

POSSIBLE FACTORS CAUSING OLDER RADIOCARBON AGE FOR BULK ORGANIC MATTER IN SEDIMENT FROM DAIHAI LAKE, NORTH CHINA

Yanhong Wu¹ • Sumin Wang² • Liping Zhou³

ABSTRACT. Many factors may influence the radiocarbon age results of lacustrine sediments, among which the hardwater effect is particularly important. Daihai Lake is a closed lake located in the semi-arid region of Inner Mongolia, China. High concentrations of HCO_3^- and CO_3^{2-} and high pH values in the lake water imply that there is a hardwater effect when using bulk lacustrine sediment samples for ^{14}C dating. To correct the apparent ^{14}C age, we present a pilot study based on a series of ^{14}C ages of lake surface sediment, lake water, submerged aquatic plant (*Myriophyllum*), fish bone (*Cyprinus carpio*), and surface soil samples from and around Daihai Lake. Assuming that the relationship between the $^{14}\text{C}/^{12}\text{C}$ ratio of DIC and of atmospheric CO_2 was constant (at 0.816), the hardwater effect ages calculated for the past 8000 yr would have varied from 949 to 1788 yr. Together with the reservoir effect and soil organic matter input, the hardwater effect is a major factor causing changes in apparent age when using bulk organic matter for ^{14}C dating.

INTRODUCTION

Radiocarbon dating is a useful method for providing a chronological sequence for Pleistocene-Holocene paleoenvironmental studies. However, many studies of lacustrine and marine sediments, and even soils, have yielded older than expected ^{14}C ages (Björck et al. 1991, 1996, 2003; Gu et al. 1993; Sun et al. 1993; Rea and Colman 1995; Colman et al. 1996; Fontes et al. 1996; Wohlfarth et al. 1998; Björck and Wohlfarth 2001; Wang et al. 2002; Hutchinson et al. 2004; Peng et al. 2005; Xiao et al. 2005). The “older” ages are caused by several factors, the most important being the reservoir effect and hardwater effect, which contribute to ^{14}C depletion during dating of materials due to old carbon incorporation (Fontes et al. 1996; Björck and Wohlfarth 2001; Hall and Henderson 2001). Bulk organic matter in lacustrine sediment is widely used as ^{14}C dating material, although pollen or other microfossils can also be used. The major source of organic matter in lakes is from endogenic aquatic plants (Meyers and Ishiwatari 1993). When the lake has been efficiently sealed off from the atmosphere by lake ice, or is mainly fed by water containing old carbon from glacier water, groundwater, or from volcanic eruption, the $^{14}\text{C}/^{12}\text{C}$ ratio of the dissolved inorganic carbon (DIC) should be lower than the $^{14}\text{C}/^{12}\text{C}$ ratio of CO_2 in the contemporaneous atmosphere. Thus, dating of aquatic plant materials that assimilate such DIC will be affected by the so-called “lake reservoir effect” (Björck and Wohlfarth 2001). In a hardwater lake rich in bicarbonate ions, aquatic plants will take up and incorporate some “dead” carbon, resulting in the hardwater effect (Björck and Wohlfarth 2001; Wu et al. 2006).

The hardwater effect and reservoir effect are crucial for past global-change research, when scientists try to correct the apparent ^{14}C ages of records such as lakes. To correct for the hardwater and reservoir effects, analyses are made of shells and terrestrial plant fossils (pollen, plant stems, etc.) and compared with the results of other dating methods, to get reservoir ages (Rea and Colman 1995; Ren 1998; Wohlfarth et al. 1998; Hall and Henderson 2001; Eiriksson et al. 2004; Hutchinson et al. 2004; Shackleton et al. 2004; Zhang et al. 2004; Bondevik et al. 2006; Reimer and Reimer 2006). Unfortunately, in many lacustrine sediments there are often few terrestrial plant fossils to resolve this problem. The more general way to estimate the effect is to determine the apparent age at zero depth according to a regression line based on a series of apparent ^{14}C ages (Fontes et al. 1996; Shen et al.

¹Institute of Mountain Hazards and Environment, Chinese Academy of Sciences, Chengdu, 610041, China. Corresponding author. Email: yhwu@imde.ac.cn.

²State Key Laboratory of Lake Science and Environment, Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, Nanjing 210008, China.

³Laboratory for Earth Surface Processes, Department of Geography, Peking University, Beijing 100871, China.

2005; Xiao et al. 2005); however, this approach does not take into account any temporal variation in the hardwater and reservoir effects. Stiller et al. (2001) calibrated the age of sediments according to the relationship between ^{14}C levels in lake water and the atmosphere in Lake Galilee during the past 10,000 yr.

Generally, ^{14}C ages are calculated according to the following equation:

$$T = 8033 \ln (A_0/A_t) \quad (1)$$

where T is the ^{14}C age of the sample (yr BP); A_0 is the initial activity of ^{14}C (cpm/g); and A_t is the ^{14}C activity measured during counting of the sample (cpm/g). The ^{14}C activity of the atmosphere is therefore regarded as A_0 . For lakes where hardwater or reservoir effects exist, the ^{14}C activity of DIC may be lower than that of atmospheric CO_2 . The hardwater age and reservoir age thus can be calculated according to the $^{14}\text{C}/^{12}\text{C}$ ratio of DIC.

In paleoenvironmental research on Chinese lake sediments, hardwater and reservoir effects have been found widely, especially in some high-salinity and high-pH lakes on the Tibetan Plateau and other arid and semi-arid areas (Gu et al. 1993; Sun et al. 1993; Fontes et al. 1996; Wang et al. 2002; Shen et al. 2005). In Daihai Lake, Inner Mongolia, north China, Cao et al. (2000) noticed large discrepancies in the sediment rate of the upper layers between ^{210}Pb results and ^{14}C dates and attributed them to the uncertainties associated with different dating methods. Xiao et al. (2005) discovered the existence of a hardwater effect in 8 ^{14}C dating results (using bulk organic matter as dating material), and deduced a hardwater age of 360 yr according to a linear regression, taking the age of 0 yr for the surface sediment. Our preliminary study on surficial samples from the same lake, however, revealed apparently more pronounced deviation from zero age, which was initially attributed to the reservoir effect (Wu et al. 2007).

Here, we present a summary of the ^{14}C dating results of surface sediment, live aquatic plants, bones of live fish, lake water, and lake beach soil samples from Daihai Lake, and discuss possible factors causing apparently older ^{14}C ages and the possible approaches to correct them.

GENERAL DESCRIPTION OF THE RESEARCH AREA

Daihai Lake (112°33'31" to 112°46'40"E, 40°29'7" to 40°37'6"N) lies about 10 km east of Liangcheng County, Inner Mongolia, in north-central China (Figure 1). It has an area of 160 km² with a maximum water depth of 16 m and an elevation of 1221 m. It is a semi-brackish closed lake, the major inflows of which are the Muhua River, forming the east bank, and the Gongba River, forming the west bank (Wang et al. 1990).

Daihai Lake is located at the transition between semi-humid and semi-arid areas in the middle temperate zone of China. The mean annual temperature is 5.1 °C with a July average of 20.5 °C and a January average of 13.0 °C (Xiao et al. 2005). The mean annual precipitation is 413 mm, about 80% of which occurs in summer. The mean annual evaporation reaches 1033 mm (Wang et al. 1990).

Because the evaporation is much higher than precipitation and the water consumption in the catchment has increased during the past several decades, the lake shrank and the salinity of the lake water increased continuously. The salinity was 2100–2400 mg/L in 1963, ~3000 mg/L in 1987 (Wang et al. 1990), and increased to 3888 to 5120 mg/L as measured in 2006 and 2007 (Table 1). The pH value also increased during the past several decades (Wang et al. 1990).

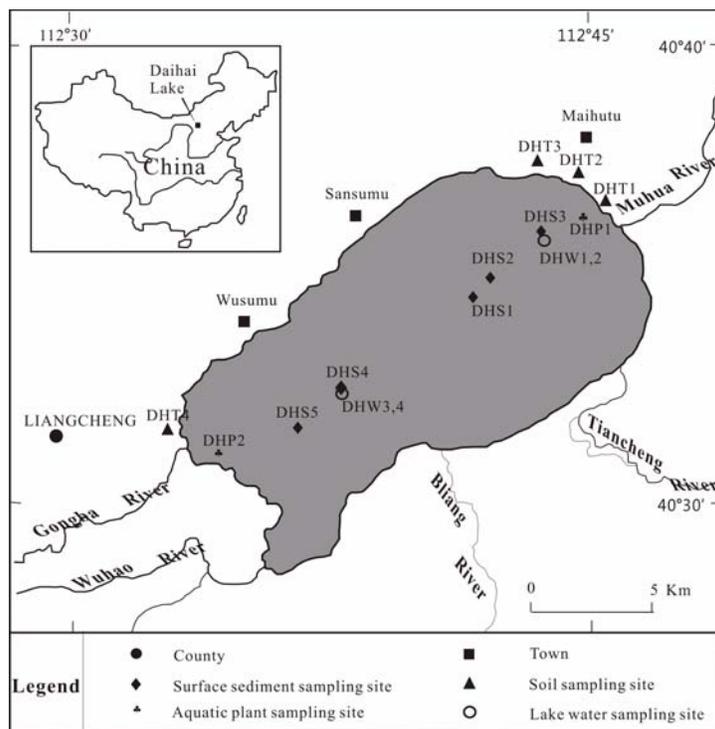


Figure 1 Map of Daihai Lake and the sampling sites

Table 1 Results of ^{14}C levels of different samples and their apparent ^{14}C ages (half-life 5730 yr).^a

Sample type	Sample nr	Material ^b	X_s	X_s / X_a	Apparent ^{14}C age (yr BP)	Age uncertainty (yr)
Surface sediments	DHS1	OM _{bulk}	1.087	0.798	1865	40
	DHS2	OM _{bulk}	1.065	0.782	2035	40
	DHS3	OM _{bulk}	1.061	0.779	2065	50
	DHS4	OM _{bulk}	1.052	0.773	2132	50
	DHS5	OM _{bulk}	1.086	0.797	1871	40
Submerged plants	DHP1	OM _{bulk}	1.003	0.737	2528	80
	DHP2	OM _{bulk}	1.086	0.798	1870	70
Water	DHW1	TIC	1.143	0.839	1450	125
	DHW2	TIC	1.088	0.799	1860	80
	DHW3	TIC	1.103	0.81	1740	70
	DHW4	TIC	1.11	0.815	1690	70
Fish bone	DHF1	OM _{bulk}	1.005	0.738	2513	70
Surrounding soil	DHT1	OM _{bulk}	1.005	0.738	2513	90
	DHT2	OM _{bulk}	1.167	0.857	1277	80
	DHT3	OM _{bulk}	1.058	0.777	2090	90
	DHT4	OM _{bulk}	1.147	0.842	1420	80

^a X_s = ^{14}C activity of measured sample; X_a = ^{14}C activity of atmosphere.

^bOM_{bulk} = bulk organic matter; TIC = total inorganic carbon.

MATERIALS AND METHODS

Three surface sediment samples (DHS1, DHS2, and DHS3) from the eastern part of Daihai Lake were collected using a gravity corer in April 2006, and undisturbed surface 2-cm sediments were used for ^{14}C dating. Another 2 samples (DHS4 and DHS5) from the western part of Daihai Lake were collected in June 2007 using the same method as the former 3 samples. Two aquatic plant (*Myriophyllum*) samples were collected from the east and west ends of the lake in April 2006. A live fish (*Cyprinus carpio*) was captured by a fisherman when we collected surface sediments in April 2006. The fish bone was collected and the flesh discarded. A water sample (DHW1) was collected 1 m below the water surface, while sample DHW2 was collected at 9 m depth, in the same location as DHW1. Another 2 water samples (DHW3 at 1 m depth and DHW4 at 7 m depth) from the same site in the western part of Daihai Lake were collected in June 2007. Four soil samples (DHT1 to DHT4) were taken from the beach 10 cm beneath the surface.

Surface sediments and soil samples were treated with 5% HCl to remove inorganic carbon after manually removing visible plant remains and roots. Samples were then burnt with excess oxygen at 400 °C to combust bulk organic matter (McGeehin et al. 2001). Aquatic plant samples and fish bones were cleaned with 5% HCl and washed with distilled water several times, then freeze-dried before burning with excess oxygen at 400 °C to extract CO_2 from the bulk organic matter. Water samples were sealed in glass vessels and added excess CaCl_2 as soon as samples were taken, and boiled thereafter to obtain crystal CaCO_3 as ^{14}C dating material.

DHS1 and DHS2 were dated at Peking University using accelerator mass spectrometry (AMS). The other samples were dated in State Key Laboratory of Lake Science and Environment, Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, using liquid scintillation counting (LSC). The measured ^{14}C activities of samples were noted as X_s and expressed as ratios relative to the ^{14}C standard specific activity of carbon of Chinese sugar (1970), which is 1.362 times the specific activity of the NBS oxalic acid standard using the ^{14}C half-life of 5730 yr (Qiu et al. 1990).

RESULTS

^{14}C activity of the sampled surface sediments, fish bones, aquatic plants, DIC of lake water and beach soil varied between 1.003 and 1.147. Their apparent ^{14}C ages were calculated according to Equation 1 (Figure 2, Table 1). The apparent ^{14}C age of the fish bone was 2513 ± 73 yr BP. Aquatic plants in the east part of Daihai Lake were much older than that in western part, being 2528 ± 84 and 1870 ± 70 yr BP, respectively. The apparent ^{14}C ages of surface sediment varied between 1865 and 2132 yr BP. The bulk soil samples' apparent ^{14}C ages varied remarkably between 1277 ± 77 and 2510 ± 90 yr BP. There was little difference in apparent ^{14}C ages at different depths in the same site, but the average apparent ^{14}C ages of lake water in the eastern and western part of Daihai Lake changed little (1655 and 1715 yr BP, respectively). All the ^{14}C data showed an "older" apparent age than expected, since the expected real age of surface sediments should be 0 yr.

DISCUSSION

Daihai Lake is a closed lake in which the residence time of lake water is very long. The evaporation is about 2.5 times the precipitation; therefore, the salinity and pH value are very high (~9.0) (Wu et al. 2007). The concentration of HCO_3^- is between 387.4 and 499.9 mg/L with an average value of 460.3 mg/L, while the concentration of CO_3^{2-} varies between 102.6 and 174.5 mg/L with an average value of 120.9 mg/L. All the lake water chemical characteristics imply that there should be a hard-water effect (Björck and Wohlfarth 2001), which is expected to be the major factor causing the older ^{14}C age of the lake sediment.

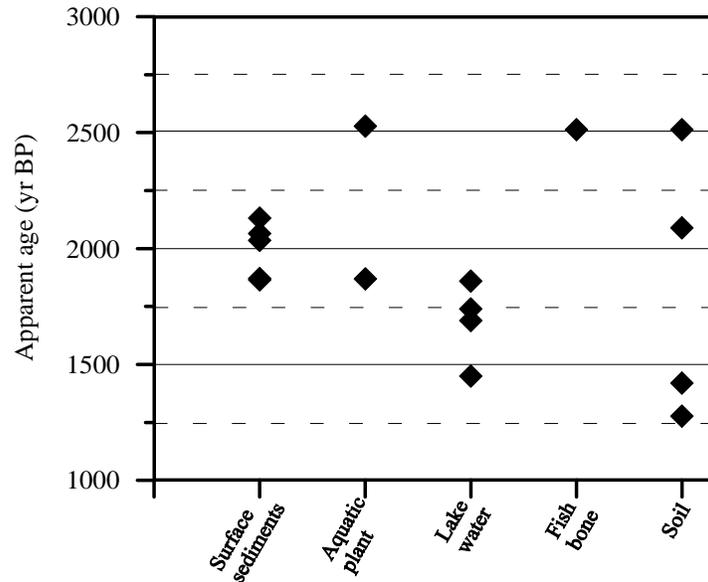


Figure 2 Apparent ^{14}C ages of surface sediment, aquatic plant, lake water, fish bone, and beach soil samples from Daihai Lake.

Based on the results of 4 lake water samples, the average ^{14}C activity level is 1.11 and the ratio of ^{14}C activity between lake water and atmospheric CO_2 (X_w/X_a) is 0.816. The hardwater effect ages of Daihai Lake can be calculated after Equation 1, as Stiller et al. (2001) have done in Lake Kinneret, on the assumption that X_w/X_a was constant in the past (Figure 3). The corresponding atmospheric ^{14}C levels are taken from Stuiver and Pearson (1993) and Pearson and Stuiver (1993). The hardwater effect ages varied between 949 to 1788 yr since 8000 yr BP, and increased notably from 5500 to 1000 yr BP.

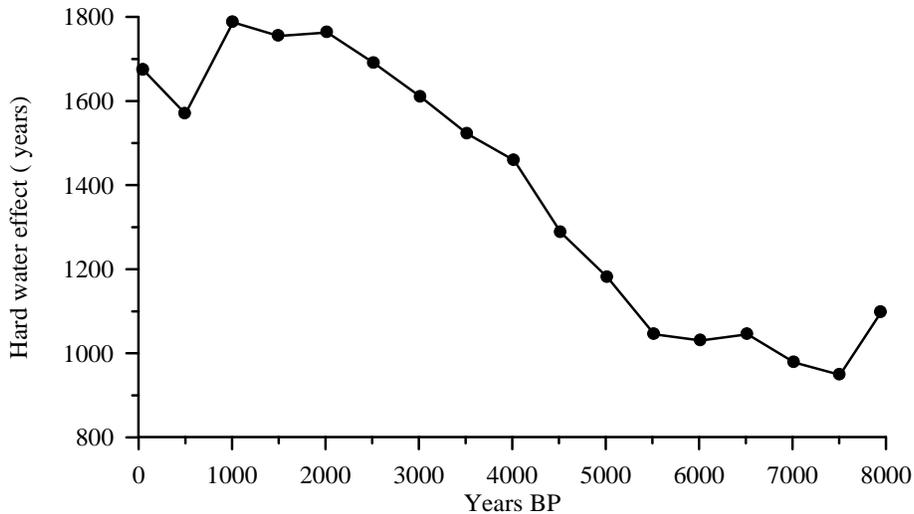


Figure 3 Inferred temporal variation of the hardwater effect in Daihai Lake since 8000 yr BP. The corresponding atmospheric ^{14}C levels are taken from Stuiver and Pearson (1993) and Pearson and Stuiver (1993).

The apparent ^{14}C ages of beach soil samples are generally older than their real ages. The possible interpretations include: 1) the sampling site was submerged in the past and the soil was formed from lake sediments according to the field investigation. Based on analyses of sedimentology, mineralogy, palynology, micropaleontology, and geochemistry of the core sediments, Wang et al. (1990) inferred that Daihai Lake maintained a high lake level at times of the Riss-Würm interglaciation and the early to middle Holocene, but assumed a general trend of contraction during the whole Late Pleistocene. Three beds of dark-colored mud deposited during the early to middle Holocene were considered to represent 3 episodes of extension of the lake around 10,000, 6500, and 4500 ^{14}C yr BP, respectively (Wang et al. 1990). The sites of 4 soil samples are close to the lake shore (Figure 1) and less than half a meter above the modern lake level, so that these sites could have been submerged during their history. 2) If the soil was formed in terrestrial conditions, the soil carbon can contain both recent terrestrial plant material and older residual carbon that is protected from decay either in aggregates or in organic-mineral complexes (Huang 2001).

Since Daihai Lake is a closed lake without any outflow, the water residence time should be very long, although no relevant study has been done thus far. The lake is covered with ~50 cm of ice from November to March (Xiao et al. 2005). We suggest that the possible factors causing the “older” ^{14}C age of lake sediments relate to the source of organic matter. The major source of organic matter in lake sediment is from endogenous plants (Meyers and Ishiwatari 1993); however, soil organic matter can be a substantial portion (Mayr et al. 2005). Thus, the hardwater effect, isotopic fractionation of aquatic plants, the reservoir effect, as well as soil organic matter together impact the ^{14}C age of lake sediments. Taking the hardwater effect and soil organic matter into account, we suggest that the correction age for lake sediment from Daihai Lake varied between 1000 and 2000 yr since 8000 BP, and stronger reservoir effects occurred since 5500 BP when the shrinkage of Daihai Lake began (Wang et al. 1990). This correction age is based on the presumption of a constant X_w/X_a . If we can get precise variation of X_w/X_a , for instance, when we can collect enough pore water for ^{14}C measurement, it should be possible to achieve a more accurate correction age.

CONCLUSION

Due to high salinity, high concentrations of HCO_3^- and CO_3^{2-} , and high pH value, we observe a hardwater effect in Daihai Lake. On the assumption that the relationship between the $^{14}\text{C}/^{12}\text{C}$ ratio of DIC and of atmospheric CO_2 was constant, the hardwater effect ages in the past can be estimated by correlation with atmospheric $^{14}\text{CO}_2$. In Daihai Lake, the X_w/X_a of present lake water is 0.816, and the hardwater effect ages during the past 8000 yr varied from 949 to 1788. We suggest that the hardwater effect is the major factor that has impacted the ^{14}C age profile of lake sediments in Daihai Lake.

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