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^{14}C GEOCHRONOLOGY AND RADIOCARBON RESERVOIR EFFECT OF REVIEWED LAKES STUDY IN CHINA

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ABSTRACT. Lacustrine sediments are important archives for paleoclimate research, but there are evident carbon reservoir effects. Radiocarbon (^{14}C) ages of lake sediments must be corrected for these effects before applying them to paleoclimate research. The authors review the lacustrine research from the last 20 years from different climatic regions in China, and systematically investigate the ^{14}C age and correction methods used in the studies of 81 lakes. It is found that the climate-vegetation cover and distribution of carbonate around lakes are dominant factor controlling radiocarbon reservoir effects. In eastern China, the average ^{14}C reservoir age is about 500 ^{14}C years and is associated with relatively dense vegetation. However, in northwest China and Qinghai-Tibet Plateau, widespread carbonate bedrock may markedly increase the radiocarbon reservoir age which frequently is about 1500 and 2500 ^{14}C years. A piecewise linear regression model provides more reliable ^{14}C reservoir age correction that accounts for sedimentary facies and sedimentation rate changes. It is worth mentioning that when analyzing ^{14}C ages deviated greatly from time sequence, the age anomalies may indicate important effects relevant to the study of climate and environmental changes.

KEYWORDS: anomalous ^{14}C, lacustrine sediments, radiocarbon reservoir effects.

INTRODUCTION

As a key archive of paleoenvironment research, lake sediment has the merits of wide spatial distribution and different sediment time span (covering multi-decadal, centennial and millennial scale), which provides valuable information for continental paleoenvironment reconstruction (Ramsey et al. 2012; Chen et al. 2016; Zhang et al. 2017; Lan et al. 2020). In recent decades, lacustrine sediments in different climatic regions of China have facilitated great progress in the study of the evolution of the mid-latitude westerly jet and the East Asian monsoon system (An et al. 2000, 2011, 2012; Chen et al. 2019, 2020). Radiocarbon (^{14}C) dating is currently the most common method in dating lacustrine sediments, which covers up to 50 ka (Hajdas et al. 1993). However, two types of ^{14}C reservoir effects should be considered in lacustrine ^{14}C chronological research. The first was discovered by Deevey et al. (1954) that the ^{14}C-deplected dissolved inorganic carbon affects the lake carbon pool and causes considerable uncertainty in the dating of fresh water (Godwin 1951; Broecker et al. 1959), the other is that lake carbon pool affected by old organic carbon revealed in recent studies (Nelson et al. 1988; Benson 1993; Moreton et al. 2004). The first phenomenon is called the "hard water effect" and the second, the "radiocarbon reservoir effect," although the second term is often use for both in the

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literature. Due to the influence of these phenomena, ^{14}C dating results of lake sediment may significantly differ from the actual age of sediments (Jull et al. 2016; Olsson 2016), which hinders the interpretation and comparison of different lake-core records (Colman et al. 1996; Björck and Wohlfarth 2002). Hou et al. (2012) noted that the ^{14}C reservoir effect in lakes from the Qinghai-Tibet Plateau may be thousands of ^{14}C years. Thus, the reconstruction of major climate events, such as the timing of the Last Glacial Maximum, and the spatial-temporal distribution of the Holocene Optimum on the Qinghai-Tibet Plateau, may incur large uncertainties. Moreover, a large number of studies have shown that the lake ^{14}C reservoir effect can significantly fluctuate with time (Olsson et al. 1969; Geyh et al. 1998; Soulet et al. 2011; Keaveney and Reimer 2012; Philippsen and Heinemeier 2013; Yu et al. 2017). Age uncertainties have often been cited as potential causes of discrepancies between findings in paleoclimate-environmental research in China (Jin et al. 2007; Dong et al. 2015; Liu et al. 2015).

Therefore, there is a pressing need to constrain and reduce age uncertainties associated with the ^{14}C reservoir effect. To achieve this goal, we review lacustrine records based on ^{14}C dates of 81 Chinese modern lakes (distinguished from ancient lakes) published in the past 20 years to make a clear understanding of the spatial distribution of lacustrine reservoir effects. The ^{14}C reservoir correction approaches used in these studies are summarized, and the effect of environmental changes on ^{14}C ages is also addressed.

SPATIAL DISTRIBUTION OF LAKE ^{14}C RESERVOIR EFFECTS IN CHINA

With more than 2759 natural lakes that exceed 1 km^2 (Wang et al. 1998), lakes are widely distributed in China. Here, we divide them into seven regions (Figure 1) according to the vegetation divisions of Zhao et al. (2009) and Zhou and Yu (2012). Qinling Mountain–Huaihe River separates North China and South China, the boundary between temperate grassland and temperate desert separates Northwest China and North China, and the 3000-m contour separates the Qinghai-Tibet Plateau, South China, and Northwest China (Chen et al. 2015), as shown in Figure S1.

In Figure 1 and Table S1, the ^{14}C reservoir ages of 81 lakes in China are reviewed based on published research from the past 20 years. The ^{14}C reservoir effect of lakes in China shows significant spatial variability, ranging from 0 ^{14}C year in Shuangchi Maar Lake (Dodson et al. 2019) in Hainan province to 23,585 ^{14}C years in Lake Qiangyong Co (Zhang et al. 2017) on the Qinghai-Tibet Plateau. On a large geographic scale (see Figure S1), the average ^{14}C reservoir effect in north China and south China is around 450 ± 260 (n = 10) and 640 ± 410 (n = 10) ^{14}C years, respectively, whereas lakes on the Qinghai-Tibet Plateau have the highest ^{14}C reservoir ages with an average reservoir age of 2370 ± 1240 (n = 48) ^{14}C years. ^{14}C reservoir ages in Northwest China fall between South China and the Qinghai-Tibet Plateau with an average reservoir age of 1420 ± 890 (n = 6) ^{14}C years. Regionally, the spatial distribution of the ^{14}C reservoir effect is Qinghai-Tibet Plateau > Northwest China > North China > South China (Table S1 and Figure S1). In addition, the average reservoir age of outflow lakes (1180 ± 720 (n = 13) ^{14}C years) is almost the same as for inflow lakes (1210 ± 670 (n = 26) ^{14}C years), indicating that the closure condition of lakes is not the dominant factor affecting the ^{14}C reservoir effects (see Figure 2a).

In terms of vegetation types, it appears that the average ^{14}C reservoir age increases from tropical monsoonal rainforest (average 0 year) to temperate steppe (average 500 ± 220

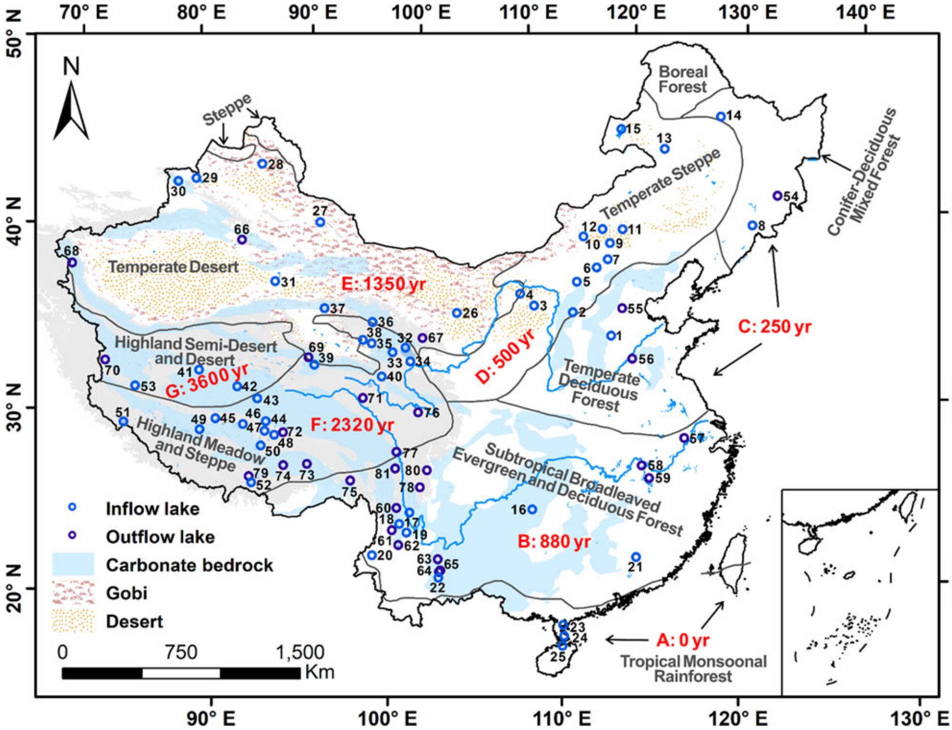

Figure 1 Spatial distribution of lake ^{14}C reservoir effect in different vegetation and bedrock zones in China. Blue circles and purple circles represent the location of inflow and outflow lakes, respectively. Lake numbers are shown here 1 and also in supplementary Table S1. A–G present different climate and vegetation zones (modified from Zhao et al. 2009). Average reservoir ages are in red.

(n = 7) ^{14}C years) and then to highland meadow and steppe (average 2320 ± 900 (n = 21) ^{14}C years) (Figure 2b). As for the subtropical broad-leaved evergreen and deciduous forest, this region has higher vegetation cover but older average lake reservoir ages (average 880 ± 590 (n = 11) ^{14}C years) than temperate steppe and temperate deciduous forest with conifer-deciduous mixed forest (average 250 ± 190 (n = 3, due to the limited amount of lake research, the statistic is for reference only) ^{14}C years). According to previous studies, widely distributed carbonate bedrock significantly affects the ^{14}C content in dissolved inorganic carbon (DIC) of lake water (Liu et al. 2017), the existence of carbonate bedrock around the lake will contribute to large ^{14}C reservoir effects (Hendy et al. 2006). So, the older reservoir ages of the subtropical broadleaved evergreen and deciduous forest probably can be attributed to the influence of karstification in the Yunnan-Guizhou Plateau. Furthermore, our statistics also reflect the fact that lakes around carbonate bedrock have higher ^{14}C reservoir ages, as shown in Figure 2c, where lake ^{14}C reservoir ages could on average reach 1430 ± 730 (n = 22) ^{14}C years in those basins with carbonate bedrock, which is about 520 ^{14}C years older than those basins without carbonate bedrock.

On the other hand, the content of total organic carbon (TOC) in sediment seems to have a possible relationship with the reservoir age. As shown in Figure 2d, when the TOC content in the sediments is high (greater than 5%), the reservoir age of the lake is not higher than 2000 years, especially less than 1000 years in non-carbonate areas. The TOC content in

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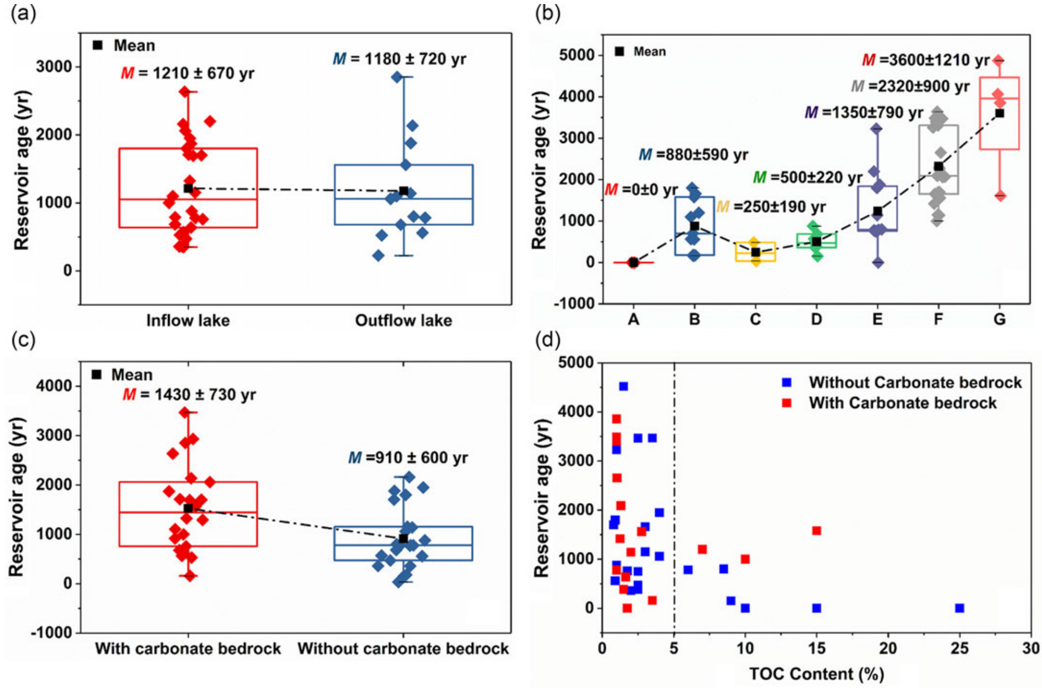

Figure 2 Statistical graph of lake reservoir effect in China: (a) box-plot of ^{14}C reservoir age in outflow and inflow lakes; (b) box-plot of lake ^{14}C reservoir age under different vegetation types (A–G, please also see Figure 1); (c) box-plot of lake ^{14}C reservoir ages in regions with carbonate and without carbonate bedrocks; (d) relationship of reservoir age verses TOC content in lake sediments. Top/bottom of boxes (a)–(c) denote upper/lower quartiles; upper/lower horizontal lines denote maximum/minimum; inner lines denote median; black squares denote arithmetic mean.

sediments of all lakes with ^{14}C reservoir age over 2000 years is less than 5%, and the ^{14}C reservoir effect fluctuates greatly within the content range of 0–5%. Thus, it seems that lakes with denser vegetation and/or higher lake productivity will have lower reservoir ages, both vegetation type and basin bedrock around the lake are jointly affecting the lake ^{14}C reservoir effect.

In summary, specifically in eastern China, given the well-developed vegetation, higher lake productivity and less carbonate bedrock, the lake ^{14}C reservoir effect is relatively small. However, in northwest China and the Qinghai-Tibet Plateau, sparse vegetation and widespread carbonate bedrock may lead to the significant regional lake ^{14}C reservoir effects. Therefore, we assume that the relationship between vegetation cover along with bedrock types around the lake is responsible for the observed differences in the ^{14}C reservoir effect in lakes of China. Follow-up research is required to verify the specific relationship.

CORRECTION OF LAKE ^{14}C RESERVOIR EFFECTS IN CHINA

^{14}C Reservoir Effect Correction Using Modern Samples

In this study, the ^{14}C age of modern samples (DOC: dissolved organic carbon, DIC: dissolved inorganic carbon, living aquatic plants, living animals, and surface sediments) were used to determine ^{14}C reservoir ages in lakes. For example, by measuring modern shells and submerged plants, Xu et al. (2015) determined that the ^{14}C reservoir age in Lake Erhai was between 523 and 610 ^{14}C years; Mischke et al. (2005) assumed currently surface living *Pisidium* shell ages as the present-day ^{14}C reservoir age of Lake Luanhaizi. However, Yang and Chen (2014) compared the ^{14}C ages of aquatic plants (*Potamogeton malaianus*) with lake water DIC and surface sediment in Dongping Lake and found that organisms may be affected by nuclear explosion events and appear to be younger, which implied that the ^{14}C reservoir age determined by post-bomb aquatic organisms may be underestimated (Philippsen 2013). Only by subtracting the effects of nuclear ^{14}C, can we obtain a reliable value of the modern ^{14}C reservoir age.

On the other hand, Peak ^{137}Cs and ^{210}Pb values in lake sediments, induced by global anthropogenic sources (nuclear tests) in the last century, can also serve as a time marker, which can be used as a standard for ^{14}C reservoir age correction by comparing the ^{14}C ages of the same sediment layers (Lan et al. 2020). For example, based on ^{14}C ages of bulk organic matter from the ^{137}Cs and ^{210}Pb peaks in 1963 CE, Kasper et al. (2012) obtained a contemporary ^{14}C reservoir age of 1420 ± 40 ^{14}C years for Lake Nam Co, and excluded some anomalous ^{14}C data at the surface layer. Based on ^{137}Cs and ^{210}Pb data, Tang et al. (2015) and Sun et al. (2018) confirmed that the ^{14}C reservoir effect in the sediments of Lake Xiari Nuur is insignificant (Xu et al. 2018).

^{14}C Reservoir Correction of Lake Sediment Sample

As lake sediments deposited over a long time, sedimentation rate and ^{14}C reservoir age can be expected to change with variations of lake hydrological conditions (Brown et al. 2000; Soulet et al. 2011). If the sedimentation rate is constant, the ^{14}C age in sediments should be linearly correlated with depth. The intercept of a linear regression equation can be taken as ^{14}C reservoir age for the whole stratum. However, the chronological framework established by this method may incur large uncertainties when the sedimentation rate is not constant. In view of this, Zhou et al. (2014, 2016) applied a method to determine the ^{14}C reservoir age of Lake Qinghai for the past 40 ka. Considering the change of sedimentation rate, average ^{14}C ages in different lithologic sections should be calculated by means of different linear regressions. The regression Eq. (1)

$$y_e = ax_d + b = y(x_d) + b\left(\overline{DCF}\right) \text{ (DCF : dead carbon fraction)} \quad (1)$$

is obtained by regression calculation between the ^{14}C age (y) measured by AMS (accelerator mass spectrometry) and depth data points (x_d). The estimated value (y_e) is the sum of both the contribution of natural decay with depth (x_d) and a constant radiocarbon reservoir effect ($\overline{DCF}$) which is implied in the intercept b of the regression equation. At the surface of the sedimentary profile, where $x_d = 0$, the contribution provided by natural decay should be 0, and only a constant contribution provided by the average old carbon effect ($y_e = b\left(\overline{DCF}\right)$). Finally, it is concluded that the average reservoir age $y(\overline{DCF})$ is the intercept (b) of the regression equation for the case where the regression equation is suitable to the sediment surface.

In the Lake Qinghai case, the lithology of 1F core is chosen for this study, the section contains clay from 0–499 cm, silty sand from 499–901 cm, and fine sand after 901–1861 cm (Zhou et al. 2014), therefore we should consider the different sedimentation rate for establishing a reliable chronology (see Figure 3). As mentioned above, we calculate the average ^{14}C reservoir age of

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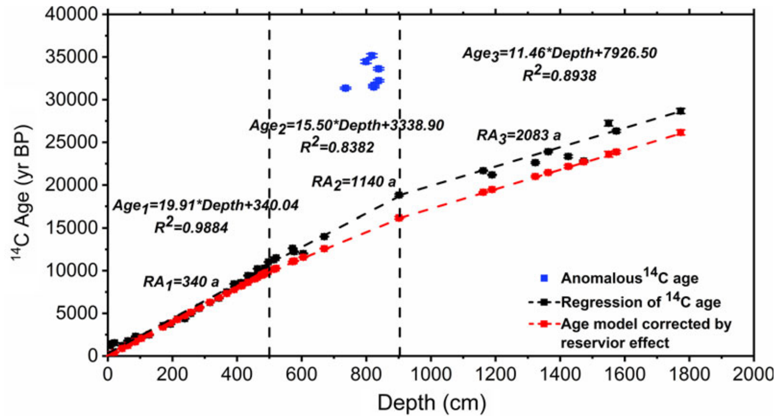

Figure 3 Age-depth model of Lake Qinghai 1F core. ^{14}C reservoir age obtained by piecewise linear regression using ^{14}C ages based on different sedimentation rate (clay from 0–499 cm, silty sand from 499–901 cm, fine sand from 901–1861 cm). RA denotes average ^{14}C reservoir age.

Lake Qinghai above 499 cm (during the Holocene) to be 340 ^{14}C years, the rest portions at 499–901 cm and 901–1861 cm to be 1140 and 2083 ^{14}C years, respectively. Li et al. (2017) also applied this method to the ^{14}C reservoir correction of Aweng Co. According to different sedimentation rates, the average ^{14}C reservoir age of the upper and lower sections of the Aweng Co core are 4066 and 3227 ^{14}C years, respectively.

In addition, Ming et al. (2020) discussed the influence of reservoir age uncertainties on climate reconstructions from Lake Bayan Nuur sediments in Inner Mongolia and put forward a paleoclimatic method to determine the ^{14}C reservoir age. They generated different Bayesian age models based on the Bacon program, varying only the ^{14}C reservoir age from its possible range (0–800 cal years), then evaluated its rationality by stratigraphic alignment. This technique allowed them to constrain the uncertainties caused by the ^{14}C reservoir age effect. The beginning and ending ages of sand layers in Lake Bayan Nuur sediments were found to be in good agreement with the timing of corresponding North Atlantic cold events (see Figure 4a).

Other Methods

Independent dating methods, such as uranium-series dating, varve counting, OSL (optically stimulated luminescence) dating and the age of terrestrial plant residues from the sediments, can also be used as reference for ^{14}C reservoir age corrections. For example, based on the results of 12 OSL and ^{14}C dating ages in Lake Zhuyeze sediments, Long et al. (2017) confirmed that the ^{14}C reservoir age in lake Zhuyeze was insignificant. However, Yu et al. (2014) compared the OSL and ^{14}C ages on bulk organic carbon and considered the ^{14}C ages appear to be anomalously old (>9000 BP). Due to the input of pre-aged carbon to the lake, they consider the reservoir age could be as old as 20,000 years. Zhang et al. (2012) found complex changes of the ^{14}C reservoir effect in Lake Lop Nuur through a comparison of OSL and ^{14}C dates on lake sediments. Zhou et al. (2009) revealed that the ^{14}C reservoir in Lake Sugan changed from 2590 to 4340 ^{14}C years through the chronology of lake varve. By comparing the age of TOC and plant residues in the same layer of Lake Bosten, Huang

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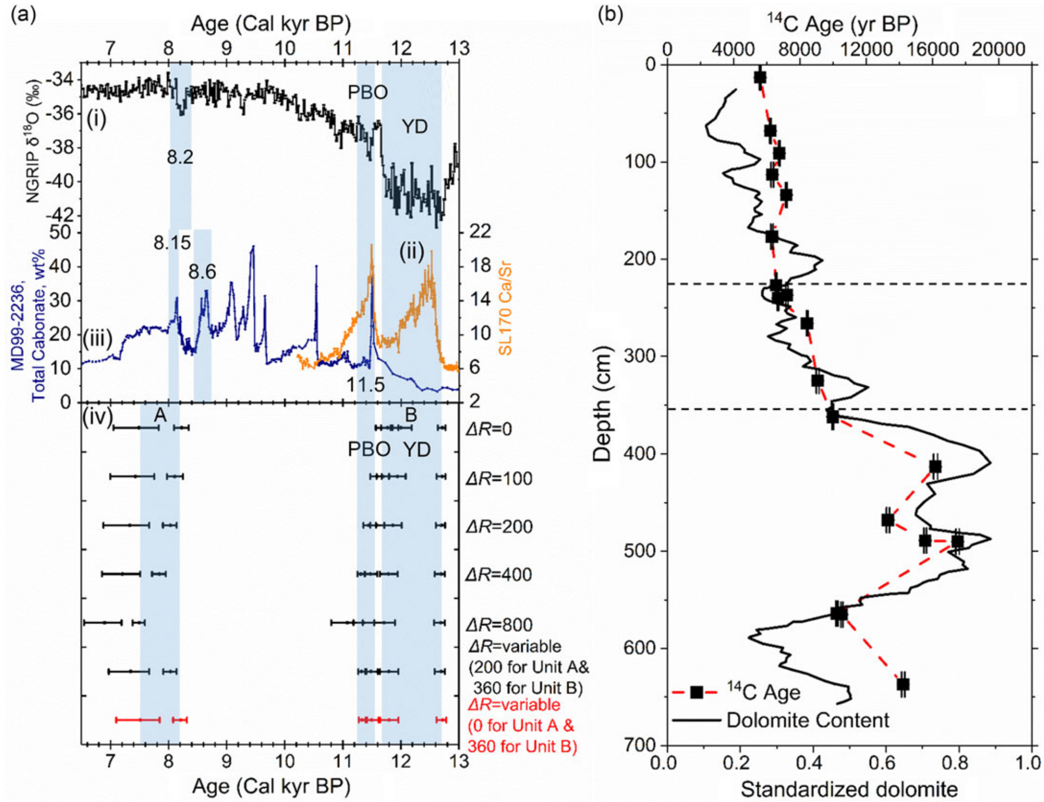

Figure 4 (a) Carbon reservoir age compared with climatic abrupt events (after Ming et al. 2020), from top to bottom: (i) NGRIP $\delta^{18}O$ record (Rasmussen et al. 2014); (ii) Ca/Sr ratios for core SL 170 in central Baffin Bay, North Atlantic (Jackson et al. 2017); (iii) detrital carbonate events recorded in MD99-2236 core, North Atlantic (Jennings et al. 2015); (iv) modeled ages for the onset and end of sand layers for the YD (Younger Dryas), PBO (pre-Boreal oscillation) and 8.2 ka events in Lake Bayan Nuur sediments. Modeled ages using different reservoir values (ΔR) are shown with calculated uncertainties; the shaded area indicates the timing of dry/cold intervals for each record; (b) standardized XRD results of allochthonous dolomite input (black line) compared with the ^{14}C ages (red dashed line) (after Lockot et al. 2015).

et al. (2009) found that the ^{14}C reservoir age was 1140 ^{14}C years age, relatively stable for Lake Bosten. By comparing the age difference of TOC and terrestrial wood fragments on the same horizon, Li et al. (2016) proposed that the average ^{14}C reservoir age of Lake Poyang was 560 ^{14}C years. In addition, considering the existence of a large number of plant macrofossils the difference between the age of TOC and plant macrofossils in the same borehole reveals that the fluctuations of lake ^{14}C reservoir age are about 8000 ^{14}C years in Lake Chenghai (Xiao et al. 2018) and 960–2200 ^{14}C years in Lake Xingyun (Zhou et al. 2015). In summary, the ^{14}C reservoir age is difference of ^{14}C time rather than calendar time, thus it is required to convert the calendar ages from other method (such as OSL) into atmospheric-derived ^{14}C age using the atmospheric calibration curve (Soulet 2015) before reservoir age calculation, referred to “decalibration” or “uncalibration” (Soulet 2015; Reimer and Reimer 2017).

Finally, for lakes with clear input and output end-members, the carbon budget can be simulated based on model calculations. The influences of each end-member and its influence on the ^{14}C reservoir effect can be evaluated in theory. For example, employing a

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two-box simulation model based on the principle of ^{14}C mass balance, Yu et al. (2007) estimated a calculated ^{14}C reservoir age value of about 1500 ^{14}C years for Lake Qinghai. However, the model approach requires detailed meteorological and hydrological data in the lake basin and its surroundings region, which limits its application.

Re-Analysis of Anomalous ^{14}C Data of Lake Sediments

When we date samples and find ages that do not correspond to their stratigraphic order, these anomalous data are often rejected in view of their implied stratigraphic inversion. However, these anomalous data may be related to changes in climate and the environment in a lake basin and should be scrutinized in this light.

For example, based on the ^{14}C ages of 1F and 2C cores in Lake Qinghai (An et al. 2012; Zhou et al. 2014, 2016), three striking anomalous ages periods were found. During the three warming intervals at 35, 19, and 14 ka BP, stalagmite records indicated that the temperature and precipitation in the monsoon region increased (Dykoski et al. 2005), which may have led to the retreat of glaciers around Lake Qinghai. Since a large quantity of meltwater flowed into the lake, a commensurate amount of old carbon would also be expected, providing a possible mechanism to explain the anomalous ^{14}C age results.

Lockot et al. (2015) found that ^{14}C age fluctuations of aquatic plants in the drilling core of Lake Heihai were highly correlated with dolomite content (see Figure 4b). During the last deglaciation period/early Holocene, the temperature gradually increased, and a large quantity of dolomite was carried into the lake by meltwater, where it dissolved into inorganic carbon (Abbott and Stafford 1995) and resulted in older ^{14}C ages. In the middle Holocene, ^{14}C dating results no longer fluctuated with the change of depth, resulting from climate and hydrological conditions generally remain stable. As for the late Holocene, the climate around lake basins experienced drought and cold temperatures. With the decrease of lake water level, old lacustrine sediments were exposed to the surface and redeposited into the lake. The ^{14}C age of sediments in this section is basically constant. From this perspective, ^{14}C dating results of sediments in Lake Heihai serve as indicators of regional climate change.

Zhang et al. (2017) conducted a comparative study on ^{14}C dating of plant residues and pollen concentrates in sediments of Lake Qiangyong Co in Qinghai-Tibet Plateau. It was found that ^{14}C dating ages of *Cyperaceae* plant residues can represent true sediment ages, while ^{14}C dating results of pollen concentrates in the same strata are greater than those of sediments from 0 to 5080 ^{14}C years. This phenomenon is called the old pollen effect (OPE). The three high OPE stages in Lake Qiangyong Co sediment correspond to the Current Warm Period, the Medieval Warm Period, and the Iron/Roman Age Optimum warm period, while the low OPE stage corresponds to the Little Ice Age, Dark Ages, and Iron Age Cold epoch. The OPE-derived cold periods are consistent with the regional glaciation processes revealed by ^{10}Be dated moraine records. Therefore, the OPE of Lake Qiangyong Co based on ^{14}C dating ages anomaly serve as a proxy for regional glacial activity.

CONCLUSION

The ^{14}C reservoir effect is widespread in lakes but varies in different regions in China. Here, we summarized and reviewed the ^{14}C reservoir effect of 81 lakes in China published in the last recent 20 years and have provided a spatial distribution of lake ^{14}C reservoir effect in

China, which displays obvious regional characteristics. These characteristics indicate that bedrock types in lake basins are a significant factor controlling the value of lake ^{14}C reservoir ages. However, considering the surrounding environment and climatic conditions, such as vegetation and rainfall, cannot be avoided. In addition, traditional methods such as modern ^{14}C reservoir age and linear regression are appropriate for lakes with small ^{14}C reservoir effect fluctuations with time. For lakes with significant changes in sedimentary facies and sedimentation rate, a piecewise linear regression method should be considered. It is worth mentioning that, when using other dating rather than ^{14}C for reservoir age correction, it requires the "decalibration" of calendar ages to obtain the corresponding atomsphere-drived ^{14}C age. In practice, some anomalous age data are rejected with good reason. However, we hold that these anomalous age data may provide valuable information about the hydrological conditions of the lake and serve as an indicator of regional environmental change. These conclusions will provide reference for correcting ^{14}C reservoir effect in future lake research.

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SUPPLEMENTARY MATERIAL

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