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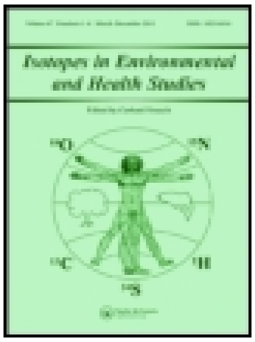

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Hydrogeochemistry and $\delta^{13}C_{DIC}$ and $\delta^{18}O_{H2O}$ composition of three Chinese Tibetan Plateau lakes

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ABSTRACT
This study established the hydrochemistry and stable isotope variations in three lakes, namely brackish Zigetang Co, freshwater Cona and Ranwu lakes located in the central and southeastern Tibetan Plateau of China. Vertical profile fluctuations in the water column, such as temperature and dissolved oxygen (DO) concentration, displayed significant differences which were linked to the thermocline. The hydrochemistry of the three lakes showed that HCO_3^- as the dominant anion, whereas Na^+ is the main cation in Zigetang Co and Cona lake and Ca^{2+} is the prevailing cation in Ranwu lake. In Zigetang Co, K^+ and Na^+ concentrations decreased by 42 % from 1999 to 2012, caused by the enlargement of the lake area, documented by field investigations carried out in 1998, 2002, 2006 and 2012. The $\delta^{13}C_{DIC}$ and $\delta^{18}O_{H2O}$ values analysed from the three lakes varied from −6.0 to 2.0 ‰, and from −14.8 to −6.4 ‰, respectively. The closed Zigetang lake showed higher $\delta^{13}C_{DIC}$ and $\delta^{18}O_{H2O}$ values compared to those of the rivers, the semi-closed Cona and open Ranwu lakes. The $\delta^{13}C_{DIC}$ values of lake water in Zigetang Co were mainly controlled by CO_2 exchange between lake water and atmosphere; the $\delta^{18}O_{H2O}$ values were dominated by the evaporation/freshwater input ratios.

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1. Introduction

The Tibetan Plateau, known as 'the roof of the world' and 'the third pole of the earth', is very sensitive to global climatic change because of its sensitivity to climate fluctuations [1–4]. There are thousands of lakes on the Tibetan Plateau, most of them are endorheic [5], forming more than 45 % of all Chinese lakes and covering an area of 36,899 km^2. The accelerated retreat of glaciers brought by climate warming led to an increase in lake areas [6,7]. Although several lakes on the plateau are remnants of late Pleistocene great lakes, each has its own evolutionary history related to their different hydrologic, ecologic and hydrochemical settings [8]. Tibetan lakes have recently become a hotspot in paleoclimate research because they are seldom influenced by human

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activity, therefore they can represent natural water bodies. The remoteness and inaccessibility of the Tibetan Plateau did not prevent the investigations of several lakes [5,9–11]. Water chemistry studies have been conducted on various water bodies located on the southern side of the Himalayas [12–15]. However, only preliminary and sporadic information on water chemistry is available for most Tibetan lakes and rivers [16–18].

Stable isotope geochemistry was first used to monitor climate change with an accurate recording of palaeoenvironment variations [19,20]. In lacustrine environments, shifts in δ^2H and $\delta^{18}O_{H2O}$ values are influenced by variations in the water source, topography, precipitation, and by hydrological and climatic processes [21]. The climate of central Asia is dominated by the Asian monsoon. The Tibetan Plateau plays a major role in the Northern Hemisphere atmospheric circulation, having a particular influence upon the Indian monsoon [22]. The summer monsoon across the Tibetan Plateau (Qinghai–Xizang) has a major impact on the precipitation gradient resulting in different lake salinities [23]. Evaporation is also an important process that regulates the chemical and isotopic composition of lake waters [24].

The carbon isotope composition of dissolved inorganic carbon (DIC) (i.e. $\delta^{13}C_{DIC}$ values) is an important tool to describe the carbon cycle in a lake system. Factors such as the isotopic compositions of the inflowing water, the CO_2 gas exchange rate between atmospheric and lake waters, and the photosynthesis and respiration of aquatic plants within the lake influence variations in $\delta^{13}C_{DIC}$ values and play an essential role in palaeolimnological studies [25–28]. During the lake closing process, the waters can interact with atmospheric CO_2 after a long residence time producing heavier $\delta^{13}C_{DIC}$ in the lake water [29]. Most northwestern China lakes are closed and semi-closed, producing lake carbonate carbon isotope values varying from −2 to +4 ‰ [28]. In the southeastern Tibet area, terrestrial plants surround the lakes, whereas there is little vegetation cover in the central and northwestern Tibetan Plateau. A high vegetation cover increases the oxidizing ability in lake waters, which can lower the $\delta^{13}C_{DIC}$ values because ^{12}C is preferably consumed during oxidation of organic matter. Of these factors, the biological productivity and oxidizing ability are important processes controlling the DIC composition of total dissolved carbonate in lakes [30].

In arid and semi-arid regions, there is a direct link between climate and surface water hydrology. The water level and chemistry of closed lakes vary in response to climatic variations [23]. In recent years, the Chinese Academy of Sciences has completed a series of systematic hydrochemical investigations on the Qinghai Lake, Pumo Yum Co, Nam Co and Serling Co lakes located on the Tibetan Plateau [3,14,16,17,31–33]. Until now, the significance of the stable isotopic compositions of different lake types has been debated. Hence, the main objectives of our study are: (i) to investigate the vertical distribution of major ions, the physicochemical properties of the water column and to provide basic information for subsequent research, (ii) to analyse the variations in stable isotope compositions (i.e. $\delta^{18}O_{H2O}$ and $\delta^{13}C_{DIC}$) of the water column and the inflowing river water, (iii) to discuss the spatial distribution and compare the $\delta^{18}O_{H2O}$ and $\delta^{13}C_{DIC}$ values among the three lakes, and (iv) to elucidate the response of the Tibetan Plateau lakes to climate change.

2. Site of study and methods of analysis

Zigetang Co is a closed lake located in the central Tibetan Plateau, while Cona lake is a semi-closed lake located in the same area. Ranwu lake is an open lake situated in the southeast Tibetan Plateau (Table 1). To improve our understanding of the lake systems, and to understand the implications of various proxy indicators obtained in the lakes, we selected three different lake types having different water input from specific regions of the Tibetan Plateau.

Zigetang Co (altitude 4561 m a.s.l.; in the Tibetan language, Co means lake) is located in the central Tibetan Plateau (Figure 1), between lat. 32°00′–32°09′ N, and long. 90°44′–90° 57′ E. Zigetang Co lies in the Dongqiao Basin in the southern Tanggula Mountains and is situated in the Amdo County of the Xizang Autonomous Region. This lake was identified as a meromictic saline endorheic lake covering an area of 191.4 km^2 [8]. Previous analyses revealed that the special topography, the shape of the lake basin and several freshwater inflows played an important role in the formation of meromixis in Zigetang Co [8]. There are several river inflows and springs at the southwestern edge of the lake, and the vegetation surrounding the lake primarily consists of alpine grasslands and meadows [34]. Meteorological records indicate an average annual temperature range from −2.0 to −1.0 °C in the basin. Precipitation (300–400 mm a^{-1}) occurs mainly during the summer, and the surface water salinity is 41.1 g l^{-1} [18,34].

Like Zigetang Co, Cona lake (altitude 4588 m a.s.l.) is also located in the central Tibetan Plateau, between lat. 31°55′–32°08′ N and long. 91°25′–91°33′ E, with a water surface area of 182.6 km^2. The lake is situated in a Quaternary basin associated with faulting and forms the upstream watershed to the Nujiang River system. The Cona lake region is characterized by a cold semi-arid climate influenced by the Indian monsoon, and the vegetation surrounding the lake is constituted of alpine grassland. The average annual temperature in this region ranges from −3.3 to −0.9 °C, and the annual precipitation is 400–500 mm. The lake water supply depends significantly on precipitation and melt water, and the surface water salinity is 0.97 g l^{-1} [35].

Ranwu lake (altitude 3850 m a.s.l.) is situated in the southeastern region of the Tibetan Plateau between lat. 29°00′–29°50′ N and long. 96°30′–97°10′ E. It is a barrier lake formed by a mountain landslide producing mud and rock flows choking the river. Ranwu lake, the largest lake in the eastern Tibetan Plateau, is located in a subtropical climatic zone

Table 1. General geographical and limnological properties of Zigetang Co, Cona and Ranwu lakes in the Tibetan Plateau.

	Zigetang Co	Cona lake	Ranwu lake
Latitude (°N)	32°00′–32°09′	31°55′–32°08′	29°00′–29°50′
Longitude (°E)	90°44′–90°57′	91°25′–91°33′	96°30–97°10′
Altitude (m a.s.l.)	4561	4588	3850
Lake type	Closed	Semi-closed	Open
Water quality	Saline lake	Freshwater lake	Freshwater lake
Surface area (km^2)	191.4	182.6	33.27
Maximal depth (m)	38.9	21.8	14.8
Annual temperature (°C)	−2.0 to −1.0	−3.3 to −0.9	−2.1 to −1.5
Annual precipitation (mm)	300–400	400–500	340–660
pH	10	8.8	8.9
Surface salinity (g l^{-1})	41.1	0.97	0.07

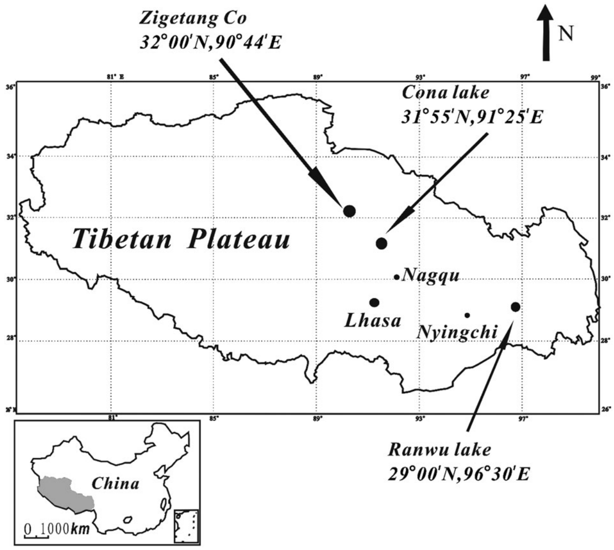

Figure 1. Location of lakes Zigetang Co, Cona and Ranwu in the Tibetan Plateau of China from which water samples were collected.

influenced by the India monsoon, and the vegetation types surrounding the lake are defined by coniferous forest, alpine vegetation and meadows. Ranwu lake is the principal upstream watershed to the Palongzangbu river, which is the tributary of the Brahmaputra river [36]. The lake water supply arrives principally from river inflow, which is mostly generated by precipitation as well as melt water in the southern portion of the lake. The average annual temperature in this region ranges from −2.1 to −1.5 °C, the annual precipitation varies from 340 to 660 mm [37], and the surface water salinity is 0.07 g l^{-1} [38]. Previous analyses revealed increasing atmospheric temperatures with a warming rate of 0.38°C $(10a)^{-1}$ from 1980 to 2005, leading to glacier retreat and a reduction in the glacierized area of the Ranwu lake valley from 496.6 to 466.9 km^2 [36]. Increasing temperatures also resulted in a higher glacier runoff, which contributed to extend the area of the Ranwu lake from 29.8 to 33.3 km^2 [36,37].

During the early stage of formation (35 ka B.P.), the Zigetang and Cona lakes were connected and formed a unique lake. The level of the lake basin was approximately 130–160 m higher than at present [35]. After the Coenozoic uplift of the Tibetan Plateau, the lake was divided and the water level declined gradually due to climate change caused by the uplift [35]. Ranwu lake is the largest lake within the outflow area of the southeast Tibetan Plateau. Glacier melt water influenced directly, the water level, water temperature, conductivity and pH [36].

Water samples from Zigetang Co were taken on 31 July, from Cona lake on 8 August and from Ranwu lake on 15 August 2012. The procedure involved sampling of the lake water column at 3 m deep vertical intervals to water depths of 28 m (Zigetang Co), 21.8 m (Cona lake) and 14.8 m (Ranwu lake), respectively. Water samples from the three lakes were collected from the centre of each lake. We also collected river samples (n = 15) and rainfall samples (n = 5) near the lakes and in the basins. All samples were analysed at the Institute of Geochemistry, Chinese Academy of Sciences.

The lake water temperature, pH, electric conductivity (EC) and concentration of DO were measured using a Yellow Spring Instrument 6600 V2 multi-sensor Sonde (YSI Inc., Yellow Springs, OH, USA; YSI6600). The principal ion concentrations were analysed after the water sample was filtered through a 0.45 µm membrane. Major cation concentrations were determined by an ICP-OES (ICP, Varian Vista Pro, Varian Inc., Palo Alto, CA, USA), and the major anions were analysed by an ion chromatograph (DIONEX ICS-90). All analyses were determined with an analytical error of 5 %. The alkalinity concentration was determined by titration using the hydrochloric acid method. For measurement of stable isotope compositions ($\delta^{13}C$) of DIC, 10 ml aliquots of filtered water samples were injected with a syringe into glass bottles that were filled beforehand with 1 ml 85 % phosphoric acid and magnetic stir bars. The CO_2 was then extracted after cryogenic removal of H_2O using a liquid nitrogen–ethanol trap and subsequently, the CO_2 was transferred cryogenically into a tube for isotope measurement. Inorganic carbon isotope analysis was performed using a Finnigan MAT 252 mass spectrometer (MF/TG-IRMS; IsoPrime; GV Instruments; UK) [39]. The results are expressed in ‰ relative to the V-PDB standard, and duplicate analyses produced an uncertainty of ±0.1 ‰. Stable isotope composition of water, that is, δ^2H and $\delta^{18}O_{H2O}$ were measured using a liquid water isotope analyzer (Model: IWA-35EP; 912-0026; Los Gatos Research, Inc. USA) equipped with a liquid auto sampler for automated injection of ~1 µl water. The precision of the chemical analyses was obtained using repeated measurements of standards. Six injections were needed for each sample and standard, due to the memory effects, whereas the first three measurements were discarded and the average of the last three measurements was used for calculation [40]. The δ^2H and $\delta^{18}O_{H2O}$ values are expressed in ‰ relative to the V-SMOW standard, the analytical precisions of δ^2H and $\delta^{18}O_{H2O}$ were ±0.5 and ±0.2 ‰, respectively.

3. Results

Water column profiles for the lakes Zigetang, Cona and Ranwu illustrate the vertical distribution in major ion concentrations and their physical characteristics (Figures 2 and 3). Water temperatures ranged from 15.4 to 2.3 °C in the Zigetang Co water column with a

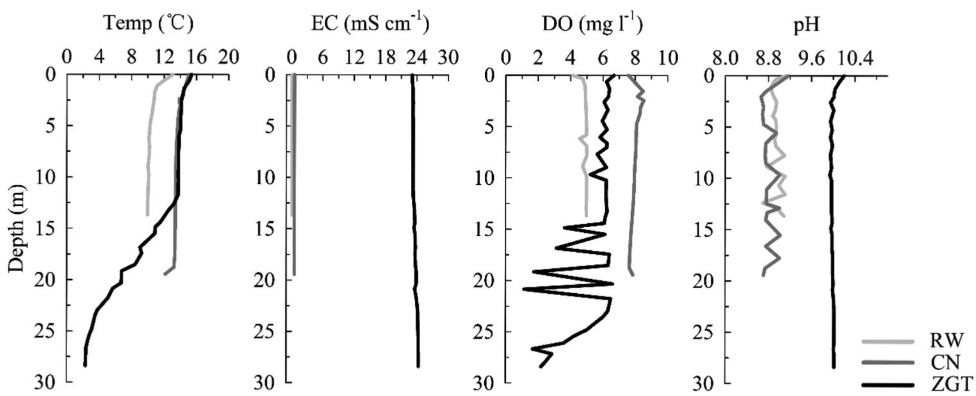

Figure 2. Depth profiles of temperature, EC, DO concentration and pH from lakes Zigetang Co, Cona and Ranwu (August 2012).

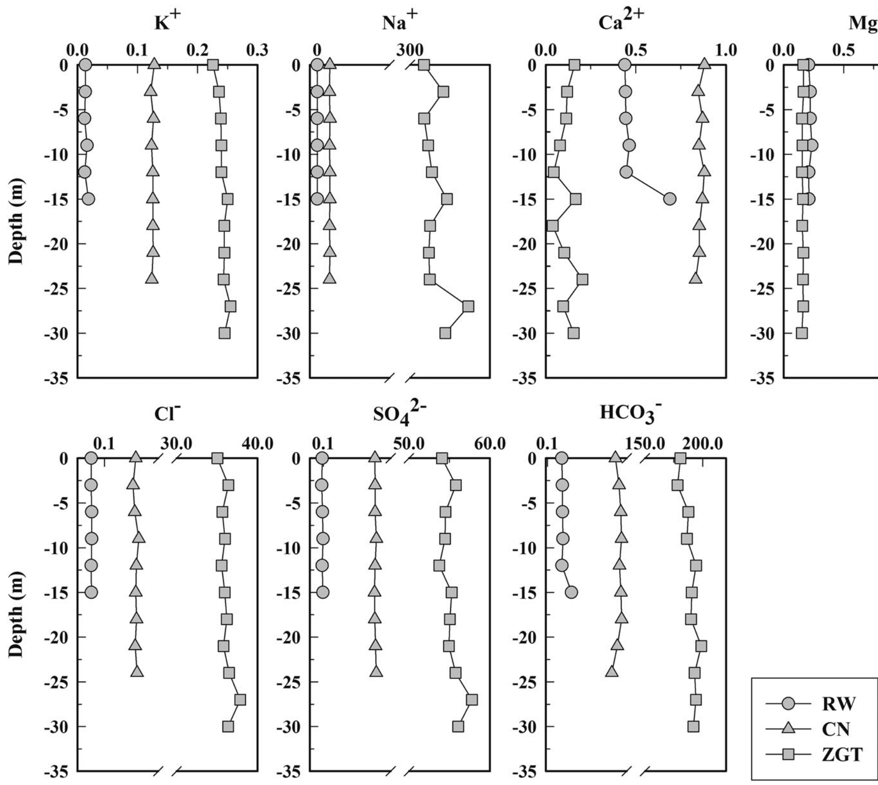

Figure 3. Vertical variation in major ion concentrations (mmol l^{-1}) in the three lake water columns (RW: Ranwu Lake, CN: Cona Lake and ZGT: Zigetang Co).

sharp decrease manifested between the upper and deeper layers. In lakes Cona and Ranwu, water temperatures diminished with depth from 15.1 to 12.2 °C and from 13.1 to 10 °C, respectively. The pH values showed slight variations in each lake water column with averages in lakes Zigetang, Cona and Ranwu of 10, 8.8 and 8.9, respectively. The value of DO concentration ranged from 6.7 to 1.1 mg l^{-1} in Zigetang Co, while it varied only slightly in lakes Cona and Ranwu with average values of 7.9 and 4.9 mg l^{-1}, respectively.

The concentrations of the principal ions including K^+, Na^+, Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-}, HCO_3^- and CO_3^{2-} are presented in Table 2. For the Zigetang Co lake water, the concentrations of

Table 2. Mean values of major cation and anion concentrations in surface Tibetan Plateau lake waters and their watersheds (mmol l^{-1}).

Water	K^+	Na^+	Mg^{2+}	Ca^{2+}	Cl^-	SO_4^{2-}	$HCO_3^- + CO_3^{2-}$
RW L	0.01	0.03	0.21	0.44	–	0.09	1.00
CN L	0.13	1.77	1.21	0.88	0.33	0.48	4.30
ZGT L	11.27	308.64	8.19	0.16	35.03	54.08	180.77
RW R	0.02	0.03	0.15	0.65	–	14.9	1.63
CN R	0.17	2.14	0.89	1.35	0.37	0.16	5.94
ZGT R	0.04	0.71	1.30	1.43	0.16	0.47	3.78
ZGT L[a]	19.39	538.69	5	0.23	42.82	85.04	298.33

Note: *L*, lake water; *R*, river water.
[a]Reported by Li et al. (2001).

cations were, in decreasing order: $Na^+ > K^+ > Mg^{2+} > Ca^{2+}$; with averages of 308.64, 11.27, 8.19 and 0.16 mmol l^{-1}, respectively. A similar order for anions leads to $HCO_3^- > SO_4^{2-} > Cl^-$, with averages of 180.77, 54.08 and 35.03 mmol l^{-1}, respectively. For the Cona lake water, cation concentrations in decreasing order were: $Na^+ > Mg^{2+} > Ca^{2+} > K^+$; with averages of 1.77, 1.21, 0.88 and 0.13 mmol l^{-1}, respectively. Anion concentrations decreased following $HCO_3^- > SO_4^{2-} > Cl^-$, with averages of 4.30, 0.48 and 0.33 mmol l^{-1}, respectively. For the Ranwu lake water cations, concentrations decreased according to the following order: $Ca^{2+} > Mg^{2+} > Na^+ > K^+$; with averages of 0.44, 0.21, 0.03 and 0.01 mmol l^{-1}, respectively. The decreasing anion concentrations followed $HCO_3^- > SO_4^{2-} > Cl^-$, with averages of 1.00, 0.09 and 0 mmol l^{-1}, respectively.

The mean $\delta^{18}O_{H2O}$ values of the monsoon rain near Ranwu, Cona and Zigetang Co were −13.0, −18.4 and −20.4 ‰, respectively; and the δ^2H values reached −103, −146 and −152 ‰, respectively. The mean $\delta^{18}O_{H2O}$ values of lake water in Ranwu lake, Cona lake and Zigetang Co were −14.8, −9.9 and −6.4 ‰, respectively; with δ^2H values of −107, −85 and −69 ‰, respectively. The mean $\delta^{18}O_{H2O}$ values of river water near Ranwu lake, Cona lake and Zigetang Co were −14.6, −14.1 and −14.5 ‰, respectively; and the δ^2H value attained −105, −116 and −115 ‰, respectively (Table 3). The $\delta^{13}C_{DIC}$ value of Ranwu lake surface water was −6.0 ‰, while reaching −6.7 ‰ at the bottom of the water column. The $\delta^{13}C_{DIC}$ values from the Cona lake and Zigetang Co water columns varied from 0.7 to 0.9 ‰ and from 2.0 to 1.9 ‰, respectively (Figure 6).

4. Discussion

4.1. Vertical distribution of the physicochemical characteristics of three Tibetan plateau lakes

Physicochemical parameters including temperature, EC, DO concentration, and pH in the lake water column of the Zigetang Co, Cona lake and Ranwu lake were observed from July to August 2012 (Figure 2). Temperature is an important physical factor, which affects other parameters such as pH, EC, dissolved gases like CO_2 and especially DO concentrations [41]. In the upper lake water layer, regular mixing of water results in minor changes in DO concentrations of the three studied lakes. But in the middle stratum – the thermocline layer – of Zigetang Co, we observed an ‘S’ curve with a rapid depletion in deeper layer reaching 0 mg l^{-1} at 22 m depth. The temperature profile showed a strong decrease at the thermocline layer from 15 to 25 m accompanied by a temperature gradient of 1.9 °C m^{-1}, while the temperature is nearly constant at depths <12.5 m. Moreover, the temperature was slowly reduced in the deeper layer, and reached 2.3 °C in the hypolimnion, although no obvious changes were observed in EC and pH values. The temperature profiles and DO

Table 3. Mean values of δ^2H, $\delta^{18}O_{H2O}$ and $\delta^{13}C_{DIC}$ in the three studied Tibetan Plateau lake water, rivers and associated rainfall.

Sample	RW lake	CN lake	ZGT lake	RW river	CN river	ZGT river	RW rain	CN rain	ZGT rain
δ^2H	−107	−85	−69	−105	−116	−115	−103	−146	−152
$\delta^{18}O_{H2O}$	−14.8	−9.9	−6.4	−14.6	−14.1	−14.5	−13.0	−18.4	−20.4
d-excess	11.1	−6.4	−15.9	12.3	−3.6	−2.4	1.6	0.5	10.8
$\delta^{13}C_{DIC}$	−6.4	0.8	2.0	−5.3	–	−6.1	–	–	–

concentrations for Zigetang Co exhibited the typical characteristics of most meromictic lakes. Previous studies [8,42,43] revealed a reduced inflow of fresh water and a sheltering due to the morphometry of the lake basin resulting in meromixis in Zigetang Co. The water temperature in the Cona lake water column showed no obvious change with depth. But the temperature in the Ranwu lake decreased rapidly from the lake surface to a depth of 1 m, then decreased slowly with increasing depth, reaching 10 °C in the bottom layer.

4.2. Major element water compositions of the three Tibetan Plateau lakes

Major ion concentrations show no vertical variation in the depth profiles for the three lakes (Figure 3). Water chemistry differs from each lake and it is necessary to investigate each one in detail [33]. The global rainfall DIC average concentration was 0.1 mmol l^{-1}, the averaged HCO_3^- concentration of rainfall in Zigetang Co (1.63 mmol l^{-1}) is significantly higher than the global rainfall value [44]. The HCO_3^- average concentration of rainfall in Cona lake (0.1 mmol l^{-1}) is similar to that of the global DIC value, whereas the Ranwu lake concentration is lower [44].

The high pH value in the lake water column indicates the basic composition of the water. The high alkalinity is caused by high carbonate concentrations, which are corroborated by the major element water composition of the lake water column. There are significant variations in the ratio of major ion concentrations that are related to differences in the geological setting. For Zigetang Co lake water, the dominant ions Na^+ and HCO_3^- account for 94.02 and 66.98 % of the total cations and anions, respectively, whereas Ca^{2+} was the principal cation in Ranwu lake waters (Table 2). Zigetang Co displayed much higher ion concentrations compared to the two other lakes (Figure 3).

Contrary to lake waters, the inflowing river waters to Zigetang Co and Ranwu lake belong to the Ca–Mg–HCO_3 type, whereas the river water flowing into Cona lake is of the Na–Ca–HCO_3 type (Table 2). Ca^{2+} concentrations in river water were higher than those measured in the lake water. This indicates the Ca^{2+} precipitated during lake sediment deposition. The biogeochemical activity could result in precipitation of Ca^{2+} as $CaCO_3$, which is a common phenomenon in the evolution of carbonate lake [18].

The major ion concentrations of the inflowing river water were lower than those of lake water. The river discharging into Ranwu lake is fed by melt water, with inherent lower concentrations of major ions. The differences in major ion concentrations presumably resulted from scant precipitation, variations in evaporation rate and the input of melt water in the lake. For instance, evaporation would generate enrichment in major ions in the lake waters, such as in the Zigetang Co water samples. Moreover, low pH values (pH = 8.8 for Cona lake and pH = 8.9 for Ranwu lake) are responsible for lower $HCO_3^- + CO_3^{2-}$ concentrations (e.g. 4.30 and 1.00 mmol l^-, respectively) relative to that of Zigetang Co water showing a very high value (180.77 mmol l^-) at a pH = 10. Higher pH values promote the conversion of gaseous CO_2 into $HCO_3^- + CO_3^{2-}$ relative to lower pH [45].

The study of endorheic lakes in closed basins of the remote Tibetan Plateau is important to the understanding of the water balance, but also provides a valuable indicator of climate change which is sensitive to variations in precipitation, evapotranspiration and glacier melting [6,46,47]. Recent investigations reveal an augmentation in the number of new lakes since the 1990s. The areas of the larger Tibetan Plateau lakes began to expand during a warmer climate period (i.e. in the 1960s), such as in Nam Co. Climate warming has been accompanied by an accelerated retreat of glaciers, and the lake area increased from 1952.5 km^2 in 1976 to 1976.9 km^2 in 2001 [6,7,14,32,48]. Ranwu lake, which was mainly supplied by meltwater, has grown by the contribution of water from retreating glaciers [36]. Little information is available on Zigetang Co, which is not supplied by glacial meltwater. However, both temperature and precipitation data collected from nearby meteorological stations showed an obvious rise since the 1970s, leading to a lake expansion of more than 20 km^2 [49]. Expansion of two steps of terrace in Zigetang Co was observed during field investigations carried out in 1998 [8], 2002 [50], 2006 [46] and 2012 (Supplemental Figure). Moreover, the concentrations of K^+ and Na^+ decreased 42 % in Zigetang Co lake water from 1999 to 2012 (Table 2). We conclude that the Tibetan Plateau lakes are highly responsive to global warming and that climatic variations have a major impact on lake levels and runoffs in this watershed.

4.3. The H and O isotope characteristics of Tibetan plateau lakes

The isotopic compositions of water, which are affected by meteorological processes, can provide a characteristic fingerprint of their origin. The δ^2H and $\delta^{18}O_{H2O}$ values of the Ranwu lake water remained low relative to those of river and rainfall (Table 3). These comparable stable isotope values could be attributed to the lake water supply which is mainly dependent on rivers fed by melting ice and snow, and effective precipitation. The isotopic composition of several rivers is a weighted average of the isotopic composition of rain water falling over their catchment [51]. Therefore, the oxygen isotopic values of hydrologically open, short-residence lakes such as Ranwu can be used as a signature tracing the composition of regional rain water.

Around Cona lake, the mean δ^2H and $\delta^{18}O_{H2O}$ values obtained from the monsoon rain were −146 and −18.4 ‰, while the water from Cona lake inflow river displayed values of −116 and −14.1 ‰, and the lake water values reached −85 and −9.9 ‰. These are slightly lower values to those analysed by Liu et al. [52] in Cona lake from 1999 to 2001. The mean δ^2H and $\delta^{18}O_{H2O}$ values measured in lake water for those years were −83 and −9.6 ‰; and −125 and −16.6 ‰ for the rain water. The oxygen isotope composition of lake waters ($\delta^{18}O_{H2O}$) is predominantly controlled by local precipitation (rain and snowfall), inflowing water, groundwater, evaporation and lake water residence times [53–55].

The $\delta^{18}O_{H2O}$ values of the Zigetang Co water samples are higher to those of Ranwu lake, and there is a similar tendency for Cona lake water. The $\delta^{18}O_{H2O}$ values did not show vertical variation in the depth profiles of the three lakes (Figure 4). Since evaporation induces fractionation of oxygen isotopes, especially in arid areas, the $\delta^{18}O_{H2O}$ values and salinity measured in closed lake water are usually higher to those of ambient water sources. Talbot [53] indicated that the value of $\delta^{18}O_{H2O}$ is related to the rate of lake water residence time to evaporation, because evaporation preferentially removes $H_2^{16}O$, enriching the remaining lake water with $H_2^{18}O$. A previous study has shown that higher evaporation/precipitation values prevail in northwestern China relative to northeastern China [28]. The monsoon rain $\delta^{18}O_{H2O}$ value near Zigetang Co was −20.4 ‰, whereas the inflowing river water value was −14.6 ‰ compared to the relatively less negative $\delta^{18}O_{H2O}$ value of −6.4 ‰ in Zigetang Co waters. These results indicate the importance of evaporation in this catchment. Since Zigetang Co is a closed lake, the lake water isotope composition depends on the evaporation and precipitation processes. Precipitation (300–400 mm a^{-1})

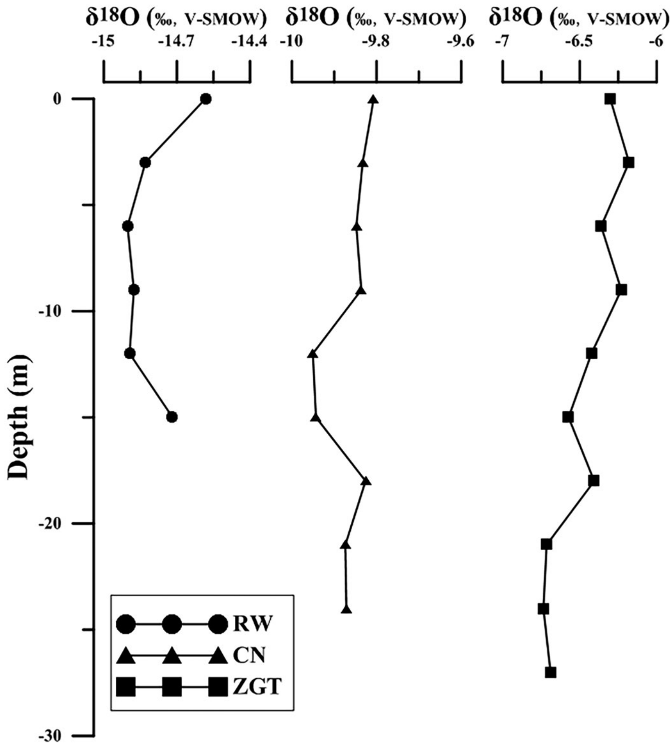

Figure 4. Variations in $\delta^{18}O_{H2O}$ values with depth in three studied lake waters.

occurred mainly during summer, while at the same time evaporation (791–1111 mm a^{-1}) was greater than precipitations [56], resulting in more positive $\delta^{18}O_{H2O}$ values of the lake water relative to those of the inflow river and precipitations.

The lake water value of Ranwu lake located in the southeast of the Tibetan Plateau was −14.8 ‰, while for the Cona lake and Zigetang Co, which are located in the central of Tibetan Plateau, $\delta^{18}O_{H2O}$ reached −9.9 and −6.4 ‰, respectively (Table 3). Zigetang Co, which is situated at an altitude of 4561 m a.s.l. in the central Tibetan Plateau presents higher $\delta^{18}O_{H2O}$ values than the Ranwu lake, located at an elevation of 3850 m a.s.l. in the southeast Tibetan Plateau. The variations in $\delta^{18}O_{H2O}$ are attributed to differences in altitude, spatial distribution and degree of evaporation [57–59]. The $\delta^{18}O_{H2O}$ values of rain water near the lakes Ranwu, Cona and Zigetang Co are −13.0, −18.4 and −20.4 ‰, respectively. Such isotopic variations resulted from the effect of altitude, that is, the $\delta^{18}O_{H2O}$ values of rainwater decrease with increasing altitude [57,58]. The effect of altitude is primarily attributed to the Rayleigh distillation process and to depleted heavy-isotope content (low $\delta^{18}O_{H2O}$ and $\delta^{2}H$ values), resulting from vapour formation and precipitation as an air mass moves over an orographic barrier and precipitates rain [58]. The $\delta^{18}O_{H2O}$ composition of river waters near the three Tibetan Plateau lakes is similar since the source of river water is comparable, corresponding to snowmelt from mountain glaciers located on the plateau. Since evaporation occurs generally at higher altitudes in northwestern China than in the northeastern sector, the $\delta^{18}O_{H2O}$ values of Zigetang Co and Cona lake waters are higher than those of Ranwu lake. The more positive $\delta^{18}O_{H2O}$ values of

Zigetang Co relative to those of Cona lake may possibly be explained by a longer water residence time and a greater evaporation rate in the former.

The distribution of stable isotopes in surface and rainwater produces a linear correlation between δ^2H and $\delta^{18}O_{H2O}$ values. This average relation for the global meteoric water line (GMWL) is expressed by a universal equation: $\delta^2H = 8\delta^{18}O + 10$ [60]. Isotopic data from the three lakes illustrate the composition of rain and lake water relative to the GMWL in δ^2H–$\delta^{18}O$ space (Figure 5). The rainwater, Zigetang Co and Cona lake water isotopic values plot below GMWL and are shifted toward heavier isotopic values relative to those expressed by the river and Ranwu lake waters. The location of Zigetang Co and Cona lake in δ^2H–$\delta^{18}O$ space is highly unusual, indicating the presence of evaporated moisture, and reflecting the correlations in isotopic values between precipitation and lake water at different latitudes. The δ^2H and $\delta^{18}O_{H2O}$ values of rainwater commonly form linear clusters in the δ^2H–$\delta^{18}O$ space close to the GMWL (Figure 5). Such clusters could be a useful indicator to describe the local isotopic input functions for hydrologic studies.

The deuterium excess (*d*-excess) is expressed by the equation: $d = \delta^2H - 8\,\delta^{18}O$ [61]. The mean values of *d*-excess in Ranwu lake water was 11.1 ‰, different from the mean value of −6.4 and −15.9 ‰ obtained for Cona and Zigetang Co lake water, respectively. Negative *d*-excess values also imply evaporation of lake water affected the isotopic compositions [62]. The *d*-excess average values in Ranwu lake river water was 12.3 ‰, again different from the average value of −3.6 and −2.4 ‰ analysed for the Cona lake and Zigetang Co river water, respectively. The rainwater *d*-excess values near the Ranwu, Cona lake and

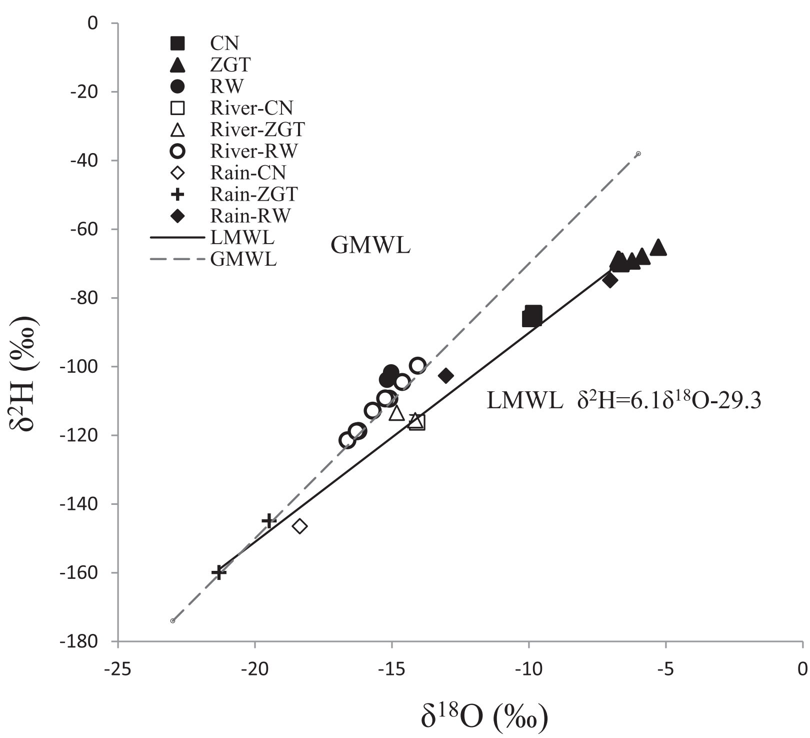

Figure 5. Stable isotope plot of δ^2H–$\delta^{18}O$ showing the isotopic signatures of rainwater, lake water and river water relative to the position of the mean GMWL.

Zigetang Co were 1.6, 0.5 and 10.8 ‰, respectively. The results suggest the isotopic compositions of lakes could be influenced by numerous factors, such as glacial melt and different water masses from river, local precipitation and evaporation effects.

4.4. Characteristic changes in the carbon isotope composition of dissolved inorganic carbon

The $\delta^{13}C$ values of lake water DIC are typically influenced by the isotopic composition of inflowing waters, exchange with atmospheric CO_2 and the photosynthesis/respiration of aquatic plants within the lakes [63–65]. The surface water $\delta^{13}C_{DIC}$ value of Ranwu lake was −6.0 ‰, while it reached −6.7 ‰ at the bottom of the water column (Figure 6). Such stable isotope variation in Ranwu lake can be related to the vegetation present in the southeastern Tibetan Plateau. This area is mountainous and characterized by high boreal forest coverage with many aquatic and terrestrial plants in and surrounding the lakes that could modify, along with a short-residence time, the lighter carbon isotope

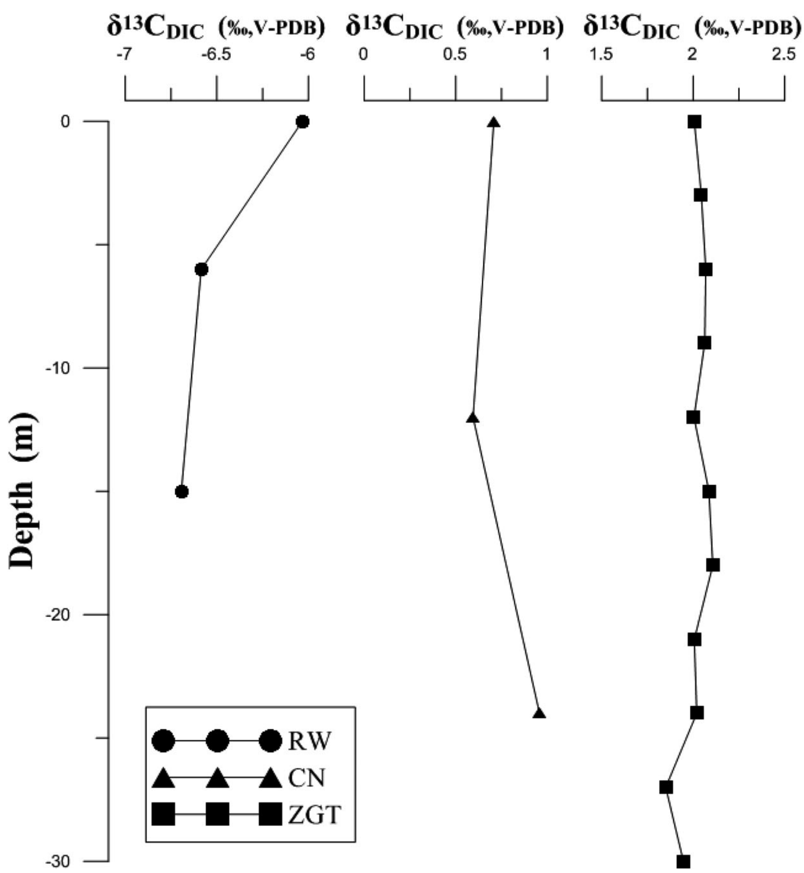

Figure 6. $\delta^{13}C_{DIC}$ in lake waters of the three Tibetan Plateau lakes (Zigetang Co, Cona and Ranwu) investigated in this study. Note only three samples were measured for the upper, middle and deeper layers in Ranwu and Cona lakes because there was no significant variation in the $\delta^{18}O_{H2O}$ values observed in the depth profiles of these two lakes.

values of Ranwu lake. Plant photosynthesis will transform the inorganic carbon into organic ^{12}C absorbed initially by the vegetation. This will enrich the lake waters with heavy isotopes while the light isotopes escape as CO_2 in gas form. Oxidation of lake waters can also change the carbon isotope composition of highly organic shallow lakes. Decomposition of organic matter in deep water can lead to negative $\delta^{13}C_{DIC}$ values due to local availability of ^{12}C. Therefore, the photosynthesis and respiration of aquatic plants within the lake is considered to be the most important controlling factor for the Ranwu open lake.

$\delta^{13}C_{DIC}$ values from the Cona lake and Zigetang Co water columns varied from 0.7 to 0.9 ‰ and from 2.0 to 1.9 ‰, respectively (Figure 6). The results showed little isotopic variation from the surface to the bottom. The $\delta^{13}C_{DIC}$ value (−6.1 ‰) of Zigetang Co river water was lower to that of Zigetang Co lake water. Zigetang Co is a closed lake; therefore, the long residence time of DIC causes gaseous CO_2 enriched in ^{12}C to escape from the lake surface water while the heavy isotope ^{13}C can stay in the water because of the very high evaporation rate. Such a process could lead to raise the contents of ^{13}C. Thus, with a low input of aquatic and terrestrial plant and a long DIC residence time, heavier $\delta^{13}C_{DIC}$ values were observed in closed and semi-closed lakes. The $\delta^{13}C_{DIC}$ values of the Zigetang Co water were significantly higher to those of inflowing rivers in Zigetang Co, but there was no significant difference between Ranwu lake and river water flowing into it (Table 3). Finally, DIC exchange with atmospheric CO_2 could increase the $\delta^{13}C_{DIC}$ values, whereas the residence time of lake waters might be the key factor for controlling the $\delta^{13}C_{DIC}$ values due to its large differences among those lakes.

5. Conclusions

This paper presents limnological investigations (including water chemistry and $\delta^{13}C_{DIC}$ and $\delta^{18}O_{H2O}$ stable isotopes) of three different types of lacustrine environments: closed (Zigetang), semi-closed (Cona) and open type (Ranwu) lakes located in the Tibetan Plateau of China.

The physicochemical characteristics of the water column in lakes Zigetang, Cona and Ranwu demonstrate that the temperature, DO concentration and pH vary periodically with water depth, whereas the concentrations of major ions display no evident vertical variation. Low concentrations of Ca^{2+} in the lake water column suggest $CaCO_3$ precipitation in the lakes. K^+ and Na^+ concentrations of Zigetang Co lake waters decreased by 42 % from 1999 to 2012, whereas the water level has risen several metres in recent years, as shown by previous studies.

$\delta^{13}C_{DIC}$ and $\delta^{18}O_{H2O}$ values of the Zigetang Co closed lake are enriched relative to that of the semi-closed lake (Cona) and open type lake (Ranwu). This is attributed to the high evaporation rate and longer water residence time occurring in the closed lakes. The differences in the $\delta^{13}C_{DIC}$ and $\delta^{18}O_{H2O}$ values in lake water and inflowing river waters are useful to estimate the source of input water. Saline water displays higher isotopic compositions than freshwater. $\delta^{13}C_{DIC}$ and $\delta^{18}O_{H2O}$ values, ion concentrations and the increase in the lake water level are useful tools to study the hydrological processes in lakes located in the Tibetan Plateau.

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Disclosure statement

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