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A SURVEY OF THE ^{14}C CONTENT OF DISSOLVED INORGANIC CARBON IN CHINESE LAKES

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ABSTRACT. We present radiocarbon (^{14}C) measurements of dissolved inorganic carbon (DIC) from surface waters of 11 lakes, widely distributed in China. Surface lake water DIC $F^{14}C$ values show distinct differences, and we relate these to the physical exchange character ("open" or "closed") of each lake. Open lakes studied here generally have lower DIC $F^{14}C$ values than closed lakes. We present a simple model of a lake water cycle to calculate an average residence time for each lake. Comparisons between lake DIC $F^{14}C$ and average residence time shows that the DIC $F^{14}C$ increases with the average residence time and reflects a steady-state.

KEYWORDS: average residence time, dissolved inorganic carbon, radiocarbon.

INTRODUCTION

The freshwater reservoir effect (FRE) is the radiocarbon (^{14}C) age difference between the atmosphere and a contemporaneous freshwater system (Godwin 1951). FRE values range from hundreds to thousands of years in different lakes and can change with time (Wu et al. 2010, 2011; Long et al. 2011; Ascough et al. 2012; Keaveney and Reimer 2012; Zhang et al. 2012; Mischke et al. 2013; Wang et al. 2013a; Yu et al. 2014; Zhou et al. 2014, 2015; Lockot et al. 2015). Dissolved inorganic carbon (DIC) is an important component of a freshwater system and influences the ^{14}C level of aquatic plants growing there (Deevey et al. 1954; Yu et al. 2007; Olsson et al. 2009). In general, three factors control the ^{14}C level of lake water DIC: (1) the rate and degree of exchange with atmospheric CO_2; (2) geological weathering; and (3) microbial decay of terrigenous organic matter (Hakansson et al. 1979; Bondevik et al. 2006; Cage et al. 2006; Hendy et al. 2006; Kritzberg et al. 2006; Hatté and Jull 2007; Olsson 2009; Kritzberg et al. 2014; Keaveney et al. 2015). DIC ^{14}C levels vary from lake to lake and the content of a particular lake will depend on its individual geographical setting. The investigation of DIC levels from a number of modern lakes is useful in identifying trends in the FRE.

A number of past studies have examined DIC in Chinese lakes. For example, at Qinghai Lake, riverine DIC has been shown to have much lower levels than the overall DIC pool in the lake. This is explained by the rapid carbon exchange that occurs between the lake and atmosphere, driven in part by its high alkalinity (Yu et al. 2007; Jull et al. 2014; Zhou et al. 2014). This effect is also seen in the DIC $\delta^{13}C$ results of different parts of Qinghai Lake and DIC $F^{14}C$ results of other lakes from the Tibetan Plateau. In many cases it has been observed that water from the center of a lake exchanges more completely with atmospheric CO_2 than with water near the shore (Li et al. 2012; Mischke et al. 2013). In a different example, DIC $F^{14}C$ time series from Lake Kinneret in Israel show temporal variations that correspond to flood discharge years, when DIC $F^{14}C$ values are relatively lower than other years (Stiller et al. 2001). Lakes on the Tibetan Plateau can be distinguished as open or closed systems according to their $\delta^{13}C_{DIC}$ characteristics. Open lakes (net water outflow from the lake) have $\delta^{13}C_{DIC}$ values that are similar to inflowing river water, while closed lakes (without water outflow from the lake) show a greater degree of exchange between lake water DIC and the atmosphere (Lei et al. 2011). By extension, the hydrological conditions in a lake can influence DIC $F^{14}C$ values.

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The average residence time of a lake can be expressed as the ratio between lake volume and rate of inflow or outflow in an equilibrium state (Yu et al. 2007; Jull et al. 2014). This quantity depends on the hydrological processes operating in a particular lake, and encompasses both open and closed lakes. Here we examine 11 lakes from different parts of China to examine the relationship between open and closed lakes and their DIC levels, as expressed by their average residence times.

LOCATION DESCRIPTION AND SAMPLING

For this study we sampled seven lakes from different parts of China and we review four additional lakes from the published literature. The lakes we sampled include Tianshuihai, Kushui Lake, Gyaring Co, Nam Co, Chenghai, Xingyun Lake, and Dalinor (Figure 1).

Tianshuihai (79°35′E, 35°27′N) and Kushui Lake (79°21′E, 35°35′N) are located in northwest China, where the climate is dominated by the westerlies, the bedrock is siltstone (Li et al. 1998). Gyaring Co (88°13′E, 31°11′N) and Nam Co (90°36′E, 30°43′N) are located in western China, in the central Tibetan plateau, the bedrock is limestone and volcanic rock, the bedrock of Gyaring Co is limestone (Zhang et al. 2011). Dalinor (116°37′E, 43°17′N) is located in northern China, near the northwestern limit of the modern East Asian monsoon domain, the bedrock is volcanic stone (Figure 1). Chenghai (100°39′E, 26°33′N) and Xingyun Lake (102°46′E, 24°19′N) are in southern China, where the climate is controlled by the Southwest Monsoon, the bedrock is limestone (Xu et al. 2015a,b) (Figure 1). Tianshuihai and Gyaring Co are open lakes (Li et al. 1998); whereas Kushui Lake, Nam Co, Dalinor, Xingyun Lake and Chenghai are closed lakes (Li et al. 1998; Zhang et al. 2011; Dong et al. 2008; Zhang et al. 2008; Hu et al. 1992).

In November 2014 and May to August 2015, we collected surface lake water from the seven lakes. In some cases we were also able to collect submerged aquatic plants. In Dalinor we collected water from the inflowing rivers, groundwater from a spring, and an air sample. All of the water samples were collected in 600-mL brown glass bottles. The sample bottles were cleaned prior to sampling

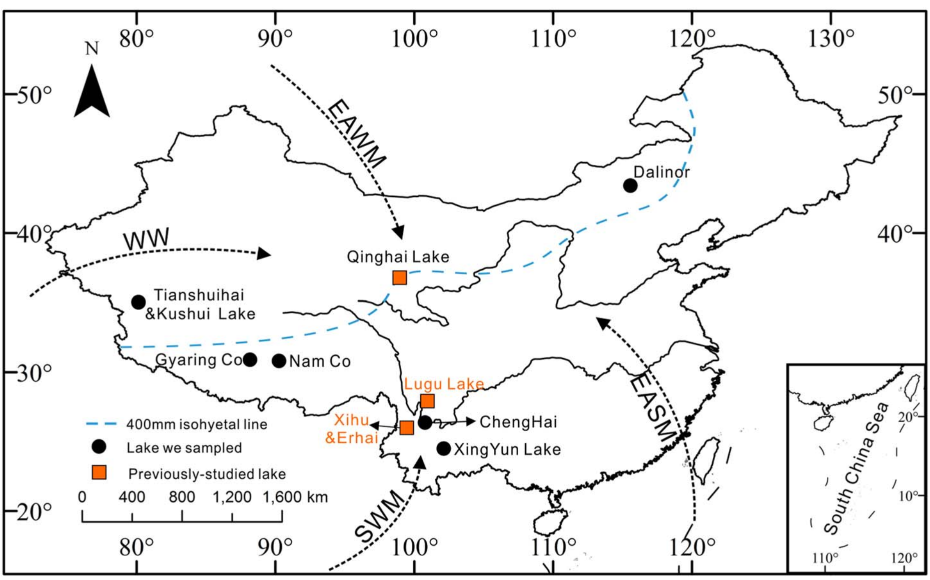

Figure 1 Location of the lakes. The circle represents the lakes we sampled; the rectangle represents previously studied lakes. EASM, EAWM, WW, and SWM are abbreviations for East Asian Summer Monsoon, East Asian Winter Monsoon, Westerly Wind, and Southwest Monsoon, respectively.

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and rinsed with lake water three times before sampling. The air samples were collected in aluminum foil gas sampling bags using an established methodology (Niu et al. 2016).

SAMPLE PREPARATION AND MEASUREMENT

All of the water samples were filtered with 0.7 µm GF/F glass-fiber filters in the laboratory. The filtered samples were placed in a Pyrex® flask and acidifed with 85% H_3PO_4 to liberate CO_2 from the DIC fraction. The GF/F glass-fiber filters and aquatic plants were treated with 1N HCl and then rinsed in distilled water and dried. The dried, pretreated samples were placed into 9-mm quartz tubes, with an appropriate amount of CuO as oxidant. They were then evacuated to $<10^{-5}$ Torr and heated to 850°C. The CO_2 produced by the combustion was divided into two parts; one is used for δ^{13}C measurement and the other was converted catalytically to graphite using a hydrogen reduction method over iron powder for measurement. Measurements were made at the 3 MV HVE Tandetron AMS (Zhou et al. 2006). δ^{13}C measurements were made with an isotope ratio mass spectrometer (MAT-252). All of the analyses were conducted at the Xi'an Institute of Earth Environment, Chinese Academy of Sciences.

The ^{14}C results are quoted as fraction of modern carbon (F^{14}C) values (Donahue et al. 1990), and the ^{13}C results are quoted as δ^{13}C values (V-PDB, ‰).

RESULTS

The results from the inflowing rivers, groundwater and air sample from Dalinor are given in Table 1. In Dalinor, the F^{14}C values of the DIC fraction from four inflowing rivers we sampled range from 0.7752 to 0.9912 (Table 1). The lowest value is from the HaoLai river and the highest is from the ShaLi river. The DIC F^{14}C of groundwater collected from the site is 0.6036. The DIC F^{14}C of surface lake water and atmospheric CO_2 are 1.0421 and 1.0096 respectively (Table 1). Hence, the DIC F^{14}C values of incoming water (river water and groundwater) are systematically lower than those of the surface lake water.

Results for Tianshuihai, Kushui Lake, Gyaring Co, Nam Co, Chenghai, Xingyun Lake, and Dalinor are shown together in Table 2. For comparison, we present published results from several other Chinese lakes in Table 3. We also indicate the open or closed nature of each lake in Tables 2 and 3. The DIC F^{14}C values of the lakes range from 0.2042 to 1.0971; DIC δ^{13}C values range from –2.71‰ to 5.34‰. The F^{14}C values of particulate organic carbon (POC) range from 0.8829 to 0.9557, and the F^{14}C values of aquatic plants in these lakes range from 0.2215 to 1.0971 (Table 2, Table 3). Figure 2 shows that F^{14}C values of aquatic plants were positively correlated with lake water DIC F^{14}C ($R^2 = 0.939$). This correlation shows that the level of aquatic plants is controlled by the level of lake water DIC, as expected. Another interesting observation is that the DIC samples from open lakes (except Kushui Lake) have lower F^{14}C

Table 1 River water, spring water, DIC and air ^{14}C results for Dalinor.

Location	F^{14}C DIC	δ^{13}C(‰) DIC	Alkalinity CO_2 collected mg/L	F^{14}C POC	δ^{13}C (‰) POC
GongGeEr R. water	0.8015 ± 0.0031	–9.93 ± 0.03	13.25	0.8132 ± 0.0024	−30.66 ± 0.19
LiangZi R. water	0.7988 ± 0.0036	–10.50 ± 0.01	13.74	0.9714 ± 0.0032	−33.58 ± 0.21
HaoLai R. water	0.7752 ± 0.0028	–11.80 ± 0.02	19.14	1.0090 ± 0.0031	−29.01 ± 0.25
ShaLi R. water	0.9912 ± 0.0033	0.45 ± 0.01	30.19	0.9887 ± 0.0027	−26.03 ± 0.14
Spring water	0.6036 ± 0.0022	–10.82 ± 0.02	33.33	—	—
Lake water	1.0421 ± 0.0043	–1.06 ± 0.02	34.7	0.8829 ± 0.0024	−27.35 ± 0.03
Air	1.0096 ± 0.0034	–11.64 ± 0.01	—	—	—

Table 2 The fraction modern carbon ($F^{14}C$) values and water residence times of the lakes we studied.

Lake	$F^{14}C$ DIC	$\delta^{13}C$ (‰) DIC	Alkalinity CO_2 collected (mg/L)	$F^{14}C$ POC	$F^{14}C$ submerged plant	pH	Surface area (km^2)	Average depth (m)	Annual mean evaporation rate (mm)	Annual mean outflow (km^3)	Lake type	Residence time (yr)
Tianshuihai	0.2215 ± 0.0014	5.34 ± 0.01	32.57	—	0.2181 ± 0.0018	6.2	4	~3[a]	2500	0.027	Open	0.32
Kushui Lake	0.2042 ± 0.0019	4.99 ± 0.03	99.34	—	—	6.7	5.2	~3[a]	2500	—	Closed	1.2
Gyaring Co	0.8172 ± 0.0029	−2.71 ± 0.01	18	0.9341 ± 0.0026	—	—	430.9	—	—	—	Open	—
Nam Co	0.9517 ± 0.0027	1.76 ± 0.02	79.35	—	—	8	2015.8[b]	45.6[b]	1184[b]	—	Closed	38.54
ChengHai	1.0288 ± 0.0035	−2.02 ± 0.01	80.8	0.9557 ± 0.0030	1.0130 ± 0.0016 (Spirulina)	9	77.2[c]	25.7[c]	2040.3[c]	—	Closed	12.62
XingYun Lake	0.9704 ± 0.0037	−0.32 ± 0.02	18.28	0.9423 ± 0.0029	0.9715 ± 0.0032	9.8	34.7[d]	7[d]	1002.2[d]	—	Closed	6.98
Dalinor	1.0421 ± 0.0043	−1.05 ± 0.02	34.7	0.8829 ± 0.0024	1.0188 ± 0.0027	9.4	224.6[e]	6.9[e]	1113.1[e]	—	Closed	6.21

The superscripts indicate the data resource of surface area, average depth and annual mean evaporation of the lakes. a. Li et al. 1998; b. Zhang et al. 2011; c. Hu et al. 1992; d. Zhang et al. 2008; e. Dong 2008; Wang et al. 2015.

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Table 3 The fraction of modern carbon ($F^{14}C$) values and water residence times of previously studied lakes.

Lake	$F^{14}C$ DIC	$F^{14}C$ submerged plant	pH	Surface area (km^2)	Average depth (m)	Annual mean evaporation rate (mm)	Annual mean outflow (km^3)	Lake type	Residence time (yr)
Qinghai Lake [a]	1.0971 ± 0.0039	1.0098 ± 0.0042	9.2	4400	21	924	—	Closed	22.73
Xihu [b]	0.8777 ± 0.0042	1.0327 ± 0.0029	8.5	4.7	2.5	1208.6	—	Open	2.07
Erhai [c]	0.9216 ± 0.0047	0.9022 ± 0.0047	8.5	249.8	10.5	1208.6	—	Open	2.75
Lugu Lake [d]	—	1.0180 ± 0.0042	8.1	50.1	40.3	920	0.052	Semi-closed (seasonal out flow Jun. to Oct.)	18.5

The superscripts indicate the data resource of the lakes. a. Jull et al. 2014; Zhou et al. 2014; b. Xu et al. 2015a; Du et al. 1998; c. Xu et al. 2015b; Fu et al. 2013; d. Chen et al. 2012; Sheng et al. 2015.

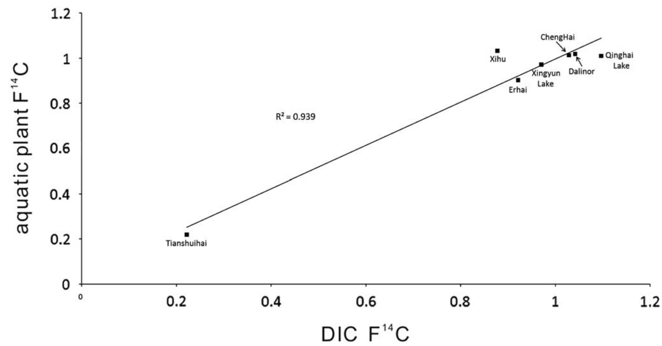

Figure 2 $F^{14}C$ of aquatic plants and DIC. The linear fit result is $y = 0.955x + 0.04$, $R^2 = 0.939$.

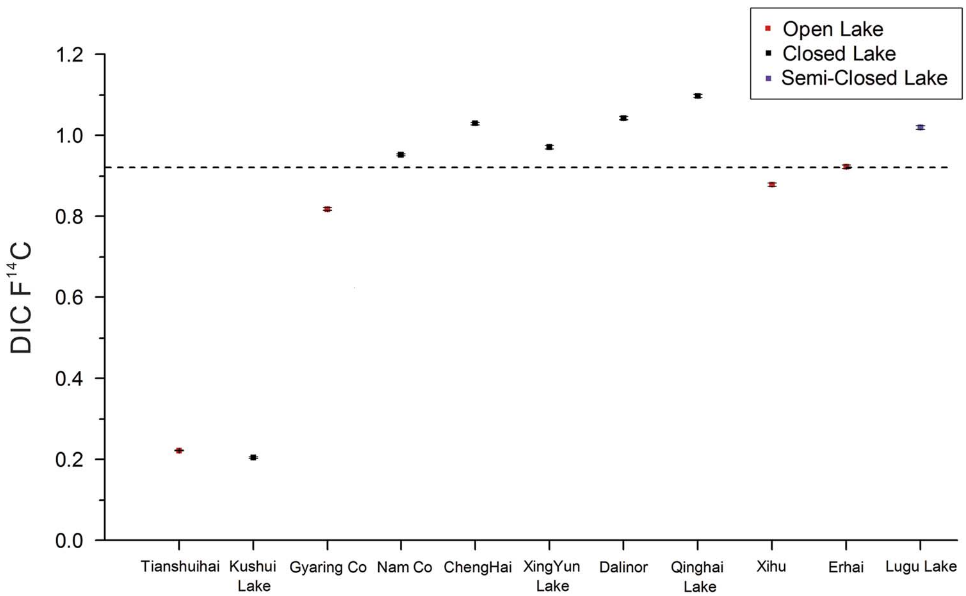

Figure 3 DIC $F^{14}C$ of different lakes. The dashed line shows the highest F_{DIC} value of open lakes. Lugu Lake uses the $F_{water\ seed}$.

values than DIC samples from closed lakes (Figure 3). This phenomenon implies that hydrological circulation influences surface lake water DIC levels.

DISCUSSION

Residence Time of Lakes

The average residence time of the water in a lake can be expressed as the ratio between lake volume and the input or output of water at steady state. This parameter reflects the hydrological processes operating in a particular lake (Yu et al. 2007; Jull et al. 2014).

In our study survey, DIC samples from open lakes (except Kushui Lake) have lower $F^{14}C$ values than DIC samples from closed lakes (Figure 3). This suggests that hydrological processes influence DIC $F^{14}C$ values. Some published research comes to the same conclusion. For example, time series DIC ^{14}C data collected from Lake Kinneret, Israel, showed that $F^{14}C$

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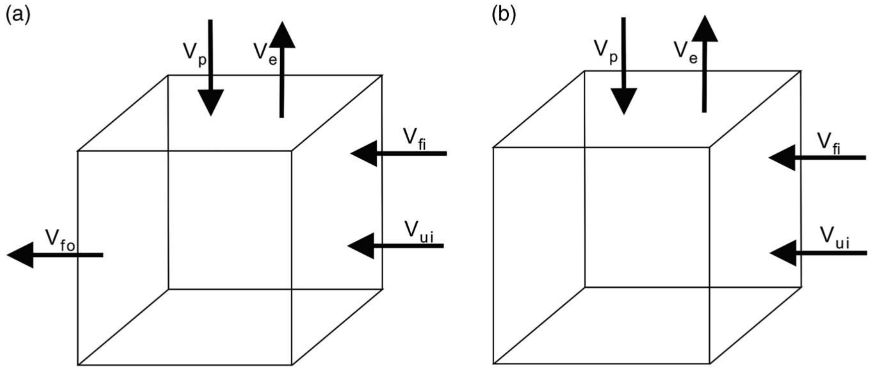

Figure 4 Model of lake water circulation (modified from Figure 1 of Yu et al. 2007). (a) open lake; (b) closed lake. V_p is the annual volume of precipitation; V_e is the annual volume of evaporation; V_{fi} is the annual water flow into the lake; V_{fo} is the annual water flow out of the lake; V_{ui} is the annual underground water flow into the lake.

values from lake water DIC were significantly reduced during years with prevalent flooding (Stiller et al. 2001). In order to further examine this influence, we calculate the average residence time for each lake. The results are shown in Tables 2 and 3.

The water balance of open and closed lakes is shown schematically in Figure 4. The steady-state water balance in a lake can be expressed as follows:

$$\text{for a closed lake:} \quad V_i = V_p + V_{fi} + V_{ui} \tag{1}$$

$$V_o = V_e \tag{2}$$

$$\text{for an open lake:} \quad V_i = V_p + V_{fi} + V_{ui} \tag{3}$$

$$V_o = V_e + V_{fo} \tag{4}$$

where V_i is annual water volume input to the lake, V_p is the annual precipitation volume to the lake, V_{fi} is the annual river inflow volume to the lake, V_{ui} is the annual underground water inflow volume to the lake, V_o is annual water output volume from the lake, V_e is the annual evaporation volume from the lake, and V_{fo} is the annual river outflow volume from the lake.

We can calculate the average residence time of a lake (τ) for a lake volume V with the following equation (Schaffner and Oglesby 1978; Jull et al. 2014):

$$\frac{dV}{dt} = \frac{1}{\tau} V \tag{5}$$

where dV/dt is the rate o f input (positive values) or output (negative values) of a lake at steady-state. As the input is more complex than the output, here we use the output to calculate the average residence time. We combine Equations (2) and (4) with Equation (5).

$$\text{for a closed lake:} \quad \tau = V / V_e \tag{6}$$

$$\text{for an open lake:} \quad \tau = V / (V_e + V_{fo}) \tag{7}$$

The volume of a lake can be expressed as

$$V = SD \tag{8}$$

$$V_e = SE \tag{9}$$

where S is the surface area of the lake, D is the average depth, and E is the annual evaporation rate. We combine Equations (8) and (9) with Equations (6) and (7).

$$\text{for a closed lake}: \ \tau = D / E \tag{10}$$

$$\text{for an open lake}: \ \tau = (SD) / (SE + V_{fo}) \tag{11}$$

In our study we use Equation (10) to calculate average residence times for all of the closed lakes. For the open lakes we calculated average residence times as follows: Tianshuihai is calculated by Equation (11), Xihu is calculated by Equation (10) due to the lack of outflow data (the actual average residence time is less than the calculated one), and Erhai and Lugu Lake average residence times are from Fu et al. (2013) and Sheng et al. (2015), respectively. As there is no published depth and outflow data for Gyaring Co, we do not calculate an average residence time. All of the average residence time results are shown in Tables 2 and 3.

Modern Water in Dalinor

GongGeEr River, LiangZi River, HaoLai River and ShaLi River are the major rivers of Dalinor; and GongGerEr River is the biggest river with 75% of total riverine inflow. The annual inflow of river and underground water accounts for almost 70% of the total annual inflow to Dalinor, and the remaining portion comes from precipitation (Dong 2008).

In Dalinor, the modern atmospheric $F^{14}C$ value is 1.0096. DIC $F^{14}C$ values of GongGeEr River (largest river at Dalinor) and spring water are 0.8015 and 0.6036 respectively. The POC $F^{14}C$ value of GongGeEr river is 0.8132 (Table 1).The surface lake water DIC $F^{14}C$ value is 1.0421 (Table 1). The $F^{14}C$ value of surface lake water DIC is similar to the atmospheric CO_2 $F^{14}C$ value, but higher than GongGeEr River (and other rivers; Table 1), and the spring DIC $F^{14}C$ value. We presume that water may exchange with atmospheric CO_2 after flowing into the lake.

The $\delta^{13}C$ value of DIC can reflect the exchange between lake water DIC and atmospheric CO_2. Carbon isotope fractionation between dissolved carbonates (HCO_3^- and CO_3^{2-}) and atmospheric CO_2, which normally has a $\delta^{13}C$ value of about −7‰, varies from 9.2‰ at 0 °C, to 6.8‰ at 30ºC (Deuser and Degens 1967; Mook et al. 1974; Myrttinen et al. 2012). Therefore, DIC in the surface lake water should have a $\delta^{13}C$ value between 0 and 2.0‰. For example, the $\delta^{13}C$ value of Qinghai Lake surface water DIC is in the range of 0.69‰ to 1.03‰ (Li et al. 2012; Wang et al. 2013b). This suggests that surface lake water there has reached a steady-state balance with atmospheric CO_2.

DIC Content of Lakes

For the lakes we studied and reviewed, surface lake water DIC $F^{14}C$ values of open lakes (except Kushui Lake) were found to be lower than surface lake water DIC $F^{14}C$ values of closed lakes (Figure 3), and the average residence times of open lakes are usually shorter than closed lakes (Figure 5). Furthermore, a comparison between surface water DIC $F^{14}C$ values and

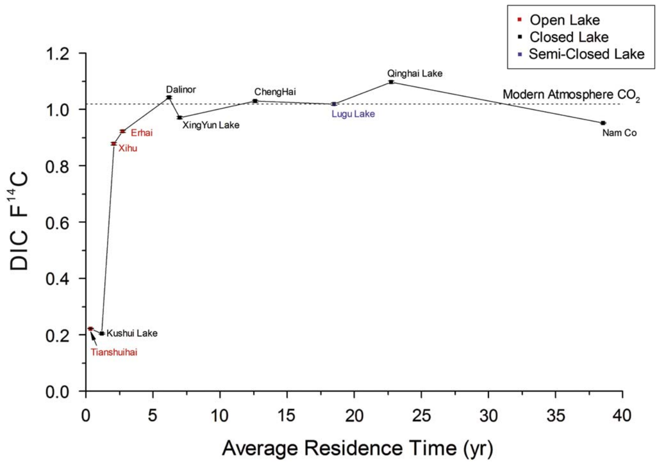

Figure 5 Surface Lake water DIC F^{14}C with lake water residence time. The dashed line shows the F^{14}C value of modern atmospheric CO_2 (Levin et al. 2013). Lugu Lake uses the F^{14}C of water seed. Gyaring Co is not shown due to a lack of residence time data.

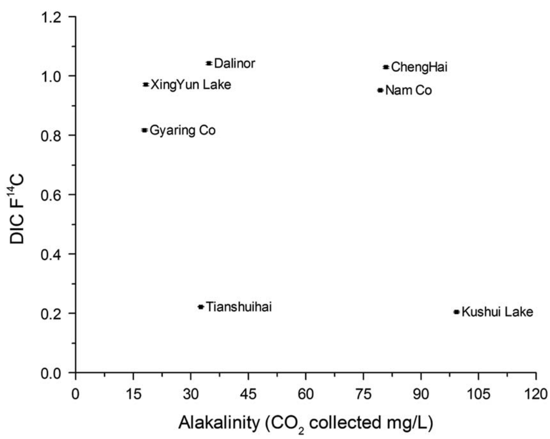

Figure 6 Alkalinity and DIC F^{14}C values of lakes.

average residence time shows that DIC F^{14}C increases with average residence time and reflects a steady-state (Figure 5).

As stated above, there are three factors that control DIC F^{14}C values in our lakes: (1) the rate and degree of exchange with atmospheric CO_2; (2) geological weathering; and (3) microbial decay of terrigenous organic matter. Ground waters and river waters generally have low DIC F^{14}C values due to the addition of "dead carbon" from rock weathering (e.g. Dalinor inflow rivers in Table 1). This effect is enhanced where large runoff occurs. The amount of "dead carbon" should be proportional to the concentration of carbon ions in the water (Mook et al. 1974). According to Keaveney et al. (2012) the reservoir offset of lakes should follow a linear relationship with alkalinity. When we plot alkalinity against DIC F^{14}C values from seven lakes however, we do not observe an obvious linear relationship (Figure 6). We notice that most lakes described by Keaveney et al. (2012) are open lakes, whereas many of the lakes studied here are

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closed lakes. Open lakes generally have lower average residence times than closed lakes, due to their relatively rapid water circulation. Open lakes have little time to exchange carbon with the atmosphere and their DIC $F^{14}C$ values are controlled primarily by the $F^{14}C$ content of source water DIC. This could explain the linear correlation between reservoir offset and alkalinity described in Keaveney et al. (2012). However, the presence of both open and closed lakes in our study leads to a much broader range of residence times, and a correlation between surface water DIC $F^{14}C$ values and average residence time (Figure 5). We are unable to quantify the impact of microbial decay of terrestrial organic carbon in our Chinese lakes since POC $F^{14}C$ values are available for only four of the lakes, and we lack DOC $F^{14}C$ data.

We find some exceptions to the general trend observed between DIC $F^{14}C$ and residence time. For example, at Nam Co, where DIC $F^{14}C$ values are much lower than atmospheric CO_2, but have a long average residence time. This difference may be due to the close proximity of the sampling site to an inflowing river. At Qinghai Lake, the DIC $F^{14}C$ value is higher than the atmospheric value, probably because of stored bomb carbon. As mentioned before, Kushui Lake is also exceptional, as a closed lake with a low DIC $F^{14}C$ value. The Kushui lake volume is very small (the average depth is only 3 m), and the evaporation in this area is very strong. These conditions lead to a very short average residence time (about one year) (Table 1). The pH of Kushui Lake is lower at 6.7 (Table 2) which leads to a lower exchange rate with atmospheric CO_2. The alkalinity of Kushui Lake is also as high as 99.34 (Table 2), which means it contains abundant carbonates from geological weathering and also may leads to rapid precipitation of carbonates. This produces a low input DIC $F^{14}C$ value, low exchange, rapid removal of surface lake water, and the observed low DIC $F^{14}C$ value in the lake.

CONCLUSIONS

We compare the DIC^{14}C characteristics of 11 lakes from different parts of China. Surface lake water DIC $F^{14}C$ values of open lakes (except Kushui Lake) are lower than the surface lake water DIC $F^{14}C$ values of closed lakes. The DIC $F^{14}C$ value of surface lake water increases with average residence time and reaches a steady-state near the modern atmospheric CO_2 $F^{14}C$ value. This phenomenon can be explained by exchange with atmospheric CO_2. The driving force (e.g. exchange with atmospheric and geological weathering) behind the freshwater reservoir effect is site-specific.

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