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Research papers

Insights into hydrological and hydrochemical processes in response to water replenishment for lakes in arid regions

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ABSTRACT

Lakes in arid regions are ecologically valuable yet highly fragile due to intense evaporation. To provide an extra water supply for maintaining water balance in lakes, the Ecological Replenishment Water Program (ERWP) in northwest China has significantly changed the hydrological and hydrochemical conditions for these lakes. Descriptive statistics and water and mass balances, together with hydrogeochemical modeling were used in this study to gain an understanding of the impacts of water replenishment (irrigation and drainage water) on evolution for Shahu Lake. A virtual sample was introduced in NETPATH hydrogeochemical modeling to compute the net chemical reactions in the lake water. Variations in TDS indicated that the lake evolved to be saline during 2004–2012 (stage I) and then tended to be fresh during 2013–2014 (stage II). Results highlighted that groundwater outflow and chemical reactions were the overriding factors controlling chemical evolution in the lake system, which greatly depend on the replenishment activities. The salinity reduction from the virtual samples to the final samples were attributed to the precipitation of calcite and dolomite, dissolution of gypsum, Na-K and Na-Ca exchange, and the CO_2 degassing in the lake system at an annual scale of 0.11 g/L in stage I and 0.15 g/L in stage II. The quality of replenishment water was as important as quantity for rehabilitating lakes, as it significantly determines the occurrence of chemical reactions in lake water. Findings from this paper can provide insight into the evolution of arid lakes in response to replenishment activities and can help contribute to better management of a valuable and fragile resource.

1. Introduction

Climate change and human activities pose great threats to global lake systems in which people rely on the natural environment for their livelihoods (Ballatore and Muhandiki, 2002). Taking the fragile environment into consideration, the situation is particularly acute in arid regions (Chen et al., 2019; Chen et al., 2018; Huo and Li, 2013; Micklin, 2007; Su et al., 2016; Williams, 2000). Many lakes of great ecological, historical, and cultural values in arid northwest China are under threat of vanishing, where surface inflow to the lakes has decreased significantly (Fang et al., 2018; Qian et al., 2013). Desiccation of Lop Nor Lake is one of the most conspicuous examples (Guo et al., 2015; Ma et al., 2010). In this regard, the authorities intervened to prevent the situation from worsening by altering the natural hydrology of lakes. The Ecological Replenishment Water Program (ERWP) has been created to increase the flow of water into lakes for rehabilitation (Chen and Qian, 2017; Jiang et al., 2018; Pang et al., 2017; Qiao et al., 2007; Tang, 2009; Wang et al., 2017; Zhang et al., 2015; Zhu et al., 2014) (Table 1). However, there are no generally accepted management practices for the replenishment of lakes in different hydrogeological settings. Changes in lake hydrology and water chemistry in response to water replenishment (both quantity and quality) have not been well understood.

Shahu Lake, the largest lake in Ningxia, northwest China, holds the highest level of tourism attraction (5A class) owing to its unique desert oasis scenery (Li et al., 2017; Wu et al., 2017). Nearly 21 million visitors traveled to the lake in 2016 (Ningxia Statistical Bureau, 2017). The lake provides economic, tangible and intangible benefits to the quality of life for three million people who live in the surrounding towns and cities. As a terminal lake in an arid environment, it is typically sensitive to the local evaporation effects and replenishment activities. Since 2000, the addition of water diverted from drainage ditches (before 2013) and the

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Table 1
Lakes subjected to water replenishment in Northwest China.

Province	Lakes	Watershed Area (km^2)	Causes of ecological deterioration	Replenishment water sources	References
Inner Mongolia	Wuliangsuhai Lake, Juyanhai Lake	~350	irrigation for agricultural, runoff decline, excess land reclamation	390 million m^3/year from the Yellow River, Heihe River, Baiyang River	Zhu et al. (2014); Qiao et al. (2007);
Xinjiang	Bositeng Lake, Ailike Lake, Ebinur Lake	~2000	continuous expansion of cultivated land and oasis, population growth and hydraulic engineering constructions	160 million m^3/year from the Yellow River, Baiyang River, reservoirs	Tang (2009); Zhang et al. (2015); Wang et al. (2017)
Ningxia	Shahu Lake, Yuehai Lake, Mingcuihu Lake	~70	intense evaporation, sewage effluent, water quality deterioration	180 million m^3/year from the Yellow River	Chen and Qian (2017)
Shaanxi	Hongjiannao Lake	~50	construction of reservoirs, groundwater level decline	1 million m^3/year from reservoirs	Jiang et al. (2018)
Gansu	Halanuoer Lake, Yueyaquan Lake,	~27	construction of reservoirs, irrigation for agricultural, groundwater level decline	70 million m^3/year from reservoirs	Pang et al. (2017)

Yellow River (till now) have helped to maintain a relatively constant lake water level. However, salinity (shown here as total dissolved solids, TDS) in the lake water unexpectedly increased at an alarming rate (0.23 g/L per year) which varied from 2.1 g/L in 2004 to 4.4 g/L in 2013. The precise cause of these changes is still unknown. Shahu Lake evolved into a saline lake (> 3g/L) by Hammer (1986). Unfortunately, a high rate of evaporation in these arid regions causes significant salt accumulation in lakes. The growing level of salinity is compounded by mass loading from dissolved salts in the additional inflow, and may have detrimental effects on the survival of a large range of aquatic organisms and plants in and around the lakes (Gutierrez et al., 2018; Wurtsbaugh et al., 2017).

For terminal lakes with no surface outlet, like Shahu Lake, a combination of processes involving evaporation, groundwater-lake exchange, various types of chemical reactions, and even water replenishment, are inextricably intertwined in the systems (Gill et al., 2013; Rodríguez-Rodríguez et al., 2006; Zhang et al., 2015; Zlotnik et al., 2009). These processes result in nonlinear dynamic behaviors of the chemical components in the lake water. Scientific knowledge concerning the variability of groundwater exchange fluxes to and from lakes and the associated effects of mass transfer has grown rapidly. This has been studied using stable isotopes and/or conservative chemical tracers such as chloride (Montalván et al., 2017; Négrel et al., 2003; Sacks et al., 2014; Villegas et al., 2018). Unfortunately, effective management policies have lagged. In most cases, lake-groundwater interactions, the importance of mineral dissolution/weathering, redox reactions and cation exchange in lakes have not been fully considered by lake management policy makers (Ali and Khairy, 2016; Ballatore and Muhandiki, 2002; Brkić et al., 2016; Burns et al., 2001; Huo et al., 2016; Rosenberry et al., 2015). Although simulations can be helpful to quantify intertwined processes, a lack of observed data in most regions can greatly hinder model development. As a result, it remains uncertain how hydrological and hydrochemical processes should be quantified during the replenishment periods even though the replenishment water on lake evolution has been widely reported. Faced with impending severe water crises, there is an urgent need to guide lake managers, as they attempt to achieve sustainable development, by providing them with a deep understanding of the causes and effects of replenishing activities on the hydrochemical behaviors of lake water.

In this study, we carried out hydrochemical analyses of water samples from the Shahu Lake region. We examined the groundwater and replenishment water by using correlation analysis, calculation of water and mass balances, and hydrogeochemical modeling. This study aimed to determine water chemistry changes in response to different replenishing activities, and ascertain the contributions of lake-groundwater interactions and chemical reactions on lake chemical evolution. This is the first systematic study to evaluate the effects of water replenishment on lake rehabilitation in an arid region, and the results can be valuable in guiding future lake sustainable management.

2. Study area

Quaternary deposits have been extensively studied on the Yinchuan Plain of Ningxia (Chen et al., 2018; Qian et al., 2012; Qian et al., 2013). The multi-layer aquifer system is composed of, in order of depth, a phreatic aquifer, an upper confined aquifer and a lower confined aquifer. These are primarily composed of fine sand, silty-fine sand and clayed sand. Quartz and plagioclase are the major minerals in the quaternary strata. In the middle reaches of the Yellow River, the plain was provided with rich water resources fed by the river for agriculture. The irrigation systems date back more than 2000 years and are still benefiting people in the plain. It is praised as "South China Beyond the Great Wall" as hundreds of lakes scattered across the plain (Chen et al., 2016; Qian et al., 2013). Shahu Lake (38°39′29.97″N, 104°04′58.66″E) is the largest lake in Ningxia with a water surface area of 13.96 km^2, and is located in the low-lying northern part of the plain at an elevation

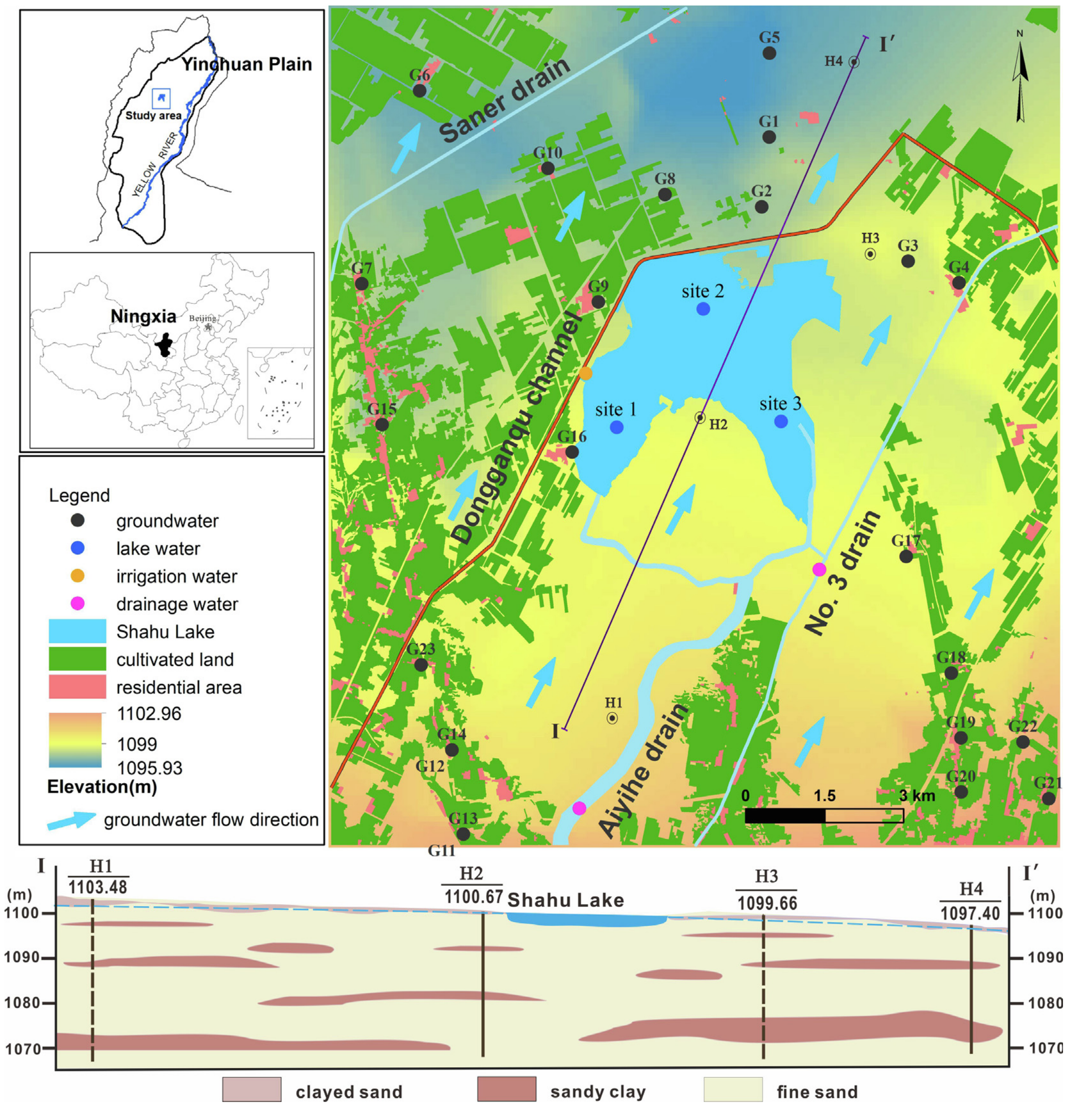

Fig. 1. Location map of Shahu Lake and related water sampling sites (I-I' hydrogeological cross section).

of 1096–1100 m above the sea level (Fig. 1). It is shallow, with a mean water depth of about 2.2 m. The lake has no natural surface runoff and outflow. In this area, groundwater in the phreatic aquifer (15–25 m thickness) generally flows from southwest to northeast (Fig. 1). Highly permeable sediments and shallow water depths, usually less than 3 m, promote a direct relationship between the lake and the groundwater system (Chen and Qian, 2017). This is the main cause for the existence of the lake in an arid region for several hundred years.

Meteorological data (2001–2016) from the Huinong meteorological Station near Shahu Lake showed that the annual average temperature was 9.6 °C, with a maximum temperature of 30.8 °C recorded in July. The annual average precipitation in the study area was 175 mm (Fig. 2a), with 75% of the rainfall falling from June to August. Evaporation observed during the same time period has an average of 1446 mm that is eight times greater than precipitation (Fig. 2b), with the maximum rates observed during the summer months (May to September). The nature of the arid environment combined with Lake Shahu's status as a terminal lake makes the added replenishment water and evaporation from lake surface dominate the gains and losses in the water budgets (Chen and Qian, 2017).

The issue of how limited water is allocated among users is rising on policy agendas, especially in arid environments (Bui et al., 2019; Gejl et al., 2019; Nethononda et al., 2019; Reynolds et al., 2007; Shaad and Burlando, 2018), and Shahu Lake is no exception. The water replenishment history of Shahu Lake can be divided into two stages due to the different water sources. The lake has received irrigation water from the Yellow River through the Dongganqu channel since 2000 to promote sustainability (Fig. 1). Drainage water, a compound mixture of irrigation return flow, groundwater, and domestic and industrial sewage (Qian et al., 2013), was also discharged into the lake via the No.3 drain and Aiyihe in the absence of sufficient supply from irrigation water (Fig. 1). Both irrigation water and drainage water were the main water sources in the first stage which ended in early 2013. After 2013, the lake was recharged solely by irrigation water, as lake managers concentrated on the ecological benefits of water transfer.

3. Material and methods

3.1. Sample collection

In this study, a total of 57 lake water samples were collected from three different sampling sites during 2012–2014 (Fig. 1). In order to describe the water-quality conditions, 23 groundwater samples and three surface water samples (from the channel and drains) were collected. The groundwater samples were taken from domestic-use wells ranging in depth from 5 to 20 m. Before sampling, the wells were

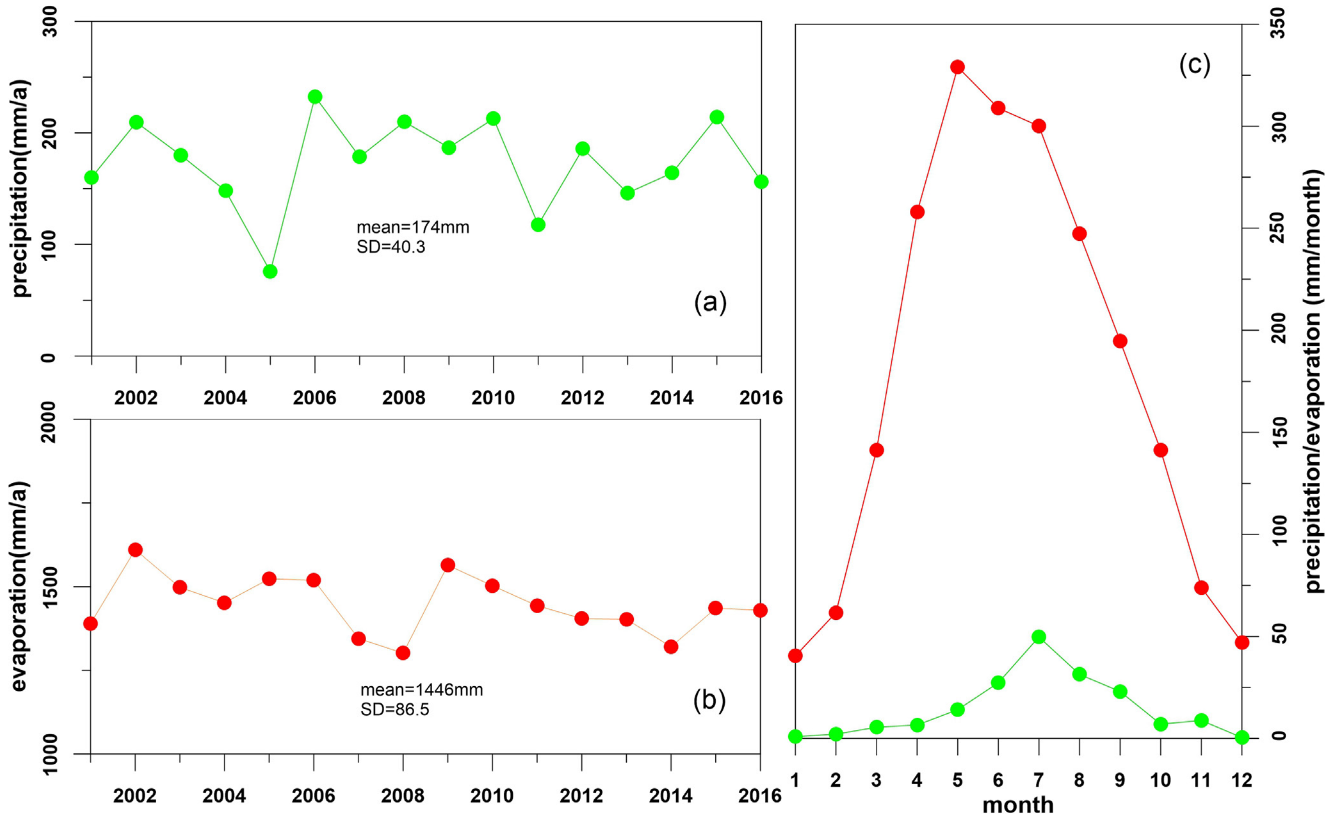

Fig. 2. Annual and monthly mean precipitation and evaporation in Shahu Lake.

pumped for 3–5 min to remove any groundwater stored in the well itself. In order to ensure that all samples represent groundwater and not water from the well bore, water temperature, EC, and pH were continuously monitored during the purging using a portable water testing kit (DDB-350, Leici company of China), and sample collection did not start until these parameters were stable. The 1000 mL polyethylene collection bottles were thoroughly rinsed 2–3 times with water to be sampled, and samples were separated into two aliquots. One aliquot was taken for cation analysis and was acidified so that the pH < 2 using 6 M ultrapure HNO_3 due to the problems of absorption or precipitation. The other aliquot was stored as collected in the polyethylene bottles for anion determination. All the samples were sealed and stored in the laboratory at 4 °C prior to analysis.

Water samples were analyzed for K^+, Na^+, Ca^{2+}, Mg^{2+}, Cl^-, SO_4^{2-}, HCO_3^-, and CO_3^{2-} concentrations at the Environmental Monitoring Station Lab in Shizuishan city, Ningxia, China. Na^+ and K^+ were analyzed using flame atomic absorption spectrophotometry. Ca^{2+} and Mg^{2+} were measured using EDTA titration; Cl^- and SO_4^{2-} were measured by ion chromatography; and HCO_3^- and CO_3^{2-} were analyzed using the potentiometric titrimetric method. TDS values of water samples were obtained based on the analysis data in the lab. Proper quality control procedures were followed to ascertain the quality of the results. The calculated ionic error balance did not exceed ± 5%.

In previous research conducted by Qian et al. (2013), the isotopic and chemical characteristics of 17 lakes in Yinchuan Plain were determined. From the previous work, the data from 2004 relevant to Shahu Lake were referenced for the present study. Additional hydrochemical data collected from the irrigation water (12 samples collected monthly during 2013) and drainage water (9 samples collected in 2004 and 2010) by the Ningxia environmental protection agency were also used for better understanding water-quality variation in the lake system. Furthermore, two soil samples were collected near the lake. Remove the roots of plants with a trowel, and take the samples from 20 to 25 cm below the surface ground. The samples were thoroughly mixed and preserved by freeze-drying for further X-ray diffraction (XRD) analysis. The XRD working condition included tube voltage of 40 KV, tube current of 20 mA, and scans a range of 2°–70° with a rate of 10°/min, in accordance with National Bureau of Standards (NBS) of the United States. The obtained curves were retrieved in the Joint Committee on Powder Diffraction Standards (JCPDs) of X-ray diffraction (XRD) to get the corresponding standard atlas (Jin et al., 2019).

3.2. Water budget calculations

The water balance equation for Shahu Lake was represented as:

$$\Delta V = RI + RD + P + GWI - E - GWO \quad (1)$$

where ΔV represents the change in water storage for the lake; *RI* represents the replenishment inflow from irrigation water (Yellow River water); *RD* represents the replenishment inflow from drainage water; *P* is the precipitation; *E* is the evaporation from the lake surface; *GWI* represents the groundwater inflow into the lake; and *GWO* represents the groundwater outflow from the lake into the aquifer.

Human intervention and natural factors maintained lake levels with nominal change over time. Because of this, the model can be simplified to a steady-state situation. In this study, terms such as precipitation, evaporation, and replenishment water recharge are measurable quantities, while groundwater seepage from and into the lake are unknown values. The direct precipitation input to the lake (*P*) and evaporation from the lake surface (*E*) were obtained for each water year using data from the Huinong meteorological Station located 5 km from the lake. Data detailing the volume of replenishment water was collected from the local Water Conservancy Bureau (Ningxia Water Conservancy Bureau, 2010-2017). Villegas et al. (2018) noted that chemical balances are helpful to determine unknown components when the concentration of a chemical of interest is not the same for inputs and outputs. The general equation for the chemical budget of Shahu Lake can be represented mathematically (Donovan, 1994):

$$\Delta(VC_L) = RI(C_{RI}) + RD(C_{RD}) + P(C_P) + GWI(C_{GWI}) - GWO(C_{GWO}) + R \quad (2)$$

where C represents a concentration of a chemical in the water. The subscript for each concentration term refers to the water balance components as defined above, while R represents chemical reactions within the lake. The chemical composition of the water discharging from Shahu Lake to groundwater was assumed to be the same as that of the lake water ($C_{GWO} = C_L$). Conservative chemicals, which are defined as nonreactive during the water cycle, are set as $R = 0$. Using this assumption, the unknown groundwater-lake exchange quality and quantity fluxes can be solved.

3.3. Hydrogeochemical modeling

Hydrogeochemical modeling is a useful tool for clarifying evolutionary process in groundwater system. We performed inverse modeling between two samples (the initial and final solution) along the flow paths that revealed the occurrence of physiochemical processes (Ahmed and Clark, 2016; Morán Ramírez and Ramos Leal, 2014; Rademacher et al., 2001). Unlike previous studies, extensive evaporation can result in salt accumulation in lakes. To compute the net chemical reactions in the lake water, a "virtual sample" was introduced in this study (Fig. 3). Given the calculated *GWI* and *GWO* from Eq. (2), the volume of water from each endmember (*RI*, *RD*, *P*, *E*, *GWI*, *GWO*, and initial water) is multiplied by each chemical parameter of that endmember, with their sum representing the virtual chemical composition of the m^{th} parameter. This demonstrates that the values of conservative parameter between virtual and real samples are the same. The ionic delta for nonconservative compositions depends mainly on the hydrochemical reactions that occurred in the lake system. The impacts of reactions in lake can be quantified using NETPATH code (El-Kadi et al., 2011), which analyzed differences between the virtual and real samples (final sample) at the same time.

4. Results

4.1. Chemical composition

Basic statistics of the physicochemical data of water samples for the lake, wells, river and discharge drains sampled within the study area are summarized in Table 2. A Kruskal-Wallis nonparametric test showed that there was no statistically significant difference in the variances of the measured parameters for the three monitoring sites in the lake ($p = 0.024$). This is consistent with the generally well-mixed water in shallow lakes (Cui et al., 2016). As shown in Table 3, the mineral composition of the soil samples was dominated mainly by quartz (40–70%), plagioclase (15–25%), Na-feldspars (5%), illite (2.5–6%), chlorite (3–5.5%), calcite and dolomite (2.2–15.5%). Small levels of 0.2% gypsum and 0.3% amphibole were also reported.

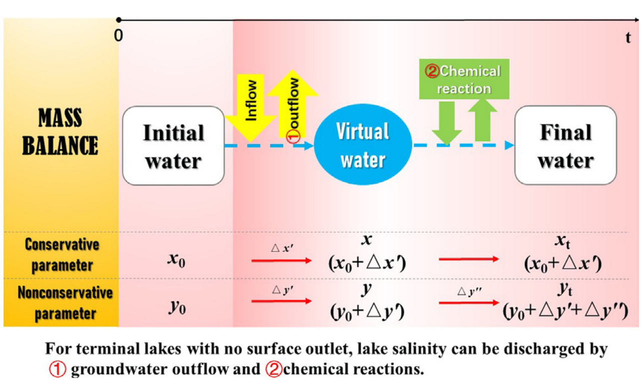

Fig. 3. Sketched map of the virtual sample in the lake system. As evaporation increased water salinity, lake salinity can be discharged by groundwater outflow and chemical reactions for terminal lakes with no surface outlet. x_0 and y_0 denote the original masses of conservative and nonconservative parameters in the initial water, respectively. The virtual water sample is only dependent on mass loading from the inflow and mass transfer with the outflow ($\Delta x'$), which can be used to quantify $\Delta y'$ and the chemical reactions ($\Delta y''$) for nonconservative parameters based on the hydrogeochemical modeling.

4.1.1. Lake water

Shahu Lake is a typical of shallow lake, as it does not have thermal stratification. Water quality data indicated that the water was mildly alkaline with pH values ranging from 8.3 to 9.2. The relative abundance of major dissolved ionic species was ($Na^+ + K^+$) > Mg^{2+} > Ca^{2+} for cations and Cl^- > SO_4^{2-} > HCO_3^- > CO_3^{2-} for anions. To reveal similarities and differences among water samples, a Piper trilinear diagram was created (Piper, 1944). As shown in Fig. 4, all the lake water fell into Cl·SO_4-Na·Mg facies even though there were significant changes in water salinity among them. The lake experienced a rapid increase in salinity with a mean annual ratio of 0.23 g/L in 2004–2013 and then tended to decline in 2013–2014 (Table 2). Minimum and maximum TDS values of the lake water were 2.1 g/L in Sep 2004 and 4.4 g/L in Dec 2013.

4.1.2. Replenishment water

The replenishment water diverted from the Yellow River (irrigation water) was fresh (HCO_3-Ca·Na and HCO_3-Na·Ca types) with a mean TDS value of 0.39 g/L. It showed little temporal variation in the chemistry (Fig. 4). The poor-quality drainage water that recharged the lake fell into Cl·SO_4·HCO_3-Na·Mg facies. The mean TDS values detected from the drainage ditches was 1.66 g/L. The mean values of the dominant ions Na^+ (0.46 g/L), Cl^- (0.48 g/L), and SO_4^{2-} (0.35 g/L) in mean drainage water were higher than those in the irrigation water.

4.1.3. Groundwater

Groundwater in the region was also characterized by weakly alkaline conditions with a mean pH of 7.6. Molar ion concentrations in the groundwater decreased from ($Na^+ + K^+$) > Ca^{2+} > Mg^{2+} for cations and HCO_3^- > Cl^- > SO_4^{2-} for anions. Beneath agricultural lands, the general zonation of groundwater type was indicative of both natural factors and human activities (Qian et al., 2013). The mean groundwater sample in Table 2 describes the origin of the chemical compositions for recharging the lake. The TDS level of mean groundwater was 0.65 g/L. As natural discharge from groundwater forms lakes in the Yinchuan Plain, one can speculate that Shahu Lake contained fresh water during the early stages of its formation.

4.2. Processes regulating lake water chemistry

4.2.1. Evaporation, precipitation and dissolution

A Gibbs diagram (Gibbs, 1970) was created to reveal the mechanism controlling the dissolved salts composition in the water. The ratios of $Na^+/(Na^+ + Ca^{2+})$ and $Cl^-/(Cl^- + HCO_3^-)$ relative to TDS commonly illustrate three-natural mechanisms in water: atmospheric precipitation, rock weathering and evaporation process (Cui et al., 2016; Elumalai et al., 2019; Ntanganedzeni et al., 2018; Prasanna et al., 2011; Xiao et al., 2015). As is evident from Fig. 5, the majority of the groundwater samples coincided with the rock dominated endmembers, but lake water samples were mainly driven by evaporation. As water evaporates, Ca^{2+}, Mg^{2+}, and HCO_3^- that were removed through carbonate precipitation is believed to be the principal reaction responsible for the lake's chemical change (Guo et al., 2015), resulting in the increased ratios of Na/(Na + Ca) and ($Cl^- + HCO_3^-$). Compared with the major inflow sources (mean groundwater and irrigation water), lower concentrations of Ca^{2+} in the lake water revealed the dominant deposition of calcium in the lake system. According to the calculation of the saturation index (SI), the values in all the lake water samples for

Table 2
Statistics overview of the hydrochemical parameters of the water samples in the Shahu Lake region. L1-L20 were lake water samples.

Water type	Unit	lake water											
		L1[a] 9/2004	L2 8/2012	L3 4/2013	L4 5/2013	L5 6/2013	L6 7/2013	L7 8/2013	L8 9/2013	L9 10/2013	L10 11/2013	L11 12/2013	L12 3/2014
Temperature	°C	23.1	25.1	7.8	11.6	18.7	27.2	26	22.3	19.6	10.0	8.0	6.2
pH	–	8.5	8.5	8.6	8.6	8.9	8.7	8.8	8.7	9.2	8.3	8.5	8.7
K^+	mmol/L	0.42	0.45	0.57	0.53	0.53	0.53	0.53	0.56	0.54	0.57	0.54	0.48
	mg/L	16.5	17.7	22.4	20.5	20.6	20.8	20.8	21.7	21.1	22.1	21.1	18.9
Na^+	mmol/L	23.8	42.3	49.0	45.1	46.3	45.8	43.6	44.2	49.0	46.1	55.7	46.4
	mg/L	548	972	1126	1037	1066	1054	1002	1016	1128	1060	1281	1068
Ca^{2+}	mmol/L	0.78	1.33	1.21	1.30	1.40	1.36	1.18	0.99	0.90	0.95	1.00	0.90
	mg/L	31.23	53.02	48.57	51.93	56.03	54.30	47.23	39.73	36.17	38.07	39.97	36.13
Mg^{2+}	mmol/L	5.00	8.67	10.1	12.7	10.5	11.1	11.5	9.13	8.17	9.88	9.33	7.46
	mg/L	120	208	242	304	252	266	276	219	196	237	224	179
Cl^-	mmol/L	13.2	26.6	28.3	31.6	29.5	29.7	28.5	28.5	28	28.9	38.5	28.3
	mg/L	470	945	1006	1122	1047	1055	1011	1013	995	1027	1366	1006
SO_4^{2-}	mmol/L	7.88	11.8	13.3	13.2	11.6	12.3	12.6	13.2	12.7	13.2	12.7	13.1
	mg/L	756	1130	1275	1263	1117	1183	1209	1267	1221	1266	1221	1256
HCO_3^-	mmol/L	6.18	10.2	9.84	8.8	9.74	8.92	8.75	8.82	8.05	8.93	8.62	9.34
	mg/L	377	619	600	537	594	544	534	538	491	545	526	570
CO_3^{2-}	mmol/L	0.09	–	0.42	0.33	0.48	0.64	0.66	0.72	0.93	0.76	0.83	0.45
	mg/L	5.5	–	25.2	19.6	28.6	38.2	39.4	43.2	55.8	45.8	49.6	27.2
TDS	mg/L	2136	3635	4045	4087	3884	3943	3872	3889	3899	4242	4466	3876
CB[b] (%)		1	2	5	4	5	5	4	−0.2	3.3	1.9	2	−1
SI (Calcite)	–	0.81	0.93	1.21	1.29	1.66	1.47	1.46	1.29	1.53	0.79	0.95	1.10
SI (Dolomite)	–	2.49	2.81	3.36	3.64	4.37	4.03	4.06	3.66	4.11	2.52	2.75	2.97
SI (Gypsum)	–	−1.51	−1.26	−1.28	−1.35	−1.35	−1.32	−1.39	−1.39	−1.47	−1.34	−1.34	−1.34

Water type	Unit	lake water								lake water source[c]		
		L13 4/2014	L14 5/2014	L15 6/2014	L16 7/2014	L17 8/2014	L18 9/2014	L19 10/2014	L20 11/2014	mean irrigation water	mean groundwater	mean drainage water
Temperature	°C	15.0	19.1	25.0	25.0	23.2	23.9	20.6	10.8	12.1	21.0	22.3
pH	–	8.5	8.7	8.5	8.8	8.8	8.7	8.7	8.6	8.1	7.6	8.1
K^+	mmol/L	0.54	0.54	0.47	0.53	0.55	0.54	0.54	0.5	0.05	0.06	0.3
	mg/L	21	21	18.3	20.6	21.3	21.1	21.2	19.6	2.0	2.3	11.7
Na^+	mmol/L	45	45	39	42.3	43.7	38.6	43.9	40.2	2.6	4.6	20.0
	mg/L	1035	1035	897	973	1005	888	1010	925	60	106	460
Ca^{2+}	mmol/L	1.01	1.01	1.22	1.14	0.98	0.93	0.72	0.78	1.52	2.10	1.21
	mg/L	40.40	40.40	48.80	45.63	39.33	37.30	28.70	31.33	60.90	83.80	48.40
Mg^{2+}	mmol/L	9.38	9.38	8.38	9.17	9.00	9.33	9.29	8.50	0.86	1.71	3.38
	mg/L	225	225	201	220	216	224	223	204	20.6	41.1	81.1
Cl^-	mmol/L	29.1	29.1	25.9	26.8	27.3	23.9	24.9	25.8	1.83	2.6	13.6
	mg/L	1032	1032	918	953	969	847	884	915	65	92	483
SO_4^{2-}	mmol/L	13.5	13.5	12.0	12.8	13.0	10.8	12.3	10.2	0.92	1.55	3.65
	mg/L	1296	1296	1150	1224	1247	1040	1182	979	88	149	350.4
HCO_3^-	mmol/L	9.34	9.34	8.18	7.66	7.61	7.52	7.61	7.62	3.18	5.82	7.38
	mg/L	570	570	499	467	464	459	464	465	194	355	450
CO_3^{2-}	mmol/L	0.54	0.54	0.47	0.77	0.74	0.72	0.74	0.75	–	–	–
	mg/L	32.40	32.40	28.20	46.00	44.20	43.40	44.60	45.20	–	–	–
TDS	mg/L	3967	3967	3512	3716	3775	3329	3626	3352	393	651	1659
CB[b] (%)		−0.1	−0.1	−0.2	2	1	5	5	3	4	3	2
SI (Calcite)	–	1.08	1.29	1.24	1.41	1.32	1.25	1.10	0.96	(0.14,1.14)	(0.08,0.86)	(-0.51,0.49)
SI (Dolomite)	–	3.13	3.61	3.45	3.86	3.73	3.62	3.40	2.90	(0.02,2.21)	(0.01,1.8)	(-0.38,1.39)
SI (Gypsum)	–	−1.33	−1.37	−1.31	−1.34	−1.39	−1.47	−1.56	−1.48	(-1.88,-1.64)	(-2.76,-0.33)	(-1.61,-1.24)

[a]Denotes the data obtained from the previous study of Qian et al. (2013), [b]denotes the charge balance error, and [c]represents that the mean groundwater, mean irrigation water and drainage water that used in the calculation and modeling.

Table 3
Analysis results of the sediments in the study area (%).

No.	quartz	plagioclase	Na-feldspar	calcite	ankerite	amphibole	gypsum	illite	chlorite
TY1	43.9	24.6	4.3	15.0	0.5		0.2	6.0	5.5
TY2	73.1	14.4	4.5	1.9	0.3	0.3		2.5	3.0

calcite and dolomite were greater than zero, indicating that they remained over-saturated. The positive correlation between HCO_3^- and Ca^{2+} (r_s = 0.56, $p < 0.05$), and HCO_3^- and Mg^{2+} (r_s = 0.47, $p < 0.05$, Fig. 6) further highlighted the precipitation of the Ca-carbonate and Mg-carbonate in the lake. Similar results were presented by Wahed et al. (2014) for Lake Qarun in Egypt. However, the molar ratios of ($Ca^{2+}+Mg^{2+}$) vs. HCO_3^- in most of lake water were above the 1:2 lines (Fig. 7a). This provides strong evidence for the additional contribution of Ca^{2+} and/or Mg^{2+} from other sources and/or the deposition of carbon.

$$CO_3^{2-} + Ca^{2+} = CaCO_3^- \downarrow \text{(calcite)} \quad \text{(Reaction 1)}$$

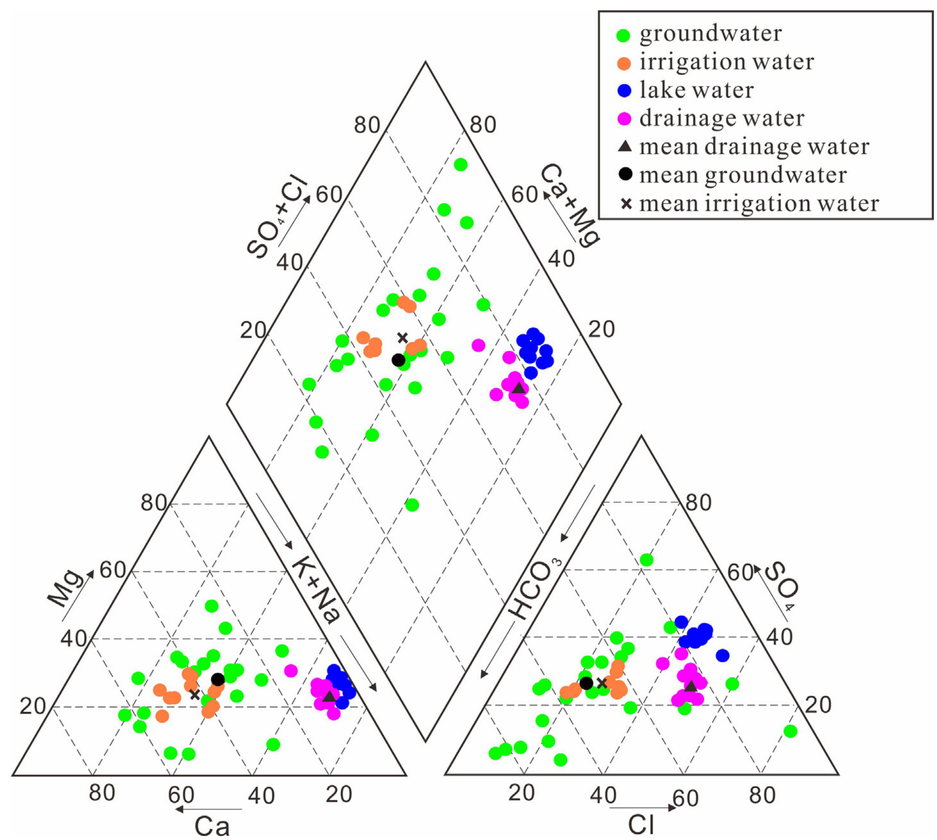

Fig. 4. Piper plots of the chemical composition of surface and groundwater samples.

$2CO_3^{2-} + Ca^{2+} + Mg^{2+} = CaMg(CO_3)_2 \downarrow$ (dolomite) (Reaction 2)

4.2.2. Cation exchange

The ionic ratios in water can provide useful information that help identify water with lithogenic origins and from anthropogenic effects (Chen et al., 2017). Lake water displayed an excess of Na^+ over Cl^- (Fig. 7b), implicating the contribution of silicate weathering and cation exchange (McNeil et al., 2005). As reported by Stallard and Edmond (1983), feldspars are more susceptible to dissolution than quartz. Despite the detected silicate mineral in the study area which implies possibility of Na-feldspar weathering (Table 3), it is unlikely sodium ions will increase under alkaline conditions. In addition, Griffioen (2001) and Thorslund et al. (2014) documented that the transport of K in water was mainly controlled by cation-exchange. These findings suggest that the additional sodium in lake water may be governed by possible Na-Ca, Na-K and Na-Mg exchanges. The exchange between Ca (and/or Mg) and other cations may occur in the lake to balance against the lost carbonate precipitation.

4.2.3. Effects on lake water salinity change

Shahu Lake receives inflow exclusively from irrigation water and

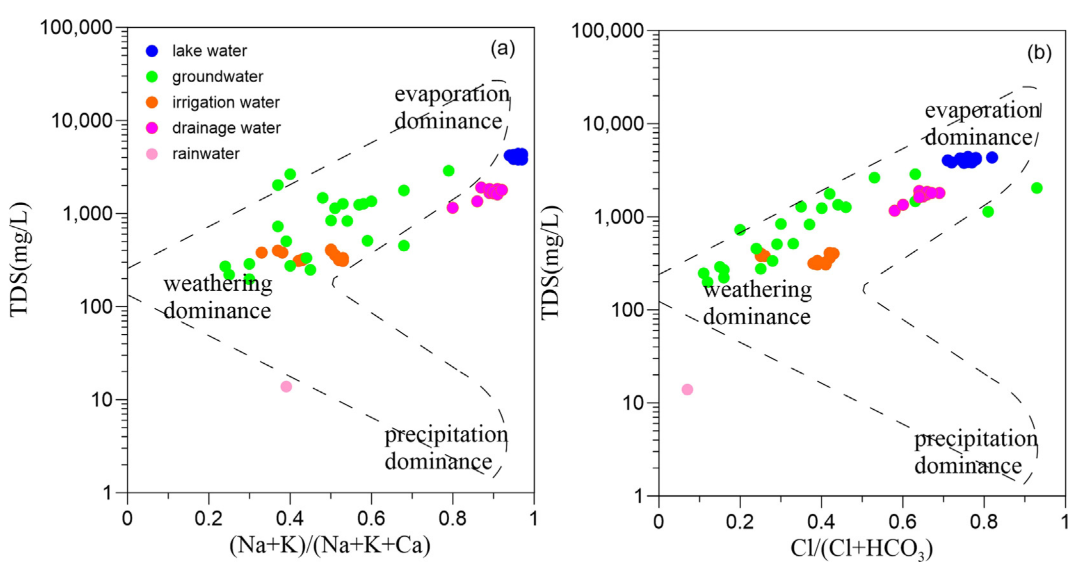

Fig. 5. Gibbs plots indicating dominant processes for formation of lake water, groundwater and other water bodies in the study area.

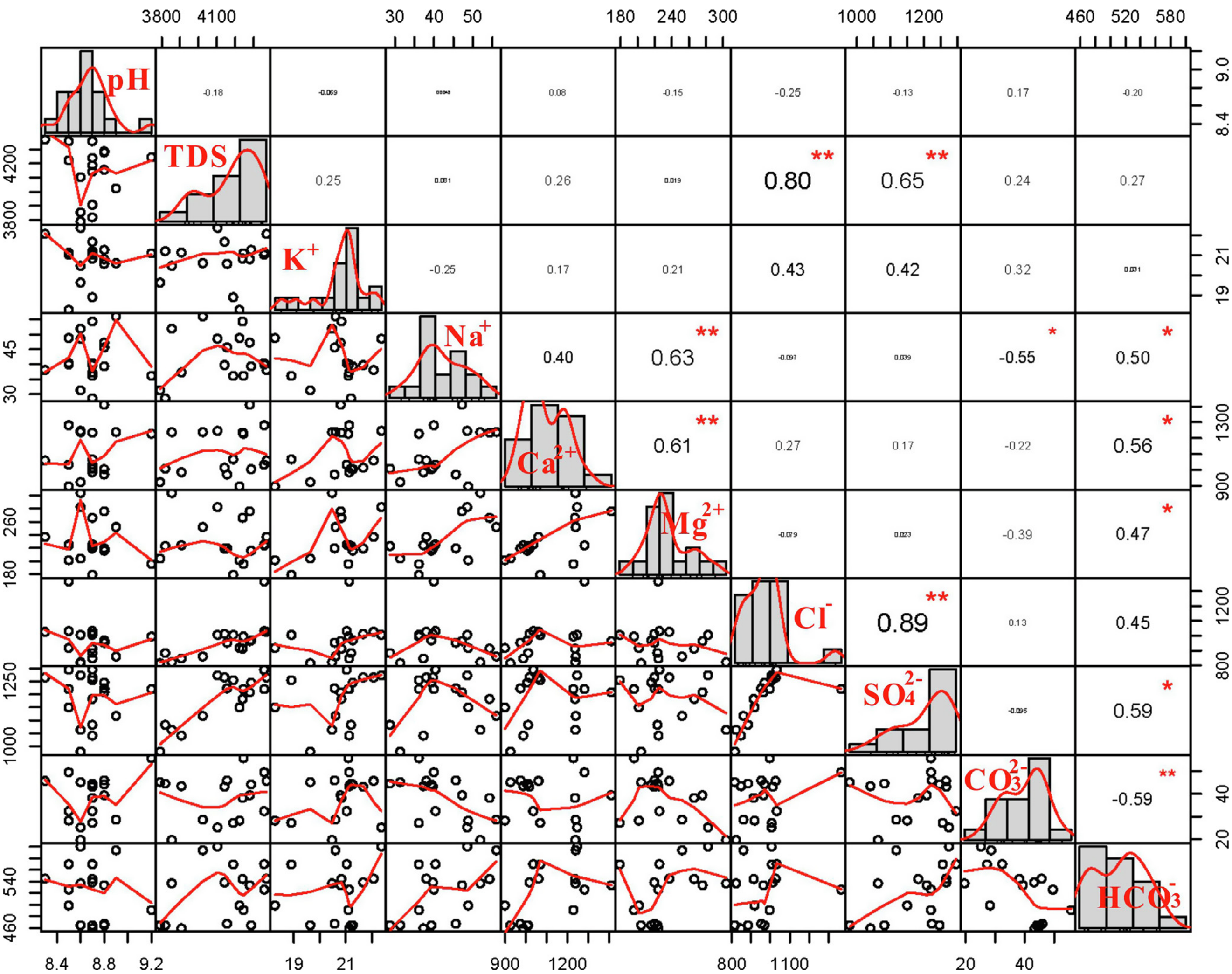

Fig. 6. Correlation relation among ions for the lake water, ** and * denote the significance level < 0.01 and < 0.05, respectively.

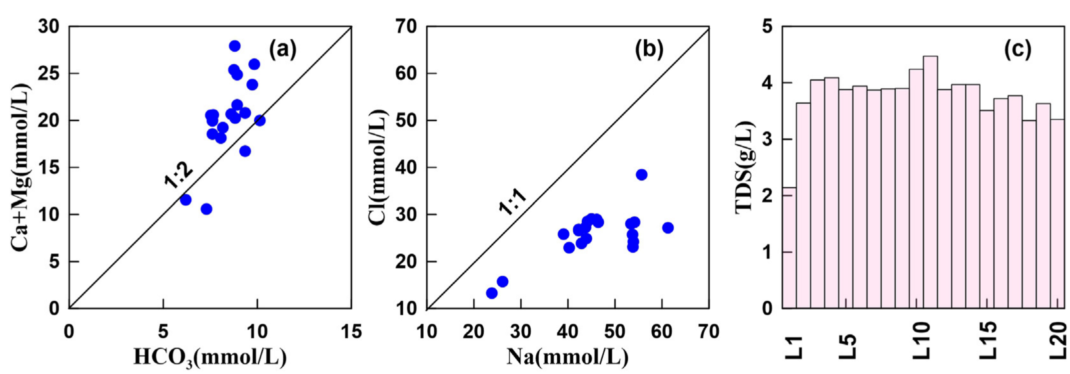

Fig. 7. Scatter diagram for the lake water samples, molar ratios of ($Ca^{2+}+Mg^{2+}$) vs. HCO_3^- (a); molar ratios of Na^+ vs. Cl (b); TDS variation (c). The detailed information about water chemistry for L1-L20 can be found in Table 2.

drainage water. Previous study has estimated the groundwater-lake water exchange in the lake system and noted that the optimal volume of replenishment water (1380×10^4 m^3/a) could sustain the lake level at a normal state (Chen and Qian, 2017). Data showed that before 2013, the annual volumes of irrigation and drainage water were in the ranges of 460–1274 $\times$ 10^4 m^3 and 0–791 $\times$ $10^4 m^3$. After 2013, it increased to 1142–2852 $\times$ 10^4 m^3 with an annual mean value of 2080 $\times$ 10^4 m^3 (Fig. 8). This shows an acceleration of the water cycle.

It is noteworthy that variations in TDS in the lake water were markedly coincident with the replenishment activities. An appreciable increase in TDS occurred, going from 2.1 g/L in 2004 to 3.6 g/L in 2012. The long-term electric conductivity (EC) trend obtained from the Ningxia Water Conservancy Bureau (2010–2017) further demonstrated the rapid water salinization during this period (Fig. 8). As the replenishment water was changed to irrigation water in 2013, lake water salinity tended to decrease. The slowed decline in TDS in 2013 indicate a time lag effect (Fig. 7c). As mentioned earlier, the drainage water inflow into the lake involved a large quantity of domestic and industrial

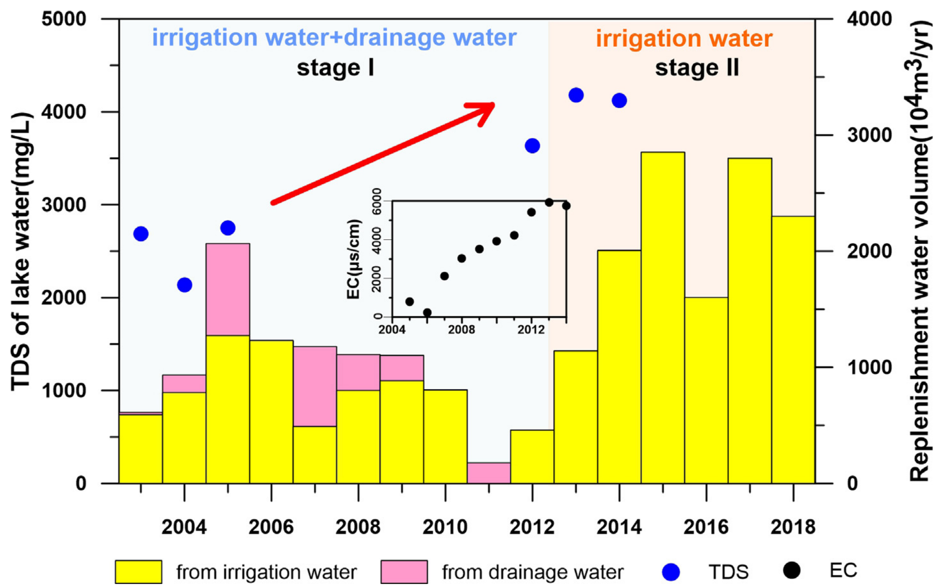

Fig. 8. The temporal variation of TDS and the volumes of replenishment water during 2003–2018 in Shahu Lake.

sewage. Given the high contents of Cl^- and SO_4^{2-} in drainage water, it profoundly affected the higher salinity in the lake before 2013. Consequently, strong positive correlations of TDS with Cl^- and SO_4^{2-} (r_s = 0.80, p < 0.01; r_s = 0.65, p < 0.01) were observed in the lake water (Fig. 6). As a nonconservative element, chemical reactions may alter the SO_4^{2-} concentrations in the lake water even though it has similar variations with TDS. Gypsum was likely to dissolve in lake water to increase SO_4^{2-} levels because of the related negative SI values (Table 2). The dissolution of gypsum may also provide more Ca^{2+} for carbonate precipitation and/or cation exchange.

4.3. Hydrological and hydrochemical processes in response to water replenishment

4.3.1. Lake-groundwater interaction

The lake has an effluent-influent behavior with the aquifer, and groundwater flows through the lake along the southwest-northeast direction (Chen and Qian, 2017). Hydrologic alterations of the lake, however, complicate groundwater movement. The fluctuations in lake-groundwater exchange have further effect on the characteristics of water and dissolved ions within, and from the lake. Two stages of hydrochemical variation in combination with replenishment activities were considered: (1) water salinization during 2004–2012 (nine hydrological years, stage I), and (2) water freshening during 2013–2014 (two hydrological years, stage II). Chloride is considered a conservative component in the lake. Assuming no chemical mass was lost in the evaporation processes and the density of lake water was constant, groundwater that flows in and out of lake during each stage could be calculated based on the combination of water budget and chloride mass balance equations (Eq. (1–2)). C_L denotes the mean value of chloride in a specific period in the lake water. The hydrochemical data for different water bodies were presented in Table 2. Specifically, water sample L1 was the initial solution in stage I. The final solution in stage I and the initial solution in stage II had the same sample of L3. The mean lake water in 2013–2014 was regarded as the final solutions in stage II.

Table 4 shows the significant difference during the hydrogeochemical evolution of the lake system. Groundwater represented as a major component of recharge to balance the water budget. In stage I, the annual groundwater inflow reached 855 × 10^4 m^3 and the outflow was 82 × 10^4 m^3 in the Shahu Lake. As a result, the lake water accumulated solutes from replenishment water and groundwater recharge under the arid environment and there was a relative amount of salinity discharge from the lake to the groundwater. In stage II, sufficient replenishment water reduced groundwater recharge (345 × 10^4 m^3/a) and accelerated the groundwater discharge (235 × 10^4 m^3). Chen and Qian (2017) explained the lake-groundwater interactions in Shahu Lake using numerical modeling. When the lake water levels declined due to a shortage of replenishment water, increased recharge from groundwater occurred. On the contrary, the large volume of replenishment water accompanied by the lake water level enhanced water discharge into the aquifer system. This suggests that groundwater-lake water exchanges are highly sensitive to changes in the volume of water replenished into the lake, which is consistent with the quantification in this study.

In addition, groundwater outflow needs to be a prominent focus for understanding salinity balance in lake management. Assuming there was no flow from the lake to groundwater (*GWO* = 0 in Eq. (1–2)), the effect on lake water level variation could have been negligible due to their small percentages (3% and 8% of the total lake water volume in stage I in stage II). Using mass balances analysis, the magnitude of the relative impact on salinity decline by groundwater discharge in the lake system was quantified. At this time, the salinity of the lake water was 5.8 g/L and 4.6 g/L at the end of stage I and stage II, respectively. These are greater than the values in the virtual samples (5.0 g/L and 3.9 g/L). This indicated that the groundwater seepage helped to reduce the salinity of the lake water at a mean annual rate of 0.09 g/L during stage I and 0.35 g/L during stage II. Therefore, a holistic water replenishment scheme considering groundwater-lake interaction can be necessary for recognizing the water and mass balance in the lake.

4.3.2. Contribution of chemical reactions

To investigate the hydrochemical evolution of Shahu Lake during the two stages, two inverse modelling scenarios were performed. Considering the conservative behavior of chloride in the lake, the selected chemical constraints (chemical elements) were K, Ca, C, Mg, Na, and S in the NETPATH models. The phases included in the modeling were selected based on the analysis of the chemical trends, calculated SI values, and the mineralogy. Because this does not preclude the possibility of reactions producing or consuming CO_2 within the lake, CO_2 was included among the plausible phases. A set of mineral and gas phases (involving calcite, dolomite, gypsum, cation exchange and CO_2) were hypothesized to be the reactive phases in the system, and then the mass transfers for possible combinations of the selected phases were simulated. In each stage, positive values refer to mass entering (dissolution), while negative values represented mass releasing

Table 4
The calculation results of lake water balance in stage I and stage II.

Stage	Year	RI	RD	P		E		calculated GWI	calculated GWO
		(10^4 m^3)	(10^4 m^3)	(mm)	(10^4 m^3)	(mm)	(10^4 m^3)	(10^4 m^3 per year)	(10^4 m^3 per year)
Stage I	2004	783	150	148	207	1452	2027	855	82
	2005	1274	791	76	106	1523	2127		
	2006	1231	0	233	325	1520	2121		
	2007	490	689	178	249	1345	1878		
	2008	800	341	210	293	1302	1817		
	2009	884	219	187	261	1564	2184		
	2010	805	0.6	213	297	1503	2098		
	2011	0	178	118	164	1443	2015		
	2012	460	0	186	259	1405	1961		
Stage II	2013	1142	0	146	204	1425	1989	345	235
	2014	2006	0	164	229	1430	1996		

Table 5
Results of mass-balance modeling of chemical processes (mmol/L per year) in lake water using NETPATH. Processes refer to: *a* Na-Ca exchange, positive sign indicated the Ca^{2+} adsorption and the release of Na^+; *b* K-Na exchange, positive sign indicated the K^+ adsorption and the release of Na^+; *c* dissolution (positive sign) or precipitation (negative sign) of dolomite, *d* dissolution or precipitation of calcite, *e* CO_2 degassing (negative sign) or dissolution (positive sign) in lake water, *f* dissolution or precipitation of gypsum.

Stage	Solution	TDS (g/L)	K^+ (mmol/L)	Na^+	Ca^{2+}	Mg^{2+}	Cl^-	SO_4^{2-}	HCO_3^-	*a* (mmol/L per year)	*b*	*c*	*d*	*e*	*f*
I	initial solution-L1	2.1	0.42	23.8	0.78	5.00	13.2	7.88	6.18						
	final solution-L3	4.0	0.57	49.0	1.21	10.08	28.3	13.3	9.84	0.07	−0.02	−0.21	−0.74	−1.2	0.04
	virtual solution	5.0	0.79	47.4	10.1	12.0	28.3	12.9	31.4						
II	initial solution-L3	4.0	0.57	49.0	1.21	10.08	28.3	13.3	9.84						
	final solution-mean water in 2013–2014	3.6	0.52	43.2	0.97	8.88	26.5	12.2	8.25	−0.57	−0.02	−0.46	−1.2	−0.32	0.03
	virtual solution	3.9	0.56	45.4	3.09	9.8	26.5	12.2	13.1						

(precipitation) into the aqueous phase. The salinity changes between the virtual samples and the final samples for each stage described the corresponding contribution of chemical reactions for the water chemistry evolution. In the lake system, only one mass-balance reaction model was consistent with the observed chemical characteristics for each stage. As shown in Table 5, the salinity reduction from the virtual samples to the final samples were attributed to the precipitation of calcite and dolomite, dissolution of gypsum, Na-K and Na-Ca exchange, and the CO_2 degassing in the lake system at an annual scale of 0.11 g/L in stage I and 0.15 g/L in stage II.

The lake water samples with a deficit of Ca^{2+} as compared to SO_4^{2-} can be explained by the prevalent precipitation of calcite. Calcite precipitation was the most abundant in the system, which occurred at mean annual ratios of 0.74 mmol/L in stage I and 1.2 mmol/L in stage II. Comparatively, the mean annual ratios of dolomite precipitation were 0.21 mmol/L in stage I and 0.46 mmol/L in stage II. Carbonate mineral precipitation and CO_2 degassing were coupled in the two stages, which is in agreement of other researches (Clark and Lauriol, 1992; Cosmo et al., 2019; Papadimitriou et al., 2004). Under the conditions, 0.04 mmol/L dissolution of gypsum occurred annually in stage I and 0.03 mmol/L in stage II, to release more Ca^{2+} into the lake water to balance the deficit.

Cation exchange represents the interchange between an ion in solution and another ion in the boundary layer between the solution and a charged surface, such as clay minerals, organic matter, or amorphous minerals (Chen et al., 2019; Edmunds et al., 2003; McNeil et al., 2005; Sposito, 2000). Helfferich (1962) noted the following selectivity sequence of $Ca^{2+} > Mg^{2+} > K^+ > Na^+$ for the common cations. In this study, only Na-K and Na-Ca exchange were identified in controlling the chemical evolution of Shahu Lake. In the study area, the fine-grained sediments in the lakebed and the abundant reeds, and phytoplankton (Chen and Qian, 2017) provided favorable conditions for the occurrence of cation exchange. The K-Na exchange explained relative excess of sodium content in the lake with 0.02 mmol/L both in stage I and stage II. Interestingly, the reversibility of Na-Ca exchange result in the Na^+ concentration in the lake water was demonstrated with an increase (0.07 mmol/L) in stage I and a deficit (0.57 mmol/L) in stage II. Although Na^+ was enriched in the lake and drainage water, the Na^+ concentration in the virtual sample was smaller than that in the final sample during stage I. Except for the carbonate precipitation and CO_2 degassing, the remaining Ca^{2+} was the primary cause for the Na^+ increase in stage I. To balance the solution, one Ca^{2+} ion would replace two Na^+ ions on soil particle surfaces to supplement the depletion of Na^+ in stage I. However, the cation exchange led to the elimination of Na^+ and the release of Ca^{2+} when large quantity of irrigation water that was enriched with Ca^{2+} recharged into the lake during stage II. This was similar with the observed processes of seawater intrusion (Mountadar et al., 2018). The release of Ca^{2+} from the surface of clay minerals into the lake water at a mean annual rate of 0.57 mmol/L was dominant in stage II. Simultaneously, the carbonate precipitation increased. This highlights the importance in the quality of replenishment water on the mineral precipitation and cation exchange processes in the lake system.

$$(1/2)CaX_2 + Na^+ \leftrightarrow NaX + (1/2)Ca^{2+} \quad \text{(Reaction 3)}$$

$$NaX + K^+ \leftrightarrow KX + Na^+ \quad \text{(Reaction 4)}$$

5. Discussion

Understanding the underlying causes of water level and water salinity variation in lakes can help guide sustainable decision making. A lake system can be compared to an iceberg which has both visible and hidden parts. To explain it more clearly, a conceptual model was developed and shown in Fig. 9. Precipitation, surface runoff, replenishment water, and evaporation (which decision makers or managers can observe) are represented by the upper portion of the iceberg. The lakes in arid regions receive low and inconstant annual rainfall and are

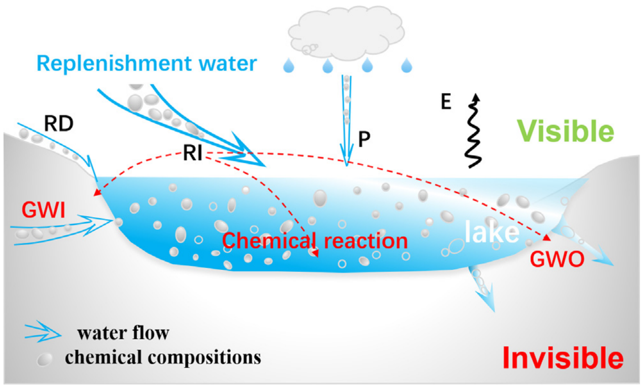

Fig. 9. Conceptual model of lake evolution. The inflow of lake includes precipitation (*P*), replenishment water from drainage water (*RD*) and irrigation water (*RI*), and groundwater inflow (*GWI*). The evaporation (*E*) and groundwater outflow (*GWO*) are the main outflow of the lake. The quantity and quality of replenishment water, precipitation and evaporation on lake water can be easily recognized. However, the mass transfer via groundwater-lake interaction and chemical reactions also plays a critical role in salinity variation even though these are generally neglected in lake management, and therefore invisible.

characterized by potential evaporation that is far greater than the precipitation. The effect of replenishment water was highlighted in lake water budgets as rainfall cannot ensure a sufficient water supply without supplementation from the other water sources. Groundwater-lake interaction and chemical reactions were represented by the much large portion of the iceberg underwater, which significantly impacts the hydrological and hydrochemical processes, respectively. Importantly, both quality and quantity of replenishment water which relied on the available water resources in the catchment, can trigger or change the water exchange between lake and groundwater as well as the corresponding chemical behaviors. The study we conducted at Shahu Lake can be considered an in-situ test for modeling the effects of water replenishment on lake rehabilitation. Understanding the invisible elements is vital for measuring effectiveness in lake management and should not be simplified despite the difficulties in their determination.

Understanding the hydrochemical behaviors of lake water was beneficial in recognizing this lake's evolution, which can be applied to other lakes, possibly bridging the knowledge gap between science and policy. For Shahu Lake, despite possible errors that might be introduced from the inter-monthly variation in water hydrochemistry, integrating the hydrochemical data with the water budget is the best available option to extract information for further management, as this study demonstrated. Results from this study show that for complex systems, lake management practices can be improved using the limited data available. Importantly, when a high rate of evaporation accumulates salt in Shahu Lake, groundwater outflow and chemical reactions contributed to 44% and 56% of salinity reduction (2.7 and 3.4 million tons each year) during the water salinization (stage I), and 70% and 30% (10.8 and 4.6 million tons each year) during the water freshening (stage II), respectively. The large quantity of saline discharge revealed possible causes for the massive ecological impacts on the lake system. The results also suggest that it is not sufficient to evaluate the effectiveness of management practices only based on changes in lake water levels. Water quantity is as important as water quality in a lake for sustaining ecological balance, even though it is indeed difficult to balance water demands among different stakeholders in arid regions. Stress on water resources due to rising freshwater supplementation and climate change have already led to water scarcity in many places (Aeschbach-Hertig and Gleeson, 2012; Mohanty, 2003). Even the Yellow River, the longest river in the north China, is facing the problems of significant reductions in streamflow and intensified competition for water resources (Miao et al., 2011; Zhang et al., 2017). This may have a significant impact on the availability of replenishment water resource for Shahu Lake and other lakes in northwest China. Therefore, to fill the gap between lake management and water resources allocation, the "invisible" elements in hydrological and hydrochemical cycles should be emphasized in further research and management, helping to prioritize lake management and remediation practices. In lakes that are experiencing water quality deterioration, additional replenishment water resources are urgently required to serve the unmet needs for maintaining the ecological balance of these lakes. Managers should understand that by adding replenishment supplies, they may induce adverse impacts on groundwater chemistry in the lake vicinity. As reported by Yechieli and Wood (2002), the increased lake inflows were the main divers of increased salinity to which the quality of the groundwater is highly attributed. To better understand the complexities and uncertainties associated with lake systems, long-term monitoring will provide valuable data which can be useful for describing, examining and documenting changes in lake regions.

6. Conclusions

This work represents the first comprehensive analysis of the hydrological and hydrochemical changes in Shahu Lake, which was impacted by replenishment water. Results showed that the lake water can be categorized as a Cl·SO_4-Na·Mg type even though the lake experienced a rapid increase and decline in salinity. The variation in TDS indicated that the lake evolved to be saline during 2004–2012 (stage I), and become increasingly fresh during 2013–2014 (stage II), which was consistent with the replenishment activities. These changes in lake water chemistry are controlled by replenishment activities, lake-groundwater interactions and chemical reactions that involved: calcite and dolomite precipitation, gypsum dissolution, ion exchange, and CO_2 degassing. As evaporation increased water salinity, groundwater outflow and chemical reactions were responsible for approximately 44% and 56% of reduction in salinity during stage I, 70% and 30% during stage II. Evaporative concentration was shown to be the major force responsible for calcium and carbon removal in lake water under arid environmental conditions. The quality of replenishment water was demonstrated to be as important as quantity for rehabilitating lakes in arid regions as it was one of the main factors controlling the variation in chemical reactions. This study demonstrates that water chemistry is an indicator that reflects the hydrologic and hydrochemical processes of lake evolution, but the "invisible" effects of groundwater-lake interactions and chemical reactions should be addressed with more research and management practices.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Author of contributing

J.C and H.Q conceived the presented idea, analysed data and co-wrote the paper. Y. G. analyzed experimental results. Y. G. and H. W. performed hydrogeochemical simulation and designed the figures. M. Z. aided in interpreting the results and worked on the manuscript. All authors discussed the results and commented on the manuscript.

References

Aeschbach-Hertig, W., Gleeson, T., 2012. Regional strategies for the accelerating global problem of groundwater depletion. Nature Geosci. 5 (12), 853.

Ahmed, A., Clark, I., 2016. Groundwater flow and geochemical evolution in the Central Flinders Ranges, South Australia. Sci. Total Environ. 572, 837–851.

Ali, E.M., Khairy, H.M., 2016. Environmental assessment of drainage water impacts on water quality and eutrophication level of Lake Idku, Egypt. Environ. Pollut. 216, 437–449.

Ballatore, T.J., Muhandiki, V.S., 2002. The case for a world lake vision. Hydrol. Process. 16 (11), 2079–2089.

Brkić, Ž., Briški, M., Marković, T., 2016. Use of hydrochemistry and isotopes for improving the knowledge of groundwater flow in a semiconfined aquifer system of the Eastern Slavonia (Croatia). Catena 142, 153–165.

Bui, N.T., et al., 2019. Groundwater sustainability assessment framework: a demonstration of environmental sustainability index for Hanoi, Vietnam. J. Environ. Manage. 241, 479–487. https://doi.org/10.1016/j.jenvman.2019.02.117.

Burns, D.A., et al., 2001. Quantifying contributions to storm runoff through end-member mixing analysis and hydrologic measurements at the Panola Mountain Research Watershed (Georgia, USA). Hydrol. Process. 15 (10), 1903–1924.

Chen, J., et al., 2019. Hydrogeochemical characteristics and quality assessment of groundwater in an irrigated region, Northwest China. Water 11 (1), 96.

Chen, J., Qian, H., 2017. Characterizing replenishment water, lake water and groundwater interactions by numerical modelling in arid regions: a case study of Shahu Lake. Hydrol. Sci. J.-J. Sci. Hydrol. 62 (1), 104–113. https://doi.org/10.1080/02626667.2015.1133910.

Chen, J., Wu, H., Qian, H., 2016. Groundwater nitrate contamination and associated health risk for the rural communities in an agricultural area of Ningxia, northwest China. Exposure and Health 8 (3), 349–359.

Chen, J., Wu, H., Qian, H., Gao, Y., 2017. Assessing nitrate and fluoride contaminants in drinking water and their health risk of rural residents living in a semiarid region of Northwest China. Exposure and Health 9 (3), 183–195.

Chen, J., Wu, H., Qian, H., Li, X., 2018. Challenges and prospects of sustainable groundwater management in an agricultural plain along the Silk Road Economic Belt, north-west China AU – Chen, Jie. Int. J. Water Resour. Dev. 34 (3), 354–368. https://doi.org/10.1080/07900627.2016.1238348.

Clark, I.D., Lauriol, B., 1992. Kinetic enrichment of stable isotopes in cryogenic calcites. Chem. Geol. 102 (1), 217–228. https://doi.org/10.1016/0009-2541(92)90157-Z.

Cosmo, R.D.P., Ressel Pereira, F.D.A., Ribeiro, D.D.C., Barros, W.Q., Martins, A.L., 2019. Estimating CO2 degassing effect on CaCO3 precipitation under oil well conditions. J. Petroleum Sci. Eng. 181. https://doi.org/10.1016/j.petrol.2019.106207.

Cui, B.-L., Li, X.-Y., Wei, X.-H., 2016. Isotope and hydrochemistry reveal evolutionary processes of lake water in Qinghai Lake. J. Great Lakes Res. 42 (3), 580–587.

Donovan, J.J., 1994. On the measurement of reactive mass fluxes in evaporative groundwater-source lakes. In: Renaut, R., Last, W.M. (Eds.), Sedimentology and Geochemistry of Modern and Ancient Saline Lakes. SEPM Society for Sedimentology Geology, pp. 33–50.

Edmunds, W., et al., 2003. Groundwater evolution in the Continental Intercalaire aquifer of southern Algeria and Tunisia: trace element and isotopic indicators. Appl. Geochem. 18 (6), 805–822.

El-Kadi, A.I., Plummer, L.N., Aggarwal, P., 2011. NETPATH-WIN: an interactive user version of the mass-balance model, NETPATH. Groundwater 49 (4), 593–599.

Elumalai, V., Nwabisa, D.P., Rajmohan, N., 2019. Evaluation of high fluoride contaminated fractured rock aquifer in South Africa – geochemical and chemometric approaches. Chemosphere 235, 1–11. https://doi.org/10.1016/j.chemosphere.2019.06.065.

Fang, L., Tao, S., Zhu, J., Liu, Y., 2018. Impacts of climate change and irrigation on lakes in arid northwest China. J. Arid Environ. 154, 34–39.

Gejl, R.N., Rygaard, M., Henriksen, H.J., Rasmussen, J., Bjerg, P.L., 2019. Understanding the impacts of groundwater abstraction through long-term trends in water quality. Water Res. 156, 241–251. https://doi.org/10.1016/j.watres.2019.02.026.

Gibbs, R.J., 1970. Mechanisms controlling world water chemistry. Science 170 (3962), 1088–1090.

Gill, L.W., Naughton, O., Johnston, P.M., Basu, B., Ghosh, B., 2013. Characterisation of hydrogeological connections in a lowland karst network using time series analysis of water levels in ephemeral groundwater-fed lakes (turloughs). J. Hydrol. 499, 289–302. https://doi.org/10.1016/j.jhydrol.2013.07.002.

Griffioen, J., 2001. Potassium adsorption ratios as an indicator for the fate of agricultural potassium in groundwater. J. Hydrol. 254 (1–4), 244–254.

Guo, M., Zhou, X., Li, J., Wu, W., Chen, Y., 2015. Assessment of the salinization processes in the largest inland freshwater lake of China. Stoch. Env. Res. Risk Assess. 29 (7), 1823–1833.

Gutierrez, M.F., et al., 2018. Salinity shapes zooplankton communities and functional diversity and has complex effects on size structure in lakes. Hydrobiologia 813 (1), 237–255.

Hammer, U.T., 1986. Saline lake ecosystems of the world, 59. Springer Science & Business Media.

Helfferich, F., 1962. Deutsche Ausfuhrlichkeit.(Book Reviews: Ion Exchange). Science, 138: 133.

Huo, A.-D., Dang, J., Song, J.-X., Chen, X.H., Mao, H.-R., 2016. Simulation modeling for water governance in basins based on surface water and groundwater. Agric. Water Manag. 174, 22–29.

Huo, A., Li, H., 2013. Assessment of climate change impact on the stream-flow in a typical debris flow watershed of Jianzhuangcuan catchment in Shaanxi Province, China. Environ. Earth Sci. 69 (6), 1931–1938.

Jiang, W., et al., 2018. Water area changing of Hongjiannao Lake and its reasons. Yellow River 4 (40), 72–75.

Jin, S., Hu, Z., Huang, Y., Hu, Y., Pan, H., 2019. Evaluation of several phosphate amendments on rare earth element concentrations in rice plant and soil solution by X-ray diffraction. Chemosphere 236, 124322. https://doi.org/10.1016/j.chemosphere.2019.07.053.

Li, P., Feng, W., Xue, C., Tian, R., Wang, S., 2017. Spatiotemporal variability of contaminants in lake water and their risks to human health: a case study of the Shahu Lake tourist area, northwest China. Exposure and Health 9 (3), 213–225.

Ma, L., et al., 2010. Hydrochemical characteristics and brine evolution paths of Lop Nor basin, Xinjiang Province, Western China. Appl. Geochem. 25 (11), 1770–1782.

McNeil, V.H., Cox, M.E., Preda, M., 2005. Assessment of chemical water types and their spatial variation using multi-stage cluster analysis, Queensland, Australia. J. Hydrol. 310 (1–4), 181–200.

Miao, C., Ni, J., Borthwick, A.G.L., Yang, L., 2011. A preliminary estimate of human and natural contributions to the changes in water discharge and sediment load in the Yellow River. Global Planet. Change 76 (3), 196–205. https://doi.org/10.1016/j.gloplacha.2011.01.008.

Micklin, P., 2007. The Aral sea disaster. Annu. Rev. Earth Planet. Sci. 35, 47–72.

Mohanty, K.K., 2003. The near-term energy challenge. American Institute of Chemical Engineers. AIChE J. 49 (10), 2454.

Montalván, F., et al., 2017. Hydrochemical and isotopes studies in a hypersaline wetland to define the hydrogeological conceptual model: Fuente de Piedra Lake (Malaga, Spain). Sci. Total Environ. 576, 335–346.

Morán Ramírez, J., Ramos Leal, J.A., 2014. The VISHMOD methodology with hydrochemical modeling in intermountain (karstic) aquifers: case of the Sierra Madre Oriental, Mexico.

Mountadar, S., Younsi, A., Hayani, A., Siniti, M., Tahiri, S., 2018. Groundwater salinization process in the coastal aquifer Sidi Abed-Ouled Ghanem (Province of El Jadida, Morocco). J. Afr. Earth Sc.

Négrel, P., Petelet-Giraud, E., Barbier, J., Gautier, E., 2003. Surface water–groundwater interactions in an alluvial plain: chemical and isotopic systematics. J. Hydrol. 277 (3–4), 248–267.

Nethononda, V.G., Elumalai, V., Rajmohan, N., 2019. Irrigation return flow induced mineral weathering and ion exchange reactions in the aquifer, Luvuvhu catchment, South Africa. J. Afr. Earth Sc. 149, 517–528. https://doi.org/10.1016/j.jafrearsci.2018.09.001.

Ningxia Statistical Bureau, 2017. Ningxia Statistical Yearbook.

Ningxia Water Conservancy Bureau, 2010-2017. Ningxia Water Resources Bulletin. In: http://www.nxsl.gov.cn/ (Ed.).

Ntanganedzeni, B., Elumalai, V., Rajmohan, N., 2018. Coastal aquifer contamination and geochemical processes evaluation in Tugela catchment, South Africa—geochemical and statistical approaches. Water 10 (6), 687.

Pang, Y., Qu, J., Niu, q., Zhang, K., Xie, S., 2017. Grain size characters of dune surface sediment in the Crescent spring scenic spot, Dunhuang. Arid Zone Research, 34: 701-706.

Papadimitriou, S., Kennedy, H., Kattner, G., Dieckmann, G.S., Thomas, D.N., 2004. Experimental evidence for carbonate precipitation and CO2 degassing during sea ice formation. Geochim. Cosmochim. Acta 68 (8), 1749–1761. https://doi.org/10.1016/j.gca.2003.07.004.

Piper, A.M., 1944. A graphic procedure in the geochemical interpretation of water-analyses. Eos, Trans. Am. Geophys. Union 25 (6), 914–928.

Prasanna, M.V., Chidambaram, S., Gireesh, T., Ali, T.J., 2011. A study on hydrochemical characteristics of surface and sub-surface water in and around Perumal Lake, Cuddalore district, Tamil Nadu, South India. Environ. Earth Sci. 63 (1), 31–47.

Qian, H., Li, P., Howard, K.W., Yang, C., Zhang, X., 2012. Assessment of groundwater vulnerability in the Yinchuan Plain, Northwest China using OREADIC. Environ. Monit. Assess. 184 (6), 3613–3628.

Qian, H., Wu, J., Zhou, Y., Li, P., 2013. Stable oxygen and hydrogen isotopes as indicators of lake water recharge and evaporation in the lakes of the Yinchuan Plain. Hydrol. Process. 28 (10), 3554–3562.

Qiao, X., Jiang, X., Chen, J., Yin, H., Chen, L., 2007. Effect of transferring water on ecological environment in east and west Juyanhai lake at the lower reaches of Heihe River. Journal of Northwest Sci-Tech University of Agriculture and Forestry.

Rademacher, L.K., Clark, J.F., Hudson, G.B., Erman, D.C., Erman, N.A., 2001. Chemical evolution of shallow groundwater as recorded by springs, Sagehen basin; Nevada County, California. Chem. Geol. 179 (1–4), 37–51.

Reynolds, J.F., et al., 2007. Global desertification: building a science for dryland development. Science 316 (5826), 847–851.

Rodríguez-Rodríguez, M., Benavente, J., Cruz-San Julián, J., Martos, F.M., 2006. Estimation of ground-water exchange with semi-arid playa lakes (Antequera region, southern Spain). J. Arid Environ. 66 (2), 272–289.

Rosenberry, D.O., Lewandowski, J., Meinikmann, K., Nützmann, G., 2015. Groundwater-the disregarded component in lake water and nutrient budgets. Part 1: effects of groundwater on hydrology. Hydrol. Process. 29 (13), 2895–2921.

Sacks, L.A., Lee, T.M., Swancar, A., 2014. The suitability of a simplified isotope-balance approach to quantify transient groundwater–lake interactions over a decade with climatic extremes. J. Hydrol. 519, 3042–3053. https://doi.org/10.1016/j.jhydrol.2013.12.012.

Shaad, K., Burlando, P., 2018. Monitoring and modelling of shallow groundwater dynamics in urban context: the case study of Jakarta. J. Hydrol.

Sposito, G., 2000. Ion exchange phenomena. Handbook of Soil Science 204, 241–263.

Stallard, R., Edmond, J., 1983. Geochemistry of the Amazon: 2. The influence of geology and weathering environment on the dissolved load. J. Geophys. Res. Oceans 88

(C14), 9671–9688.
Su, X., Cui, G., Du, S., Yuan, W., Wang, H., 2016. Using multiple environmental methods to estimate groundwater discharge into an arid lake (Dakebo Lake, Inner Mongolia, China). Hydrogeol. J. 24 (7), 1707–1722.
Tang, S., 2009. Relationship between environmental change and ecological water replenishment for Ailike Lake. Xinjiang Water Resour. 5, 28–31.
Thorslund, J., et al., 2014. Geochemical controls on the partitioning and hydrological transport of metals in a non-acidic river system. Hydrol. Earth Syst. Sci. Discuss. 11 (8), 9715–9758.
Villegas, P., Paredes, V., Betancur, T., Taupin, J.D., Toro, L.E., 2018. Groundwater evolution and mean water age inferred from hydrochemical and isotopic tracers in a tropical confined aquifer. Hydrol. Process. 32 (14), 2158–2175.
Wahed, M.S.A., Mohamed, E.A., El-Sayed, M.I., M'nif, A., Sillanpää, M., 2014. Geochemical modeling of evaporation process in Lake Qarun, Egypt. J. Afr. Earth Sci. 97, 322–330.
Wang, Y., Liu, Z., Yao, J., Bayin, C., 2017. Effect of climate and land use change in Ebinur Lake Basin during the past five decades on hydrology and water resources. Water Resour. 44 (2), 204–215.
Williams, W.D., 2000. Dryland lakes. Lakes Reservoirs Res. Manage. 5 (3), 207–212.
Wu, J., Xue, C., Tian, R., Wang, S., 2017. Lake water quality assessment: a case study of Shahu Lake in the semiarid loess area of northwest China. Environ. Earth Sci. 76 (5), 232.
Wurtsbaugh, W.A., et al., 2017. Decline of the world's saline lakes. Nat. Geosci. 10 (11), 816.
Xiao, J., Jin, Z., Wang, J., Zhang, F., 2015. Major ion chemistry, weathering process and water quality of natural waters in the Bosten Lake catchment in an extreme arid region, NW China. Environ. Earth Sci. 73 (7), 3697–3708.
Yechieli, Y., Wood, W.W., 2002. Hydrogeologic processes in saline systems: playas, sabkhas, and saline lakes. Earth Sci. Rev. 58 (3–4), 343–365.
Zhang, F., et al., 2015. The influence of natural and human factors in the shrinking of the Ebinur Lake, Xinjiang, China, during the 1972–2013 period. Environ. Monit. Assess. 187 (1), 4128.
Zhang, Q., Liu, J., Singh, V.P., Shi, P., Sun, P., 2017. Hydrological responses to climatic changes in the Yellow River basin, China: climatic elasticity and streamflow prediction. J. Hydrol. 554, 635–645. https://doi.org/10.1016/j.jhydrol.2017.09.040.
Zhu, D., Ryan, M.C., Sun, B., Li, C., 2014. The influence of irrigation and Wuliangsuhai Lake on groundwater quality in eastern Hetao Basin, Inner Mongolia, China. Hydrogeol. J. 22 (5), 1101–1114.
Zlotnik, V.A., Olaguera, F., Ong, J.B., 2009. An approach to assessment of flow regimes of groundwater-dominated lakes in arid environments. J. Hydrol. 371 (1–4), 22–30.