Hydrogeochemical Characteristics and Formation Processes of Ordovician Limestone Groundwater in Zhuozishan Coalfield, Northwest China

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Abstract

Groundwater is essential for water supplies in arid and semi-arid region. Analyzing the characteristics and formation mechanisms of groundwater in mining area is crucial for coal mine water prevention and rational groundwater resources management. This study reached the hydrogeochemical characteristics and evolutions of Ordovician groundwater in the Zhuozishan coal mine, northwest China. 34 groundwater samples were collected for hydrogeochemical analyses and piper trilinear diagram, gibbs diagram, ion ratio diagram were used to research groundwater formation processes. The results indicate that the concentration of $\text{Na}^+ + \text{K}^+$, HCO_3^- , Cl^- , SO_4^{2-} , TDS, and pH increase from the recharge area to the discharge area, however, that of Ca^{2+} and Mg^{2+} decreases. And each indicator varied greatly in the north and south of the coalfield. Hydrogeochemical types of the runoff from Zhuozishan Mountain to Gongdeer Mountain and then to the south-north directional change obviously. And the groundwater process is controlled by rock weathering action and cation exchange. Whereas the hydrochemical type of the runoff from Zhuozishan Mountain to the south is always $\text{SO}_4\text{--Cl--Ca--Na}$ type, and groundwater process is controlled from rock weathering action to evaporation concentration and reverse cation exchange. When it comes to ion source, Ca^{2+} and Mg^{2+} mainly origin from the silicate dissolution, while Na^+ and K^+ are mainly derived from cation exchange and barely from the halite dissolution. In summary, in the northern part of the coalfield, due to geological structure, a retention area is formed with groundwater, and the runoff process is disordered with a complicated formation process. The middle part is not blocked by the geological structure, the runoff direction do not change, so the formation of groundwater is simple. And in the southern part, due to the deepening stratum and fault blocking, a retention zone is formed, making groundwater formation process more complicated. This study will contribute to groundwater resources management and mine water inrush control in this coalfield and at other mines.

Introduction

In recent times, the global demand for fresh groundwater has increased because of population increase, industrial progress and intensive irrigation activities (Rajesh et al. 2015). However, in many counties groundwater is suffering from water table decline and quality contamination through natural processes and anthropogenic mining activities, such as India (Singh et al. 2012), Bangladesh (Islam et al. 2017), Turkey (Rajesh et al. 2015), China (Li et al. 2018a) and so on. During the coal mining, huge quantities of groundwater should be discharged from mine ditch and pits which leads to groundwater recession. Meanwhile, hydraulic connectivity of different aquifers caused by groundwater discharge will result in water quality contamination. Generally, mining activities have noticeable impacts on groundwater quantity and quality (Vidic et al. 2013). Groundwater geochemistry characteristics and its process can provide a good understanding of groundwater circulation, quality and possible changes in it (Huang et al. 2018). As such, studying Hydrogeochemical characteristics and formation process of groundwater in mining areas is essential.

China is rich in coal resources.

China, with abundant coal resources, is the largest coal producer and consumer country in the world (Wu et al. 2018). Coal consumption was 2.22 billion tons, accounting for 57.7% of its total consumption in 2019(Wang et al. 2020). However, hydrogeologic conditions of most coal mines in China are complicated. Hence, China has suffered more mine water inrush than other countries. It is reported that over 4500 deaths due to mine water inrush from 2000 to 2014(Wu et al. 2018). The Zhuozishan coalfield owned the westernmost part of Ordos Basin, China, has large coal reserves. The coal seams are mainly found in carboniferous and Permian aged formations. Mine water inrush from overlying Ordovician limestone aquifer is the biggest hazard threatening the safety mining in this area. In 2010, a particularly serious water inrush occurred in Luotuoshan Mine(belong to Zhuozishan coalfield), with a peak water inrush of 60036.0 m³ /h, which caused huge economic losses and endangered human lives. If the hydrochemical characteristics of groundwater are mastered in advance, we can exploit it to identify mine water source to prevent mine water inrush. Therefore, studying the hydrochemical characteristics and its formation processes of Ordovician limestone aquifer in Zhuozishan Coalfield is of great significance.

The Zhuozishan coalfield, owned Wuhai administrative region, lies within a arid to semi-arid area. And the annual precipitation and evaporation rate in this area is 140 mm and 3600 mm, respectively per year. In addition, surface water is scarce. Thus, groundwater is the most valuable resource for drinking, domestic and production water in Wuhai city. It is reported that 42.4% of Wuhai municipal water supply come from groundwater, and 8% of groundwater is from drainage of Ordovician limestone in Zhuozishan Coalfield(Wuhai Water Resources Bulletin). And with the intensive mining activities, drainage water of Ordovician limestone is increasing. Therefore, in the future, the Ordovician limestone water from Zhuozishan Coalfield is an important groundwater source for the economic development of Wuhai city. However, coal mining activity can inevitably affect the hydrochemical characteristics and the groundwater evolution, which in turn influences how water quality evolves (Li et al.2018b). Hence, it is always indispensable and essential to gain a full understanding of hydrogeochemical characteristics and formation process of Ordovician limestone groundwater in Zhuozishan Coalfield. It will be conducive to scientific planning, effective utilization, and protection of groundwater resources in Zhuozishan coalfield even Wuhai city.

Study Area

Location and Climate

The Zhuozishan coal mine is an important coal district of Ordos Basin. It is located between Zhuozi mountain and Gangdeer mountain, and extends between longitude 107°2'15"–107°5'45" and latitude 39°21'15"–39°22'41". It is located at the western continental margin of the North China Platform, and is characterized by rolling hilly landscape. Recently, Zhuozishan coalfield consists of 10 mining areas named Kabuqi, pinggou, Luotuoshan, Baiyinwusu, Dilibangwusu, Tianyu, Hairong, Zhengfeng, Limin and Qipanjiang, respectively.

This area is in the arid to semiarid region, and climate is characterized by large temperature differences. The annual average temperature is 7.6°C, with a daily highest and lowest air temperature of 36.7°C and – 31.6°C. The annual precipitation is 140mm and annual evaporation is 3600 mm per year. Obviously, the annual evaporation is about 25 times as the annual precipitation. In addition, surface water resources are scarce. Hence, the groundwater resources are more important in this area.

Geology and Hydrogeology

The main outcropped strata in Zhuozishan Coalfield are Quaternary and Neogene formations. And main coal seams are mainly found in Permian (Shanxi unit P_1^s) and Carboniferous (Upper Taiyuan unit C_2^{t2}) aged formations. Based on hydrogeological drilling in this area, the formations, from old to new, are Zhuozishan unit of Ordovician Series, Taiyuan unit of the Carboniferous Series, Shanxi unit of the Permian Series, Palaeogen and Quaternary. When it comes to water-bearing system, Zhuozishan Coalfield is an independent karst groundwater system, which belongs to the Cambrian-Ordovician carbonate karst groundwater system in Ordos Basin. According to karst groundwater hydraulic connection, water quantity characteristics and runoff, discharge conditions, it is divided into four relatively independent water-cycling systems, including karst water subsystem in Qianlishan (图), karst water subsystem in Gongdeer Mountain (图), karst water subsystem in the northern part of Zhuozishan Mountain (图), and karst water subsystem in the southern part of Zhuozishan Mountain (图), as shown in Fig. 1.

The study area is located in Northwest China, where surface water is scarce and precipitation is main recharge source of Ordovician limestone groundwater. The precipitation mainly recharges the aquifer in the outcrop area of the Ordovician limestone in Zhuozishan Mountain, with the runoff going westwards, and passing through the syncline in Kabuqi to the eastern margin of Gongdeer Mountain. It is blocked by water-blocking fracture, and changes to north-south directional runoff in the syncline, which is finally discharged in the form of spring or flows to the Yellow River and discharged sideways, so that the syncline in Kabuqi divides Zhuozishan Mountain into karst water subsystems in the south and north. On the other hand, groundwater will also flow southward from Zhuozishan Mountain, but due to the south strata extension and the blocking of Zhengyiguan strike-slip fault, a retention zone will be formed, and it will also be finally discharged in the form of spring or flows to the Yellow River and discharged sideways. Due to the limitation of groundwater recharge in Ordovician limestone, groundwater circulation is relatively slow, and strong runoff zone will form only in karst faulted structure and deep karst gully.

Samples Collection and Analysis

34 Ordovician limestone groundwater samples were collected randomly from the 10 mining areas belonged to Zhuozishan coalfield. And the specific sampling locations used are shown in Fig. 1. The sampling and preservation methods and processes observe national standard (Ministry of Environmental

Protection of the PR China 2009). Prior to water sampling, bottles were rinsed for three times, and at every sampling location, two 1-L plastic bottles were filled. One was used for cations and anions tests and the other was used for in-situ test. A multiparameter probe device was taken for total dissolved solids (TDS) and pH. Then the sampling bottles were labelled, sealed and carried to the laboratory of coal mine water hazard prevention and control technology in Shaanxi Province for cations and anions determinations within 24 hours. The concentrations of anions (Cl^- , SO_4^{2-}) were determined by anion chromatograph with anion columns (Thermo Company, Waltham, USA), and cations (Ca^{2+} , Mg^{2+} , Na^+ , K^+) were measured by anion chromatograph with cation columns. Besides, HCO_3^- was analyzed through titrimetric method.

Results And Discussion

General Hydrochemistry

According to the detection results, Seven conventional ions (Ca^{2+} , Mg^{2+} , Na^+ , K^+ , Cl^- , SO_4^{2-} , HCO_3^-) concentration, pH and TDS contour maps were drawn respectively (Fig. 2). The runoff-recharging directions of Ordovician limestone water can be divided into two types: one is from the Zhuozishan Mountain to Gongdeer Mountain, and then to south-north runoff, and the other is the runoff from the Zhuozishan Mountain to the south. Obviously, the distribution characteristics of hydrochemical indicators are related with the direction of the groundwater runoff. The concentration of Na^+ + K^+ , HCO_3^- , Cl^- , SO_4^{2-} , TDS, and pH generally increase from the recharge area to the discharge area, whereas Ca^{2+} and Mg^{2+} showed the opposite distribution characteristic. Especially in the central region, this distribution is more conspicuous. The contour distribution of various indicators are relatively dense in the northern and southern parts, indicating that the hydrochemical indicators varied greatly.

Hydrochemical Types

Piper trilinear diagram is favourable for determining connections of different dissolved components and types of groundwater according to its hydrochemical characters (Tiwari et al. 2017, Shakya et al. 2019). As indicated by the Piper diagram (Fig. 3). The hydrochemical types of Ordovician limestone water in the Zhuozishan Coalfield mainly included $\text{SO}_4\text{--Cl--Ca--Na}$, $\text{HCO}_3\text{--SO}_4\text{--Cl--Ca--Na}$, $\text{HCO}_3\text{--Cl--Ca--Na}$, and $\text{HCO}_3\text{--Ca--Na}$ type. According to the direction of groundwater runoff, the changes in hydrochemical types can be divided into two types, as shown in the figure: ① from the Zhuozishan Mountain to the Gongdeer Mountain and then to the north-south directional runoff, with the hydrochemical type changing from $\text{SO}_4\text{--Cl--Ca--Na}$ type in the upstream region to $\text{HCO}_3\text{--SO}_4\text{--Cl--Ca--Na}$, $\text{HCO}_3\text{--Cl--Ca--Na}$, or $\text{HCO}_3\text{--Ca--Na}$; ② the runoff from the Zhuozishan Mountain to the south, and the hydrochemical type is always the $\text{SO}_4\text{--Cl--Ca--Na}$ type. According to TDS contour map, the TDS concentration of runoff from the Zhuozishan Mountain to the Gongdeer Mountain and then to the north is 1,200–1,400 mg/L, and that of the runoff to the south ranges from 600–1,000 mg/L. The TDS concentration of the runoff from the Zhuozishan Mountain to the south is 1,200–2,800 mg/L, with a gradual increase. Obviously, the proportion of HCO_3 (mg equivalent)

increases, whereas the TDS concentration changes only slightly in the runoff process from the Zhuozishan Mountain to the Gongdeer Mountain and then to the south or north. In the runoff process from the Zhuozishan Mountain to the south, the hydrochemical type remains unchanged but the TDS concentration gradually increases, indicating that hydrochemical reactions occur in the groundwater during the runoff process.

Hydrochemical Process

1. Rock weathering action

The Gibbs diagram (Gibbs 1970) was an important means primitively to study the mechanisms surface water evolution, and now it is widely used in groundwater researches (Li et al. 2016a). Generally, the main control factors can be divided into three types: evaporation and concentration dominance, rock weathering dominance, and precipitation dominance (Pehlivan et al. 2020). The analytical results show that groundwater hydrochemical process includes two types: the runoff from Zhuozishan Mountain and then to north-south directional runoff, and groundwater control action is mainly rock weathering action the runoff from Zhuozishan Mountain to the south, and the control action of groundwater transforms from the rock dominance to the evaporation dominance from the upstream to downstream. However, the Ordovician limestone groundwater is deeply buried, which will not be subjected to evaporation and concentration. Therefore, it can be speculated that the reverse cation exchange effect may occur in the runoff direction from Zhuozishan Mountain to the south, resulting in the increase of some ions concentration.

2. Cation Exchange

Cation exchange is also a vital process affecting groundwater chemical feature (Zhang et al. 2020; Shi et al. 2017). Typically, two chloro-alkaline indices (CAI-1 and CAI-2) proposed by Schoeller (1965) were important way to research the occurrence of cation exchange, where the units of all ions are meq/L. When CAI-1 and CAI-2 are negative, it indicates that an ion exchange effect occurs, in which Na^+ and (or) K^+ in water-bearing media are replaced by Ca^{2+} and (or) Mg^{2+} in groundwater. Whereas CAI-1 and CAI-2 are positive, the reverse cation exchange effect occurs (Qian et al. 2016). The calculation equations of CAI-1 and CAI-2 can be estimated as follows (Li et al. 2014):

$$\text{CAI} - 1 = (\text{Cl}^- - (\text{Na}^+ + \text{K}^+))/\text{Cl}^- \quad (1)$$

$$\text{CAI} - 2 = (\text{Cl}^- - (\text{Na}^+ + \text{K}^+))/(\text{HCO}_3^- + \text{SO}_4^{2-} + \text{CO}_3^{2-} + \text{NO}_3^-) \quad (2)$$

In this study, north and central parts of Zhuozishan Coalfield groundwater samples are drawn in the lower left area, whereas south part samples are mostly in the upper right area of Fig. 5a. This suggests that an ion exchange effect (Eq. (3)) occurs in the runoff from Zhuozishan Mountain to Gongdeer Mountain and then to south-north directional and the reverse cation exchange (Eq. (4)) occurs the runoff from

Zhuozishan Mountain to the south. In other words, groundwater cation exchanges of these two parts occur reverse.

To verify the hypothesis of cation exchange reactions, the relationship between $(\text{Na}^+ + \text{K}^+ - \text{Cl}^-)$ and $[(\text{Ca}^{2+} + \text{Mg}^{2+}) - (\text{HCO}_3^- - \text{SO}_4^{2-})]$ was examined (Fig. 5b). Na^+ and Cl^- come from halite dissolution, so $(\text{Na}^+ + \text{K}^+ - \text{Cl}^-)$ values shows the amount of Na^+ gains or lose to that caused by halite dissolution (Li et al. 2018b). And, $[(\text{Ca}^{2+} + \text{Mg}^{2+}) - (\text{HCO}_3^- - \text{SO}_4^{2-})]$ values shows the amount of Ca^{2+} and Mg^{2+} gains or lose to that caused by gypsum, calcite and dolomite dissolution (Farid et al. 2013). Figure 5a illustrates that all groundwater samples are distributed along or closed to the line with -1 slope indicating that the Ordovician limestone groundwater is all affected by cation exchange. The samples from coal mines located in the northern and central parts of Zhuozishan Coalfield are mainly distributed in I quadrant of the coordinate axis, whereas those in the south part are mainly distributed in III quadrant of the coordinate axis. Besides, the farther it is from Zhuozishan Mountain (groundwater recharging area), the farther the water sampling point is from the origin of coordinates, which indicates that the farther it is from the recharging area, the stronger the cation exchange effect.

3. Main ion sources

$\text{Na}^+ + \text{K}^+$ vs Cl^- can be used to study the sources of Na^+ and K^+ in groundwater. When the samples plots near the 1:1 equiline, it shows that Na^+ and K^+ mainly originate from halite dissolution (Li et al. 2016b). Figure 6a indicates that groundwater samples from the south and north of Zhuozishan Coalfield plot far from the 1:1 line, indicating that Na^+ and K^+ originate from halite dissolution, barely be affected by cation exchange.

Previous researches have shown that if the sulphate rocks dissolution are the main processes in a groundwater system, the $(\text{Ca}^{2+} + \text{Mg}^{2+}) / \text{SO}_4^{2-}$ ratio will be or closed to 1 (Li et al. 2018c). Figure 6b shows that most of the water samples are distributed near 2:1 above the 1:1 line, indicating that Ca^{2+} and Mg^{2+} in the Ordovician limestone groundwater scarcely comes from sulfate minerals, and they mainly comes from carbonate or silicate dissolution

Besides, if the carbonate dissolution is the main processes in a groundwater system, the $(\text{Ca}^{2+} + \text{Mg}^{2+}) / \text{HCO}_3^-$ ratio will be or closed to 1. Figure 6c indicates that only few groundwater samples plot near the 1:1 line. The south and north of the coalfield water samples plot above the 1:1 aquiline, Whereas samples from the middle part of the coalfield are mainly distributed below the 1:1 line. Combined with Fig. 4b and 4c, Ca^{2+} and Mg^{2+} are mainly from the silicate dissolution, with a minority from the other kinds of rocks dissolution.

Mechanisms of Hydrogeochemistry

According to previous sections, the Ordovician groundwater runoff in Zhuozishan Coalfield can be divided into two directions: one is from Zhuozishan Mountain to Gongdeer Mountain, and then to the north-south directional runoff; the other is from Zhuozishan Mountain to the south runoff. And according to the hydrochemical analysis above, we divided Zhuozishan Coalfield into three characteristic regions including “northern”, “central” and “southern” parts. Hydrochemical mechanism of each region is summarized, respectively as follows:

1. Northern part

In the northern part, groundwater flows westward after being recharged from Zhuozishan Mountain. It passes through the Kabuqi syncline to the eastern margin of Gongdeer Mountain, which is blocked by water-blocking fracture, and changes to north-south directional runoff in the syncline. Due to the sudden change in the direction of groundwater runoff, the retention area of groundwater is formed in the Kabuqi syncline. Besides, due to the constant recharge of groundwater, the direction of groundwater runoff in the retention area is disordered (Fig. 7). Thus, groundwater flows to the downstream area for a long time, resulting in serried and non-uniform contours of hydrochemical indices (Fig. 2), and the hydrochemical types and the hydrochemical process are more complicated in turn.

2. Central part

In the central part, groundwater comes from the runoff in the north or Zhuozishan Mountain recharge area, and then it flows downstream to the southern part of Zhuozishan Coalfield along the fault zone. Therefore, the groundwater in the central part has not been blocked by geological structure, and the runoff direction remains unchanged. Thus the contours of hydrochemical indices (Fig. 2) regularly change, and the hydrochemical type is dominated by $\text{HCO}_3\text{--Cl-Ca-Na}$ (Fig. 3). The hydrochemical process is controlled by rock weathering (Fig. 3) action and cation exchange (Fig. 6)

3. Southern part

In the southern part, groundwater comes from the runoff in the north or Zhuozishan Mountain recharge area. However, due to the gradual deepening of strata in the south and the blocking of Zhengyiguan strike-slip fault, a retention zone has been formed (Fig. 7). Then, it runoff to the southwest. Therefore, affected by the above two factors, the contours of hydrochemical indices is dense and regular (Figure 2). The hydrochemical type is always dominated by $\text{SO}_4\text{--Cl-Ca-Na}$ type (Fig. 3), with a stronger reverse cation exchange effect (Fig. 6).

Conclusion

Groundwater sample were carried out in the Zhuozishan mine, northwest China, to study characteristics of Ordovician limestone groundwater. In addition, Gibbs diagram, ion relationship were used to explore

the dominant hydrochemical processes and mechanism in depth. The conclusions can be summarized as follows.

- Hydrogeochemical characteristics of Ordovician limestone groundwater in Zhuozishan are closely related to direction of groundwater runoff. In the mass, the concentration of $\text{Na}^+ + \text{K}^+$, HCO_3^- , Cl^- , SO_4^{2-} , TDS, and pH increase from the recharge area to the discharge area, however, that of Ca^{2+} and Mg^{2+} decreases. Concentration contour of $\text{Na}^+ + \text{K}^+$, HCO_3^- , Cl^- , SO_4^{2-} , Ca^{2+} , Mg^{2+} , TDS and pH densely distribute in the northern and southern of Zhuozishan indicating that these chemical indices vary greatly.
- Hydrogeochemical type change of two runoff directions (from Zhuozishan Mountain to Gongdeer Mountain and then to the south-north directional runoff and that from Zhuozishan to the south) are also different. Hydrogeochemical types of previous runoff direction (from Zhuozishan Mountain to Gongdeer Mountain and then to the south-north directional runoff) change obviously, whereas the latter is always dominated with $\text{SO}_4\text{--Cl--Ca--Na}$. This indicates different hydrochemical effects have occurred in their runoff process. Besides, according to analyse of Gibbs diagram and Chloro-alkaline index, it can be concluded that the formation processes of groundwater in the two runoff directions were controlled by different actions. Specifically, the former is affected by rock weathering action and cation exchange, whereas the latter is controlled from rock weathering action to evaporation concentration and reverse cation exchange. When it comes to ion source, Ca^{2+} and Mg^{2+} mainly origin from the silicate dissolution, while Na^+ and K^+ are mainly derived from cation exchange and barely from the halite dissolution.
- Three hydrochemical characteristic areas have formed during to two runoff directions in the study area.
- In the northern part, due to geological structure, the groundwater forms a retention area, the runoff direction is disordered, and the hydrochemical types and processes are relatively complicated. The central part is not blocked by geological structure and the runoff direction remains unchanged, thus the hydrochemical type is relatively simple, which is affected by rock weathering action and cation exchange. In the southern part, due to strata deepening and fault blocking, leading to the hydrochemical type similar to that in the recharge area and strong reverse cation exchange.

Declarations

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Figures

Figure 1

Overview of Ordovician limestone groundwater in Zhuzishan Coalfield

Figure 2

Concentration contours of hydrochemical indicators

Figure 3

Piper trilinear diagram of Ordovician limestone water in Zhuozishan Coalfield

Figure 4

Chloro-alkaline indices of Ordovician limestone water

Figure 5

$(Ca^{2+}+Mg^{2+}-HCO_3^- -SO_4^{2-})/(Na^{++}K^{+}-Cl^{-})$ and chloro-alkaline indices of water

Figure 6

Ratio relation of main ions in Ordovician limestone water

Figure 7

Schematic diagram for the formation of Ordovician groundwater in Zhuozishan Coalfield