Study on hydrogeochemical connection and water quality assessment of subsidence lake and shallow groundwater in Luling coal-mining area of the Huaibei coalfield, Eastern China

Jiying Xu a, Herong Gui a,*, Yuting Xia b, Honghai Zhao c, Chen Li c, Jiayu Chen b, Chunlei Wang b and Chen Chen b

a National Engineering Research Center of Coal Mine Water Hazard Controlling (Suzhou University), Suzhou, Anhui 234000, China

b School of Earth and Environment, Anhui University of Science and Technology, Huainan, 232001, Anhui, China

c Hydrological exploration team of Anhui Coalfield Geology Bureau, Suzhou, 234000, China

*Corresponding author. E-mail: guiherong@163.com

ABSTRACT

The surface water from subsidence lake and shallow groundwater play an important role for agriculture, industry and local communities in coal mining areas. The connection between two type water and quality, however, remains unclear. In this study, 37 samples were collected from subsidence lake and shallow groundwater around Luling coal mine. The hydrogeochemistry data and formation mechanisms were analyzed to reveal the connection proof. Compared with shallow groundwater, the surface water from subsidence lake have a higher content of Na$^+$ and HCO$_3^-$ for two type water samples, Na$^+$ and Mg$^{2+}$ are two most abundant cations, followed by Ca$^{2+}$. The contents of anions followed the same order: HCO$_3^-$ > SO$_4^{2-}$ > Cl$^-$. The water samples were controlled by NaHCO$_3$ (99.3%) type. The chemical composition of two type water were similar, which are all mainly influenced by silicate minerals weathering and ion-exchange interactions, among which ion-exchange interactions was more intense than in shallow groundwater. The recharging source of two type water mainly came from atmospheric precipitation. The two water in the study area, in general, showed the risk of high salt and low-moderate alkali damage. The outcomes provide a better understanding of subsidence lake and groundwater and it will help for utilization of water resources.

Key words: connection, formation mechanism, hydrogeochemical characteristics, Luling coal mining area in Huaibei Coalfield, shallow groundwater, subsidence lake caused by mining

HIGHLIGHTS

- First analysis on the connection between subsidence lake caused by coal mining and shallow groundwater.
- Formation mechanisms of subsidence lake for Coal mining area.
- Recharging source of subsidence lake.
- Water quality for Irrigation purpose of subsidence lake.

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INTRODUCTION

The hydrogeochemical connection and water quality of groundwater and surface water is a key issue observed in hydrologic cycle and water resource management, which plays an important role in maintaining the water renewable capacity and ecosystem stability (Abbasnia et al. 2019). The coal reserves of the Huaibei Coalfield are abundant and the annual production is high. A large number of coal mining has brought a series of ecological and environmental problems (Yang et al. 2018), among which ground subsidence is one of the most serious problems. In coal mining areas, the mining of underground coal mines caused the overlying strata of coal seams to move downward and forms a subsidence basin, commonly known as subsidence area (Zhang et al. 2020). The large-scale collapse caused by mining activities had created large-scale subsidence lake. The surface subsidence lake is natural water storage area, which are the main sources for local residents, mining production, aquaculture and agricultural irrigation. Besides atmospheric precipitation and surface runoff, the main supply source of subsidence lake is shallow groundwater. During flood season, subsidence lake can recharge groundwater, while during drought and dry period groundwater can act as an important source to feed the subsidence lake. Consequently, groundwater and subsidence lake are closely linked components of the hydrologic system. Therefore, the connection between subsidence lake and groundwater and the environmental quality of both are highly concerned.

It is generally believed that hydrochemical composition records the formation process and runoff path, and environmental isotopes tracking have been utilized to assess the source and transformation during the hydrologic cycle (Najafi Saleh et al. 2020), which are also effective tracers to explain the connection between surface water and groundwater. Numerous studies have been conducted to understand this connection, including the radio of hydrochemical parameter (such as cations and anions), the environmental isotopic tracers, and exchange characteristics of water quantity and quality (Kshetrimayum 2015; Yang et al. 2016). In the northern Pakistan, Rashid et al. (2018) studied the fate, source distribution and hydrogeochemistry of the groundwater and surface water hydrogeochemical profile, which was based on chemical parameters such as pH, TDS, Na⁺ and SO₄²⁻, etc. Gu et al. (2017) analyzed the differences in hydrochemical and isotopic composition between surface water and groundwater to clarify the transformation relationship in Liu-jiang Basin, Hebei province, China. Zhao et al. (2018) evaluated the groundwater discharged and associated chemical inputs through the analysis of stable isotopes, ²²²Rn measurements, and corresponding models. In addition, water quality assessment is of great significance for environment protection and sustainable utilization of water resources. Misaghi et al. (2017) used improved water quality index to evaluate irrigation water quality of Ghezel Ozan river. However, for the subsidence lake around the coal mine, the hydrogeochemical connection between surface water local shallow and quality remain unclear, which need to be address urgently.

Hence, the objective of this paper are as follows: (1) Investigate the connection between surface water from subsidence lake and shallow groundwater based on hydrogeochemical characteristics, formation mechanism and source analysis. (2) assess the suitability for irrigation purpose. The outcomes of this research provide a better understanding of subsidence lake and groundwater and it will help for environmental protection and rational utilization of water resources.
METHODS

Study area

Luling coal mine is located at southeast of Suzhou City (Figure 1(a) and 1(b)) in the Huaibei coalfield (117°06'30"E, 33°35'59"N), with an area of 33.877 km². It was built and put into operation in 1970, with actual annual production capacity of 2.3 million tons per year. In the mining area, except for the subsidence lake formed by mining, the minefield is farmland with flat terrain, which tends to be higher in the West and lower in the East, with an elevation of +22–25 m. The climate in this area is mild, belonging to the north temperate monsoon area marine continental climate. The climate change is obvious, with four distinct seasons. The average annual rainfall is 766 mm, most of which are concentrated in July and August. The area is suitable for the comprehensive development of agriculture. The planting industry is dominated by wheat, soybeans, corn and lettuces.

The surface water and shallow groundwater systems of Luling coal mine are relatively abundant. The surface water system in the mine field is mainly subsidence lake and several artificial rest ditches. The Luling coal-mining subsidence lake is formed by coal-mining collapse, which was formed about 50 years, and its shape is irregular with area nearly 1,000 hm², the accumulation water is mainly derived from atmospheric precipitation and shallow groundwater. The average water depth is about 6 m, and the maximum water depth is 8.33 m. There is a lot of coal gangue buried along the coast (Figure 2). The shallow groundwater system is composed by thick loose layer with a thickness of about 200–300 meters. It is divided into four aquifers and three aquicludes (Gui et al. 2018). Among them, the 1st aquifer is located in the top layer, which is in direct contact with the bottom of subsidence lake. It belongs to phreatic water type, the thickness of aquifer is about 30 m, and the buried depth of water level is 3–5 m. The lithology of 1st aquifer is mainly composed of sandy soil and sandy clay with thin layer of fine silt, containing abundant calcareous nodules and humic acid.

Sampling and testing

On the basis of hydrogeological survey, in order to reveal the transformation relationship between coal mine subsidence lake and shallow groundwater, a total of 37 water samples were collected in May 2018, including 31 samples from 3 subsidence lakes and 6 samples from shallow groundwater. The location of the sampling points is shown in Figure 1(c). According to the
technical specification for Surface Water and Sewage Monitoring (HJ-T91-2002, China), the water samples 0.5 m below the surface of the subsidence lake were taken from four subsidence lakes in Luling coal mine. The groundwater is taken from the 1st aquifer with a depth of about 35 m. The samples were collected by pre-cleaned HDPE bottles. Before sampling, the samples were moistened with water samples for 3 times and were sealed with sealing film on site. The electrical conductivity (EC), pH, and total soluble solids (TDS) of all samples were measured by portable devices from OHAUS corporation (Shanghai, China). Portable instruments to test EC, pH and TDS were ST20R, ST20 and ST20T-B, respectively. The measurement accuracy of ST20R, ST20 and ST20T-B reached 1 μs/cm, 0.01 mg/L, 0.01. Then, the collection samples were immediately filtered with 0.45 μm filter paper within 24 h after collected, and the water samples were kept under low temperature conditions.

The contents of main cations (Na⁺, K⁺, Ca²⁺, Mg²⁺) and main anions (SO₄²⁻, Cl⁻) in samples were measured by Ion chromatograph (ICS-600-900, USA). The contents of CO₃²⁻ and HCO₃⁻ were determined by acid-base titration (analysis error of anion and cation should be controlled within 5%). The stable isotopes of hydrogen and oxygen (δD, δ¹⁸O) were measured by isotope analyzer (LGR-LWIA-45EP, USA) of LGR company. The measurement accuracy of δD and δ¹⁸O was 1.0 ‰ and 0.2 ‰ respectively. Before all samples were tested, the stability of the test instrument was tested with standard samples, and parallel samples were set, and the relative deviation of parallel samples was less than 5%.

Software
The Pearson’s correlation analysis was carried out to display the relation of each hydrochemical parameters in subsurface water and shallow groundwater separately. All samples were analyzed with IBM SPSS Statistics (version 26, IBM, USA) and graphs were generated by Origin 9.0 (version 9.1, Originlab, USA).

Methods and principles
The Chadha diagram can be used to determine the hydrochemical type of water and explain its source (Chadha 1999). The Gibbs diagram can effectively determine the control mechanism of chemical components (Gibbs 1970). The relationship between the ion proportion coefficient and TDS is generally used to decide the origin of different water samples, the sources
of chemical components and the formation process (Jin et al. 2018). The variation of hydrogen and oxygen isotopic composition of water during evaporation follows the principle of rayleigh fractionation, which can be used as a natural tracer to study the runoff transformation process of surface water and groundwater (Bu et al. 2018).

**Index of ion exchange reaction**

Ion exchange reaction can be studied by choro-alkaline index (Jiang et al. 2016), and calculation formula is as follows (1) and (2).

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

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

while Ca\(^{2+}\) or Mg\(^{2+}\) of aquifer reacts with Na\(^+\) or K\(^+\) in the surrounding medium, the choro-alkaline index is negative. On the contrary, if the reverse ion exchange reaction occurs, the choro-alkaline index is positive.

**Isotope exchange equilibrium reaction**

If the groundwater is in full contact with the silicate minerals from loose soil or rock fissure for a long time, the oxygen isotope exchange equilibrium reaction will occurs (Jiang et al. 2016), which was called silicate exchange and reaction equation was as follows:

\[
Si^{18}O_2 + 2H_2^{16}O \leftrightarrow Si^{16}O_2 + 2H_2^{18}O \tag{3}
\]

Due to biogenic products, coal and coal gangue contain various hydrocarbon groups and other hydrogen containing substances. While water is in full contact with coal bearing strata, hydrogen isotope exchange reaction is easily formed, which is referred to as hydrocarbon exchange reaction (Bowman et al. 2009) and equation was as follows:

\[
H_2O + D_{Coal} \leftrightarrow HDO + H_{Coal} \tag{4}
\]

**Water quality evaluation of irrigation**

Due to high salinity or alkalinity will affect the growth of crops and soil quality, it is necessary to evaluate water quality of irrigation. At present, the commonly evaluation criteria of agricultural irrigation water quality mainly include Sodium Adsorption Ratio (SAR), Sodium percentage (% Na), Residual sodium carbonate (RSC) and Permeability index (PI).

Soil adsorption ratio, abbreviated as SAR, is an important indicator to indicate the content of sodium ion in irrigation water or soil solution and to measure the degree of soil alkalization caused by irrigation water. The higher the SAR value, the stronger the ability of soil to adsorb Na\(^+\), which will lead to destruction of soil aggregate structure, poor permeability and

| Table 1 | Classification of evaluation index of irrigation water |
|---------|-------------------|-------------------|-------------------|-------------------|
| Index   | Grade             | SAR > 26          | 118 < SAR < 26    | 10 < SAR < 18     | SAR < 10          |
|         | unsuitable        | basically suitable| suitable          | suitable          | very suitable     |
| %Na     | %Na > 60\%        | 30% < %Na < 60%   | %Na < 30%         | –                 | –                 |
|         | unsuitable        | basically suitable| suitable          | –                 | –                 |
| RSC     | RSC > 2.5         | 1.25 < RSC < 2.5  | RSC < 1.25        | –                 | –                 |
|         | unsuitable        | basically suitable| suitable          | –                 | –                 |
| PI      | PI < 25%          | 25% < PI < 75%    | PI > 75%          | –                 | –                 |
|         | unsuitable        | basically suitable| suitable          | –                 | –                 |
hydraulic conductivity. The calculation formula (Yousefi et al. 2017) is as follows:

\[
SAR = \frac{Na^+}{\sqrt{(Ca^{2+} + Mg^{2+})/2}}
\]  

\(\%Na\) is used to represent the risk of alkali damage caused by irrigation water. The higher the value of \(\%Na\), the greater the risk of alkali damage. Assuming that the groundwater with high \(\%Na\) value is used for irrigation, cation exchange may occur on the surface of soil. As a result, \(Na^+\) is adsorbed by soil, and \(Ca^{2+}\) and \(Mg^{2+}\) are released, which reduces the soil permeability and leads to poor drainage in the soil. The calculation formula (Asghari et al. 2018) is as follows:

\[
\%Na = \frac{Na^+}{Ca^{2+} + Mg^{2+} + Na^+ + K^+} \times 100\%
\]  

Residual Sodium Carbonate, abbreviated as RSC, is an important indicator of alkali damage degree. If RSC is negative, it means that the content of \(CO_3^{2-}\) and \(HCO_3^-\) is smaller than that of \(Ca^{2+}\) and \(Mg^{2+}\), and there is no excess carbonate to react with \(Na^+\), so alkali damage will not be caused. On the contrary, if the RSC is positive, it is likely to cause alkali damage, and the greater the RSC value, the greater the possibility of alkali damage. The calculation formula (Soleimani et al. 2018) is as follows:

\[
RSC = (CO_3^{2-} + HCO_3^-) - (Ca^{2+} + Mg^{2+})
\]  

Permeability index, abbreviated as SI, is affected by long-term irrigation and the contents of \(Na^+, Ca^{2+}, Mg^{2+}\) and bicarbonate in soil. The calculation formula is as follows:

\[
PI = \frac{Na^+ + \sqrt{HCO_3^-}}{Ca^{2+} + Mg^{2+} + Na^+}
\]  

The unit of all parameters in formula (5)–(8) is meq/L, and the classification of evaluation index (Asghari et al. 2018) is shown in Table 1.

RESULTS AND DISCUSSION

Hydrogeochemical characteristics

Through the statistical analysis of the \(Na^+, Mg^{2+}, Ca^{2+}, K^+, Cl^-, SO_4^{2-}, HCO_3^-, CO_3^{2-}, pH, Ec, TDS\) of the samples of the subsidence lake and shallow groundwater, the relevant parameters was shown in Table 2. The ANOVA analysis was used to compare characteristics of samples, which can be seen in Figure 3.

Table 2 and Figure 3(a) show that, the range values of pH, EC, TDS of subsidence lake are 8.59–8.78, 734–784 μs/cm and 453.82–470.64 mg/L, while the range values in shallow groundwater are 7.20–8.23, 812–1,176 μs/cm and 534–682.53 mg/L, respectively. The result of pH value shows in this area was alkali, and the values of pH in subsidence lake was found slightly higher than that of shallow groundwater due to evaporation and human activities, which belong to low variance (CV < 15%). The value of TDS in subsidence lake was lower than shallow groundwater, which indicate that the water in this area was low salinity fresh water and shallow groundwater possessed fine solution filtration conditions. The value of EC from surface water to shallow groundwater showed an increasing trend, and the spatial variability was low, which can be seen that there was transformation relationship between the them, which was mainly due to the slow runoff of shallow groundwater and the dissolution of more minerals.

The range values of cations of \(Na^+, Mg^{2+}, Ca^{2+}\) in subsidence lake is 119.25–124.55 mg/L, 23.96–24.69 mg/L and 16.98–19.79 mg/L, while in shallow groundwater is 126.64–197.57 mg/L, 24.13–35.55 mg/L and 18.22–32.12 mg/L. (Table 2, Figure 3(b)). The mean concentrations and of \(Na^+, Mg^{2+}, Ca^{2+}\) in subsidence lake are all lower than them in shallow groundwater, and \(Na^+, Mg^{2+}, Ca^{2+}\) in shallow groundwater (16%, 30.69%, 18.45%) belonged to medium variation (15% < CV < 36%). The range values of anions of \(Cl^-, SO_4^{2-}\) and \(HCO_3^-\) in subsidence lake is 49.16–55.75 mg/L, 65.39–70.62 mg/L and
299.39–340.07 mg/L, while in shallow groundwater is 11.71–42.22 mg/L, 25.53–87.50 mg/L, 549.74–707.19 mg/L (Table 2, Figure 3(c)). The mean concentrations of Cl⁻/C₀ and SO₄²⁻/C₀ in subsidence lake were larger than shallow groundwater, however, the mean concentration of HCO₃⁻/C₀ in subsidence lake were lower than shallow groundwater. The CV of Cl⁻ (49.53%) and

| Table 2 | Statistical analysis of main ions of Subsidence lake and shallow groundwater |
|---------|--------------------------------------------------------------------------|
| samples of Subsidence lake | Na⁺ (mg/L) | K⁺ (mg/L) | Ca²⁺ (mg/L) | Mg²⁺ (mg/L) | Cl⁻ (mg/L) | SO₄²⁻ (mg/L) | HCO₃⁻ (mg/L) | CO₃²⁻ (mg/L) | EC (μs/cm) | pH- | TDS (mg/L) |
| Min     | 119.25 | 3.50 | 16.98 | 23.96 | 49.16 | 65.39 | 299.39 | 5.40 | 734.00 | 8.59 | 453.82 |
| Max     | 124.55 | 3.79 | 19.79 | 24.69 | 55.75 | 70.62 | 340.07 | 22.00 | 784.00 | 8.78 | 470.64 |
| Mean    | 121.13 | 3.63 | 19.15 | 24.46 | 54.13 | 67.94 | 317.44 | 13.12 | 764.74 | 8.67 | 462.28 |
| CV(%)   | 0.90 | 1.34 | 2.93 | 0.66 | 2.16 | 1.93 | 2.99 | 27.15 | 1.69 | 0.60 | 1.03 |
| samples of shallow groundwater | Na⁺ (mg/L) | K⁺ (mg/L) | Ca²⁺ (mg/L) | Mg²⁺ (mg/L) | Cl⁻ (mg/L) | SO₄²⁻ (mg/L) | HCO₃⁻ (mg/L) | CO₃²⁻ (mg/L) | EC (μs/cm) | pH- | TDS (mg/L) |
| Min     | 126.64 | – | 18.22 | 24.13 | 11.71 | 25.53 | 549.74 | – | 812 | 7.20 | 534.00 |
| Max     | 197.57 | – | 32.12 | 53.55 | 42.22 | 87.50 | 707.19 | – | 1,176 | 8.23 | 682.56 |
| Mean    | 160.89 | – | 25.47 | 37.04 | 24.68 | 46.28 | 635.49 | – | 996.83 | 7.66 | 612.67 |
| CV(%)   | 16.00 | – | 18.45 | 30.69 | 49.53 | 47.68 | 10.61 | – | 12.57 | 5.50 | 8.47 |

![Box plots for pH, Ec, TDS, Na⁺, Mg²⁺, Ca²⁺, Cl⁻, SO₄²⁻, HCO₃⁻, CO₃²⁻ in subsidence lake and shallow groundwater.](image-url)

299.39–340.07 mg/L, while in shallow groundwater is 11.71–42.22 mg/L, 25.53–87.50 mg/L, 549.74–707.19 mg/L (Table 2, Figure 3(c)). The mean concentrations of Cl⁻ and SO₄²⁻ in subsidence lake were larger than shallow groundwater, however, the mean concentration of HCO₃⁻ in subsidence lake were lower than shallow groundwater. The CV of Cl⁻ (49.53%) and
SO$_4^{2-}$ (47.68%) in shallow groundwater was attributed to high variance (CV > 36%). The result showed that both of subsidence lake and shallow groundwater were all weakly alkalinity with low salinity, Na$^+$ and Mg$^{2+}$ are two most abundant cations, followed by Ca$^{2+}$, while the concentrations of anions are all following order: HCO$_3^-$ > SO$_4^{2-}$ > Cl$^-$.  

**Hydrochemical type**

The Chadha diagram is the modified form of piper diagram. It reveals an association between cations and anions of surface water and groundwater. According to Chadha diagram, four different water type were formed, including NaHCO$_3$, CaHCO$_3$, NaCl and Ca-Mg-Cl, and the water data were plotted as the difference between (HCO$_3^-$)-(SO$_4^{2-}$ + Cl$^-$) against (Ca$^{2+}$ + Mg$^{2+}$)-(Na$^+$ + K$^+$), expressed as a percentile (Chadha 1999).

Almost all water samples ($n = 36$) located in Field-4 and revealed 99.3%, only one shallow groundwater sample in Field-1 and revealed 0.7%, there has no sample located in Field-2 and Field-3. The result showed that all subsidence lake and most shallow groundwater samples was the type of NaHCO$_3$. The formation of these water type mainly resulted from ion exchange processes. It showed a higher concentration of Na$^+$ and HCO$_3^-$ in subsidence lake and shallow groundwater. The halite, albite, carbonate and calcite minerals were the dominating minerals to form NaHCO$_3$, CaHCO$_3$ and NaCl water type. There was a large amount of coal gangue buried along the subsidence lake (Figure 2), whose main components were silicate and carbonate minerals. The water type revealed that the coal gangue around the subsidence lake was mostly dissolved within the lake and aquifer. Also, HCO$_3^-$ was derived from the dissolution of CO$_2$ in the atmosphere and the dissolution and leaching of various carbonates and weathering materials. In addition, the variation in the formation of subsidence lake was minor, which was relatively obvious in shallow groundwater. The phenomenon was mainly due to the fact that the runoff velocity of subsidence lake was faster than that of shallow groundwater, and hydrological cycle period was shorter than that of shallow groundwater. It indicated that the chemical composition of shallow groundwater was affected by human factors to a certain extent, showing obvious spatial differences.

**Formation mechanisms of chemical components**

The Gibbs diagram is widely used to distinguish the main factor controlling the formation mechanisms of chemical components. The relationships between the gibbs ratios, Na$^+$/ (Na$^+$ + Ca$^{2+}$) for cation and Cl$^-$/ (Cl$^-$ + HCO$_3^-$) for anion are usually used to analyze the dominance factors that influence water composition, including evaporation, rock and precipitation effect (Gibbs 1970). In Gibbs diagram, evaporation factor is located at the upper right corner with TDS value above

![Figure 4](image-url)  

**Figure 4** | Chadha diagram of subsidence lake and shallow groundwater in study area. Field-1 represents CaHCO$_3$ water type reveals weathering of host rock and recharge processes; Field-2 represents Ca-Mg-Cl water type reveals reverse ion exchange processes; Field-3 represents NaCl water type indicates evaporation and mixing with sea or ocean water; Field-4 represents NaHCO$_3$ water type shows base ion exchange processes.
10,000 mg/L and Cl⁻/(Cl⁻ + HCO₃⁻) value above 0.5. The precipitation controlling field, located at the lower right, has low value of TDS and Cl⁻/(Cl⁻ + HCO₃⁻) value above 0.5. The rock weathering dominance field is located in the middle of the diagram (Figure 5). The data points of both subsidence lake and shallow groundwater fell into the rock dominance field, with high values of Na⁺/(Na⁺ + Ca²⁺) more than 0.8 and low values of Cl⁻/(Cl⁻ + HCO₃⁻) less than 0.3, implying formation mechanisms of chemical components in the study is mainly influenced by rock dominance. Meanwhile, the data points fell outside the solid line, which indicated that there were other factors, such as ion-exchange interactions and human factors.

The sources of Na⁺, Ca²⁺, Mg²⁺, HCO₃⁻ can be estimated by the ratios of (Mg²⁺/Na⁺)/(Ca²⁺/Na⁺) and (HCO₃⁻/Na⁺ vs Ca²⁺/Na⁺). It is determined that high values of the ratio indicates the domain of the bicarbonate dissolution, while low values of this ratio mean the dissolution of silicates (Simsek et al. 2007). The processes controlling the two waters were discussed in detail in Figure 6. Through the use of bivariate diagrams of Figure 6, it indicated that all the subsidence lake and shallow groundwater samples were mainly affected by silicate weathering, with ratios of (Mg²⁺/Na⁺)/(Ca²⁺/Na⁺) approximately equal to 1 and (HCO₃⁻/Na⁺)/(Ca²⁺/Na⁺) slightly more than 1. The samples of subsidence lake were concentrated

**Figure 5** | Gibbs diagrams (a) TDS versus Na⁺/(Na⁺ + Ca²⁺) and (b) TDS versus Cl⁻/(Cl⁻ + HCO₃⁻).

**Figure 6** | The normalized diagram (a) Mg²⁺/Na⁺ vs Ca²⁺/Na⁺ and (b) HCO₃⁻/Na⁺ vs Ca²⁺/Na⁺.
and mainly affected by silicate weathering, there was a partial influence of evaporate dissolution of the cations. The cations of shallow groundwater were dispersed, showing the influence of both evaporate dissolution and silicate weathering. The subsidence lake is a perennial lake located in the discharge zones of the shallow groundwater flow system. The hydrochemical formation mechanism and source analysis of subsidence lake was similar to that of shallow groundwater. These similarities were clear evidence of local subsidence lake and shallow groundwater interaction, followed by subsequent evaporation and silicate weathering affecting both groundwater and subsidence lake.

The negative CAI-I and CAI-II values suggest Ca\(^{2+}\) in samples have been exchanged with Na\(^+\) in surrounding material. Besides, the larger absolute values of CAI-I and CAI-II are, the stronger the ion-exchange interaction is. In this study, the CAI-I and CAI-II of subsidence lake ranged from −1.54 to 1.22 and −0.19 to −0.16. In shallow groundwater, their CAI-I and CAI-II were range from −12.55 to −2.68 and −0.31 to −0.14 (Table 3). The subsidence lake and shallow groundwater all show negative CAI-I and CAI-II value, implying Ca\(^{2+}\) in this study have been exchanged by Na\(^+\) in surrounding rock. Moreover, shallow groundwater has higher absolute values of CAI-I and CAI-II than those of subsidence lake, suggesting ion-exchange interactions were more intense in subsidence lake.

**Source analysis of chemical components**

Studies shown that there is a significant correlation between the contents of chemical components, which can reflect that each component has the same source or geochemical process. If there is a significant and extremely significant correlation between the elements, it indicates that the elements generally have a certain homologous relationship or belong to the compound formation environment.

The correlation of hydrochemical parameters has been given in Figure 7. The correlation of cations and anions in subsidence lake and groundwater was similar, which the highly significant correlation for subsidence lake and shallow groundwater were be found as follows: Cl\(^-\) and SO\(_4^{2-}\), Cl\(^-\) and Na\(^+\), Na\(^+\) and Mg\(^{2+}\), Mg\(^{2+}\) and Ca\(^{2+}\), indicating that there was a hydraulic connection between the subsidence lake and the surrounding shallow groundwater. The Cl\(^-\) and SO\(_4^{2-}\), Cl\(^-\) and Na\(^+\) showed highly significant correlation, which indicated the weathering of calcite and dolomite minerals. The highly significant occurs between the cations of Na\(^+\), Ca\(^{2+}\), Mg\(^{2+}\), which may be attributed to cation exchange. However, in subsidence lake, a highly significant relationship existed between Cl\(^-\) and Mg\(^{2+}\), HCO\(_3\) and Ca\(^{2+}\), a slightly better relationship exists between SO\(_4^{2-}\) and Mg\(^{2+}\), SO\(_4^{2-}\) and Ca\(^{2+}\), while there is no correlation between them in shallow groundwater. There is a large area of agricultural activity around Luling coal mine area, then the subsidence lake is significant affected by human activities. It is therefore inferred that Cl\(^-\), HCO\(_3\), Mg\(^{2+}\), Ca\(^{2+}\) and SO\(_4^{2-}\) are mostly originated from the human activities and fertilizer (Leone et al. 2009).

**Analysis of recharging sources**

**Characteristics of stable isotope**

Generally, the composition of chemical composition is restricted by the source of replenishment, evaporation and mixing, which reflect different characteristics of isotopes. The hydrogen and oxygen isotopic compositions of surface water and

| ID | CAI-I | CAI-II | ID | CAI-I | CAI-II | ID | CAI-I | CAI-II | ID | CAI-I | CAI-II |
|----|-------|--------|----|-------|--------|----|-------|--------|----|-------|--------|
| S-1 | −1.22 | −0.17 | S-11 | −1.29 | −0.17 | S-21 | −1.32 | −0.18 | S-31 | −1.37 | −0.19 |
| S-2 | −1.31 | −0.18 | S-12 | −1.29 | −0.17 | S-22 | −1.28 | −0.18 | G-1  | −9.31 | −0.22 |
| S-3 | −1.30 | −0.17 | S-13 | −1.32 | −0.18 | S-23 | −1.34 | −0.18 | G-2  | −4.88 | −0.18 |
| S-4 | −1.25 | −0.16 | S-14 | −1.27 | −0.18 | S-24 | −1.54 | −0.19 | G-3  | −12.55| −0.21 |
| S-5 | −1.29 | −0.17 | S-15 | −1.30 | −0.18 | S-25 | −1.35 | −0.18 | G-4  | −2.68 | −0.14 |
| S-6 | −1.33 | −0.19 | S-16 | −1.34 | −0.18 | S-26 | −1.32 | −0.18 | G-5  | −12.84| −0.31 |
| S-7 | −1.32 | −0.17 | S-17 | −1.33 | −0.18 | S-27 | −1.22 | −0.17 | G-6  | −2.29 | −0.15 |
| S-8 | −1.30 | −0.18 | S-18 | −1.30 | −0.18 | S-28 | −1.25 | −0.17 |       |       |       |
| S-9 | −1.31 | −0.18 | S-19 | −1.36 | −0.18 | S-29 | −1.23 | −0.17 |       |       |       |
| S-10| −1.23 | −0.16 | S-20 | −1.28 | −0.18 | S-30 | −1.33 | −0.18 |       |       |       |
groundwater can indicate the difference of recharge source and water circulation process, and reflect the interaction between them partly. The $\delta D$ and $\delta^{18}O$ are used as an ideal indicator to trace water recharge and discharge within different flow system. During the water circulation process, due to the equilibrium fractionation and thermal fractionation of isotopic composition, there is a linear relationship between stable isotopes of $\delta D$ and $\delta^{18}O$ in global precipitation, which is expressed as $\delta D = 8\delta^{18}O + 10$ and defined as the Global Meteoric Water Line (GMWL) by Craig (1961). $\text{D-excess (d) = } \frac{\delta D}{\delta^{18}O}$ can reveal the degree of imbalance of evaporation and condensation process of regional atmospheric precipitation, as well as the characteristics of water environment. With strongly evaporated water, the isotope fractionation occurred, which leads to the decrease of D-excess, indicating the stronger evaporation.

The hydrogen-oxygen stable isotope $\delta$ water samples of the subsidence lake ($n = 5$) and the shallow groundwater ($n = 6$) in Luling coal mining area is chosen. The numerical results was calculated shown in Table 4. In general, the isotopic values of subsidence lake samples ranged from $-41.02\%$ to $-25.00\%$ for $\delta D$, and from $-4.69\%$ to $-2.15\%$ for $\delta^{18}O$ in subsidence lake. It ranged from $-56.01\%$ to $-47.10\%$ for $\delta D$, and from $-7.54\%$ to $-6.16\%$ of for $\delta^{18}O$ in shallow groundwater. The $\delta D$ and $\delta^{18}O$ Mean values of shallow groundwater are relatively poorer than those of subsidence lake. The results also show that the D-excess value of subsidence lake ($-2.93\%$) is smaller than that of shallow groundwater ($4.03\%$), indicating that the evaporation of subsidence lake is stronger.

Recharging sources
The values of $\delta D$ and $\delta^{18}O$ described in the $\delta D$-$\delta^{18}O$ coordinate system composed of the Global Meteoric Water line (GMWL), the Local Meteoric Water line of the China (LMWL) and the regional Local Evaporation line (LEL), as shown in Figure 8.

All samples were located below the Global Meteoric Water line (GMWL), the Local Meteoric Water line of the China (LMWL), indicating that subsidence lake and shallow groundwater were mainly from atmospheric precipitation. The values of $\delta D$ and $\delta^{18}O$ were fitted by linear regression, and the equations were as follows: $\delta D = 5.21\delta^{18}O - 15.66(n = 5, R^2 = 0.84)$, $\delta D = 5.21\delta^{18}O - 15.36(n = 6, R^2 = 0.55)$. The $d$ values of Subsidence lake and shallow groundwater was significantly smaller than that of the Global Meteoric Water Line ($10\%$), revealing that different degrees of evaporation.

### Table 4 | Content characteristic statistics of $\delta D$ and $\delta^{18}O$

| Parameters | Unit | Subsidence lake | | Shallow groundwater | |
|---|---|---|---|---|---|
| | | Min | Max | Mean | Min | Max | Mean |
| $\delta D$ | $\%$ | $-41.02$ | $-25.02$ | $-33.69$ | $-56.01$ | $-47.10$ | $-51.60$ |
| $\delta^{18}O$ | $\%$ | $-4.69$ | $-2.15$ | $-3.84$ | $-7.54$ | $-6.16$ | $-6.95$ |
| D-excess | $\%$ | $-7.99$ | $1.04$ | $-2.93$ | $0.81$ | $8.21$ | $4.03$ |
The two slope values of the fitting equation were both less than 8 (GMWL) and 7.83 (LMWL), indicating $\delta$D and $\delta^{18}$O enrichment occurred during evaporation due to dynamic fractionation. The subsidence lake samples were distributed approximately along LEL, and the content of $\delta$D and $\delta^{18}$O is higher than that of shallow groundwater, indicating that secondary evaporation of the lake water is stronger than that of the shallow groundwater, which led to the excessive fractionation of $\delta$D and $\delta^{18}$O in the lake water. In addition, a large amount of coal gangue was accumulated around the subsidence lake shore, and the subsidence lake water was in fully contact with silicate minerals of the coal gangue for a long time, resulting in oxygen isotope exchange equilibrium reaction, which was called silicate exchange and reaction equation was (3). Due to the existence of various hydrocarbon groups and other hydrogen bearing materials in coal bearing strata, hydrogen isotope exchange equilibrium reaction is produced on the basis of oxygen isotope exchange equilibrium, which is referred to as hydrocarbon exchange reaction and equation was (4). The sufficient exchange of silicate and hydrocarbon groups in the subsidence lake resulted in the enrichment of $\delta$D and $\delta^{18}$O in the lake compared with that in the shallow groundwater.

Water quality for irrigation use

There are many kinds of utilization forms of subsidence lake, such as industry and agriculture, breeding, landscape and so on. In this study area, agricultural activities are more intensive around the subsidence lake, and the shallow groundwater is also the main body of agricultural irrigation. Therefore, it is necessary to study the water quality of the shallow lake for irrigation.

Single factor evaluation of irrigation water quality

The SAR, %Na, RSC and PI were used to evaluate the single index irrigation water of the subsided pond and shallow groundwater. The statistical summary of results are shown in Table 5 calculated by formula (1), (2), (3) and (4).

**Table 5** Statistical summary of irrigation quality indexes of subsidence lake and shallow groundwater

| Index | Type   | Min   | Max   | Mean  | SD    | Unsuitable | Basically suitable | Suitable | Very suitable |
|-------|--------|-------|-------|-------|-------|-------------|-------------------|----------|---------------|
| SAR ((meq/L)$^{1/2}$)       | SL     | 4.25  | 4.52  | 4.30  | 0.05  | 0           | 0                 | 0        | 100           |
|       | SG     | 3.16  | 6.28  | 4.90  | 1.28  | 0           | 0                 | 0        | 100           |
| %Na (%) | SL     | 63    | 65    | 63.10 | 0.00  | 100         | 0                 | 0        | 0             |
|       | SG     | 48    | 71    | 61.67 | 0.09  | 50          | 50                | 0        | 0             |
| RSC (meq/L)     | SL     | 2.47  | 2.88  | 2.65  | 0.11  | 100         | 0                 | 0        | –             |
|       | SG     | 4.17  | 8.67  | 6.06  | 1.83  | 100         | 0                 | 0        | –             |
| PI (%)       | SL     | 90    | 95    | 91.35 | 0.01  | 0           | 0                 | 100      | –             |
|       | SG     | 75    | 100   | 90.33 | 0.11  | 0           | 0                 | 100      | –             |

Type: SL represents subsidence lake, SS represents Shallow groundwater.
According to the evaluation results of SAR and PI, the SAR index of all subsidence lake and shallow groundwater samples was at very suitable grade and PI was at suitable grade. However, RSC and %Na have different evaluation results as follows: the RSC index of all samples was unsuitable grade; the %Na of all subsidence lake samples were in unsuitable state, and 50% of shallow groundwater were unsuitable and 50% are basically suitable. The results of evaluation indexes are obviously different, which is mainly due to the different emphasis of evaluation. The RSC was characterized by \( \text{CO}_3^{2-}, \text{HCO}_3^- \), \( \text{Ca}^{2+} \) and \( \text{Mg}^{2+} \) to define alkali damage, and SAR, PI and %Na emphasize the characterization of \( \text{Na}^+ \) to alkali damage. Whether the water quality is suitable for irrigation, it is necessary to be further analyzed by applied comprehensive indexes.

**USSL and Wilcox diagram of irrigation water quality**

The U.S. Department of agriculture utilized the U.S. salinity laboratory’s diagram (USSL diagram) to evaluate the quality of irrigation water, which combined the effects of SAR and EC values on soil (Richards 1954). According to the degree of salinization damage, irrigation water is divided into C1 low salinization (EC < 250 μS/cm), C2 medium salinization (250–750 μS/cm), C3 high salinization (750–2,250 μS/cm), C4 high salinization (>2,250 μS/cm). Then, according to the degree of alkalinity damage, irrigation water also can be divided into four types: S1 low degree alkali damage (SAR < 10 (meq/L)^{1/2}), S2 medium degree alkali damage (10(meq/L)^{1/2}–18(meq/L)^{1/2}), S3 high degree alkali damage (18(meq/L)^{1/2}–26(meq/L)^{1/2}), S4 very high alkali damage (>26(meq/L)^{1/2}). Therefore, the irrigation water can be divided into 16 categories by USSL.

The samples collected from subsidence lake and shallow groundwater in the study area were plotted in the USSL diagram. As shown in Figure 9, the distribution of EC in subsidence lake is concentrated with an average value of 764.74 μS/cm. The EC of shallow groundwater varied significantly, ranging from 812 to 1,176 μS/cm, with an average of 996.83 μS/cm. All water samples are located in C3 area. Combined with the SAR value on the ordinate, most of the water samples are distributed in C3S1 and C3S2 areas, indicating the risk of high salt and low-moderate alkali damage. If the soil is excellent at leaching and drainage, irrigation can be carried out. Otherwise, the water should be treated and then irrigated, or plants with good salt tolerance should be selected to reduce the risk of salinity damage.

The Wilcox diagram was characterized by %Na and EC, divided into five regions as follows: Excellent to good, Go to permission, Permission to doubleful, Double to unsuitable and Unsuitable (Wilcox 1955). Assuming that the sample plotted in Excellent to good and Go to permissive region, such water for agricultural irrigation will not bring salt or alkali damage.

**Figure 9 | USSL diagram.**
While the sample plotted in Permissible to doubtful region, the water may lead to a minor risk of alkali damage, which can be prevented by adopting appropriate measures. Also, there would be risks of salt and alkali damage plotted in Doubtful to unsuitable region. The water plotted in Unsuitable region is not suitable for irrigation, which will bring serious salt and alkali damage.

The samples collected from subsidence lake and groundwater in the study area were plotted in the Wilcox diagram. As shown in Figure 10, almost all the samples were distributed in the Permissible to doubtful area, and only one sample was plotted in the Go to permission area, which indicating subsidence lake and groundwater for irrigation may lead to minor risk of alkali damage and can be prevented by adopting appropriate measures.

Strengths and limitations

In this study, the analysis on the connection between subsidence lake caused by coal mining and shallow groundwater was firstly carried out. Moreover, the formation mechanisms and source were discussed based on Gibbs, correlation and stable isotope. In addition, water quality for Irrigation purpose of two type water were assessed. The outcomes of this research provide a better understanding of subsidence lake and groundwater and it will help in securing safe irrigation water sources for local agriculture. However, the various factors such as test data, reference values of atmospheric line, variability of samples and seasonal changes caused uncertainty result, the study of subsidence lake and shallow groundwater need to further studied.

CONCLUSIONS

Based on comparative analysis on hydrogeochemical characteristic, formation mechanism of chemical composition and sources of subsidence lake and shallow groundwater were studied, and the suitability of irrigation water was evaluated. The results are as follows:

1) In both subsidence lake and shallow groundwater, Na\(^+\) and Mg\(^{2+}\) are two most abundant cations and Ca\(^{2+}\) is the second abundant cation. The contents of anions in two type water follow the same order: HCO\(_3^-\) > SO\(_4^{2-}\) > Cl\(^-\). The hydrochemical facies types of two type water samples are controlled by NaHCO\(_3\) (99.3%). compared with the shallow groundwater, the surface water from subsidence lake have a higher content of Na\(^+\) and HCO\(_3^-\), due to the dissolution of silicate minerals from coal gangue buried along the subsidence lake.
(2) The chemical composition of surface water in subsidence lake and shallow groundwater are similar, which are all mainly influenced by silicate minerals weathering and ion-exchange interactions. In addition, ion-exchange in subsidence lake is more intense than in shallow groundwater.

(3) The values of δD and δ18O of samples follow the line: δD = 5.21δ18O – 13.66 and δD = 5.21δ18O – 15.56, implying two kinds of water were mainly from atmospheric precipitation. Besides, The two slope values of the fitting equation were both less than 8 (GMWL) and 7.83 (LMWL), indicating the enrichment of δD and δ18O is related to dynamic fractionation. These similarities were clear evidence of connection between local subsidence lake and shallow groundwater. The evaluation for irrigation purpose suggest that there are high salt and low-moderate alkali damage in the study area, which can be prevented by adopting appropriate measures.

(4) In this research, the accuracy of test data, reference values of atmospheric line, variability of samples and seasonal changes lead to different degrees of uncertainty to results, which need to be further studied in the future.

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CONFLICTS OF INTEREST
The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT
All relevant data are included in the paper or its Supplementary Information.

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