Ref.: " ***Evaluation of Lacustrine Groundwater Discharge, Hydrologic Partitioning, and Nutrient Budgets in a Proglacial Lake in Qinghai-Tibet Plateau: Using 222Rn and Stable Isotopes***" (Dr. Xin Luo)

Editor

I received comments from two reviewers, both suggested "minor revision". The reviewers are generally positive to the manuscript, but pinpoint some revisions needed to be done before its acceptance. After receiving the comments, I carefully read the manuscript again, and concur with the reviewers. Please reply to the comments/questions in detail and revise your manuscript accordingly.

- We appreciate the overall positive comments for this study. In the revised MS, we have addressed all the comments raised by the two reviewers in details as shown below.
- We have also checked the MS carefully for typos, missing authors, and their affiliations, terminology, updates of data in tables, and updates of variables in equations.

Reviewer #1

General comments: This paper is focusing on the evaluation of LGD and its related nutrient budgets and hydrologic partitioning in proglacial lake of QTP. The work is great and the paper is overall well organized. Anyway, I have the following comments for the authors to consider.

Reply to General comments:

- We appreciate the overall positive comments for this study.

Specific comments

1. Authors should address more about why it's important to study proglacial lake, especially the ones in QTP, in the introduction part.

- Well taken. We have added more description of the proglacial lakes, and lake dynamics in QTP under the influence of climate change and global warming. Some latest lake studies in the QTP were also reviewed (lines 81-104).

2. The primary productivity is calculated based on the dissolved inorganic nutrient budgets. Authors should be careful to do so. Did the authors consider the transformation between dissolved inorganic and particulate inorganic forms? Redfield ratio usually works in oceanic aquatic system. In lakes, the ratio is fairly variable.

- Good question, indeed, we noticed that the DIN and DIP relation cannot be used to quantify the primary production in the fresh lacustrine systems due to the high variability of Redfield ratios and the possibility of transformation between DIN/DIP. Based on the measurement results, DIN: DIP ratios in the lake water and groundwater are all much large than 16:1, indicating the phosphate limited conditions.
- As indicated by previous studies of glacial melting water bodies in the QTP, Arctic and Antarctic, the dominant form of dissolve phosphate is DIP, while the DOP contributes less than 10 % of the dissolved phosphate (Hawkings et al. 2016, Hodson 2007, Hodson et al. 2005, Liu et al. 2011, Mitamura et al. 2003). In the freshwater system, particular phosphate is highly bounded. Therefore, it is reasonable to assume that primary production in the lake

water is limited by DIP, and to assign glacial melting water as phosphate limited condition in this study.

- ➢ The primary producers in the lake system consume the nutrient production under variant N: P ratios as indicated by previous studies (Downing and McCauley 1992). To avoid the ambiguous statements and conclusions, we discussed this term as the biological uptake/transformation of nutrients, and removed the ambiguous statements on primary production throughout the MS (lines 756-800).

3. Radon in air is important term to do balance calculation. Was the Rn in air measured? I did not see the information or data about this term in the manuscript or the SI.

- ➢ Yes, we placed RAD7 for air radon-222 measurement at the lake shore for about 4 hours, and the mean activity of the lake area is 1.51 ±0.97 Bq m^{-3}. We added the ambient air radon-222 activities in Table 1 and some descriptions in the methodology part (lines 268-271).

4. Line 53-60, these two sentences are both started with the locations. Please revise them.

- ➢ Well taken, these sentences were revised as suggested (lines 76-77).

5. Line 208, how often were the Ra-226 samples collected? Or just one sample, and you assume Ra-226 is constant? 6. Line 279, how long is ïA¸Dˇ t? 7. Line 327, figure 5 is not attached.

- ➢ Due to the bad weather condition and large sampling volume, we could only obtain one ^{226}Ra sample. However, radium in the lacustrine system is significantly lower compared to ^{222}Rn, therefore, the decay input of ^{222}Rn from ^{226}Ra is minimal and negligible. Thus, the spatial heterogeneity of radon-222 will mount minimal effects on the final ^{222}Rn mass balance models.
- ➢ The time step is set to be 5 min, in consistent with the ^{222}Rn record interval. More statements were added in the revised MS (lines 365-366).
- ➢ Figure 5 was attached in the revised MS.

Technical corrections

1. Line 144, 0.7 or 7?

- ➢ Well taken, this is 0.7 m s^{-1}, and change was made in the revision MS (line 205).

2. Line 195, the unit should be L min-1.

- ➢ Well taken, unit was change to L min-1 (line 256).

3. Line 230, change “recently” to “recent”.

- ➢ Well taken, change was made (line 302).

4. Line 280, should be Equation (2).

- ➢ Well taken, change was made (line 367).

5. Line 383, two 18O?

- ➢ Typo and change was made (line 477).

Reviewer #2

General comments: This is an interesting and generally well-written paper that makes a good contribution to understanding the groundwater surface water interactions and estimating the lacustrine groundwater discharge in mountainous proglacial lakes in the QTP. The abstract is correctly informative with some remarks (see below). The introduction and the site description take into account previous papers in exhaustive way. The methodological approach for data analysis is modern without particular novelties. High-resolution 222Rn activities, water level, both in water temperature and, wind speed together with stable isotopic data are quite impressive. Many studies have done to explain the high single digits in the whole paper.

- We appreciated the overall positive comments.

4. Discussion

Do the adjoining lacustrine aquifers receive ('recharge') to sustain the inferred rate of groundwater discharge? And is the inferred width of the zone of lacustrine groundwater discharge compatible with the physics of the groundwater flow system and hydrological cycle?

- The regional precipitation recorded by the Jiuzhi station is to be around 90 mm d^{-1} during Aug, 2015. When deploying an empirical infiltration coefficient of 0.2 for the lake basin, the aquifer recharge rates are yielded up to 18 mm d^{-1}, which is sufficient to maintain the water balance within the lacustrine aquifers. Moreover, as indicated by previous studies in an interior lake of the QTP, Nam Co, a lake located at the area with relatively high evaporation and lower precipitation, its LGD is estimated to be 5-8 mm d^{-1} and is comparable to the results of this study. This also indirectly implicate that the LGD in this study is tenable and can be balanced by the recharge of the lacustrine aquifers, as Ximen Co basin is influenced by rather larger precipitation and lower evaporation compared to Nam Co.
- The inferred width of the zones of lacustrine groundwater discharge is also regarded as the seepage face. Previous studies have indicated that the groundwater seepage areas are mostly located along the transect within 10-20 m across the lakeshore (Luo et al. 2016, Luo et al. 2017, Rosenberry et al. 2015, Schafran and Driscoll 1993). While the deep groundwater system is rather constrained by the Precambrian bedrocks (Einarsdottir et al. 2017), and the LGD occurrence is considered to be constrained within the seepage faces along the lakeshores, and within the bathymetry of epilimnion.

Did you consider the lag time between recharge and chemical changes in the lacustrine aquifers?

- The lag time between the recharge and the chemical changes in the lacustrine aquifers is not considered in this study for the following reasons: (1) For ^{222}Rn, the equilibrium state is assigned as ^{222}Rn will reach equilibrium states within short distance (sever centimeters) and elapsed time after the infiltration (Ku et al. 1992, Porcelli 2008). (2) Stable isotopes generally behave rather conservatively after entering the aquifer and there is negligible fractionation during the transport in the aquifer between the recharge and discharge. (3) The groundwater sampling locations were located at the immediate zones of the lake shore, and therefore, the dynamics the flow length and recharge lag time is minimal and negligible for the reactive solutes of DIN and DIP, similar as suggested in many previous studies (Dimova and Burnett 2011, Dimova et al. 2013, Kluge et al. 2012, Luo et al. 2016).

Please consider the relationship between Fig 5 and Fig 6 to give a relevant illustration on chemical components and isotopic data.

- We are sorry that we forgot to attach Figure 5 in the previous version. This figure was used to give a relevant illustration on chemical components and isotopic data.

Fig. 6 The conceptual model of ^{222}Rn transient model looks well. But the associated illustration in the text is not convincing on the flow pathways for the 222Rn sources. Clearly some components of the conceptual understanding are not supported by the data. The manuscript would also benefit greatly from a more thorough literature review, which in-turn will help establish the objectives of the work. My main concern with the paper is with the 222Rn analysis that I don't think is well enough explained to be convincing. Doing a more thorough job on this will add material.

- If we understood properly, this comment has two points: reliability of some components and literature review. The reviewer did not specify which components that were no supported by the data. We guess they could be lake evaporation and riverine inflow. To take account this comment, we have reviewed more relevant literatures and added more discussion on lake evaporation and riverine inflow (lines 684 to 713).

Conclusions

This section just summarizes the main findings of the project. In the introduction you make some general statements about the need to understand processes in these impacted lacustrine aquifers in general. In this section explain in more detail how your project helps us to understand processes in these environments more broadly; the paper will have more impact if researchers from elsewhere in the world can see relevance to their studies and a paper in a major international journal such as HESS needs to have broad appeal

- To stress the research significance of this study, we have added more discussions to explain how the results of this study facilitate the understanding the environments more broadly (lines 857-863). We hope the updated MS can meet the board research interest of HESS.

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Revised MS submitted to HESS

Evaluation of Lacustrine Groundwater Discharge, Hydrologic Partitioning, and Nutrient Budgets in a Proglacial Lake in Qinghai-Tibet Plateau: Using ^{222}Rn and Stable Isotopes

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Abstract

Proglacial lakes are good natural laboratories to investigate groundwater and glacier dynamics under current climate condition and to explore biogeochemical cycling under pristine lake status. This study conducted a series of investigations of ^{222}Rn, stable isotopes, nutrients and other hydrogeochemical parameters in Ximen Co Lake, a remote proglacial lake in the east of Qinghai-Tibet Plateau (QTP). A radon mass balance model was used to quantify the lacustrine groundwater discharge (LGD) of the lake, leading to an LGD estimate of 10.3 ± 8.2 mm d^{-1}. Based on the three end member models of stable ^{18}O and Cl^{-}, the hydrologic partitioning of the lake is obtained, which shows that groundwater discharge only accounts for 7.0 % of the total water input. The groundwater derived DIN and DIP loadings constitute 42.9 % and 5.5 % of the total nutrient loading to the lakes, indicating the significance of LGD in delivering disproportionate DIN into the lake. This study presents the first attempt to evaluate the LGD and hydrologic partitioning in the glacial lake by coupling radioactive and stable isotopic approaches and the findings advance the understanding of nutrient budgets in the proglacial lakes of QTP. The study is also instructional in revealing the hydrogeochemical processes in proglacial lakes elsewhere.

Keywords: Proglacial lake; ^{222}Rn; lacustrine groundwater discharge; hydrologic partitioning; nutrient budgets

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1. Introduction

High altitude and latitude areas are intensively influenced by the melting of glaciers due to climatic warming. Of particular importance are the proglacial areas, such as proglacial lakes and moraines, because they are particularly affected by climatic change induced glacier retreating and thawing of permafrost (Heckmann et al., 2016;Barry, 2006;Slaymaker, 2011). The proglacial lakes are usually located close to ice front of a glacier, ice cap or ice sheet, with the vicinity to the ice front sometimes defined as the areas with subrecent moraines and formed by the last significant glacier advances at the end of the Little Ice Age (Heckmann et al., 2016;Barry, 2006;Slaymaker, 2011;Harris et al., 2009). These lakes are located in the transition zones from glacial to non-glacial conditions, and can serve as natural laboratories to explore hydrological processes, biogeochemical cycles and geomorphic dynamics under current climatic conditions (Dimova et al., 2015;Heckmann et al., 2015).

The Qinghai-Tibet Plateau (QTP), the third pole of the world, serves as the water tower of most of the major rivers in Asian (Qiu, 2008). Unique landscapes such as endorheric lakes, permafrost, glaciers, and headwater fluvial networks are developed due to the intensive interaction between the atmosphere, hydrosphere, biosphere and cryosphere (Lei et al., 2017;Zhang et al., 2017a;Zhang et al., 2017b;Yao et al., 2013;Yao et al., 2012). Distributed

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mountainous glaciers and lakes are the most representative landscapes and are highly sensitive to the climate changes. In the past decade, the lakes in the interior of the QTP show overall expanding with respect to an overall increase of precipitation, accelerated glacier melting and permafrost degradation (Zhang et al., 2013;Zhang et al., 2017b;Zhang et al., 2017a;Heckmann et al., 2016;Yang et al., 2014). Some latest studies have made effects to depict the hydrologic partitioning of the majority of the lakes in the QTP based on long term observation of climatological parameters, and remote sensing approaches. However, so far a quantitative evaluation of the water balance and hydrologic partitioning, especially the groundwater component of the lakes in the QTP is limited due to the scarcity of observational data. Therefore, there is a great need to conduct refined and systematical field observation to provide groundtruth dataset and tenable models to depict the water balance and hydrologic partitioning of the lakes, especially proglacial lakes in the QTP (Yang et al., 2014;Zhang et al., 2017b).

Mountainous proglacial lakes, formed by glacial erosion and filled by melting glaciers, are widely distributed in the Qinghai-Tibet Plateau (QTP), especially along the substantial glacier retreating areas of Himalaya Mountains (MT.), Qilian MT., Tienshan MT., etc. Characterized by higher elevations, small surface areas but relatively large depths, mountainous proglacial lakes in QTP lack systematic field-based hydrological studies due to their remote locations and difficulty in

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conducting field work (Yao et al., 2012;Farinotti et al., 2015;Bolch et al., 2012).

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There has been extensive recognition of the importance of groundwater discharge to various aquatic systems for decades (Dimova and Burnett, 2011;Valiela et al., 1978;Johannes, 1980). Very recently, the topic of 'lacustrine groundwater discharge (LGD)', which is comprehensively defined as groundwater exfiltration from lake shore aquifers to lakes (Lewandowski et al., 2015;Rosenberry et al., 2015;Blume et al., 2013;Lewandowski et al., 2013), has been introduced. LGD is analogous of in submarine groundwater discharge (SGD) in coastal environments. LGD also plays a vital role in lake hydrologic partitioning, which is defined as the separation of groundwater discharge/exfiltration, riverine inflow, riverine outflow infiltration, surface evaporation and precipitation for the hydrological cycle of the lake (Luo et al., 2017;Good et al., 2015). LGD also serves as an importance component in delivering solutes to lakes since groundwater is usually concentrated in nutrients, CH_4, dissolved inorganic/organic carbon (DIC/DOC) and other geochemical components (Paytan et al., 2015;Lecher et al., 2015;Belanger et al., 1985;Dimova et al., 2015). Nutrients and carbon loading from groundwater greatly influences ratios of dissolved inorganic nitrogen (DIN) to dissolved inorganic phosphate (DIP) (referred as N: P ratios thereafter), ecosystem structure and the primary productivity of the lake aquatic system (Nakayama and Watanabe, 2008;Belanger et al., 1985;Hagerthey and Kerfoot, 1998).

The approaches to investigate LGD include 1) direct seepage meters (Shaw and Prepas, 1990;Lee, 1977), 2) geo-tracers such as radionuclides, stable ^{2}H and ^{18}O isotopes (Gat, 1995;Kluge et al., 2007;Kraemer, 2005;Lazar et al., 2008), 3) heat and temperature signatures (Liu et al., 2015;Sebok et al., 2013), 4) numerical modeling (Winter, 1999;Smerdon et al., 2007;Zlotnik et al., 2009;Zlotnik et al., 2010) and 5) remote sensing (Lewandowski et al., 2013;Wilson and Rocha, 2016;Anderson et al., 2013). Recently, some researchers started to investigate groundwater dynamics in peri- and proglacial areas, mostly based on the approaches of numerical modeling (Lemieux et al., 2008b;Lemieux et al., 2008c;Andermann et al., 2012;Scheidegger and Bense, 2014;Lemieux et al., 2008a). However, the quantification of groundwater and surface water exchange in proglacial lakes is still challenging due to limited hydrogeological data and extremely seasonal variability of aquifer permeability (Dimova et al., 2015;Callegary et al., 2013;Xin et al., 2013).

^{222}Rn, a naturally occurring inert gas nuclide highly concentrated in groundwater, can be more applicable in fresh aquatic systems and has been widely used as a tracer to quantify groundwater discharge in fresh water lakes (Luo et al., 2016;Corbett et al., 1997;Dimova et al., 2015;Dimova and Burnett, 2011;Dimova et al., 2013;Kluge et al., 2007;Kluge et al., 2012;Schmidt et al., 2010) and terrestrial rivers and streams

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(Burnett et al., 2010;Cook et al., 2003;Cook et al., 2006;Batlle-Aguilar et al., 2014). Of particular interest are investigations of temporal ^{222}Rn distribution in lakes, since it can be used to quantify groundwater discharge and reflect the locally climatological dynamics (Dimova and Burnett, 2011;Luo et al., 2016). Temporal radon variations give high resolution estimates of groundwater discharge to lakes over diel cycles, allowing evaluation of LGD and the associated chemical loadings. However, there has been no study of radon-based groundwater discharge in mountainous proglacial lakes, especially for those lakes in the QTP.

This study aims to investigate the groundwater surface water interactions for the proglacial lake of Ximen Co, by estimating the LGD and evaluating the hydrologic partitioning of the lake. LGD is estimated with ^{222}Rn mass balance model, and the hydrologic partitioning of the lake is obtained with the three endmember model coupling the mass balance of water, stable isotopes and Cl^-. Moreover, LGD derived nutrients are estimated and the nutrient budgets of the lake are depicted. This study, to our knowledge, makes the first attempt to quantify the LGD, hydrologic partition, and groundwater borne nutrients of the proglacial lake in QTP and elsewhere via the approach integrating multiple tracers. This study provides insights of hydrologic partitioning in a typical mountainous proglacial lake under current climate condition and reveals groundwater borne chemical loadings in this proglacial lake in QTP and

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2. Methodology

2.1 Site descriptions

The Nianbaoyeze MT., located at the eastern margin of the QTP and being the easternmost part of NW-SW trending Bayan Har Shan, is situated at the main water divide of the upper reaches of Yellow River and Yangtze River (Figure 1). With a peak elevation of 5369 m, the mountain rises about 500-800 m above the surrounding peneplain and displays typical Pleistocene glacial landscapes such as moraines, U-shaped valleys and cirques (Lehmkuhl, 1998;Schlutz and Lehmkuhl, 2009;Wischnewski et al., 2014). The present snow line is estimated to be at an elevation of 5100 m (Schlutz and Lehmkuhl, 2009). Controlled by the South Asia and East Asia monsoons, the mountain has an annual precipitation of 975 mm in the southern part and 582 mm in the northwestern part, with 80 % occurring during May and October (Yuan et al., 2014;Zhang and Mischke, 2009). The average temperature gradient is about 0.55 ^{o}C per 100 m, and the closest weather station, locating in Jiuzhi town (N: 33.424614°, E: 101.485998) at the lower plains of the mountain, recorded a mean annual temperature of 0.1 ^{o}C. Snowfalls occur in nearly 10 months of the entire year and there is no free-frost all year around (Böhner, 1996, 2006;Schlutz and

Lehmkuhl, 2009). The precipitation, daily bin-averaged wind speed and temperature in Aug, 2015 were recorded to be 90 mm, 0.7 m s^{-1} and 9.5 ^{o}C from Jiuzhi weather station (Figure 2). The water surface evaporation was recorded to be 1429.8 mm in 2015 from Jiuzhi weather station.

Among the numerous proglacial lakes developed in the U-shaped valleys of the Nianbaoyeze MT., Ximen Co lake is located at the northern margin of the mountain with an elevation of 4030 m asl, and is well studied and easily accessible (Lehmkuhl, 1998;Zhang and Mischke, 2009;Schlutz and Lehmkuhl, 2009;Yuan et al., 2014). The lake was formed in a deep, glacially eroded basin with a catchment area of 50 km^2, and has a mean and a maximum depth of 40 m and 63.2 m, and a surface area of 3.6 km^2. The vegetation around the lake is dominated by pine meadows with dwarf shrubs, rosette plants and alpine cushion (Schlutz and Lehmkuhl, 2009;Zhang and Mischke, 2009;Yuan et al., 2014). Mostly recharged by the glacial and snowpack melting water and regional precipitation, the lake is stratified with an epilimnion depth about 4.4 m in the summer time. The lake is usually covered by ice in the winter time (Zhang and Mischke, 2009). The superficial layer within the U-shaped valley is characterized by peat, clay and fluvial gravels with a depth about 1-3.5 m. Discontinuous and isolated permafrost is present at the slope of the valley above the elevation of about 4150 m. The maximum frozen depth is about 1.5 m for the seasonal frozen ground around the lake. The seasonal frozen ground serves as an unconfined aquifer during the unfrozen months from July to October, and groundwater discharges into the epilimnion of the lake (Wang, 1997;Schlutz and Lehmkuhl, 2009;Zhang and Mischke, 2009).

2.2 Sampling and field analysis

The field campaign to Ximen Co Lake was conducted in August, 2015, when it is warm enough to take the water samples of different origins as the studied site is seasonally frozen. A ^{222}Rn continuous monitoring station was setup at the southeast part of the lake, where is fairly flat for setting up our tent and monitoring system. Surface water samples were collected around the lake, rivers at the upstream and downstream. Porewater samples were collected at one side of the lake as the other side is steep and rocky. The basic water quality parameters of conductivity (EC), dissolved oxygen (DO), TDS, ORP, and pH in the water were recorded with the multi-parameter meter (HANNA, Co.). Relative humidity was recorded with a portable thermo-hydrometer (KTH-2, Co.). Lake water samples were taken with a peristaltic pump into 2.5 L glass bottles for ^{222}Rn measurement with the Big Bottle system (Durridge, Co.). Surface water samples were filtered with 0.45 μm filters (Advantec, Co.) in situ and taken into 5 ml, 15 ml, 15 ml and 50 ml Nalgene centrifugation tubes for stable isotope, major anion, cation and nutrient analysis.

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Porewater samples were taken from the lakes shore aquifers with a push point sampler (M.H.E, Co.) connected to peristaltic pump (Solinst, Co.) (Luo et al., 2014;Luo et al., 2016). 100 ml raw surface water or porewater was titrated with 0.1 μM H_2SO_4 cartridge (Hach, Co.) in situ to measure total alkalinity (Hasler et al., 2016;White et al., 2016;Warner et al., 2013). Porewater was filtered with 0.45 μm syringe filters in situ and taken into 5 ml, 15 ml, 15 ml and 50 ml Nalgene centrifugation tubes for stable isotope, major anion, cation and nutrient analysis. 250 ml porewater was taken for ^{222}Rn measurement with RAD7 H_2O (Durridge, Co.). Samples for major cation analysis were acidified with distilled HNO_3 immediately after the sampling.

^{222}Rn continuous monitoring station was set up at the northwest of the lake, close to the downstream of the lake (Figure 1b). Lake water (about 0.5 m in depth) was pumped with a DC pump (12 V) driven by lithium batteries (100 Ah) and sprinkled into the chamber of RAD7 AQUA with a flow rate > 2 L min^{-1}, where ^{222}Rn in water vapor was equilibrated with the air ^{222}Rn. The vapor in the chamber was delivered into two large dry units (Drierite, Co) to remove the moisture and circulated into RAD7 monitor, where ^{222}Rn activities were recorded every 5 mins. A temperature probe (HOBO$^{@}$) was insert into the chamber to record the temperature of the water vapor. The monitoring was performed from 11: 31 am, Aug 22nd to 6: 30 am, Aug 24th, 2015. During the period of 1:50-4:30 pm on Aug 22nd, a sudden blizzard occurred,

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leading to an hourly precipitation about 0.6 mm to the lake area. Daily and hourly climatological data such as wind speed, air temperature and precipitation were retrieved from the nearest weather station in Jiuzhi town (N: 33.424614°, E: 101.485998). Moreover, another RAD7 was placed at the lakes hore to measure ^{222}Rn in the ambient air around the lake. Due to extremely low activities, the monitoring period was conducted only for 4 hours, and the mean activity was adopted as the background radon-222 activity to be used in the mass balance model. Water level and temperature fluctuations were recorded with a conductivity-temperature-depth diver (Schlumberger, Co.) fixed at about 20 cm below the lake surface and calibrated with local atmospheric pressure recorded by a baro-diver (Schlumberger, Co.) above the lake. To correct for dissolved ^{226}Ra supported ^{222}Rn, one radium sample was extracted from 100 L lake water with MnO_2 fiber as described elsewhere (Luo et al., 2014;Moore, 1976).

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2.3 Chemical analysis

Major ions were measured with ICS-1100 (Dionex. Co.) in the Department of Earth Sciences, the University of Hong Kong. The uncertainties of the measurements are less than 5 %. Nutrients, DIN and DIP were analyzed with flow injection analysis equipped with auto-sampler (Lachat. Co.) in the School of Biological Sciences, the

University of Hong Kong. Stable ^{18}O and ^{2}H isotopes were measured with MOA-ICOS laser absorption spectrometer (Los Gatos Research (LGR) Triple Isotope Water Analyzer (TIWA-45EP)) at State Key Laboratory of Marine Geology, Tongji University, Shanghai. The stable isotopic standards and the recovery test have been fully described elsewhere (Luo et al., 2017). The measurement uncertainty is better than 0.1 % for ^{18}O and 0.5 % for ^{2}H. ^{226}Ra was detected with RAD7 with the method described elsewhere (Kim et al., 2001;Lee et al., 2012;Luo et al., 2018).

2.4 Radon transient model

Previous studies employed a steady state radon-222 mass balance model to quantify LGD to lentic system such as lakes and wetlands (Dimova and Burnett, 2011;Luo et al., 2016). This model assumes that radon input derived from groundwater inflow, diffusion and river inflow are balanced by the radon losses of atmospheric evasion, decay and river outflow. However, recent studies revealed that the steady state is mainly reached after 2-15 days of constant metrological conditions, and most lentic system cannot be treated as steady state due to rapid radon-222 degassing to the atmosphere driven by wind-induced turbulence (Gilfedder et al., 2015; Dimova and Burnett, 2011).

Ximen Co lake is demonstrated to be highly stratified with an epilimnion of 4.4

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m (Zhang and Mischke, 2009). The lake was formed by glacier erosion and the lakebed is characterized by granite bedrock with a thin sedimentary clay layer. Previous studies have indicated that sediment consisting of clay, soils and gravels has been developed on the bedrock and forms the lake shore aquifer with a thickness of 0.7-3.3 m (Schlutz and Lehmkuhl 2009). Porewater sampled in the aquifer immediate behind the lake shore can well represent groundwater discharging into the lake, as suggested previously (Lewandowski et al., 2015;Rosenberry et al., 2015;Schafran and Driscoll, 1993). LGD has been widely considered to occur within the first few meters of the lake shore (Schafran and Driscoll, 1993;Rosenberry et al., 2015;Lee et al., 1980) and groundwater is considered to predominately discharge into the epilimnion since deep groundwater flow is highly limited by the Precambrian bedrock (Einarsdottir et al., 2017). Due to negligible hydrological connection between the epilimnion and hypolimnion, ^{222}Rn mass balance model is established to quantify LGD to the epilimnion from the lake shore.

The governing equation of radon-222 transient mass balance model within a 1 x 1 x z cm (where z is the depth in cm) can be expressed as (Gilfedder et al., 2015):

$$z\frac{\partial I_w}{\partial t} = F_{gw} + (I_{^{226}Ra} - I_w) \times z \times \lambda_{222} + F_{diff} - F_{atm} \quad (1)$$

where F_{gw}, F_{diff}, F_{atm} [Bq m^{-2} d^{-1}] are ^{222}Rn loadings from LGD, water-sediment diffusion and water-air evasion, respectively; z [m] is the lake water level depth

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recorded by the diver. λ_{222} is the decay constant of ^{222}Rn with a value of 0.186 d^{-1}. $\lambda_{222} \times I_{^{226}Ra}$ and $\lambda_{222} \times I_w$ account for the production and decay of ^{222}Rn [Bq m^{-2} d^{-1}] in the water column, respectively. I_w and $I_{^{226}Ra}$ [Bq m^{-2}] represent ^{222}Rn and ^{226}Ra inventories in the epilimnion, and are expressed as: $I_w = H \times C_w$ and $I_{^{226}Ra} = H \times C_{^{226}Ra}$, respectively; where H [m] is the depth of the epilimnion; C_w and $C_{^{226}Ra}$ is the ^{222}Rn and ^{226}Ra activity [Bq m^{-3}], respectively.

The model is valid under the following assumptions: 1) The epilimnion is well mixed which is the actual condition for most natural boreal and high altitude glacial lakes (Zhang and Mischke, 2009;Åberg et al., 2010). 2) ^{222}Rn input from riverine water inflow, and loss from the lake water outflow and infiltration into the lake shore aquifer is negligible compared to the groundwater borne ^{222}Rn, because ^{222}Rn concentration of groundwater is 2-3 orders of magnitude larger than that of lake water (Dimova and Burnett, 2011;Dimova et al., 2013). Generally, ^{222}Rn in the epilimnion is sourced from LGD and decay input from parent isotope of ^{226}Ra under secular equilibrium, and is mainly lost via atmospheric evasion and radioactive decay.

F_{atm} is the key sinking component of the transient model and is finally a function of wind speed and water temperature, both of which are temporal variant variables (Supplementary information). Lake water level z is also a temporal variant variable which represents the fluctuations of water volume of the epilimnion. This equation is discretized by the forward finite difference method, and the groundwater flux at each time step can be solved as follow

$$[{}^{222}Rn_{t+\Delta t}] = \frac{[z \times {}^{222}Rn_t + [F_{diff} + F_{gw} - F_{atm} - {}^{222}Rn_t \times \lambda \times z] \times \Delta t}{z} \quad (2)$$

where ${}^{222}Rn_{t+\Delta t}$ and ${}^{222}Rn_{t+\Delta t}$ [Bq m^{-3}] is the ^{222}Rn activity at current time step and at the previous time steps, respectively, and Δt [min] is the time step which is set to be 5 min in consistence with the ^{222}Rn record interval. With the inverse calculation based on Equation (2), the groundwater inflow at each time step can be obtained. However, large errors of the final LGD calculation will be induced by even a small amount of noise in the measured ^{222}Rn data due to the ${}^{222}Rn_{t+\Delta t} - {}^{222}Rn_t$ term being with the measure uncertainty. To reduce the random errors of the measured ^{222}Rn concentrations, the time window with a width of 1 hour is proposed to smooth the curve (Supplementary information).

3. **Results**

3.1 Time series data

Figure 2 shows the basic climatological parameters of the lake catchment during the campaign month. There are discrete rainfall events occurring throughout the month with an average rainfall of 3.1 mm d^{-1}. The temperature during the month ranges from 5.0 – 12.5 ^{o}C within an average of 9.3 ^{o}C. The daily averaged wind speed ranges from 0.7 – 2.5 m s^{-1}, with an average of 1.7 m s^{-1}. ^{222}Rn temporal distribution

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and other time series data are shown in Figure 3a and listed in Supplementary Table 1. Generally, ^{222}Rn concentration varies from 32.2 to 273 Bq m^{-3}, with an average of 144.2 ± 27.7 Bq m^{-3}. ^{222}Rn over the monitoring period shows typical diel cycle, much higher at nighttime and lower in the day time. Figures 3b-3d show the time series data of temperature (5 mins interval), nearshore lake water level (1 min interval), and wind speed (1 hour interval). Temperature and lake water level also show typical diel cycles, but with antiphase fluctuations with each other. Temperature is higher during the daytime and lower at nighttime. However a sudden decrease of temperature was recorded due to the sudden blizzard (Figure 3b). Water level is higher at nighttime and lower during the daytime, with a strong fluctuation due to the turbulence caused by the blizzard (Figure 3c). The variability might reflect the dynamics of groundwater input and surface water inflow. The air temperature of the lake area is in phase with the water temperature. Wind speed is normally higher during the daytime and lower at nighttime (Figure 3d).

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The variation of ^{222}Rn is nearly in antiphase with the fluctuations of lake water temperature and air temperature, indicating that the dominated controlling factors of ^{222}Rn fluctuations are water temperature and wind speed (Figure 3a). This phenomenon is reasonable as lake water ^{222}Rn is predominately lost via atmospheric evasion, which is the function of wind speed and water temperature (Dimova et al., 2015;Dimova and Burnett, 2011;Dimova et al., 2013). High water temperature and wind speed leads to elevated atmospheric evasion and causes the decline of ^{222}Rn concentration in the lake water. However, there is a sudden reduction of radon activity from 2: 00 pm to 4: 00 pm on Jul 22nd, 2015, when the snow event led to a sudden decrease of water temperature, increase of wind speed, and large surface water turbulence as indicated by water level fluctuations (Figures 3a-3d). ^{222}Rn in the porewater is 2-3 orders of magnitude larger than ^{222}Rn in the lake water, suggesting that ^{222}Rn is an ideal tracer to estimate the LGD (Supplementary Table 1). ^{222}Rn concentrations in surface water range from 22.2 to 209 Bq m^{-3}, with an average of 92.5 Bq m^{-3} (n = 12), which is in the range of ^{222}Rn continuous monitoring results, suggesting reliable ^{222}Rn measurements (Supplementary Table 2).

3.2 Geochemical results

The results of major ions, nutrients and stable isotopes in different water endmembers are shown in Figures 4 and 5. Cl^- ranges from 0.6 to 2.1 mg L^{-1} in the surface water (including riverine inflow water, lake water and downstream water), 0.4 to 2.7 mg L^{-1} in porewater and has a much higher concentration of 5.9 mg L^{-1} in rainfall water. Na^+ ranges from 1.6 to 3.4 mg L^{-1} in the surface water, 1.2 to 4.4 mg L^{-1} in porewater and has a concentration of 4.4 mg L^{-1} in rainfall water. SO_4^{2-} ranges

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from 1.2 to 2.3 mg L^{-1} in the surface water, 0.4 to 1.7 mg L^{-1} in porewater and has a significant low concentration of 0.01 mg L^{-1} in rainfall water. Ca^{2+} ranges from 3.0 to 12.4 mg L^{-1} in lake water, 3.4 to 12.5 mg L^{-1} in porewater and has a significantly high concentration of 20.5 mg L^{-1} in rainfall water. Other concentrations of major ions are listed in Supplementary Table 2. As shown in Figure 4d and Supplementary Table 2, $\delta^{18}O$ in the lake water ranges from - 13.06 ‰ to - 12.11 ‰, with an average of - 12.41 ‰ (n = 7), and $\delta^{2}H$ ranges from - 91.83 ‰ to - 87.47 ‰, with an average of - 89.0 ‰ (n = 7). $\delta^{18}O$ in the riverine inflow water ranges from - 13.44 ‰ to - 13.29 ‰, with an average of – 13.37 ‰ (n = 2), and $\delta^{2}H$ ranges from - 93.25 ‰ to – 91.92 ‰, with an average of - 92.59 ‰ (n = 2). $\delta^{18}O$ in the downstream water ranges from - 12.51 ‰ to - 12.18 ‰, with an average of - 12.35 ‰ (n = 3), and $\delta^{2}H$ ranges from - 88.96 ‰ to - 87.1 ‰, with an average of - 87.98 ‰ (n = 3). $\delta^{18}O$ in the porewater ranges from - 12.66 ‰ to - 11.52 ‰, with an average of - 11.97 ‰ (n = 8), and $\delta^{2}H$ ranges from – 91.3 ‰ to -82.87 ‰, with an average of - 85.5 ‰ (n = 8). DIN in the surface water (including riverine inflow water, lake water and downstream water) ranges from 6.6 to 16.9 µM, with an average of 10.3 µM, and DIP from 0.36 to 0.41 µM, with an average of 0.38 µM. DIN for the porewater ranges from 0.7 to 358.8 µM, with an average of 92.8 µM, and DIP from 0.18 to 0.44 µM with an average of 0.31 µM (Figure 5).

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4. Discussion

4.1 Proglacial hydrologic processes and geochemical implications

Generally, major ion concentrations in the lake water and porewater of Ximen Co lake are significantly lower than those in major rivers, streams and other tectonic lakes in the QTP (Yao et al., 2015;Wang et al., 2010;Wang et al., 2016b), and are similar to those of snow and glaciers (Liu et al., 2011), suggesting that the lake water is mainly originated from glacier and snow melting. Ion concentrations in the lake and porewater of Ximen Co lake are much lower than those of rainfall collected in Jiuzhi town. This suggests that lake water is less influenced by precipitation (Figures 4a-4c). The concentrations of major ions in the porewater are high compared to the lake water, indicating weathering affects from the aquifer grains. The ratios of Ca^{2+}/Na^{+} in the porewater and groundwater is >1, also suggesting influences of weathering digenesis of major ions from the seasonal frozen ground at the lake shore aquifer (Weynell et al., 2016;Yao et al., 2015;Wang et al., 2010).

The isotopic compositions of the lake water and porewater are significantly isotopically depleted, with values close to the compositions of glaciers and surface snow in the QTP, suggesting the lake is dominantly recharged from snow and glacier melting (Cui et al., 2014;Wang et al., 2016a;Zongxing et al., 2015). The relation of

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$\delta^{18}O$ versus δ^2H for the lake water is $\delta^2H = 4.25 \times \delta^{18}O - 35.99$, with a slope much lower than that of the global meteoric water line (GMWL) (Figure 4d), suggesting the effects of lake surface evaporation. The relation of $\delta^{18}O$ versus δ^2H for the porewater is $\delta^2H = 6.93 \times \delta^{18}O - 2.67$, overall on GWML (Figure 4d). Deuterium excesses is defined as $\Delta D = \delta D - 8 \times \delta^{18}O$ (Dansgaard, 1964). The value of ΔD is dependent on airmass origins, altitude effect and the kinetic effects during evaporation (Hren et al., 2009). Global meteoric water has a ΔD of + 10 ‰. In the QTP, glacier/snowpack melting water usually has large positive ΔD, while the precipitations derived from warm and humid summer monsoon has lower ΔD (Ren et al., 2017;Ren et al., 2013). In this study, ΔD of surface water, lake and porewater ranges from + 8.5 to + 11.8 ‰, closed to the glacier melting water but much smaller than that of the local precipitation of + 18.8 ‰, This indicates the stream and lake water are mainly originated from glacial/snowpack melting rather than precipitation (Gat, 1996;Wang et al., 2016a;Lerman et al., 1995). The slopes of δ^2H versus $\delta^{18}O$ in lake water and porewater are 4.25 and 6.93, both of which are lower than that of GMWL due to surface evaporation. Lake water is more intensively influenced by evaporation compared to porewater. The plots of $\delta^{18}O$ versus Cl^-, and δ^2H versus Cl^- are well clustered for porewater end member (orange area), lake water end member (blue area), riverine inflow water end member (yellow area), and precipitation water (Figures 4e

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and 4f), suggesting stable $\delta^{18}O$ and $\delta^{2}H$ isotopes and Cl^- can serve as tracers to quantify the hydrologic partitioning of the lake by setting three endmember models.

The concentrations of DIN and DIP are all within the ranges of other glacial melting water and proglacial lake water (Hawkings et al., 2016;Hodson, 2007;Hudson et al., 2000;Tockner et al., 2002;Hodson et al., 2005). Briefly, rainfall and upstream lake water such as YN-4 have the highest DIN concentrations, indicating the glacier melting and precipitation could be important DIN sources in proglacial areas (Dubnick et al., 2017;Anderson et al., 2017). DIN in porewater is overall higher compared to the lake water, suggesting the porewater to be DIN effective source; and DIP concentration is higher in the lake water compared to porewater, suggesting the porewater is a DIP sink (Figure 5). The N: P ratios in the lake water and porewater are averaged to be 27.1 and 320.5, respectively, both much larger than the Redfield Ratio (N: P = 16:1) in water and organism in most aquatic system and within the range of other proglacial lakes (Anderson et al., 2017). This also suggests that the lake water and porewater are under phosphate limited condition. N: P ratio in the rainfall water is 30.4, similar to the lake water. The average N: P ratio of porewater is much higher than that of lake water, indicating DIN enrichment in the lake shore aquifers (Figure 5). In pristine groundwater, NO_3^- is the predominated form of dissolved nitrogen and is highly mobile within the oxic aquifers, leading to much higher DIN concentrations

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in the porewater; DIP has high affinity to the aquifer grains, resulting in much lower DIP concentrations in the porewater (Lewandowski et al., 2015;Rosenberry et al., 2015;Slomp and Van Cappellen, 2004). Thus, in analogous to surface runoff from glacier/snowpack melting, LGD can be also regarded as an important DIN source for the proglacial lakes. Because of very high DIN and N: P ratios in the porewater, a relatively small portion of LGD delivers considerable nutrients into the glacial lake, shifting the aquatic N: P ratios and affecting the proglacial aquatic ecosystem (Anderson et al., 2017).

4.2 Estimation of LGD

Figure 6a shows all the sinks and sources of radon with the epilimnion of the lake. Within ^{222}Rn transient mass balance model, the dominant ^{222}Rn loss is atmospheric degassing/evasion. Generally, ^{222}Rn degassing rate is the function of the radon-222 concentration gradient at the water-air interface and the parameter of gas piston velocity *k*, which is finally the function of wind speed and water temperature (Dimova and Burnett, 2011;Gilfedder et al., 2015). To evaluate ^{222}Rn evasion rate, this study employs the widely used method proposed by MacIntyre et al. (1995) which is also detailed described in supplementary information. Based on the field data of ^{222}Rn concentration in the lake water, wind speed and temperature log, the radon degassing

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rate is calculated in a range of 0.8 to 265.2 Bq m^2 d^{-1}, with an average 42.0 of Bq m^2 d^{-1}.

In addition to the atmospheric loss and sedimentary diffusion inputs, ^{222}Rn is also sinked via radioactive decay, and sourced from decay of parent isotope of ^{226}Ra. The decay loss of ^{222}Rn fluctuates in phase with the distribution of ^{222}Rn concentration monitored by RAD 7 AQUA. The equations to estimate benthic fluxes are shown in supplementary information. The decay loss is calculated to be 26.4 to 223.4 Bq m^{-2} d^{-1}, with an average of 118.0 ± 22.7 Bq m^{-2} d^{-1}. ^{226}Ra concentration is 0.01 Bq m^{-3} for the lake water. Under secular equilibrium, the ^{226}Ra decay input can be calculated by multiplying ^{226}Ra concentration in the lake water with λ_{222} (Corbett et al., 1997;Kluge et al., 2007;Luo et al., 2016). ^{226}Ra decay input is calculated to be 0.83 Bq m^{-2} d^{-1}, which is significantly low compared to other ^{222}Rn sources to the epilimnion.

With the obtained sinks and sources of ^{222}Rn in the lake, and the constants given in Table 1, LGD rate can be obtained by dividing the groundwater derived ^{222}Rn with its concentration in groundwater endmember. The obtained LGD rate, ranges from -23. 7 mm d^{-1} to 90.0 mm d^{-1}, with an average of 10.3 ± 8.2 mm d^{-1} (Figure 7). The LGD rate range is relatively smaller than the daily lake water level variations (≈ 50 mm), indicating that the lake water level variation could be a combined effect of surface runoff and LGD (Hood et al., 2006). The negative values of LGD rate reflect the

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return groundwater flow due to infiltration into the porewater. Normally, the dominant values are positive, indicating LGD rate is significant compared to water infiltration into lake_shore aquifer. The temporal variation of LGD rate could be attributed to the fluctuations of the hydraulic gradient in the proglacial areas (Hood et al., 2006;Levy et al., 2015). As indicated by ΔD (mostly > 10) of surface water, the lake and the upstream water is considered to be mainly recharged from glacial/snowpack melting rather other precipitations.

To assess the magnitude of uncertainty of ^{222}Rn transient model, the sensitivity of estimated LGD to changes in other variables is examined. A sensitivity coefficient *f* is proposed to evaluate this uncertainty according to Langston et al. (2013)

$$f = (\Delta F_{LGD} / F_{LGD}) / (\Delta y_i / y_i) \quad (3)$$

where ΔF_{LGD} is the amount of change in F_{LGD} from the original value. Δy_i is the amount of change in the other variable of y_i from the original value. Thus, higher *f* indicates a large uncertainty of final LGD estimate. The uncertainty mainly stems from ^{222}Rn measurements in different water endmembers, the atmospheric loss and water level record. The uncertainties of ^{222}Rn measurement are about 10 % and 15-20 % in groundwater and lake water endmember, respectively. The uncertainty of atmospheric loss is derived from uncertainty of ^{222}Rn in lake water (with an uncertainty of 15-20 %), temperature (with an uncertainty ≈ 5 %) and wind speed

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(with an uncertainty ≈ 5 %). Thus, the final LGD estimate has an integrated uncertainty of 35-40 %.

4.3 Hydrologic partitioning

Compared to the groundwater labeled radionuclide of ^{222}Rn, stable $^{18}O/^{2}H$ isotopes are advantageous in the investigation of evaporation processes due to their fractionations from water to vapor and have been widely used to investigate the hydrologic cycle of lakes in various environments (Stets et al., 2010;Gat, 1995;Gonfiantini, 1986;Gibson et al., 1993). With the field data of stable isotopic composition and Cl^- concentrations in different water endmembers, groundwater input, surface water input, lake water outflow and infiltration, and evaporation can be partitioned by coupling stable isotopic mass balance model with Cl^- mass balance model (Figure 6b).

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The model, consisting of the budgets of stable isotopes and Cl^-, and water masses for the epilimnion, is used to quantify riverine inflow, lake water outflow and infiltration, and evaporation (LaBaugh et al., 1995;LaBaugh et al., 1997;Gibson et al., 2016). The model is valid under the following assumptions: (1) constant density of water; (2) no long-term storage change in the reservoir; (3) well-mixed for the epilimnion (Gibson, 2002;Gibson et al., 2016;Gibson and Edwards, 2002;LaBaugh et al., 1997). The above assumptions are reasonably tenable during the short monitoring period. The model can be fully expressed as

$$F_{in} + F_{LGD} + F_p = F_E + F_{out} \quad (4)$$

$$F_{in} \times \delta_{in} + F_{LGD} \times \delta_{gw} + F_p \times \delta_p = F_E \times \delta_E + F_{out} \times \delta_L \quad (5)$$

$$F_{in} \times [Cl^-]_{in} + F_{LGD} \times [Cl^-]_{gw} + F_p \times [Cl^-]_p = F_{out} \times [Cl^- \quad (6)$$

where F_{in} [mm d^{-1}] is the surface water inflow to the lake; F_{gw} [mm d^{-1}] is LGD rate. F_p [mm d^{-1}] is the mean daily rainfall rate during the sampling period. F_E [mm d^{-1}] is the lake evaporation. F_{out} [mm d^{-1}] is the lake water outflow via runoff and infiltration into the lake shore aquifer. δ_{in}, δ_{gw},, δ_E and δ_p are the isotopic compositions of surface water inflow, LGD, and evaporative flux, respectively. The values of δ_{in}, δ_{gw} and δ_p are obtained from field data and the composition of δ_E are cacluated as shown in supplementary information. $[Cl^-]_{in}$, $[Cl^-]_{gw}$, $[Cl^-]_L$ and $[Cl^-]_p$ are the chloride concentrations in the inflow water, porewater, lake water and precipitation, respectively.

The components of the mass balance model can be obtained from the field data of isotopic composition and Cl^- concentrations in different water endmembers. The average ^{18}O composition -13.37 ‰ of riverine inflow water is taken as the value of the input parameter δ_{in}. $\delta^{18}O$ and δ^2H in the groundwater endmember and lake water end member are calculated to be -12.41 ‰ and -87.18 ‰, respectively. $\delta^{18}O$ and δ^2H

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in the rainfall are measured to be -5.47 ‰ and -24.98 ‰, respectively. With the measured values of δ_L, h, δ_{in}, and the estimated ε and δ_a, the isotopic composition of δ_E is calculated to be -35.11 ‰, which is in line with the results of alpine and arctic lakes elsewhere (Gibson, 2002;Gibson et al., 2016;Gibson and Edwards, 2002). The values of $[Cl^-]_{in}$, $[Cl^-]_{gw}$, and $[Cl^-]_L$ are calculated to be 0.91 mg L^{-1}, 1.48 mg L^{-1} and 1.02 mg L^{-1}, respectively. All the parameters used in the model are shown in Table 2.

According to Equations 4-6, the uncertainties of calculations of F_{in}, F_{out} and E are mainly derived from the uncertainty of F_{LGD} and the compositions of Cl^-, δD and $\delta^{18}O$ in different water endmembers as suggested in previous studies (Genereux, 1998;Klaus and McDonnell, 2013). The compositions of Cl^-, δD and $\delta^{18}O$ in surface water, groundwater endmembers have an uncertainty of 5 %. The uncertainty of δ_E is reasonably assumed to be ≈ 20 % . Thus, considering the uncertainty propagation of all the above parameters, the uncertainties of F_{in}, F_{out} and E would be scaled up to 70-80 % of the final estimates.

4.4 The hydrologic partitioning of the glacial lake

Based on the three endmember model of ^{18}O and Cl^-, the riverine inflow rate was calculated to be 135.6 ± 119.0 mm d^{-1}, and the lake outflow rate is estimated to be

141.5 ± 132.4 mm d^{-1}; the evaporation rate is calculated to be 5.2 ± 4.7 mm d^{-1}. The summary of the hydrologic partitioning of the lake is shown in Figure 8a. Generally, the proglacial lake is mostly recharged by the riverine inflow from the snowpack or the glacier melting. The groundwater discharge contributes about only 7.0 % of the total water input to the lake, indicating groundwater input does not dominate water input to the proglacial lake. The recent review on LGD rate by Rosenberry et al. (2015) suggests that the median of LGD rate in the literatures is 7.4 mm d^{-1} (0.05 mm d^{-1} to 133 mm d^{-1}), which is about 2/3 of LGD rate in this study. This difference may be due to the hydrogeological setting of the lake shore aquifer. This aquifer is formed by grey loam, clayey soil and sand (Lehmkuhl, 1998;Schlutz and Lehmkuhl, 2009), which is with relatively high permeability.

Previous studies have indicated that groundwater forms a key component of proglacial hydrology (Levy et al., 2015). However, there have been limited quantitative studies of groundwater contribution to hydrologic budget of proglacial areas. This study further summarizes the groundwater discharge studies over the glacial forefield areas. Based on long term hydrological and climatological parameter monitoring on the Nam Co lake in the QTP, Zhou et al. (2013) estimated the LGD to be 5-8 mm d^{-1} , which is comparable to the surface runoff input and LGD of this study. Brown et al. (2006) investigated the headwater streams at the proglacial areas of

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Taillon Glacier in French and found that groundwater contributes 6-10 % of the stream water immediate downwards of the glacier. Using water mass balance model, Hood et al. (2006) shows that groundwater inflow is substantial in the hydrologic partitioning of the proglacial Lake O'Hara in front of Opabin Glacier in Canada and comprised of 30 -74 % of the total inflow. Roy and Hayashi (2008) studied the proglacial lakes of Hungabee lake and Opabin lake at glacier forefield of Opabin Glacier and found that groundwater component is predominant water sources of the lakes and consisted of 35-39 % of the total water input of the lakes. Langston et al. (2013) further investigated a tarn immediate in front of Opabin Glacier and indicated the tarn is predominantly controlled by groundwater inflow/outflow, which consisted of 50-100 % of total tarn volume. Magnusson et al. (2014) studied the streams in the glacier forefield of Dammagletscher, Switzerland and revealed that groundwater contributed only 1-8 % of the total surface runoff. Groundwater contribution in this study is similar to those obtained the mountainous proglacial areas in Europe, but much lower than those obtained in the proglacial areas of polar regions. It is concluded that proglacial lakes/streams in front of mountainous glaciers are mainly recharged by surface runoff from glacier/snowpack melting. This might be due to well-developed stream networks and limited deep groundwater flow (Einarsdottir et al., 2017;Brown et al., 2006;Magnusson et al., 2014). However, proglacial tarns and lakes in the polar areas are predominantly controlled by groundwater discharge, due to less connectivity of surface runoff and high shallow and deep groundwater connectivity (Langston et al., 2013;Hood et al., 2006;Roy and Hayashi, 2008).

The evaporation constitutes relatively small ratio (≈ 3.5 %) of total water losses. The annual evaporation rate was recorded to be 1429.8 mm (equivalent to 3.92 mm d^{-1}) in 2015 by the Jiuzhi weather station, lower than the obtained evaporation in this study. This may be due to much higher evaporation in August during the monitoring period. The estimation of evaporation in this study generally represents the upper limit of the lake, as the sampling campaign was conducted during the summer time when the highest evaporation might occur. The lake surface evaporation derived from the pan evaporation in the QTP ranges from ~ 700 mm yr^{-1} in the eastern QTP to over 1400 mm yr^{-1} in the interior lakes of the QTP (Zhang et al., 2007;Ma et al., 2015;Yang et al., 2014). The evaporation of this study is rather in line with the previous evaporation observation in the eastern QTP, stressing the tenability of evaporation in this study. The runoff input is predominated recharge component (> 90 %) compared to other components, with an area normalized value comparable to previous studies of runoff input in other glacial melting dominant lakes in the QTP (Zhou et al., 2013;Zhang et al., 2011;Biskop et al., 2016). The runoff input and the lake evaporation of the study area, however, are subject to highly daily, seasonal and inter-annual variability as

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indicated by previous studies in the QTP (Zhou et al., 2013;Lei et al., 2017;Ma et al., 2015;Lazhu et al., 2016). Therefore, further investigations of long term and high resolution climatological and isotopic data are required to provide precise constraints of hydrologic partitioning of the lakes in the QTP.

4.5 LGD derived nutrient loadings, nutrient budget and ecological implications

Compared to extensive studies of SGD derived nutrient loadings in the past decade (Luo and Jiao, 2016;Slomp and Van Cappellen, 2004), studies of LGD derived nutrient loadings have received limited attention, even given the fact that groundwater in lake shore aquifers is usually concentrated in nutrients (Lewandowski et al., 2015;Rosenberry et al., 2015). Even fewer studies focus on chemical budgets in the proglacial lakes which are often difficult to access for sampling. Groundwater borne DIN and DIP across the sediment-water interface in this study are determined with an equation coupling the advective or LGD-derived, and diffusive solute transport (Lerman et al., 1995;Hagerthey and Kerfoot, 1998)

$$F_j = -nD_j^m \frac{dC_j}{dx} + v_{gw}C_j \qquad (7)$$

where $-nD_j^m \frac{dC_j}{dx}$ is the diffusion input and $v_{gw}C_j$ is the LGD derived fluxes, F_j [μM m^{-2} d^{-1}] is the mol flux of nutrient species *j* (representing DIN or DIP). *n* is the sediment porosity. D_j^m is the molecular diffusion coefficient of nutrient species *j*,

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which is given to be 4.8 x 10^{-5} m^2 d^{-1} for DIP (Quigley and Robbins, 1986), and 8.8 x 10^{-5} m^2 d^{-1} for DIN (Li and Gregory, 1974), respectively. C_j [μM] is the concentration of nutrient species *j*. *x*[m] is the sampling depth. v_{gw} is LGD rate estimated by ^{222}Rn mass balance model and has a value of 10.3 $\pm$8.2 mm d^{-1}. $\frac{dC_j}{dx}$ is the concentration gradient of nutrient species *j* across the water-sedimentary interface.

Substituting the constants and the field data of DIN and DIP in to Equation 6, LGD derived nutrient loadings are calculated to be 954.3 μmol m^{-2} d^{-1} and 3.2 μmol m^{-2} d^{-1} for DIN and DIP, respectively. Riverine inflow brings 1195.0 μmol m^{-2} d^{-1} DIN, 52.9 μmol m^{-2} d^{-1} DIP into the lake. Lake water outflow derived nutrient loss is estimated to be 1439.9 μmol m^{-2} d^{-1} and 54.7 μmol m^{-2} d^{-1} for DIN and DIP, respectively. Nutrients in the lake can be also sourced from atmospheric deposit (mostly in form of precipitation). With the nutrient concentrations in the rain water during the monitoring period, the wet deposit is calculated to be 76 μmol m^{-2} d^{-1} and 2.5 μmol m^{-2} d^{-1}, for DIN and DIP, respectively. The loadings of DIN to the lakes are mainly from surface runoff and LGD, which comprised of 42.9 % and 53.7 % of the total DIN loadings. Groundwater derived DIP input, however, constitutes only 6.3 % of the total DIP inputs to the lake, indicating groundwater borne DIP is less contributive to the nutrient budget of the lake compared to DIN. Very recent studies on polar regions have indicated that the glacier/snowpack water is the main N sources to the proglacial

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lakes (Anderson et al., 2013;Dubnick et al., 2017). However, they do not consider the contribution of groundwater borne N, in spite of the high groundwater connectivity in the proglacial areas (Roy and Hayashi, 2008). This study stresses that groundwater borne DIN could be comparable to the surface runoff derived DIN.

Based on nutrient results, the lake is considered to be an oligotrophic lake, similar to other glacier melting dominant lakes in the QTP (Mitamura et al., 2003;Liu et al., 2011). Phytoplankton is good dissolved organic phosphate (DOP) recyclers and will overcome inorganic P limitation though DOP cycling in most template lakes (Hudson et al., 2000). However, this may be not applicable for the glacial melting water and the peri/pro-glacial lake water. Previous studies show that phosphate nutrients are dominated by DIP and particulate phosphate, and the DOP contributes less than 10 % of the dissolved phosphate (Cole et al., 1998;Hawkings et al., 2016;Hodson, 2007). Thus DOP recycling is not likely to low N: P ratio under these conditions. Thus, the primary production (PP) is therefore considered to be controlled by the DIP loadings. The sum of DIN and DIP inputs minus the calculated DIN and DIP outputs leads to surpluses of 785.4 µmol m^{-2} d^{-1} and 3.9 µmol m^{-2} d^{-1} for DIN and DIP, respectively. The surpluses are expected to be consumed by the phytoplankton and converted into the PP under phosphate limited conditions. As primary producers in the fresh lacustrine system consume the nutrient under variant N: P ratios (7.1 to 44.2, mean:

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22.9) (Downing and McCauley, 1992), the biological uptake of DIN is roughly estimated to be 89.3 $\mu M\ m^{2}\ d^{-1}$. Therefore, the nutrient budgets for DIN and DIP can be finally conceptualized in Figures 8b and 8c.

4.6. Implications, prospective and limitations

Mountainous proglacial lakes are readily developed in glacier forefields of QTP and other high mountainous glacial such as Europe Alps and Pamir at central Asian (Heckmann et al. 2016). The proglacial lakes are always trapping system of sediment and sinks for water and chemical originated from glacier/snowpack melting and groundwater. In analogous to cosmogenic isotopes such as ^{10}Be serving as a tool to quantify the sediment sources, approaches integrating ^{222}Rn and stable isotopes provides both qualitatively and quantitatively evaluations of groundwater contributions and hydrologic partitioning in these remote and untapped lacustrine systems. Thus, it is expected that the multiple aqueous isotopes is considered to be effective tools to investigate the LGD and hydrologic partitioning in other proglacial lakes. This study is mainly limited by the relatively short sampling and monitoring period. As a special hydrologic regime, the lake shore aquifers of the proglacial lakes are experiencing frozen-unfrozen transition seasonally, and the dominant recharge of glacial melting could be fluctuated significantly due to air temperature variation. Therefore, future groundwater and hydrological studies can be extended to longtime

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sampling and monitoring of stable isotopes and ^{222}Rn in different water endmembers to reveal the seasonally hydrological and hydrogeological dynamics and their impacts on local biogeochemical cycles and ecological systems. Special concerns would be placed on how surface/groundwater interactions and the associated biogeochemical processes in response to the seasonal frozen ground variations and glacier/snowpack melting intensity.

5. Conclusion

A ^{222}Rn continuous monitoring is conducted at Ximen Co Lake, a proglacial lake located at the east QTP. A dynamic ^{222}Rn mass balance model constrained by radium mass balance and water level fluctuation is used to quantify temporal distribution of LGD of the lake. The obtained LGD over the monitoring time ranges from – 23. 7 mm d^{-1} to 80.9 mm d^{-1}, with an average of 10.3 ± 8.2 mm d^{-1}. Thereafter, a three endmember model consisting of the budgets of water, stable isotopes and Cl^{-} is used to depict the hydrologic partitioning of the lake. Riverine inflow, lake water outflow via surface runoff, and surface evaporation are estimated to be 135.6 mm d^{-1}, 141.5 mm d^{-1} and 5.2 mm d^{-1}, respectively. LGD derived nutrient loading is estimated to be 785.4 μmol m^{-2} d^{-1} and 3.2 μmol m^{-2} d^{-1} for DIN and DIP, respectively. This study also implicates that LGD constitutes relatively small portion of the proglacial hydrologic

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partitioning, however, delivers nearly a half of the nutrient loadings to the proglacial lake.

This study presents the first attempt to quantify LGD and the associated nutrient loadings to the proglacial lake of QTP. To our knowledge, there is almost no study on the groundwater-lake water interaction in the high altitude proglacial lakes in QTP. This study demonstrates that ^{222}Rn based approach can be used to investigate the groundwater dynamics in the high altitude proglacial lakes. The method is instructional to similar studies in other proglacial lakes in the QTP and elsewhere. For a comprehensive understanding the hydrological and biogeochemical dynamics in the QTP, interdisciplinary and multi-approach integrated studies are in great need. Of particular importance are the lake hydrology and groundwater surface water interaction studies based on multiple approaches such as remote sensing products, long term and high resolution observation of climatological parameters and isotopic data.

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Figure captions

Figure 1 The geological and topographic map of the Yellow River Source Region, Nianbaoyeze glacial mountains (**a**), and the sampling settings of the Ximen Co Lake (**b**), with the bathymetry map of the lake (**d**). (**c**) Photograph of the Ximen Co Lake and the surrounding geomorphic settings looking northeast direction on 22 Aug 2015, showing the late-laying snowpack in the U-shaped valleys of the north part of Nianbaoyeze MT.

Figure 2 The climatological parameters (wind speed, air temperature, and precipitation) in the Aug, 2015 recorded from Jiuzhi weather station.

Figure 3 The temporal distributions of ^{222}Rn (**a**), water temperature (**b**), water level fluctuation recorded by the divers (**c**), and hourly wind speed and air temperature recorded in Jiuzhi weather station (**d**).

Figure 4 The cross plots of Cl^- versus Na^+ (**a**), SO_4^{2-} versus Cl^- (**b**), Ca^{2+} versus Cl^- (**c**); The relations of ^2H versus ^{18}O (**d**), Cl^- versus ^2H (**e**), and Cl^- versus ^{18}O (**f**).

Figure 5 Cross plots of ^{222}Rn versus DIN (**a**) and DIP (**b**).

Figure 6 The conceptual model of ^{222}Rn transient model (**a**), and three endmember model (**b**).

Figure 7 The results of the final LGD derived from ^{222}Rn transient model.

Figure 8 The hydrologic partition of the proglacial lake of Ximen CO (**a**), and the budgets of DIN (**b**) and DIP (**c**).

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Figure 1

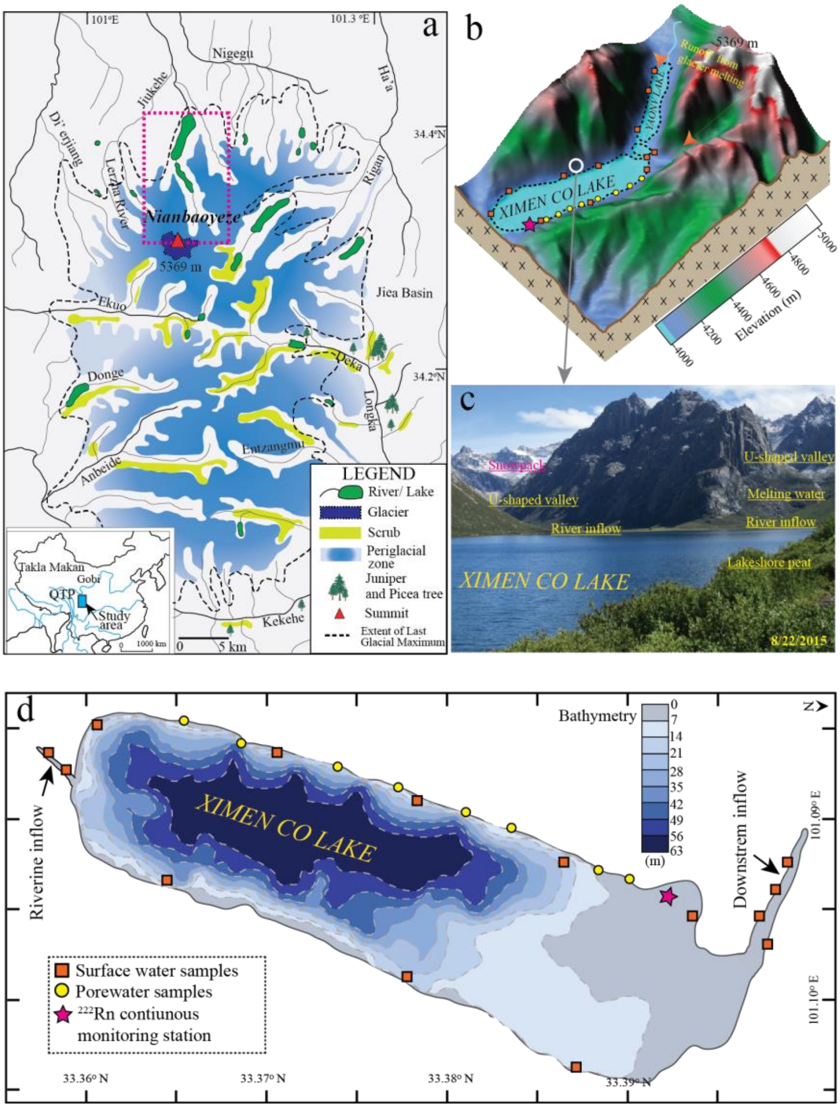

a
b
c
d
101°E
101.3 °E
34.4°N
34.2°N
Nigegu
Jiukehe
Di' erjiang
Lerzha River
Ha'a
Rigan
Nianbaoyeze
5369 m
Ekuo
Jiea Basin
Deka
Donge
Longka
Entzangmu
Anbeide
Kekehe
0
5 km
Takla Makan
Gobi
QTP
Study area
1000 km
LEGEND
River/ Lake
Glacier
Scrub
Periglacial zone
Juniper and Picea tree
Summit
Extent of Last Glacial Maximum
5369 m
Runoff from glacier melting
XIMEN CO LAKE
Elevation (m)
4000
4200
4400
4600
4800
5000
Snowpack
U-shaped valley
River inflow
U-shaped valley
Melting water
River inflow
Lakeshore peat
XIMEN CO LAKE
8/22/2015
Bathymetry
0
7
14
21
28
35
42
49
56
63
(m)
N
Riverine inflow
Downstrem inflow
XIMEN CO LAKE
101.09° E
101.10° E
33.36° N
33.37° N
33.38° N
33.39° N
Surface water samples
Porewater samples
²²²Rn continuous monitoring station

Figure 2

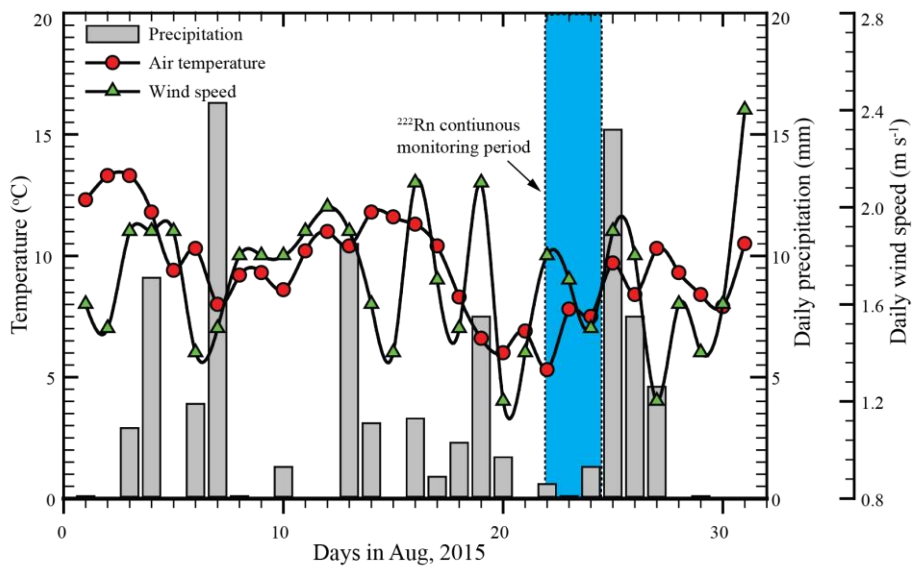
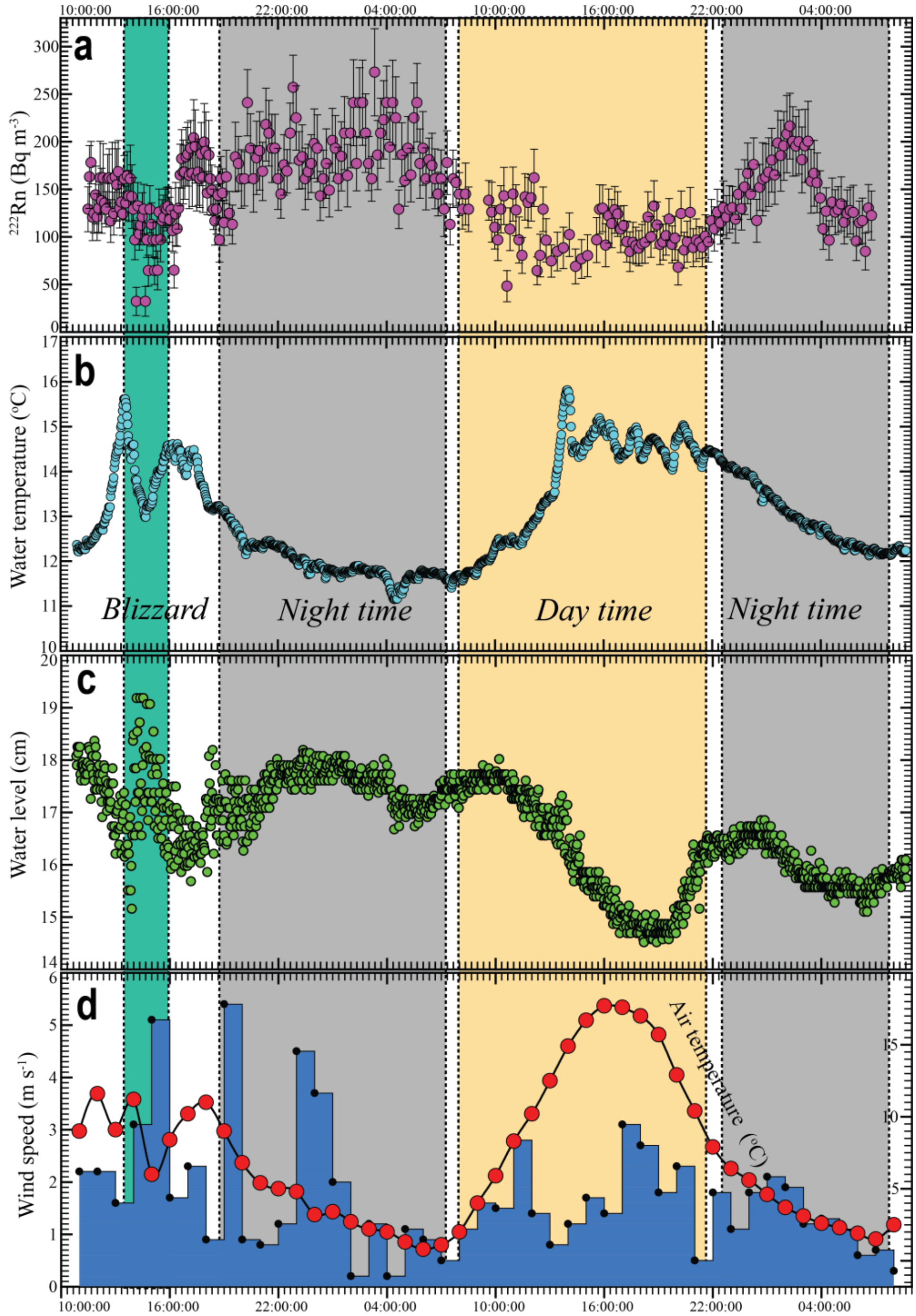
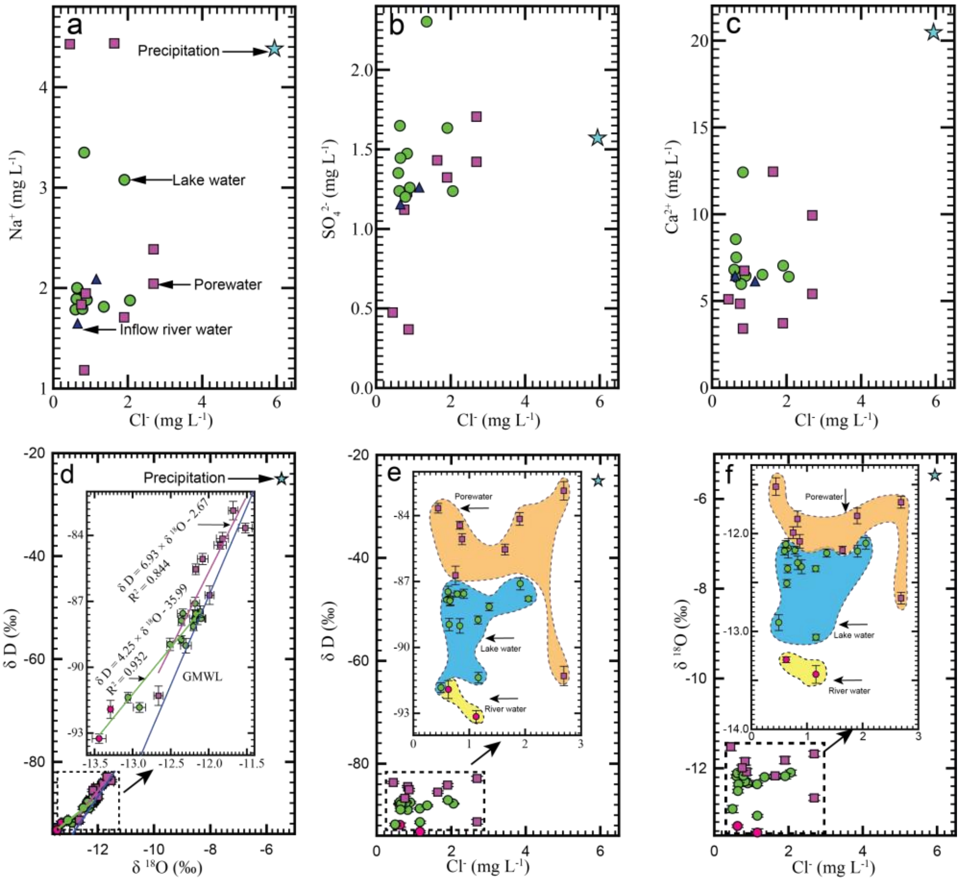

Precipitation
Air temperature
Wind speed
222Rn contiunous
monitoring period
Temperature (°C)
Daily precipitation (mm)
Daily wind speed (m s-1)
Days in Aug, 2015
0
10
20
30
0
5
10
15
20
0.8
1.2
1.6
2.0
2.4
2.8

Figure 3



10:00:00
16:00:00
22:00:00
04:00:00
a
b
c
d
^{222}Rn (Bq m^{-3})
Water temperature (°C)
Water level (cm)
Wind speed (m s^{-1})
Air temperature (°C)
Blizzard
Night time
Day time
Night time

Figure 4



a
Precipitation
Lake water
Porewater
Inflow river water
Na+ (mg L-1)
Cl- (mg L-1)
b
SO4 2- (mg L-1)
Cl- (mg L-1)
c
Ca2+ (mg L-1)
Cl- (mg L-1)
d
Precipitation
δD = 6.93 × δ18O - 2.67
R2 = 0.844
δD = 4.25 × δ18O - 35.99
R2 = 0.932
GMWL
δ D (‰)
δ 18O (‰)
e
Porewater
Lake water
River water
δ D (‰)
Cl- (mg L-1)
f
Porewater
Lake water
River water
δ 18O (‰)
Cl- (mg L-1)

Figure 5

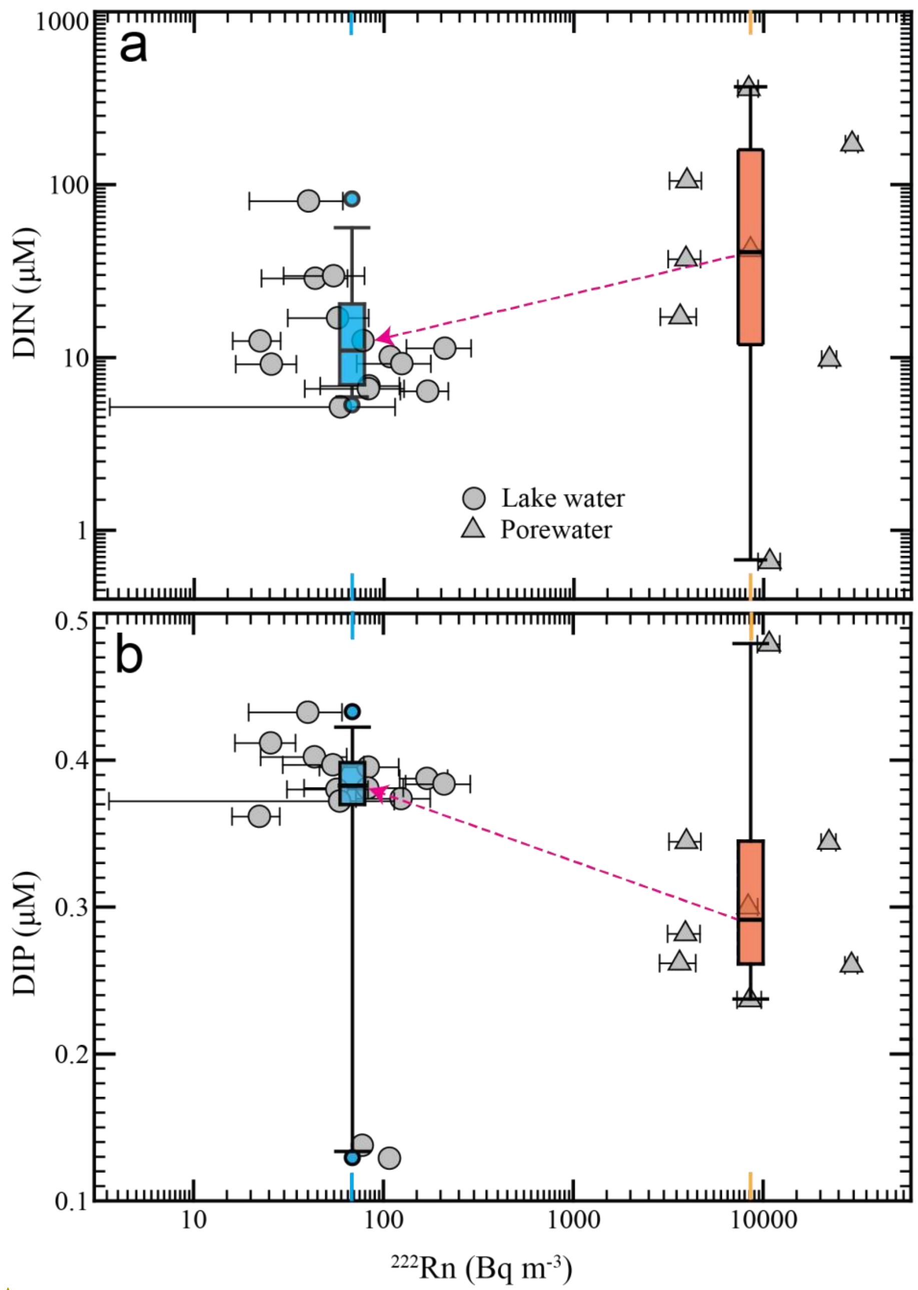


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Figure 6

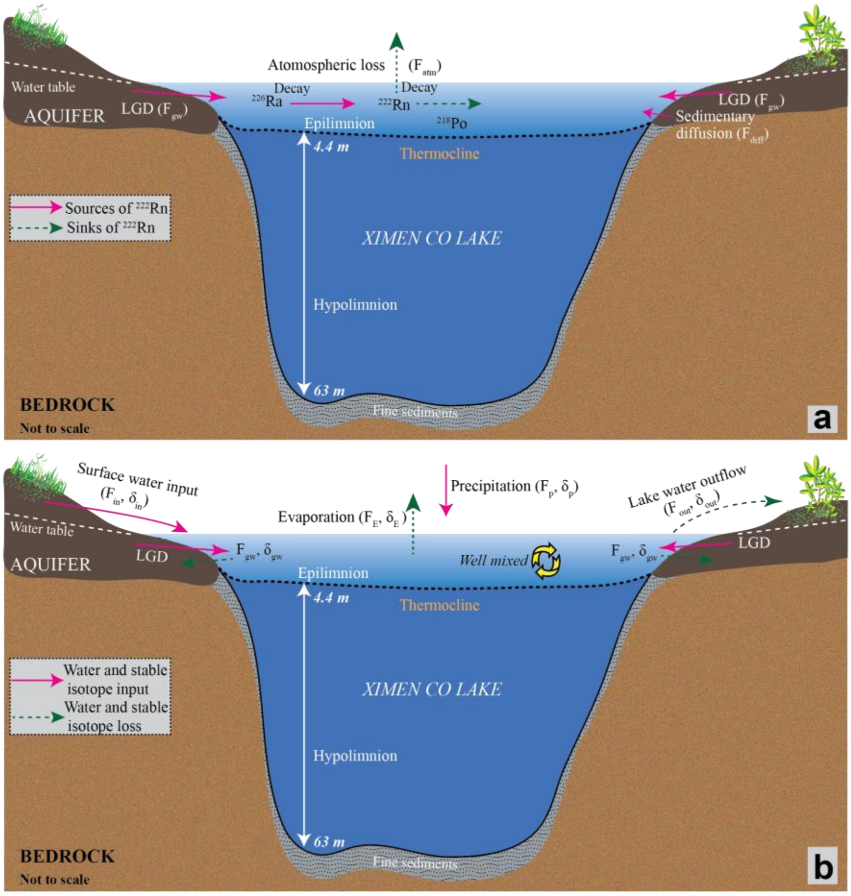

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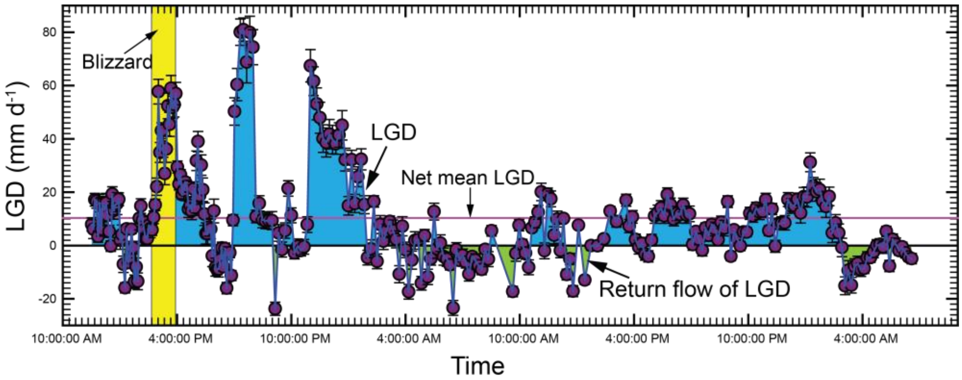
Figure 7


Blizzard
LGD
Net mean LGD
Return flow of LGD
LGD (mm d-1)
80
60
40
20
0
-20
10:00:00 AM
4:00:00 PM
10:00:00 PM
4:00:00 AM
10:00:00 AM
4:00:00 PM
10:00:00 PM
4:00:00 AM
Time

1396 Figure 8

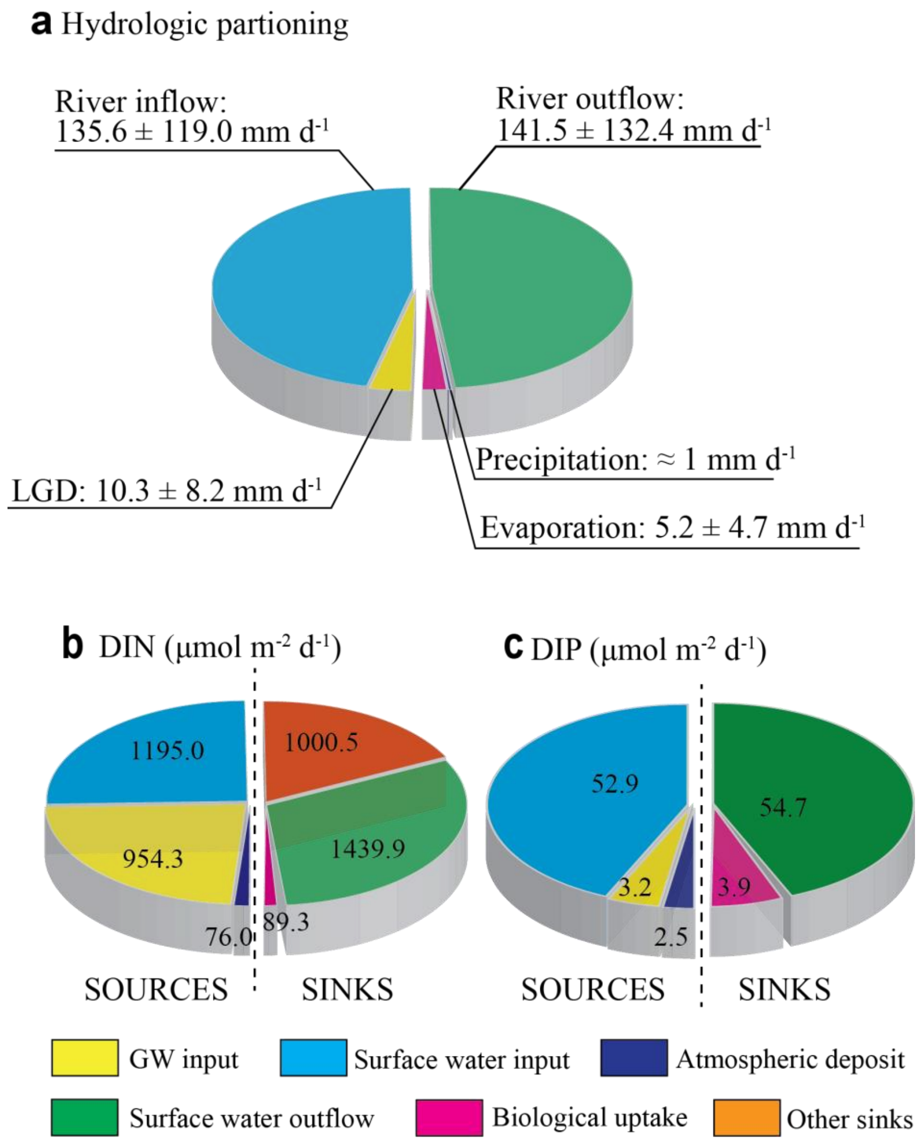


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Table 1 Parameters used to establish the mass balance model of ^{222}Rn in Ximen Co Lake.

Parameters	Units or values	Estimated Uncertainty (%)	Evaluation
Wind speed ($\boldsymbol{\omega_{10}}$)	0.2 - 5.4 m s^{-1}	n.a	From Jiuzhi weather stations
Water-air temperature	11.2 - 15.6 ^{o}C	n.a	Recorded with probe in the chamber; sensitive to temperature results
Molecular diffusion of ^{222}Rn in water ($\boldsymbol{D_m}$)	9.2×10^{-6} - 1.0×10^{-5} cm^2 s^{-1}	n.a	1.16×10^{-6} at 20 ^{o}C; adjustable for temperature
Molecular diffusion of ^{222}Rn in sediments ($\boldsymbol{D_s}$)	2.2×10^{-6} - 2.5×10^{-5} cm^2 s^{-1}	n.a	Adjusted for temperature, sediment porosity
Dynamic viscosity ($\boldsymbol{\mu}$)	1.1×10^{-3} - $1.3\times10^{-3}cm^2$ s^{-1}	n.a	Calculated based on water temperature, density and salinity
Schmidt number ($\boldsymbol{S_c}$)	1078.6 - 1371.6 [-]	n.a	Calculated as the ratio of $\boldsymbol{\upsilon}$ to $\boldsymbol{D_m}$
Water depth ($\boldsymbol{H}$)	4.4 m	n.a	Epilimnion depth of Ximen Co Lake
Decay constant ^{222}Rn ($\boldsymbol{\lambda_{222}}$)	0.186 d^{-1}	n.a	Constant
Groundwater endmember ^{222}Rn ($\boldsymbol{C_{gw}}$)	11200 $\pm$1200 Bq m^{-3}	8 sites~~s~~ ~~dependant~~dependent for	Measured; final result for water flux inversely proportional to ^{222}Rn groundwater concentration
Lake water endmember ^{222}Rn ($\boldsymbol{C_l}$)	21.6- 418.8 Bq m^{-3}	15-25%	Measured with RAD 7 AQUA
Ambient air 222Rn (C_{air})	1.51 $\pm$0.97 Bq m^{-3}	15-25 %	Measured with RAD 7 under open loop conditions
Atmospheric ^{222}Rn ($\boldsymbol{C_a}$)	1.5 $\pm$1.0 Bq m^{-3}	20-25%	Measured or assumed value, model not sensitive to radon in air variation
$K_{air/water}$	0.29 - 0.33 [-]	n.a	Calculated based on temperature in the chamber and salinity in lake water
Porosity $\boldsymbol{n}$	0.31	n.a	Assumed based on literatures
Tortuosity $\boldsymbol{\theta}$	2.05	n.a	Calculated based on porosity
Piston velocity ($\boldsymbol{\kappa}$)	0.004 - 1.11 m d^{-1}	20-25%	Calculated from Equation 3 in supplementary information
^{226}Ra concentration in lake waters ($\boldsymbol{C_{226Ra}}$)	0.01 Bq m^{-3}	≈10%	Measured with RAD7
Diffusive flux of ^{222}Rn ($\boldsymbol{F_{diff}}$)	0.68 - 213.5 Bq m^{-2} d^{-1}	n.a	Calculated from Equation 9 in supplementary information
Atmospheric flux of ^{222}Rn ($\boldsymbol{F_{atm}}$)	0.7 - 213.5 Bq m^{-2} d^{-1}	n.a	Calculated from Equation 1 in supplementary information
Groundwater flux of ^{222}Rn ($\boldsymbol{F_{gw}}$)	14.7 - 349.8 Bq m^{-2} d^{-1}	n.a	Calculated from Equation 1
Inventory of ^{222}Rn ($\boldsymbol{I}$)	Bq m^{-2}	n.a	Measured with RAD7 AQUA
Groundwater discharge ($\boldsymbol{Q_{gw}}$)	10.3 $\pm$8.2 (3.5-38.6) mm d^{-1}	n.a	Calculated from Equation 1

Table 2 Input parameters for the three endmember model of Ximen Co Lake

Input parameter	Description	Values (using ^{18}O as a tracer)	Parametric sources
h	Relatively humidity	0.63	Measured by the humidity meter
T (°C)	Water temperature	15.66	Monitored with divers
$\delta_{surface}$ (^{18}O) ‰	Surface water isotopic compositions	-12.45	Average value of surface inflow samples
δ_{gw} (^{18}O) ‰	Groundwater isotopic compositions	-11.97	Average value of porewater samples
δ_L (^{18}O) ‰	Lake water isotopic compositions	-12.54	Average value of Ximen Co Lake water samples
F_{gw} (mm/d)	LGD rates	14.18	Calculated based on ^{222}Rn mass balance model
$\varepsilon*$ (^{18}O) ‰	Effective equilibrium isotopic enrichment factor	10.12	Equations 13-14 in supplementary information
C_k (^{18}O) ‰	Kinetic constant for ^{18}O	14.2	Constants based on evaporating experiment
ε_k (^{18}O) ‰	Kinetic enrichment factor	5.2	From Equation 15 in supplementary information
ε (^{18}O) ‰	Total isotopic enrichment factor	15.33	The sum of $\varepsilon*$and C_k
$\alpha*$ (^{18}O) ‰	Effective isotopic equilibrium factor	1.01	$\alpha*=1+\varepsilon*$
δ_a (^{18}O) ‰	Isotopic composition of ambient air	-23.12	Estimated with δ_{in} and δ_a
δ_{in} (^{18}O) ‰	Isotopic composition of surface inflow water	-13.41	Average value of surface inflow water
δ_E (^{18}O) ‰	Isotopic compositions of evaporating vapor	-35.1	From Equation 12 in supplementary information
$[Cl^-]_{in}$ (mgL^{-1})	Chloride concentrations in surface inflow water	0.91	Filed data
$[Cl^-]_L$ (mgL^{-1})	Chloride concentrations in lake water	1.02	Filed data
$[Cl^-]_{gw}$ (mgL^{-1})	Chloride concentrations in groundwater	1.48	Filed data