Hydrochemical characteristics and influencing factors of mine water in Tangjiahui mining area

Wang Shidong\textsuperscript{1,2}, Tang Hongwei\textsuperscript{1,2}, Yang Zhibin\textsuperscript{1,2}, Liu Ji\textsuperscript{1,2}, Zhu Hongjun\textsuperscript{1,2}, Xu Feng\textsuperscript{1,2}, Zhu Kaipeng\textsuperscript{1,2}, Fan Juan\textsuperscript{1,2}, Fang Gang\textsuperscript{1,2}

\textsuperscript{1}Xi’an Research Institute of China Coal Technology & Engineering Group Corp, Xi’an, Shaanxi, 710054, China; \textsuperscript{2}Key Laboratory of Coal Mine Water Hazard Prevention and Control Technology in Shaanxi Province, Xi’an, Shaanxi, 710054, China;

Abstract: In water-deficient areas, the reuse of water discharged from coal mining is highly desirable. In order to study the water quality in the Tangjiahui mining area in Jungar Coalfield (Inner Mongolia, China), 34 groups of mine water samples collected at various stages during coal mining process were analyzed for their hydrochemical characteristics using conventional and statistical methods. A Piper trilinear diagram was used to analyze the main ion composition characteristics and the hydrochemical type of the mine water. Gibbs map and ion correlation methods were used to investigate the sources and influencing factors of the main ions in mine water. The results showed that the TH of the mine water in the study area was in the range of 219.52–390.6 mg/L with an average of 315.04 mg/L, which can be classified as slightly hard/ hard water. The TDS was in the range of 926.61–1889.56 mg/L with an average of 1514.31 mg/L, which mostly belongs to brackish water. The cation content in the mine water was ranked from the highest to the lowest as Na\textsuperscript{+} > Ca\textsuperscript{2+} > Mg\textsuperscript{2+}, while the anion content was ranked as Cl\textsuperscript{−} > HCO\textsubscript{3}− > SO\textsubscript{4}2−. The Na\textsuperscript{+} mass concentration was in the range of 179.00–523.06 mg/L with an average of 399.77 mg/L, while the Cl\textsuperscript{−} mass concentration was in the range of 207.10–812.63 mg/L with an average of 550.88 mg/L. The hydrochemical type of the mine water was Cl-Na. According to the correlation matrix of the various chemical indicators in the mine water, the TDS was significantly positively correlated with Na\textsuperscript{+}, Ca\textsuperscript{2+}, Cl\textsuperscript{−}, and SO\textsubscript{4}2−. Of these, the main sources of TDS were Na\textsuperscript{+} and Cl\textsuperscript{−}, as these had correlation coefficients > 0.9. The hydrochemical characteristics of the mine water were mainly controlled by the condensation-crystallization and anti-cation exchange, which indicated the main ions were largely derived from the dissolution of halite. Due to its high TDS, EC, SAR, and Na\% values, the mine water in the study area was not suitable for human consumption and agricultural irrigation. These results can provide a reference towards water resource management and the sustainable use of mine water by local governments.

Keywords: mine water, Tangjiahui mining area, hydrochemical characteristics, Piper trilinear diagram, Gibbs map, ion source
Introduction

Coal mining plays an important role in the economic development of many countries worldwide (Schneider et al. 2012; Li et al. 2015). According to information disclosed by the Energy Information Administration (EIA 2019) of the United States of America, the total global coal output in 2019 was around 7.6 billion tons, of which China’s contribution was around 3.7455 billion tons, or nearly 50% of the total (Yu et al. 2020). Coal has always accounted for over 60% of China’s energy production and consumption, and is therefore the main energy source in the country (Liu et al. 2020). However, within the country, coal and water resources exhibit a reverse distribution—that is, areas with abundant coal resources are basically lacking in water resources (Wu et al. 2017). For example, the water-scarce arid and semi-arid regions of western China have extremely abundant coal resources. These water shortages are exacerbated by the coal mining process, as in order to prevent flood accident occurrence, a large amount of mine water is required to be drained before or during coal mining (Huang et al. 2017; Xu et al. 2019). Therefore, research on mine water reuse is critical towards finding ways of mitigating this problem. As the quality of the mine water directly affects its usefulness in different applications, the development of quality evaluation protocols is of great importance towards the sustainable utilization of mine water resources in western China (Li 2018; Liu et al. 2020).

The hydrochemical characteristics of groundwater can directly reflect its quality, and possible changes in the groundwater quality can be indirectly understood via analysis of the factors influencing these characteristics, making this a valuable area of study (Li et al. 2018; Liu et al. 2019a). To date, therefore plenty of research has been carried out on the hydrochemical characteristics of groundwater. The conventional Piper trilinear diagram (Piper 1944) and Gibbs map (Gibbs 1970) have been widely used to determine the hydrochemical characteristics and influencing factors of the groundwater. In recent years, it has been found that a combination of statistical and conventional methods is more effective compared to traditional approaches (Redwan et al. 2016; Wang et al. 2017; Li et al. 2018; Sefie et al. 2018). Li et al. (2013a; 2013b) used a correlation analysis method in conjunction with the Piper trilinear diagram and Gibbs map to determine the hydrochemical characteristics and influencing factors for the shallow groundwater in both Pengyang County and Dongsheng Coalfield in China. Cloutier et al. (2008) used principal component analysis (PCA) and other conventional methods to determine the hydrochemical processes occurring in the groundwater in Basses-Laurentides, Canada.

The Tangjiahui coal mining area is part of the Jungar Coalfield, which is located in the arid and semi-arid regions of Inner Mongolia. The continuation of coal seam mining in Tangjiahui is threatened by the Ordovician limestone karst fissure aquifer, which is the main local water resource, as a large amount of mine water is required to be drained before and during the mining process (Li et al. 2020). If the mine water cannot be
recycled, this equates to wastage of the valuable water resources. For this reason, this area has been selected as the object of research in the present study, with the aim of providing a scientific basis for local water resource management and rational utilization of mine water in the area. To this end, Ordovician underground mine water samples were analyzed at different stages of the mining process, and the hydrochemical characteristics and influencing factors of the mine water were investigated.

**Overview of the study area**

The Tangjiahui coal mine area is located in the central part of the Jungar Coalfield in Ordos City (Fig. 1). The area of the mining field is 28.58 km², and the minable coal seams are Nos. 4, 5, 6, 9 upper, and 9 lower, with a resource reserve of 0.805 billion tons. Of these, the main coal seam is No. 6, with a design capacity of 6 million tons/year. The coal mine was constructed in October 2010 and put into operation in 2013. The study area is arid and semi-arid with occasional rain. The average annual precipitation is around 400 mm, and the rainfall is mainly concentrated in the three summer months of July, August, and September, which account for 60–70% of the annual precipitation. The evaporation is intense, with average annual evaporation > 2000 mm, resulting in a relative lack of local groundwater resources. The surface drainage in the study area is mainly to the Yellow River. Riverside source fields (the main water source is the Ordovician aquifer) were constructed in areas which the Yellow River flows through, providing domestic water for residents.

The mine formations may be ranked from oldest to newest, as follows: Middle Ordovician Majiagou Formation (O₂m), Upper Carboniferous System Taiyuan Formation (C₂t), Lower Permian Shanxi Formation (P₁s), Lower Shihezi Formation (P₁x), Upper Shihezi Formation (P₂s), Shiqianfeng Formation (P₂sh), Lower Cretaceous Zhidan Group (K₁zh), Neogene Pliocene (N₂), and Quaternary System (Q). The main coal seam in the study area is located at the Taiyuan Formation stratum, which is mainly threatened by the Ordovician limestone aquifer with high water pressure and abundant water during the seam mining process. The depth of the rock aquifer is > 500 m, and it contains topographic features such as corrosion fissures, karst caves, and dissolved pores. The unit water inflow is 0.024–34.321 L/s m, and the water abundance varies from weak to very strong. Therefore, in order to prevent the occurrence of water hazard accidents, the pressure and amount of water in the aquifer should be reduced to safe levels through drilling drainage before mining.

**Sample collection and testing**
In total, 34 mine water samples were collected from the drainage boreholes in situ during different mining stage. For each water sample, 1 L was collected. Prior to sample collection, the container was rinsed with the sampled water 3 times. After collection, the water sample container was immediately covered, sealed, and labeled with the sample collection time, sampling number, surveillance project number, and name of the sample collector.

The test indicators for the water sample include pH, cations (K+, Na+, Ca2+, Mg2+, NH4+, and Fe3+), anions (Cl−, SO42−, HCO3−, CO32−, NO2−, and NO3−), total hardness (TH), and total dissolved solids (TDS). Of these, Ca2+ and Mg2+ were measured using an EDTA complexometric titration method, Cl− was measured using a silver nitrate titration method, and SO42− was measured using a barium sulfate gravimetric method. The total dissolved solids (TDS) value was obtained by calculating the mass concentrations of various ions.

In order to ensure the accuracy of the sample test results, the main anion-cation equilibrium in the water quality analysis results was calculated. When the equivalent concentration difference ratio is within 5%, it can be considered that ionic equilibrium has been reached and that the test results are valid. Otherwise, the test results are considered invalid (Ghahremanzadeh et al. 2018; Adimalla and Qian 2019).

In this study, the SPSS software package (IBM) was used for the statistical analysis of the water sample data. The hydrochemical software package AquaChem (Waterloo Hydrogeologic) was used to plot the Piper trilinear diagram for hydrochemical type analysis. In addition, the factors influencing the hydrochemical characteristics of the groundwater were analyzed using ion correlation analysis.

**Fig. 1** Location of the study area.

### Results and discussion

### Hydrochemical characteristics

#### Ion composition characteristics

The statistics of the hydrochemical parameters for the 34 mine water samples are shown in Table 1. As the table showed, the TH of all the samples was in the range of 219.52–390.6 mg/L with an average of 315.04 mg/L, corresponding to slightly hard or hard water based on the classification of Chinese Standards for Drinking Water Quality (Ministry of Health of the P. R. China 2006; Li et al. 2010). The TDS was in the range of 926.61–1889.56 mg/L with an average of 1514.31 mg/L, revealing that most of the water samples were brackish (WHO...
These results indicated that most of the water samples were not potable. The standard deviations of the TH and TDS values were 37.39 and 200.61 mg/L, respectively, with the much greater spread in the TDS values indicating that the geohydrologic conditions had a greater influence on the TDS of mine water (Liu et al. 2019b).

The cation content in the mine water was ranked in the following order (from highest to lowest): Na$^+$ > Ca$^{2+}$ > Mg$^{2+}$. The Na$^+$ mass concentration was in the range of 179.00–523.06 mg/L with an average of 399.77 mg/L, the Ca$^{2+}$ mass concentration was in the range of 66.43–114.47 mg/L with an average of 93.25 mg/L, while the Mg$^{2+}$ mass concentration was in the range of 9.3–34.73 mg/L with an average of 19.95 mg/L. The anion content in the mine water was ranked from highest to lowest as Cl$^-$ > HCO$_3^-$ > SO$_4^{2-}$. The Cl$^-$ mass concentration was in the range of 207.10–812.63 mg/L with an average of 550.88 mg/L. The SO$_4^{2-}$ mass concentration was in the range of 105.33–249.85 mg/L with an average of 710.39 mg/L. The HCO$_3^-$ mass concentration was in the range of 219.73–343.22 mg/L with an average of 279.71 mg/L. These results showed that all the mine water samples were dominated by Na$^+$ and Cl$^-$ with high standard deviations and TDS values, indicating that the update capacity of the groundwater in the study area was weak, and that the acceptance of fresh water recharge was poor (Li et al. 2018; Liu et al. 2019b).

Table 1 Descriptive statistics of main hydrochemical parameters.

| Hydrochemical type                           |
|---------------------------------------------|

The use of a Piper trilinear diagram is conducive to understanding the main ion composition and hydrochemical type of the groundwater (Singh et al. 2010; 2012). Here, the Piper trilinear diagram was plotted according to the content of the main ions from the mine water in the study area (Fig. 2). As the figure showed, all mine water samples were close to the Cl$^-$ end member and far away from the SO$_4^{2-}$ and CO$_3^{2-}$ + HCO$_3^-$ end members. The anion was mainly Cl$^-$ (anion triangular chart at the lower right corner). In the cation triangular chart, the water samples were distributed close to the Na$^+$ end member and far away from the Ca$^{2+}$ and Mg$^{2+}$ end members. These ions were probably obtained from the weathering dissolution of the carbonate and evaporite (Tiwari et al. 2017). All the water samples were located in the upper diamond shaped I region, indicating that the amount of alkali metals exceeded that of alkaline-earth metals, that the strong acid exceeded the weak acid, and that the main hydrochemical type of the mine water was Cl-Na.

Fig. 2 Piper trilinear diagram of mine water.
Correlation between the chemical indicators

The ions in groundwater do not exist in isolation, as they interact with each other to some extent, and the correlation relationships between them reflect the main sources of the ions (Zaidi et al. 2019). The interrelation matrix of various chemical indicators in the mine water (Table 2) showed that, remarkably, positive correlations ($P > 0.01$) existed between TDS and $\text{Na}^+$, $\text{Ca}^{2+}$, $\text{Cl}^-$, and $\text{SO}_4^{2-}$, indicating that these ions contributed to TDS. In particular, the correlations between TDS with $\text{Na}^+$ and $\text{Cl}^-$ were $> 0.9$, indicating that these two ions were the main sources of TDS. However, there were no obvious correlations between TDS and $\text{Mg}^{2+}$ and $\text{HCO}_3^-$, indicating that some chemical reactions might take place. For example, cation exchange, precipitation and crystallization can disturb the ion contents, which caused a weak correlation between TDS and $\text{Mg}^{2+}$ as well as $\text{HCO}_3^-$ (Li et al. 2018). In addition, $\text{Na}^+$ and $\text{Cl}^-$ significantly correlated ($r = 0.943$), and the concentration were both high, indicating that these ions were most likely derived from the dissolution of a single substance such as halite, whereas $\text{HCO}_3^-$ was insignificantly correlated with $\text{Ca}^{2+}$ and $\text{Mg}^{2+}$, suggesting that the dissolution of limestone does not play a significant role. Further, the correlation coefficient between $\text{SO}_4^{2-}$ and $\text{Ca}^{2+}$ was 0.161, which was not significant, implying that gypsum was not the main source of $\text{SO}_4^{2-}$.

Table 2 Correlation matrix of main hydrochemical parameters.

Analysis of influencing factors

Rock weathering

During the runoff process, the groundwater continuously reacts with the surrounding rock minerals and is also influenced by atmospheric precipitation and evaporation condensation (Utom et al. 2013). Studying these influencing mechanisms therefore sheds light on the hydrochemical origins of groundwater. Gibbs (1944) first proposed the use of correlations between TDS and $\text{Na}^+$, $\text{Ca}^{2+}$, $\text{Cl}^-$, and $\text{HCO}_3^-$ to study the formation mechanisms of the river surface water, including evaporative crystallization, rock weathering, and atmospheric precipitation effects. In recent years, the Gibbs map has been widely used to investigate the formation mechanism of groundwater (Tiwari et al. 2017; Liu et al. 2020).

Here, a Gibbs map was used to analyze all the mine water samples (Fig. 3). The results showed that the TDS values of the samples were relatively high (>1000 mg/L), and that the $\text{Na}^+/(\text{Na}^+ + \text{Ca}^{2+})$ ratios and $\text{Cl}^-/(\text{Cl}^- + \text{HCO}_3^-)$ ratios were $> 0.6$. The mine water samples therefore mainly occurred in the evaporation dominance area, indicating that the formation mechanism of the mine water in the study area was through condensation
crystallization, whereas the contributions from rock weathering and atmosphere precipitation were small.

Fig. 3 Gibbs map of mine water in the study area.

*Cation exchange*

Cation exchange is another important process in the formation of groundwater hydrochemical components. Schoeller (1965) proposed the use of two indicators, CAI-1 and CAI-2, to determine the occurrence of cation exchange in the groundwater. The relevant equations are:

\[
CAI-1 = \frac{Cl^- - (Na^+ + K^+)}{Cl^-} \quad (1)
\]

\[
CAI-2 = \frac{Cl^- - (Na^+ + K^+)}{HCO_3^- + SO_4^{2-} + CO_3^{2-} + NO_3^-} \quad (2)
\]

If the two indicators are positive, it indicates that cation exchange has occurred, as follows:

\[
2Na^+ + (Ca, Mg)X_2 = (Ca, Mg)^{2+} + 2NaX \quad (3)
\]

If, however, the two indicators are negative, it indicates the occurrence of inverse cation exchange, as follows:

\[
(Ca, Mg)^{2+} + 2NaX = 2Na^+ + (Ca, Mg)X_2 \quad (4)
\]

Fig. 4a shows that both CAI-1 and CAI-2, were negative, indicating that inverse cation exchange occurred in the mine water. Further, the relationship between \((K^+ + Na^+ + Cl^-)\) and \((Ca^{2+} + Mg^{2+} + SO_4^{2-} + HCO_3^-)\) is also normally used to verify the presence of cation exchange in the groundwater, with a slope of around \(-1\). In this case, the line of best fit between these quantities in the mine water samples was \(Y = -0.9825X - 0.0183\) (\(R^2 = 0.9962\)) (Fig. 4b); hence, the inverse cation exchange did occur in the study area.

Fig. 4 Relationships between (a) CAI-1 and CAI-2 and (b) \((K^+ + Na^+ + Cl^-)\) and \((Ca^{2+} + Mg^{2+} + SO_4^{2-} + HCO_3^-)\).

*Source analysis of main ions*

The results presented thus far showed that the mine water in the study area was controlled by a combination of weathering dissolution and condensation crystallization (Gan et al. 2018; Mahato et al. 2018). Figure 5 shows the relation diagram of \(Ca^{2+}/ Na^+\), \(HCO_3^-/ Na^+\), and \(Mg^{2+}/ Na^+\) of the evaporite, silicate, and carbonate, whence it can be seen that the mine water samples were distributed between the silicate rock and evaporate, but were closer to the latter, indicating that the ion components in the mine water were mainly derived from the dissolution of evaporite. This seemed to contradict the data seen earlier which indicated that
the mine water samples mainly came from Ordovician limestone water. Through calculation of the saturation levels of the dolomite, calcite, gypsum, and halite (Fig. 6), it was found that the saturations of dolomite and calcite were both positive, while those of gypsum and halite were both negative. This indicated that the former two minerals were excessively dissolved and exhibited a saturated state with the occurrence of crystallization, whereas the latter two did not saturate and can therefore be further dissolved. Hence, the supposition that Na+ and Cl− are the main ions in the mine water is correct.

Additionally, the proportional relationships between the ions in the mine water (Fig. 7) can be used to determine their main sources (Li et al. 2018; Liu et al. 2019b). Fig. 7a showed that the Na+/Cl− values of the mine water samples were close to 1, further indicating that the dissolution of halite was the main source of the Na+ and Cl− in the mine water. When \( \frac{(\text{HCO}_3^- + \text{SO}_4^{2-})}{(\text{Ca}^{2+} + \text{Mg}^{2+})} \) was equal to 1, it indicates that the HCO₃⁻, SO₄²⁻, Ca²⁺, and Mg²⁺ ions in the groundwater were derived from the weathering dissolution of dolomite, calcite, and gypsum. The ratios of \( \frac{(\text{HCO}_3^- + \text{SO}_4^{2-})}{(\text{Ca}^{2+} + \text{Mg}^{2+})} \) in the mine water samples were all > 1 (Fig. 7d), indicating that gypsum was not the main source of these ions. The ratio of Ca²⁺ and HCO₃⁻ as well as that of Ca²⁺ + Mg²⁺ and HCO₃⁻ can be used to qualitatively determine whether the ions were derived from dolomite and calcite. These ratios were mostly < 1 in the mine water samples (Fig. 7b and 7c), indicating that the dissolution of carbonate was not the main source of Ca²⁺ and Mg²⁺.

**Fig. 5** Relationships between Ca²⁺ and Na⁺, HCO₃⁻ and Na⁺, and Mg²⁺ and Na⁺.

**Fig. 6** Relationships between the saturation of: (a) dolomite and calcite, (b) gypsum and halite.

**Fig. 7** The correlation between mine water ions in the study area: (a) Na⁺-Cl⁻, (b) HCO₃⁻-Ca²⁺, (c) HCO₃⁻-(Ca²⁺+Mg²⁺), and (d) (SO₄²⁻+HCO₃⁻)-(Ca²⁺+Mg²⁺).

**Suitability for human consumption**

The mine water samples were compared against the drinking water standards stipulated by the World Health Organization (WHO 2017) as well as by the Chinese government (Ministry of Health of the P. R. China 2006). Of the 34 samples, TDS was < 1000 mg/L in only two, while all the others exceeded the quality limits
for drinking water in China, making them unsuitable for human consumption without treatment. The TH values of the analyzed water samples were within the slightly hard/hard range. The high TH, which is mainly due to Ca\(^{2+}\) and Mg\(^{2+}\), may be attributed to the existence of alkaline soil. However, the TH values in all mine water samples were < 450 mg/L, which is the allowable limit for drinking water in China.

**Suitability for irrigation**

*Alkali and salinity hazard*

The electrical conductivity (EC) and Na concentrations are of great importance in the classification of irrigation water (Ayers and Westcot 1985; Singh et al. 2010). Salts not only directly affect the growth of plants, but also affect the structure, permeability, and aeration of soil, thereby indirectly influencing plant growth (Collins and Jenkins 1996). The total concentration of soluble salts in irrigation water can be classified as low (EC =< 250 µS cm\(^{-1}\)), medium (250–750 µS cm\(^{-1}\)), high (750–2250 µS cm\(^{-1}\)), and very high (2250–5000 µS cm\(^{-1}\)) (Richards 1954; Singh et al. 2010). The high EC leads to the formation of saline soil, while the high sodium concentration produces alkaline soil. In semi-arid regions, water losses through evaporation are a serious issue, as they often result in the presence of excess solute in irrigation water (Li et al. 2018; Liu et al. 2019a; Singh et al. 2010). These types of problems most commonly occur in areas with poor drainage, which causes the water level of groundwater to rise up to the vicinity of the plant rooting zone, leading to sodium salt accumulation by capillary ascent and evaporation (Mahato et al. 2018; Singh et al. 2010).

The sodium adsorption ratio (SAR) or the alkali hazard is determined by both the absolute and relative concentration of the cations, which can be estimated using the following equation: SAR = Na\(^+\) / [(Ca\(^{2+}\) + Mg\(^{2+}\))/2]\(^{0.5}\). According to the SAR value, the irrigation water can be classified into four alkalinities (S1-S4): low (0-10), medium (10-18), high (18-26), and very high (> 26) (Richards 1954; Singh et al. 2010). The SAR of the mine water samples was in the range of 4.5–12.4. In the US salinity diagram (Fig. 8), EC represents the salinity hazard and SAR represents the alkaline hazard, indicating that most of the mine water samples were in the C4S2 region, meaning that the mine water was not suitable for irrigation.

*Fig. 8* US salinity diagram for classification of irrigation waters.

_Electrical conductivity (EC) and sodium percentage (Na\%)_
The sodium content (Na%), in the mine water samples was calculated according to the equation \( \text{Na\%} = \frac{(\text{Na}^+ + \text{K}^+)}{(\text{Ca}^{2+} + \text{Mg}^{2+} + \text{Na}^+ + \text{K}^+)} \times 100 \) (Wilcox 1955). The Na% values in the analyzed mine water samples were in the range of 57.1–78.9%. A high sodium content results in soil overturn and a decrease in permeability. The plot of EC vs. NA% (Fig. 9) showed that most of the mine water samples were in the doubtful/unsuitable region, indicating that the water was not suitable for irrigation (Fig. 9).

**Fig. 9** Plot of NA% vs. EC for the mine water samples.

**Conclusions**

In this paper, 34 mine water samples were collected from the Tangjiahui coal mine area, and the hydrochemical characteristics and influencing factors of mine water were determined by combining traditional with statistical methods. Meanwhile, the suitability for domestic consumption and irrigation was evaluated. The main conclusions which may be drawn from the results presented herein are as follows:

1. The mine water in the study area may be classified as slightly hard/hard and brackish. The main cation and anion in the mine water were Na\(^+\) and Cl\(^-\), respectively, and the main hydrochemical type was Cl-Na.

2. TDS showed a remarkably positive correlation with Na\(^+\), Ca\(^{2+}\), Cl\(^-\), and SO\(_4^{2-}\). Na\(^+\) was significantly correlated with Cl\(^-\), while HCO\(_3^-\) did not show significant correlation with Mg\(^{2+}\), SO\(_4^{2-}\), or Ca\(^{2+}\).

3. The mine water ions in the study area were mainly derived from condensation crystallization and inverse cation exchange effects in the groundwater. The saturations of dolomite and calcite in the mine water were positive, while those of gypsum and halite were negative. The dissolution of halite was the main source of the ions in the mine water.

4. Due to its high TDS, EC, SAR, and Na\% values, the mine water in the study area was not suitable for human consumption or agricultural irrigation.

The results are significant to mine water management in the study area. Due to the poor quality, the mine water can not be discharged at random, which needs to be properly treated and reasonably managed by the government.

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