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Monitoring of flow field based on stable isotope geochemical characteristics in deep groundwater

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Abstract The water circulation in deep aquifers controls not only chemical composition of the groundwater, but also stable isotope composition. In order to analyze the flow field in the process of the deep groundwater circulation in different aquifers, specimens belonging to the fourth aquifer in the Quaternary (the fourth aquifer for short), the coal and sandstone cranny aquifer in the Permian, and Carboniferous (the coal catena aquifer for short), the Taiyuan group limestone aquifer in the Carboniferous (the Taiyuan limestone aquifer for short), and the limestone aquifer in the Ordovician (the Ordovician limestone aquifer for short) were gained from the top down in Renlou colliery and local Linhuan coalmine district, northern Anhui, China, in the study. δD, $\delta^{18}O$, and the content of tall dissolve solids (TDS for short) of these specimens were tested. The experimental results had revealed that the groundwater in the fourth aquifer and the Taiyuan limestone aquifer takes on ^{18}O excursion and the coal catena aquifer takes on D excursion in Linhuan coalmine district, while excursion characteristic in the Ordovician limestone aquifer is not evident in the coalmine district. By analysis, $\delta^{18}O$ and the content of TDS are in negative relationship in the groundwater of the fourth aquifer and the Taiyuan limestone aquifer in Linhuan coalmine district, yet δD and the content of TDS are in positive relationship in the coal catena aquifer. Mining greatly influences the fourth aquifer and the coal catena aquifer so the groundwater in the fourth aquifer flows from northwest and southeast to mining areas and the groundwater in the coal catena aquifer flows from around to mining areas. However, mining does not influence the Taiyuan limestone aquifer evidently so the groundwater flows from east to west still.

Keywords Hydrogen and oxygen stable isotopes · Deep groundwater · Excursion · Flow field · Aquifer

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Introduction

Stable isotopes have been widely applied in hydrological studies, for their ability to provide a sharper focus on some of the underlying processes that control chemical and physical behavior of elements and compounds in the environment.

Stable isotopes of particular interest for hydrological studies include ^{18}O and ^{2}H of water, which are incorporated within the water molecule ($H_2^{18}O$, $^{1}HD^{16}O$), and exhibit systematic spatial and temporal variations as a result of isotope fractionations that accompany water-cycle phase changes and diffusion. Isotope fractionation produces a natural labeling effect within the global water cycle that has been applied to study a wide range of hydrological processes at the local, regional, and global scales. An extensive review of the application of isotope tracers to hydrological studies was recently published (Mook 2000; Longinelli and Selmo 2003; Long and Putnam 2004; Barbieri et al 2005). Herein, we focus on reviewing applications of the stable water isotopes (^{1}H, ^{2}H, ^{16}O, ^{18}O), which are the most universal tracers in hydrological research, and among the most commonly applied in recent studies. The use of stable oxygen and hydrogen isotopes as tracers in hydrologic studies has expanded over the past five decades following the initial description of systematic variations in world precipitation (Craig 1961; Dansgaard 1964), development of theory describing isotopic fractionation during evaporation (Craig and Gordon 1965), and testing and validation under a range of field conditions (Fritz and Fontes 1980; Gat and Gonfiantini 1981; Gat 1996; Clark and Fritz 1997; Kendall and McDonnell 1998; Gibson and Prowse 2000).

Isotopic compositions are expressed conventionally as δ values, representing deviation in per mil (‰) from the isotopic composition of a specified standard, such that δD or $\delta^{18}O = 1000 \times \left[\left(R_{\text{sample}}/R_{\text{standard}}\right) - 1\right]$, where R refers to the $^{2}H/^{1}H$ or $^{18}O/^{16}O$ ratios in both sample and standard. The most widely used standard in hydrological applications is the Vienna standard mean ocean water (V-SMOW), which approximates the bulk isotopic composition of the present-day global ocean reservoir, and hence has δD and $\delta^{18}O$ values both defined to be exactly 0‰. This is a logical datum for hydroclimate studies, since evaporation from the oceans is the fundamental source of global atmospheric moisture, which provides the precipitation input for continental water cycling, and the isotopic composition of the oceans is more-or-less invariant on human time scales. Use of the δ scale referenced to V-SMOW also implies that most precipitation and continental waters will have negative values, indicating a lower heavy isotope content compared with the world oceans.

Hydrochemical methods are commonly applied to study groundwater flow (Freeze and Cherry 1979; Lee and Krothe 2001; Raffaele et al. 2008). Chemical processes may provide evidence for groundwater flow. When stable isotopes and hydrochemical data are examined for both surface water and groundwater systems, important information about processes related to water cycling and rock–water interaction that may affect water quality are readily discernable (Marfia et al. 2004). Stable isotopes of hydrogen and oxygen are particularly useful as tracers because the isotopic composition of hydrogen and oxygen in groundwater does not change as a result of rock–water interactions at low temperatures (Sidle 1998).

In recent years, people think much more of hydrogen and oxygen stable isotopes and characteristics of hydrochemistry in surface water or shallow underground water and obtain a lot of research achievements (Leybourne et al. 2006; Song et al. 2002; Garcia et al. 1997; Li et al. 2006; Gui et al. 2005; Duan et al. 1994). Very little attention was concentrated on the study in deep groundwater, especially in deep groundwater in multi-aquifer system. In fact, it is very difficult to get the groundwater specimens in various deep aquifers together from 300 to 1,000 m below surface. However, it is an advantage of the mining area that a lot of deep groundwater specimens in various aquifers can be obtained. So it is possible to carry out an intensive research into flow field in deep groundwater by analyzing the composition of hydrogen and oxygen stable isotopes and the content of tall dissolve solids (TDS for short) in deep aquifers.

Taking Renlou colliery and local Linhuan coalmine district, Huaibei City, Anhui province (including Linhuan colliery, Haizi colliery, Tongting colliery, Xutuan colliery, Wugou colliery, and so on), for instance, this article focuses on the composition of hydrogen and oxygen stable isotopes and the content of TDS, on the basis, analyzes the flow field distribution in the deep groundwater of various aquifers. From upward, Linhuan coalmine district includes the following

Fig. 1 Distribution of sampling sites. **a** Location of Linhuan coalmine district in China. **b** Distribution of sampling sites in Linhuan coalmine district. **c** Distribution of sampling sites in Renlou colliery

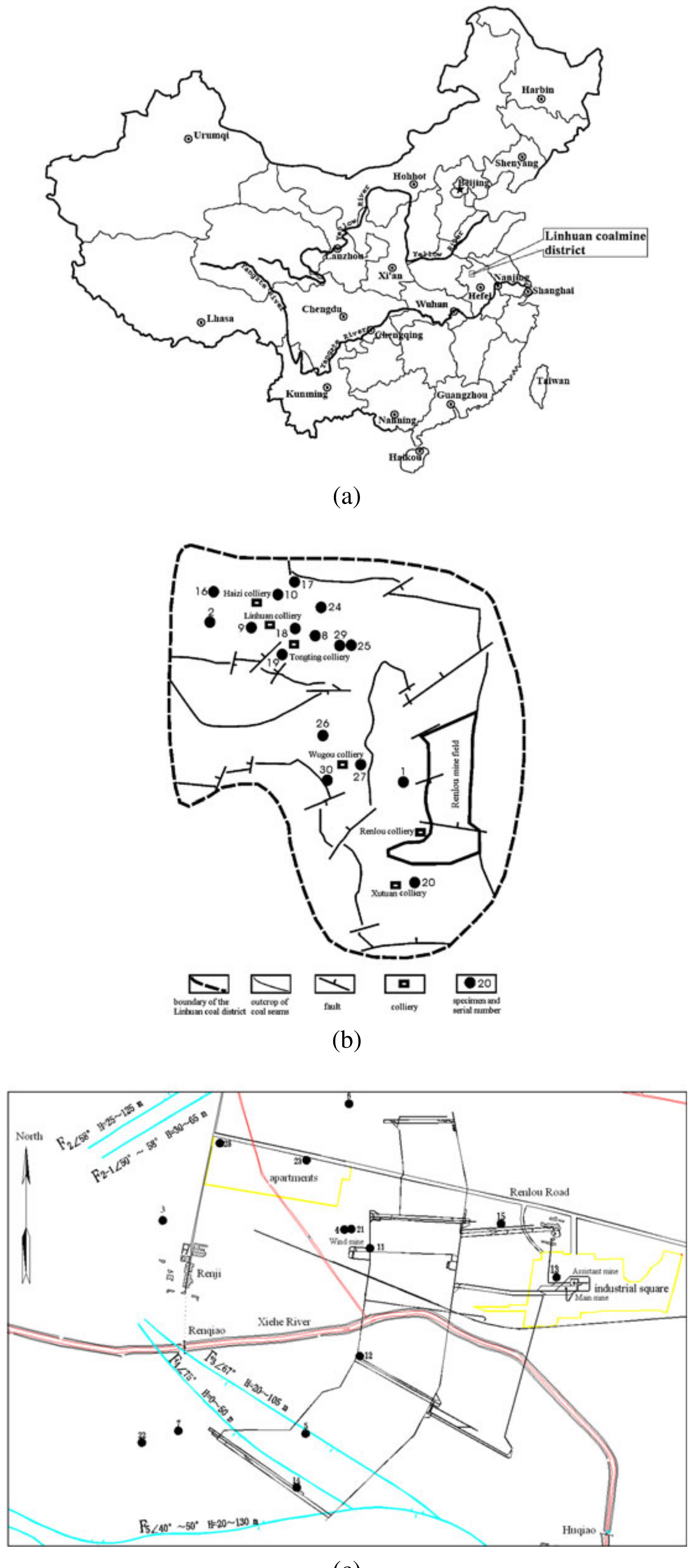

Table 1 Testing results of deep groundwater specimens of surface water

No.	Sampling location	δD (‰)	$\delta^{18}O$ (‰)	TDS (mg/L)
1	Xiehe River	−42.1	−5.8	–
2	Mining subsidence in Linhuan	−43.2	−6.1	–
3	Mining subsidence in Renlou	−46.1	−6.5	–

Table 2 Testing results of deep groundwater specimens of the fourth aquifer

No.	Sampling location	δD (‰)	$\delta^{18}O$ (‰)	TDS (mg/L)
4	Water-hole 1 in Renlou	−59.3	−7.57	780
5	Water-hole 5 in Renlou	−68.5	−8.23	1,558
6	Water-hole 5 in Renlou	−57.1	−8.62	1,588
7	Water-hole 5 in Renlou	−55.1	−7.41	789
8	Tongting	−68.8	−7.08	1,132
9	Haizi	−66.2	−8.95	1,041
10	Linhuan	−66.7	−9.48	1,423

Table 3 Testing results of deep groundwater specimens of the coal catena aquifer

No.	Sampling location	δD (‰)	$\delta^{18}O$ (‰)	TDS (mg/L)
11	Drill hole 1 in Renlou	−61.0	−8.45	2,539
12	Drill hole 2 in Renlou	−67.6	−8.48	2,806
13	−720 m main stone door in Renlou	−60.3	−8.55	2,435
14	Underground substation in Renlou	−75.7	−9.08	2,232
15	‖$7_2$22 wind lane in Renlou	−65.6	−8.83	3,079
16	Linhuan (|)	−78.7	−8.81	2,208
17	Linhuan (‖)	−83.4	−8.90	483
18	Haizi	−67.0	−9.02	1,943
19	Tongting	−67.3	−8.62	2,813
20	Xutuan	−73.0	−9.72	1,632

Table 4 Testing results of deep groundwater specimens of the Taiyuan limestone aquifer

No.	Sampling location	δD (‰)	$\delta^{18}O$ (‰)	TDS (mg/L)
21	Water-hole 8 in Renlou	−63.1	−8.30	1,759
22	Water-hole 14 in Renlou	−52.5	−6.79	435
23	Water-hole 7 in Renlou	−74.9	−8.77	1,231
24	Haizi	−59.2	−8.43	1,121
25	Tongting	−55.9	−7.54	1,326
26	Wugou	−54.4	−5.52	579
27	Wugou	−63.0	−9.23	938

Table 5 Testing results of deep groundwater specimens of the Ordovician limestone aquifer

No.	Sampling location	δD (‰)	δ^{18}O (‰)	TDS (mg/L)
28	Water-hole 6 in Renlou	−45.8	−4.51	228
29	Tongting	−63.7	−7.14	462
30	Wugou	−50.0	−6.03	337

aquifers: the fourth aquifer in the Quaternary (the fourth aquifer for short, 300 m in depth), the coal and sandstone cranny aquifer in the Permian, and Carboniferous (the coal catena aquifer for short, related with the depth of the working seam), the Taiyuan group limestone aquifer in the Carboniferous (the Taiyuan limestone aquifer for short, 600 m in depth), and the limestone aquifer in the Ordovician (the Ordovician limestone aquifer for short, 900 m in depth). They are the main water inrush aquifers because they are deep, complicated in regional structure, and hydrological condition (Gui and Chen 2007). With the decreasing of the coal resources in shallow bed, mining will shift to the deep coal seams. Therefore, it is of great importance to make a research into the flow field in the deep groundwater in related aquifers (Xu and Qian 2004). For this purpose, the studying team had got some surface water specimens and deep groundwater specimens in all Linhuan coalmine district, especially in Renlou colliery. These groundwater specimens were ranged from the fourth aquifer to the Ordovician limestone aquifer by the observation holes or drain openings of each aquifer. Through testing these specimens, the δ value of hydrogen and oxygen stable isotopes and the content of TDS were obtained. Based on these data, the composition characteristic of hydrogen and oxygen stable isotopes and the law of flow field distribution are analyzed in this article, being beneficial to draw a scientific conclusion for mining area's anti-water-disaster designing, and also to provide scientific basis for the forecast of water inrush in deep aquifers.

Sampling and testing

In this study, 30 specimens belonging to the fourth aquifer, the coal catena aquifer, the Taiyuan limestone aquifer and the Ordovician limestone aquifer were gained from the top down by surface water and the long-term observation bores and mine streaming water in Renlou colliery (see Fig. 1b and c) and local Linhuan coalmine district, northern Anhui, China (see Fig. 1a and b), including Linhuan colliery, Haizi colliery, Tongting colliery, Xutuan colliery, Wugou colliery, and so on.

Some of groundwater specimens from every sampling site were sent to the State Key Laboratory of Geological Processes and Mineral Resources, China University of Geosciences (Wuhan) to test δ^{18}O and δD. CO_2–H_2O equilibrium method was introduced to process the ^{18}O specimen, and uranium reduction process method was introduced for preparation of the D specimen. δ^{18}O and δD were analyzed by mass spectrometry. Table 1 shows the testing results. Other groundwater specimens were sent to the Groundwater Testing Center of Renlou colliery to have a instant test of the content of TDS. Tables 1, 2, 3, 4, and 5 show the testing results.

Composition of ^{18}O and D in surface water

Like local mining subsidence, water in Xiehe River, which flows through Linhuan coalmine district, is controlled by atmospheric precipitation, so is considered as surface water. It is directly or indirectly supplied by atmospheric precipitation after evaporation effect. Because the evaporation effect of precipitation is a process of high fractionation, the remained water is full of heavy hydrogen and oxygen stable isotopes. Based on the testing results, through linear regression analysis, it is easy for us to get the evaporation line of hydrogen and oxygen stable isotopes in Linhuan coalmine district, which is

$$\delta D = 5.8895 \cdot \delta^{18}O - 7.6804 \quad (1)$$

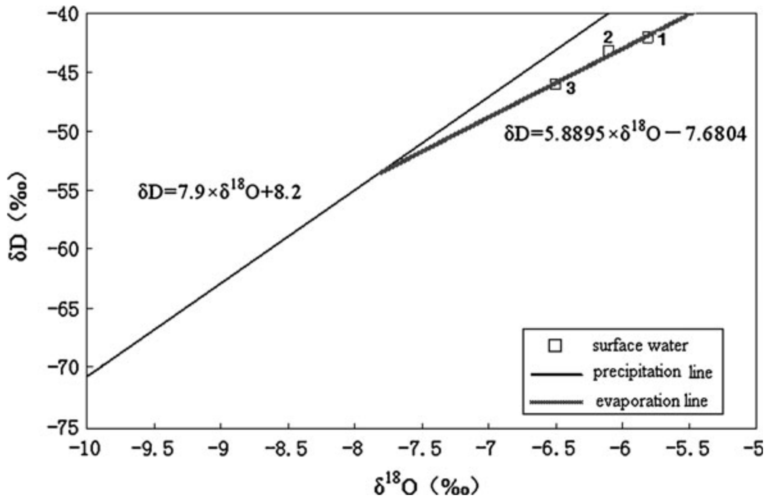

Fig. 2 Precipitation line in China and evaporation line of δD and δ^{18}O in Linhuan coalmine district

Figure 2 shows the evaporation line of δD and δ^{18}O (SMOW) in Linhuan coalmine district and the precipitation line in China (Rozanski et al. 1992).

It can be seen from Fig. 2 that the slope of the evaporation line is throughout less than that of the precipitation line in China. This suggests that evaporation effect exists through the process of atmospheric precipitation turning into surface water, conforming to evaporating feature of surface water in the temperate zone. According to isotope hydrogeology, the average composition of hydrogen and oxygen stable isotopes in local atmospheric precipitation is nearly equal to the junction value of the precipitation line and the evaporation line in Fig. 2 (which is $\delta^{18}O = -7.90‰$, $\delta D = -54‰$).

Composition of ^{18}O and D and their excursion characteristics

For the groundwater of the fourth aquifer in Renlou colliery, its δ^{18}O varies from −8.62‰ to −7.41‰, and has an average of −7.96‰, while its δD varies from −68.5‰ to −55.1‰, and has an average of −60.0‰. For in the whole Linhuan coalmine district, its δ^{18}O varies from −9.48‰ to −7.08‰, and has an average of −8.19‰, while its δD varies from −68.8‰ to −55.1‰, and has an average of −63.1‰. δ^{18}O and δD in the fourth aquifer in Renlou colliery are close to the annually average values of precipitation in Linhuan coalmine district. From the relation graph of δD and δ^{18}O, for the most groundwater specimens of the fourth aquifer in Renlou colliery (see the nos. 4, 6, and 7 in Fig. 3a), the composition of hydrogen and oxygen stable isotopes possesses an ^{18}O excursion characteristic. δD and δ^{18}O on the junction point between the excursion line and the precipitation line in China are close to local annually average value of precipitation. This indicates that there is some indirect hydrological connection between the fourth aquifer and precipitation because of mining and exploitation. Similar to Renlou colliery, the composition of ^{18}O and D in the groundwater of the fourth aquifer in Linhuan colliery, Haizi colliery, and Tongting colliery takes on ^{18}O excursion (see the nos. 8, 9, and 10 in Fig. 3a), but δD and δ^{18}O on the junction point between the excursion line and the precipitation line are less than local annually average value of precipitation. This explains that the groundwater of the fourth aquifer in these three collieries flows very slowly, and does not have a good hydrological connection with precipitation.

For the groundwater of the coal catena aquifer in Renlou colliery, its δ^{18}O varies from −9.08‰ to −8.45‰ and has an average of −8.68‰, while its δD varies from −75.7‰ to −60.3‰, and has an average of −66.04‰. For in the whole Linhuan coalmine district, its δ^{18}O varies from −9.72‰ to −8.45‰, and has an average of −8.85‰, while its δD varies from −83.4‰ to −61.0‰, and has

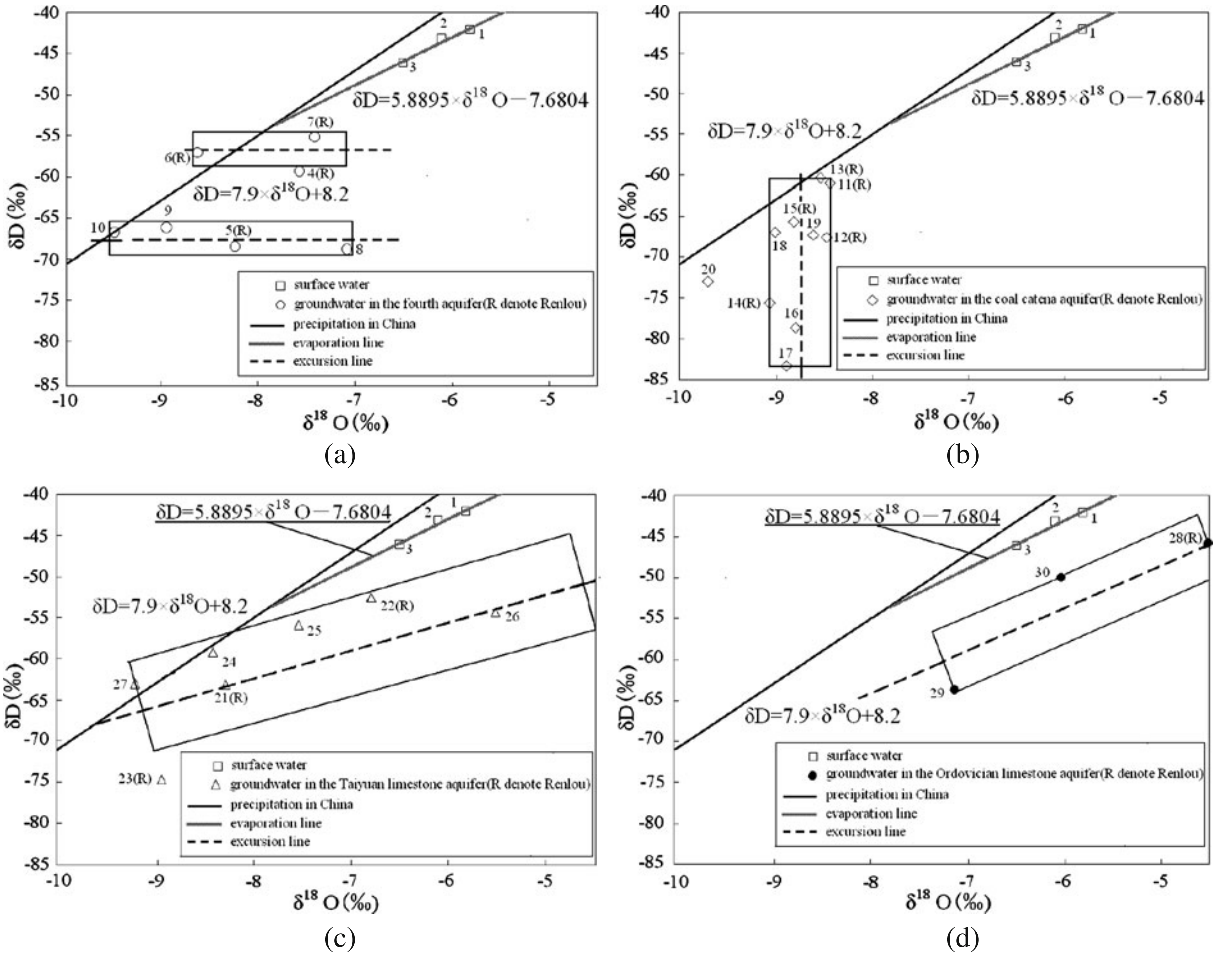

Fig. 3 Relation graph of δD and δ^{18}O in deep groundwater. **a** The fourth aquifer. **b** The coal catena aquifer. **c** The Taiyuan limestone aquifer. **d** The Ordovician limestone aquifer

an average of −69.9‰. In the whole Linhuan coalmine district, the coal catena aquifer is relatively closed and does not have good hydrological connection with precipitation, and the composition of ^{18}O and D in the aquifer takes on a D excursion (see in Fig. 3b).

For the groundwater of the Taiyuan limestone aquifer in Renlou colliery, its δ^{18}O varies from −8.77‰ to −6.79‰ and has an average of −7.95‰, while its δD varies from −74.9‰ to −52.5‰, and has an average of −65.50‰. For in the whole Linhuan coalmine district, its δ^{18}O varies from −9.23‰ to −5.52‰, and has an average of −7.80‰, while its δD varies from −74.9‰ to −52.5‰, and has an average of −60.4‰. The values of δ^{18}O and δD of most specimens in the whole Linhuan coalmine district take on great differences, and the general tendency line in the relation graph of δD and δ^{18}O is nearly parallel to the evaporation line (see in Fig. 3c), slightly suggesting ^{18}O excursion. This indicates that the groundwater flows fast in the Taiyuan limestone aquifer.

There is only one observation hole for the Ordovician limestone aquifer in Renlou colliery because the aquifer is buried deep. δ^{18}O in the groundwater of the aquifer is −7.48‰, while δD is −45.8‰. In the whole of Linhuan coalmine district, δ^{18}O in the groundwater of the aquifer varies from −7.14‰ to −4.51‰, and has an average of −5.89‰, while δD in the groundwater of the aquifer varies from −63.7‰to −50.0‰, and has an average of −53.17‰. As well as the Taiyuan limestone aquifer, $\delta^{18}O$ and δD of the most groundwater specimens of the aquifer in the whole Linhuan coalmine district take on great differences, and the general tendency line in the relation graph of δD and $\delta^{18}O$ is nearly parallel to the evaporation line (see in Fig. 3d), but it does not take on evident ^{18}O or D excursion, indicating that the groundwater of the Ordovician limestone aquifer flows faster than in the Taiyuan limestone aquifer.

Influence on composition of excursion isotopes in process of salinization in deep groundwater

During the course of its migration and saltilization, the chemical composition of groundwater is the product of geological history, and is also the results of ions' migration and transformation under the action of physical and chemical equilibrium. Common ions (such as Ca^{2+}, Mg^{2+}, K^{+}, Na^{+}, SO_4^{2-}, Cl^{-}, and HCO_3^{-}) in groundwater are playing an important part in the equilibrium. TDS is a comprehensive reflection of common ions' accumulation in groundwater, and is also an important index for groundwater's saltilization. Usually, the TDS of groundwater increases in the direction of runoff. Therefore, based on the theory of hydrogen and oxygen stable isotopes, the relationship between TDS and hydrogen or oxygen stable isotope is of great practical importance to analyze the distribution of the groundwater flow field.

The groundwater of the fourth aquifer and the Taiyuan limestone aquifer in Linhuan coalmine district takes on an ^{18}O excursion, and $\delta^{18}O$ generally has a negative relationship with TDS (see in Fig. 4). On the one hand, groundwater had dissolved oxygen-bearing minerals and aggravated the groundwater's salinization during the process of flowing (Li et al. 2006). On the other hand,

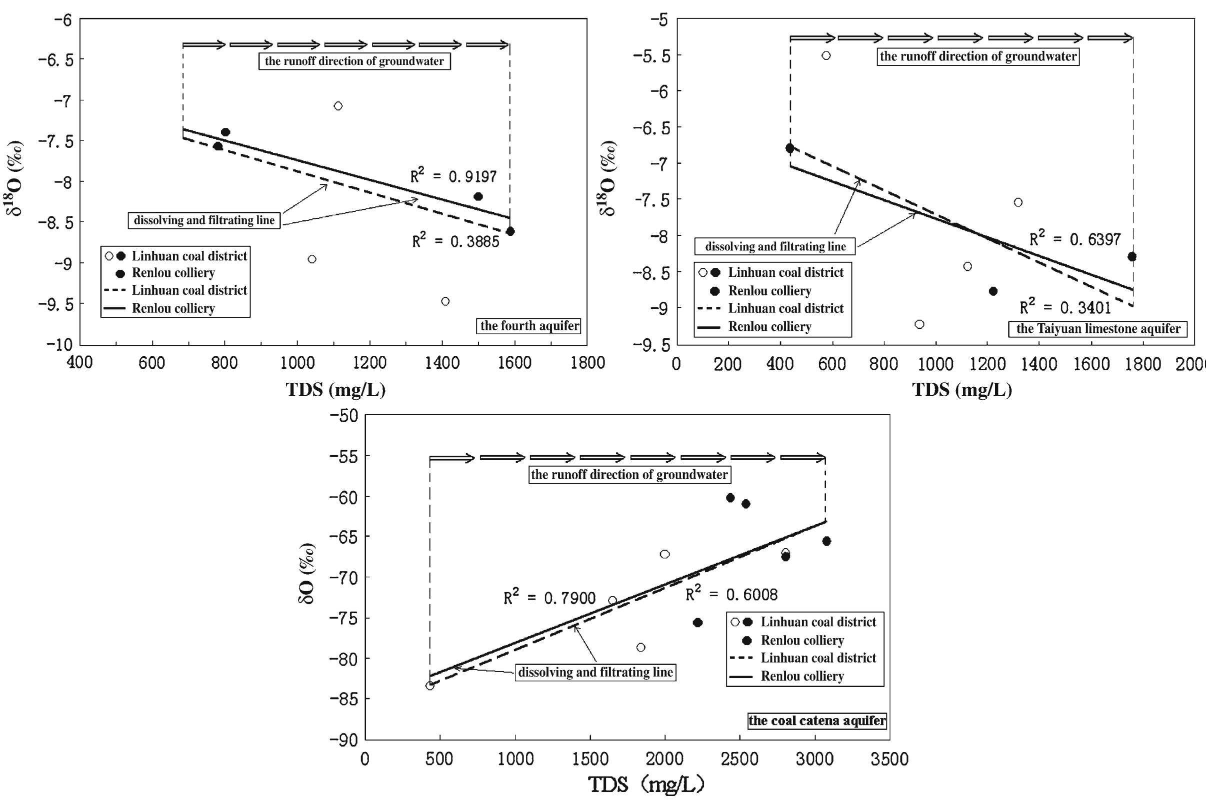

Fig. 4 Relation graph between δD or $\delta^{18}O$ and TDS in deep groundwater

with the dissolution of oxygen-bearing minerals in groundwater, the lighter oxygen ions (^{16}O) are easy to produce isotope exchange reaction with groundwater containing a lot of the heavier oxygen ions (^{18}O) after experiencing evaporation effect. So, the isotope exchange equilibrium equations move leftward (see Eqs. 2–5) and $\delta^{18}O$ decreased in accordance with the increase of TDS value, that is, $\delta^{18}O$ decreases in the runoff direction.

$$CaCO_2^{18}{}_{\text{(calcite, dolomite and limestone)}} + H_2O \leftrightarrow CaCO_3 + H_2^{18}O \quad (2)$$

$$SiO^{18}O_{\text{(quartz and calcedony)}} + H_2O \leftrightarrow SiO_2 + H_2^{18}O \quad (3)$$

$$CaAl_2Si_2O_7^{18}O_{\text{(feldspar)}} + H_2O \leftrightarrow CaAl_2Si_2O_8 + H_2^{18}O \quad (4)$$

$$14\left(Mg, Fe\right)_5 Al_2Si_3O_{10}\left(OH\right)_7\left(^{18}OH\right)_{\text{(chlorite)}} + H_2O \leftrightarrow 14\left(Mg, Fe\right)_5 Al_2Si_3O_{10}\left(OH\right)_8 + H_2^{18}O \quad (5)$$

According to Fig. 4, making a comparison on the R^2 value between the dissolving and filtrating line of the Linhuan coalmine district and Renlou colliery, we can easily conclude that intensive research in Renlou colliery shows a more evident negative relationship than a large-scale comprehensive research in Linhuan coalmine district.

Likewise, the groundwater of the coal catena aquifer in Linhuan coalmine district takes on a D excursion and δD generally demonstrates a positive relationship with TDS (see in Fig. 4). On the one hand, much hydrogen-bearing minerals (such as H_2S, CH_4, and the organic materials that is easy to be dissolved) in aquifer was dissolved into groundwater during the course of flowing and the groundwater's saltilization was aggravated (Li et al. 2006). On the other hand, with the hydrogen-bearing mineral's dissolution into the groundwater, the lighter hydrogen ions (H) are easy to be absorbed into the clay mineral of coal-bearing strata (containing a large amount of mudstone and shale except sandstone and coal), the heavier hydrogen ions (D) are easy to produce isotope exchange reaction with groundwater containing less the lighter hydrogen ions (H). So, the isotope exchange equilibrium equation moves rightward (see Eq. 6) and δD increased in accordance with the increase of TDS value, that is, δD increases in the runoff direction.

$$H_2O + D_{\text{(hydrocarbonic materials)}} \leftrightarrow HDO + H_{\text{(hydrocarbonic materials)}} \quad (6)$$

According to Fig. 4, making a comparison on the R^2 value between the dissolving and filtrating line of Linhuan coalmine district and Renlou colliery, we can easily conclude that intensive research in Renlou colliery shows a more evident positive relationship than a large-scale comprehensive research in Linhuan coalmine district.

Analysis of deep groundwater flow field in Renlou colliery

The basic cause of excursion of hydrogen and oxygen stable isotopes is the isotope exchange reaction between the groundwater and the water-bearing medium. Therefore, the excursion characteristic of hydrogen and oxygen stable isotopes in recharge area is nearly inconspicuous, but in runoff area and drainage area, the characteristic is evident because of long-time isotope exchange reaction between groundwater and water-bearing medium. With the narrowing of the research area, the excursion becomes much clear. So, it is feasible to speculate upon flow field of the main water inrush aquifers within a colliery, according to the changing characteristic of the excursion isotopes in the runoff direction and the spatial distribution of the $\delta^{18}O$ or δD value in the process of the groundwater saltilization (Gui and Chen 2004).

From the $\delta^{18}O$ isoline in the groundwater of the fourth aquifer in Renlou colliery of Fig. 5a, the gradient of $\delta^{18}O$ varies relatively greatly, and the minimum value comes into being between the no. 36 exploration line and the no. 40 exploration line in the north of the colliery and between the

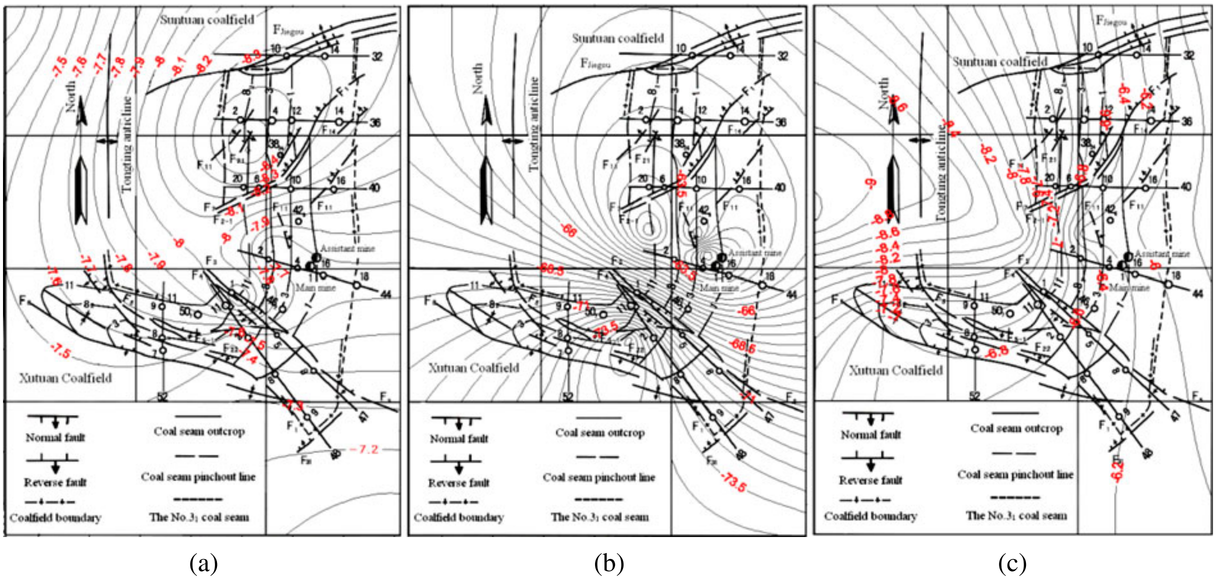

Fig. 5 Isoline of δD or δ^{18}O in deep groundwater. **a** The fourth aquifer (δ^{18}O). **b** The coal catena aquifer (δD). **c** The Taiyuan limestone aquifer (δ^{18}O)

no. 44 exploration line and the no. 48 exploration line in the south of the colliery. The minimum value of δ^{18}O is corresponding to mining area and mining has great influence on the groundwater of the fourth aquifer. It can be concluded that the groundwater of the fourth aquifer flows from northwest and southeast to mining area.

From the δD isoline in the groundwater of the coal catena aquifer in Renlou colliery of Fig. 5b, the gradient of δD varies relatively greatly, and the one maximum value of δD comes into being in three areas between the no. 40 exploration line and the no. 44 exploration line in the middle of the colliery, and the other maximum value of δD comes into being between the no. 48 exploration line and the no. 52 exploration line in the south part of the colliery. The maximum value of δD is corresponding to mining area and mining has great influence on the groundwater of the coal catena aquifer. It can be concluded that the groundwater of the coal catena aquifer flows from the around to mining area.

From the δ^{18}O isoline in the groundwater of the Taiyuan limestone aquifer in Renlou colliery of Fig. 5c, the gradient of δ^{18}O varies slightly by comparison of the fore-mentioned two, and changes monotonously, only in the middle part of the colliery, an abnormal δ^{18}O area appears. According to the spatial distribution of δ^{18}O, we can speculate that mining has only limited influence on the Taiyuan limestone aquifer, and the groundwater in the aquifer flows from east to west.

Conclusion

The following conclusion can be drawn from this study:

1. According to δD–δ^{18}O relationship, the groundwater of the fourth and Taiyuan limestone aquifer takes on ^{18}O excursion in the process of circulation. However, the groundwater of the coal catena aquifer takes on D excursion, and the groundwater of the Ordovician limestone aquifer does not have the evident excursion of hydrogen and oxygen stable isotopes.

2. For the groundwater of the fourth and the Taiyuan limestone aquifer, $\delta^{18}O$ is generally in negative relation with TDS, while for the coal catena aquifer δD is generally in positive relation with TDS.
3. According to the changing rule of hydrogen and oxygen stable isotopes in the runoff direction in the process of saltilization and the spatial distribution of the $\delta^{18}O$ or δD value, we can conclude that mining in Renlou colliery has so great influence on the fourth aquifer and the coal catena aquifer that the groundwater of the fourth aquifer flows from northwest and southwest to mining areas and the groundwater of the coal catena aquifer flows from the around to mining areas. However, mining in Renlou colliery has very little influence on the Taiyuan limestone aquifer so the groundwater of the aquifer flows from east to west still.

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