

APPLICATION OF HYDROCHEMISTRY FOR INRUSH WATER SOURCE IDENTIFICATION IN COAL MINE: APPROACH BASED ON STATISTICAL ANALYSIS

Linhua SUN*

School of Resources and Civil Engineering, National Engineering Research Center of Coal Mine Water Hazard Controlling, Anhui 234000, China

Abstract: The source of chemical constituents in the groundwater and the source of inrush water are two important issues related to the hydrochemical evolution and safety of coal mining in coalfield of China, respectively. In this study, major ion concentrations of thirty-four groundwater samples from three representative aquifers in northern Anhui province, China have been analyzed by a series of statistical methods for tracing the sources of major ions and inrush water. The differences of major ion concentrations in groundwater from different aquifers indicate that they have undergone different types and degrees of water rock interactions, and provide the possibility for water source identification based on major ions. Factor analysis has identified two potential sources responsible for the major ion concentrations of the groundwater, including dissolution of carbonates and evaporates and the weathering of silicate minerals, which was further confirmed by the Unmix model analysis. Discriminant analysis can classify the sources of groundwater with high efficiency, similar to the results obtained based on the source contributions of the Unmix model analysis. In summary, different with those of factor and discriminant analysis, the Unmix model analysis can provide information about source of major ions and water simultaneously.

Keywords: *inrush water, coalmine, source identification, statistical analysis, hydrochemistry*

1. INTRODUCTION

To be one of the most important water components, the groundwater constitutes about 2/3 of the freshwater resources of the world (Lvovitch, 1970), and has played

Corresponding author: sunlinh@126.com (L. Sun)

important roles in the human history: not only the use of domestic, but also the agricultural and industrial purpose. However, as for the coalmines, groundwater is a double-edged sword: firstly, it is important for the water supply in coal mining areas because most of these areas are lacking of water (Gui et al., 2011); secondly, the groundwater is an important threaten for the safety of coal mining as it can brought to humans with great losses (Wu et al., 2013). And therefore, how to control the water hazards related to the groundwater and utilization of them simultaneously is an important issue in the current coal mine production of China.

During the last two decades, as one of the most important and effective tools, hydrochemistry has played important roles for understanding the groundwater evolution and application in coal mines, especially the inrush water source identification and water resource utilization. In the early stage, most of the studies related to the source identification of inrush water based on only the differences of the hydrochemical signatures of the groundwater from different aquifers, and the methods applied include discriminant and neural network et al (Chen et al., 2009; Sun and Gui, 2012), and the reasons for why these differences occur (related to the water rock interaction) have not been considered. Later, the hydrochemical research focused on the water rock interaction has been brought to light, and the information about the source of the major ions and the hydraulic connection between aquifers have been obtained (Sun and Gui, 2015; Sun, 2014).

Previous studies revealed that groundwater chemistry is a function of recharge, runoff and discharge, however, the mineral compositions of the aquifers, and the degrees of water rock interaction is considered to be the major process because solid phases are the primary sources and sinks of dissolved constituents of groundwater (Sarkar et al., 2007). And therefore, the differences of mineral compositions of different aquifers are the main reasons for the hydrochemical differences of the groundwater from the different aquifers. In other words, the differences of groundwater hydrochemistry can be used for understanding the differences of aquifer compositions (Liu and Sun, 2015).

In this study, a new approach based on the source approximation of major ions has been applied for inrush water source identification. Through this method, not only the qualitative information (always obtained from the factor analysis), but the quantitative contribution ratios about the sources of major ions can be achieved. Most importantly, these contribution ratios have been applied for water source identification.

2. MATERIALS AND METHODS

2.1. HYDRO-GEOLOGICAL BACKGROUND

The Huaibei coalfield is located in the eastern part of Yu-huai depression zone, the southeast margin of North China Craton. There are three-four aquifer systems related to the safety of coal mining from shallow to deep in the area: the loose layer (QA),

coal bearing (CBA) and limestone aquifer systems (TA&OA, including Carboniferous and Ordovician limestone aquifer systems) (Figure 1).

All of these aquifer systems have different hydrological features, e.g. the loose layer aquifer system is mainly composed of conglomerate and loose sediments, and the hydrological condition is open as it can be recharged by precipitation or surface water. Whereas the coal bearing aquifer system is mainly composed of hard sandstones with cracks and coal seams, and the hydrological condition is relative close under the condition that there is no natural fault or cracks produced by coal mining activities. The limestone aquifer system is mainly composed of carbonate rocks, to a lesser extent, clastic rocks, and the hydrological condition is also open, as it can be recharged by surface water because the limestone strata in the area have outcrops (Fig. 1).

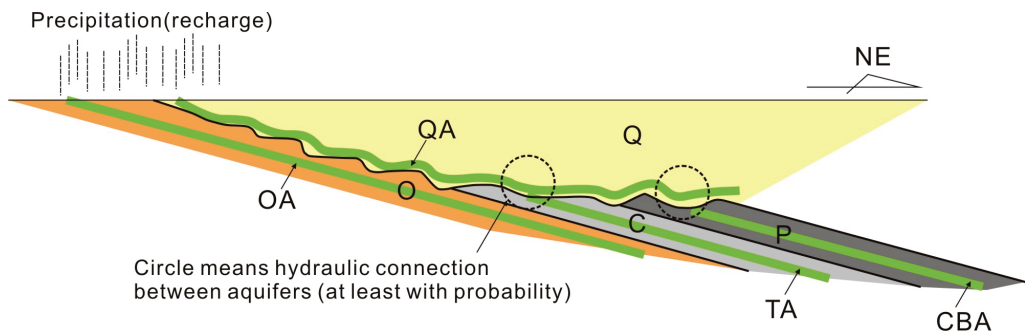


Fig. 1. Schematic diagram showing the distribution of aquifer systems and hydraulic connections between aquifer systems (QA, CBA, TA and OA mean loose layer, coal bearing sandstone, Carboniferous and Ordovician limestone aquifer systems, respectively) (after Liu and Sun, 2015)

2.2. METHODS

Major ion concentrations ($K^+ + Na^+$, Ca^{2+} , Mg^{2+} , Cl^- , SO_4^{2-} , HCO_3^- and CO_3^{2-}) of 34 groundwater samples have been collected from three coal mines in the area (the Wolonghu, Baishan and Qianyingzi coalmines), and 14, 11 and 9 from the LA, CBA and TA&OA, respectively. And the statistical analyses are processed as follows.

Firstly, all of the data were processed by Microsoft Excel for calculating the mean concentrations of the groundwater samples from each aquifer, which can provide a glance at the differences between aquifers. Then, all of the data were analyzed by traditional statistical methods (Mystat 12), including factor and discriminant analyses, the former is applied for identifying the potential sources of the major ions, and the latter is applied for establishment of water source identification model. Finally, the Unmix model has been applied for quantifying the source of major ions and to obtain the contributions of each source, and the source aquifer of the samples was evaluated

according to the contribution ratios. Then the results about the source of major ions and groundwater obtained by the Unmix model were compared with the results obtained by factor and discriminant analyses.

The Unmix model is a mathematical receptor model used for quantifying the sources of contaminants. It based on the reducing of the large number of variables in complex analytical data sets to combinations of species called source types and contributions (Lewis et al., 2003; Zhang et al., 2012; Lang and Yang, 2014). The source types are identified by comparing them to measured profiles, whereas the source contributions are used to determine how much each source contributed to a sample. The basic idea of the model is as follows:

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

C_m is the measured concentration of sample, S_n is the profile contributed by any types of sources, X_n is the portion of S_n , and $(X_n \times S_n)/\sum(X_n \times S_n)$ is the contribution of source n for the C_m .

The hydrochemistry of the groundwater is also a multi-source contribution issue, the different types of water rock interactions can be considered as sources, whereas the degrees of water rock interaction can be considered as contributions.

3. RESULTS AND DISCUSSION

3.1. STATISTICAL DIFFERENCES OF GROUNDWATER HYDROCHEMISTRY

Mean concentrations of major ions in the groundwater samples from different aquifers are synthesized in the Table 1. As can be seen from the table, the chemical compositions of the groundwater from the three aquifers vary considerably: the groundwater samples from the CBA possess the highest mean concentrations of $\text{Na}^+ + \text{K}^+$, HCO_3^- and CO_3^{2-} relative to other aquifer systems, whereas the groundwater samples from the TA&OA have the relatively highest Ca^{2+} , Mg^{2+} , Cl^- , and SO_4^{2-} concentrations. It is also noticed that samples from the LA have lowest $\text{Na}^+ + \text{K}^+$ and Cl^- concentrations, samples from the CBA have the lowest Ca^{2+} , Mg^{2+} and SO_4^{2-} concentrations, whereas the samples from the TA&OA have lowest HCO_4^{2-} and CO_3^- concentrations. These differences suggest that the types and degrees of water rock interactions in these aquifers are different.

Table 1. Mean concentrations (mg/l) of major ions in the groundwater samples from different aquifers

Aquifer	$\text{Na}^+ + \text{K}^+$	Ca^{2+}	Mg^{2+}	Cl^-	SO_4^{2-}	HCO_3^-	CO_3^{2-}
LA ($n = 14$)	294	71.8	53.9	149	325	621	4.50
CBA ($n = 11$)	600	6.29	4.86	211	19.8	1070	103
TA&OA ($n = 9$)	354	301	179	286	1600	234	3.28

3.2. POTENTIAL SOURCES OF MAJOR IONS – FACTOR ANALYSIS

Factor analysis is a statistical method used to describe variability among observed, correlated variables in terms of a potentially lower number of unobserved variables, which has long been used for environmental studies for tracing the source of pollutants (Liu et al., 2003; Romić and Romić, 2003; Almasoud et al., 2015). In this study, two factors have been obtained based on the factor analysis with eigenvalue higher than one after varimax rotation (Table 2).

Table 2. Results of factor analysis

Species	Na ⁺ + K ⁺	Ca ²⁺	Mg ²⁺	Cl ⁻	SO ₄ ²⁻	HCO ₃ ⁻	Eigenvalue	Explained
Factor 1	-0.097	0.985	0.944	0.768	0.986	-0.798	4.072	67.9%
Factor 2	0.981	-0.045	-0.242	0.539	0.009	0.545	1.61	26.8%

As can be seen from the table, the first factor, which accounts for 67.9% of the total variance explanation, has high positive loadings of Ca²⁺, Mg²⁺, Cl⁻ and SO₄²⁻, whereas the second factor with 26.8% of the total variance explanation, has high positive loadings of Na⁺ + K⁺ and HCO₃⁻. Based on the investigations and previous studies (Sun and Gui, 2015), these two factors can be explained to be the dissolution of carbonates and evaporates (Factor 1) and the weathering of silicate minerals (Factor 2). This consideration is consistent with the characteristics of major ion concentrations of the groundwater from each aquifer (Table 1), that the groundwater samples from the CBA (sandstone dominant) show high Na⁺ + K⁺ and HCO₃⁻ concentrations, whereas the groundwater samples from the TA&OA with high SO₄²⁻ and Cl⁻ might be an indication of high contents of evaporate minerals.

3.3. SOURCE IDENTIFICATION OF INRUSH WATER – THE DISCRIMINANT ANALYSIS

Discriminant analysis provides linear or quadratic functions of the variables that “best” separate cases into two or more predefined groups, which has long been applied for water source identification (Sun and Gui, 2012) and the ore exploitation (Geranian et al., 2016). In this study, the groundwater samples collected from the LA, CBA and TA&OA are signed to be group 1, 2 and 3, respectively. The classical method of Mstat (version 12) has been chosen for calculation and the results are shown in Table 3.

As can be seen from the Table, the calculated distance between S1 and group 1, 2 and 3 are 2.10, 41.9 and 94.9. According to the principal of the discriminant analysis, the sample should be classified to be the group with the lowest distance, and therefore,

the samples S1 is classified to be group 1 (LA), consistent with its source aquifer. Similarly, all of the samples are classified to be the groups consistent with their source aquifer (Table 3). Such results suggest that the discriminant analysis is effective for water source identification.

Table 3. Result of discriminant analysis

ID	Source	Predicted	Distance 1	Distance 2	Distance 3
S1	LA	1	2.10	41.9	94.9
S2	LA	1	1.77	43.6	90.7
S3	LA	1	5.54	50.7	66.8
S4	LA	1	1.58	34.4	98.8
S5	LA	1	0.83	41.8	90.7
S6	LA	1	1.87	41.3	88.2
S7	LA	1	5.84	53.0	73.1
S8	LA	1	3.72	37.1	96.4
S9	LA	1	10.3	73.8	51.5
S10	LA	1	3.45	49.5	85.4
S11	LA	1	15.9	87.1	91.3
S12	LA	1	3.55	48.4	81.9
S13	LA	1	3.74	48.1	81.6
S14	LA	1	2.60	53.2	71.6
S15	CBA	2	63.3	6.03	240
S16	CBA	2	27.5	6.64	182
S17	CBA	2	23.5	6.43	176
S18	CBA	2	83.7	21.4	268
S19	CBA	2	43.5	0.82	210
S20	CBA	2	45.3	0.84	215
S21	CBA	2	79.7	8.53	262
S22	CBA	2	65.8	7.74	253
S23	CBA	2	67.3	7.25	257
S24	CBA	2	34.1	1.91	200
S25	CBA	2	38.7	1.01	205
S26	TA&OA	3	157	325	16.2
S27	TA&OA	3	73.8	201	3.93
S28	TA&OA	3	67.0	193	9.44
S29	TA&OA	3	84.6	222	3.98
S30	TA&OA	3	90.5	233	3.53
S31	TA&OA	3	69.5	208	4.23
S32	TA&OA	3	81.7	224	4.92
S33	TA&OA	3	63.7	198	4.25
S34	TA&OA	3	73.8	213	4.15

3.4. SOURCE OF MAJOR IONS – THE UNMIX MODEL APPROACH

Based on the calculation of the Unmix model, two sources have been identified and the results are listed in the Table 4, and their contributions are shown in Figure 2. These two sources have Min Rsq = 0.96 and Min Sig/Noise = 8.60, higher than the minimum requirement of the model (Min Rsq > 0.8 and Min Sig/Noise > 2), suggesting that the modeling is efficient (Ai et al., 2014).

Weathering of silicate minerals, dissolution of carbonate and evaporate minerals are the main processes controlling the groundwater chemistry in the coal mining related aquifers (Sun and Gui, 2015). Therefore, the Source 2 dominated by $\text{Na}^+ + \text{K}^+$, and HCO_3^- should be explained to be the weathering of silicate minerals, and the Source 2 dominated by Ca^{2+} , Mg^{2+} and SO_4^{2-} can be explained to be the dissolution of evaporate minerals, to a lesser extent, carbonate minerals. Such considerations can be demonstrated by the contributions of these sources (Fig. 2), as the samples from the CBA have highest contributions from the Source 2, consistent with the truth that this aquifer system is rich in sandstone, whereas the samples from the TA&OA with high contents of carbonates and evaporates have highest contributions from the Source 1. Such a result is also consistent with the result obtained by factor analysis.

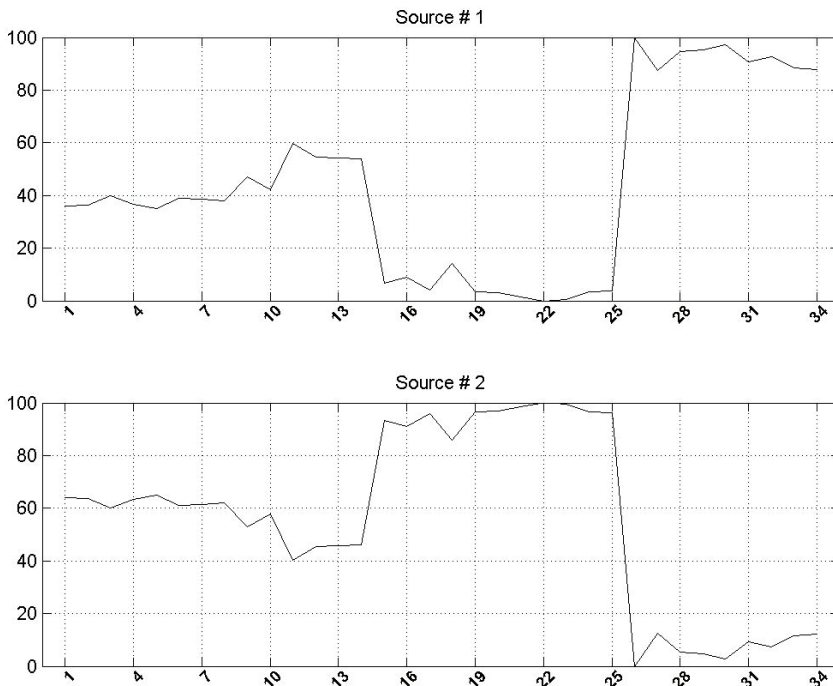


Fig. 2. Variations of source contributions

Table 4. Source compositions (mg/l)

Species	Source 1	Source 2
Na ⁺ + K ⁺	109	314
Ca ²⁺	111	-2.1
Mg ²⁺	62.3	3.56
Cl ⁻	98	107
SO ₄ ²⁻	590	-26.9
HCO ₃ ⁻	39.4	618

3.5. SOURCE OF INRUSH WATER – THE UNMIX MODEL APPROACH

As mentioned in the introduction, several methods have been applied for water source identification in coal mines. However, hydrochemical analysis is the most popular and reliable method, because water flow over the rocks can inherit the chemical signatures of the rocks, and the rocks in the different aquifer systems are different. In this study, the main rocks in the LA, CBA and TA&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. Factor analysis confirmed that the groundwater samples collected from the LA, CBA and TA&OA have undergone different types and degrees of water rock interactions. And the source aquifers of all of the samples can be identified correctly based on discriminant analysis. However, can similar result be obtained from the Unmix model?

The following information can be obtained from Figure 2: (1) According to the contributions of the Source 1, the CBA samples have low contributions from this source (<20%), which is different with those of the samples from the LA (30–60%) and TA&OA (>80%). (2) According to the contributions of the Source 2, the samples from the CBA have high contributions from this source (>80%), different with those of the samples from the LA (40–70%) and TA&OA (<20%). These obvious differences suggest that the contributions of the sources can be used for water source identification efficiently. In comparison with the discriminant analysis, the method with the Unmix model has advantages as it considers not only the statistical similarity, but also the water rock interactions as revealed by hydrochemistry.

4. CONCLUSION

Based on the analyses of major ion concentrations of groundwater samples collected from the representative aquifers in northern Anhui province, China, the following conclusions have been obtained:

- (1) Groundwater samples from different aquifers have variable concentrations of major ions, which suggest that the types and degrees of water rock interactions in these aquifers are different, and provide possibility for water source identification based on major ions;
- (2) Factor analysis has identified two potential sources responsible for the major ion concentrations of the groundwater, including dissolution of carbonates and evaporates (Factor 1) and the weathering of silicate minerals (Factor 2), discriminant analysis can classify the sources of groundwater with high efficiency;
- (3) Unmix model analysis has identified two sources similar to the factor analysis, and all of the groundwater samples with different sources can also be classified correctly. Different with those of factor and discriminant analysis, it can provide information about source of major ions and water simultaneously.

ACKNOWLEDGEMENTS

This work was financially supported by the Academic Funding for Top-talents in Disciplines of Universities in Anhui Province (gxbjZD48), the Foundation of Scholarship Leaders (Reserve) in Suzhou University (2018XJHB08).

REFERENCES

- AI J.C., WANG N., YANG J., 2014, *Source apportionment of soil heavy metals in Jiapigou goldmine based on the UNMIX model*, Environmental Science, 35 (9), 3530–3536.
- ALMASOUD F.I., USMAN A.R., AL-FARRAJ A.S., 2015, *Heavy metals in the soils of the Arabian Gulf coast affected by industrial activities: analysis and assessment using enrichment factor and multivariate analysis*, Arabian Journal of Geosciences, 8 (3), 1691–1703.
- CHEN H.J., LI X.B., LIU A.H., 2009, *Studies of water source determination method of mine water inrush based on Bayes' multi-group stepwise discriminant analysis theory*, Rock and Soil Mechanics, 30 (12), 3655–3659.
- GERANIAN H., TABATABAEI S.H., ASADI H.H., CARRANZA E.J.M., 2016, *Application of discriminant analysis and support vector machine in mapping gold potential areas for further drilling in the Sari-Gunay Gold Deposit, NW Iran*, Natural Resources Research, 25 (2), 145–159.
- GUI H.R., YAO E.Q., SONG X.M., YUAN Z.H., ZHANG G., 2011, *Research on Recycling Technology of CoalmineWater*, China University of Mining and Technology Press, Xuzhou.
- LANG Y.H., YANG W., 2014, *Source apportionment of PAHs using Unmix model for Yantai costal surface sediments, China*, Bulletin of Environmental Contamination and Toxicology, 92 (1), 30–35.
- LEWIS C.W., NORRIS G.A., CONNER T.L., HENRY R.C., 2003, *Source apportionment of Phoenix PM_{2.5} aerosol with the Unmix receptor model*, Journal of the Air & Waste Management Association, 53 (3), 325–338.
- LIU C.W., LIN K.H., KUO Y.M., 2003, *Application of factor analysis in the assessment of groundwater quality in a blackfoot disease area in Taiwan*, Science of the Total Environment, 313 (1), 77–89.
- LIU X.H., SUN L.H., 2015, *Chemical variations of groundwater in coal bearing aquifer in northern Anhui Province, China: study based on Unmix model and major ions*, Electronic Journal of Geotechnical Engineering, 20 (15), 6549–6557.

- LVOVITCH M.I., 1970, *World water balance (general report)*, World Water Balance: Proceedings of the Reading Symposium.
- SUN L.H., 2014, *Statistical analysis of hydrochemistry of groundwater and its implications for water source identification: a case study*, Arabian Journal of Geosciences, 7 (9), 3417–3425.
- SUN L.H., GUI H.R., 2012, *Establishment of water source discrimination model in coal mine by using hydrogeochemistry and statistical analysis: a case study from Renlou Coal Mine in northern Anhui Province, China*, Journal of Coal Science and Engineering (China), 18 (4), 385–389.
- SUN L.H., GUI H.R., 2015, *Hydro-chemical evolution of groundwater and mixing between aquifers: a statistical approach based on major ions*, Applied Water Science, 5 (1), 97–104.
- ROMIC M., ROMIC D., 2003, *Heavy metals distribution in agricultural topsoils in urban area*, Environmental Geology, 43 (7), 795–805.
- SARKAR D., DATTA R., HANNIGAN R., 2007, *Concepts and applications in environmental geochemistry*, Elsevier, Netherlands.
- WU Q., ZHAO Q., SUN W.J., CUI F.P., WU C., 2013, *Classification of the hydrogeological type of coal mine and analysis of its characteristics in China*, Journal of China Coal Society, 38 (6), 901–905.
- ZHANG Y., GUO C.S., XU J., TIAN Y.Z., SHI G.L., FENG Y.C., 2012, *Potential source contributions and risk assessment of PAHs in sediments from Taihu Lake, China: comparison of three receptor models*, Water Research, 46 (9), 3065–3073.