundwater samples collected from the LA, CA, TA and OA have undergone different types and degrees of water rock interactions. The source aquifer system of the unknown samples has been identified to be the TA based on discriminant analysis with high extent of probability. However, can similar result be obtained from the EPA Unmix model?

Here the analysis is focused on the source contributions (Figure 4), and the analysis is step by step (see also in Figure 5(a) and 5(b)):



**Figure 5** | Step by step water source identification. UN–unknown source.

Step 1: according to the contributions of the Source 1 (silicate weathering), the CA samples have high contributions from this source, which is different with those of the samples from the LA, TA, OA and unknown source. Therefore, CA is not considered to be the source for the unknown samples, and the probability sources are LA, TA and OA.

Step 2: according to the contributions of the Source 2 (carbonate dissolution), the OA samples have low contributions from this source, different with those of the samples from the LA, TA and unknown source. Therefore, OA is not considered to be the source for the unknown samples, and the probability sources are LA and TA.

Step 3: according to the contributions of the Source 3 (evaporite dissolution), the LA samples have low contributions from this source, different with those of the samples from the TA and unknown source. Therefore, LA can be ruled out, and TA is considered to be source for the unknown source samples.

No matter which source considered firstly, the results are similar to each other (see in Figure 5(a) and 5(b)). Moreover, the results are also consistent with the results obtained by statistical analyses (including cluster and discriminant analyses), which suggests that the method is applicable and believable.

Moreover, although the statistical methods have long been used for water source identification in coal mines (Sun 2014; Ma *et al.* 2014; Sun & Gui 2015b), which have played important roles for water hazards controlling and management. However, the water source discrimination model established for one coal mine based on statistical differences cannot be migrated to another coal mine easily, because of the lack of the understanding about the water rock interactions. The method with EPA Unmix model has advantages relatively to previous statistical methods, because it considers not only the statistical similarity, but also the wall rock compositions as revealed by hydrochemistry.

## CONCLUSIONS

Based on the analyses of major ion concentrations of groundwater samples collected from four representative aquifer systems in the Baishan coal mine, northern Anhui Province, China, the following conclusions have been obtained:

1. The major ion concentrations of the groundwater samples are different with each other, and then provided the possibility of water source identification.
2. Factor analysis suggests that weathering of silicate minerals and dissolution of carbonates, sulfates and chlorides are the main factors controlling the groundwater chemistry.
3. Cluster analysis and discriminant analysis indicate that the TA is the source aquifer system for the unknown source samples.
4. The analysis based on EPA Unmix model identified three sources (weathering of silicate minerals, dissolution of carbonate and evaporate minerals) responsible for the hydrochemistry of the

groundwater, and show that their contributions for the groundwater in different aquifer systems vary considerably.

5. Based on the variations of contributions of each source and step by step analysis, the TA is determined to be the source aquifer system for the groundwater samples with unknown source.
6. EPA Unmix model can be applied for water source identification in coal mine, as it can provide information about water rock interaction and water source identification simultaneously.

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