# An organic carbon isotopic record of glacial-postglacial change in atmospheric $pCO_2$ in the sediments of Lake Biwa, Japan

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#### ABSTRACT

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A 12 m piston core of sediment from Lake Biwa, Japan, provides a high-resolution record of sedimentary organic matter accumulation over the Pleistocene-Holocene transition. Organic carbon concentrations are generally 1-2% in sediments deposited since 15 kyr ago. C/N ratios indicate that the organic matter is predominantly from aquatic production. A shift in organic carbon isotopic composition from -21% to -25% occurs over the glacial-postglacial boundary. Changes in lake productivity, organic matter preservation, sources of organic matter, and water temperature can be eliminated as probable causes of the observed organic carbon isotopic shift. Annual overturn of lake waters and rapid dissolution of biogenic carbonates maintain equilibrium between atmospheric  $CO_2$  and dissolved  $CO_2$  in this lake. The isotopic composition of organic matter buried in the sediments of Lake Biwa consequently appears to record the shift in atmospheric  $pCO_2$  from 200 ppm to 280 ppm which occurred at the Pleistocene–Holocene transition.

#### Introduction

The concentration of CO2 has varied in the atmosphere by 75-100 ppm between glacial and interglacial times. Neftel et al. (1982) measured concentrations of CO2 in air bubbles trapped in an ice core from Greenland and Barnola et al. (1987) made similar measurements in the Vostok core from East Antarctica to determine that atmospheric pCO<sub>2</sub> was about 200 ppm during the last glacial period, as opposed to about 280 ppm before the industrial revolution. Furthermore, the icecore record from Vostok reveals that the penultimate glacial atmosphere (160 kyr) also contained about 200 ppm. These records, from both boreal and austral locations, verify that glacial-interglacial fluctuations in atmospheric pCO<sub>2</sub> occur on a global scale. Such fluctuations in concentration are likely to affect carbon isotope fractionation by aquatic biota, which is dependent on dissolved  $CO_2$  concentration (Broecker, 1982). We present here evidence that sedimentation of the organic matter produced by freshwater organisms in Lake Biwa, Japan, over the past 15 kyr has produced a record of change in atmospheric  $pCO_2$ .

#### Samples and analysis

Sample location

Lake Biwa is located on the island of Honshu between Kyoto and the Sea of Japan (Fig. 1). The lake has an area of 674 km<sup>2</sup> and a maximum water depth of 104 m. The main North Basin has two depressions over 70 m deep and a mean depth of 41 m, making much of the this basin deep enough that bottom sediments are not easily disturbed by wave turbulence. Detailed paleolimnological investigations have been done on sediment floral and

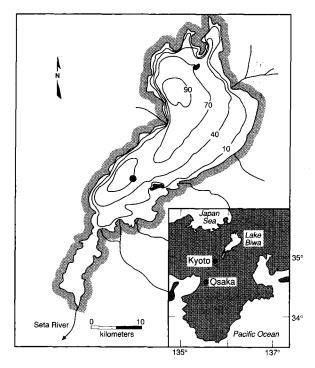


Fig. 1. Location of Lake Biwa on Honshu, Japan. Position of 1967 piston core site at 70 m water depth is shown. Depth contours given in meters. Mountains border the lake on the west and northwest; broad plains are present along most of the eastern shore. The major rivers entering the lake and the Seta River, the only river draining the lake, are shown.

faunal fossil contents, grain size distributions, mineralogy, physical and geophysical properties, and geochemistry (e.g., Horie, 1991). The sediment samples for our study were taken from a piston core which recovered 12 m of sediment in 1967 from a location in 70 m of water in the southern depression of the North Basin (Fig. 1). The sediments consisted of fine-grained clays which were apparently deposited under relatively uniform deepwater conditions. Biogenic carbonates do not accumulate in the sediments of Lake Biwa because of their rapid dissolution within the water column and at the sediment-water interface. Annual overturn of lake waters and the carbonate dissolution maintain equilibrium between atmospheric CO<sub>2</sub> and the dissolved CO<sub>2</sub> in this lake. Sedimentation of organic matter is the only significant process which incorporates carbon into the paleolimnologic record of Lake Biwa.

#### Sediment dating

The ages of three sediment horizons were estimated using radiocarbon dating of organic matter. Two of these dates are from a piston core obtained in 1965, and one is from the 1967 core which provided our samples. Both cores are from the same location (Fig. 1). Furthermore, seismic profiles show that layers of the upper 250 m of sediment are continuous across the area from which the two different piston cores were taken (Takemura, 1990). We have therefore assigned the ages from the 1965 core to the same depth horizons in the 1967 core. The radiocarbon ages for the 0.8 m and 4.5 m horizons of the 1965 core are 1430  $\pm$  95 and 3650  $\pm$  105 yr, respectively. The age for the 11.5 m horizon of the 1967 piston core is 14,980 + 460 yr (Horie et al., 1971). These ages are uncorrected for the recycling of detrital organic carbon in lakes which commonly renders <sup>14</sup>C ages 1000-2000 yr older than actual sediment ages (e.g. Stuiver, 1975; Rea et al., 1980). Extrapolation of the measured radiocarbon ages to the sediment-water interface gives an age of 1200 yr B.P. for modern sediments. We have not attempted to use this figure to correct the core ages because it is unlikely that the amount of detrital carbon has remained constant over the glacial-postglacial transition in this lake.

#### Organic carbon and C/N analyses

Sediment samples from the 1967 Lake Biwa piston core contain less than 0.2% CaCO<sub>3</sub> (Horie et al., 1971); consequently no effort was made to remove inorganic carbon from our samples. The sediment samples were simply dried at room temperature and coarsely ground in preparation for subsequent analyses. Measurements of total organic carbon and of elemental C/N ratios were made by high-temperature combustion using a Carlo Erba 1108 CHNS-O analyzer. Data have been corrected for minor amounts of contamination determined by blank runs and for response factors determined from analyses of authentic CHNS standards.

#### Organic carbon isotope analyses

Carbon isotope measurements were done in the Stable Isotope Laboratory at The University of Michigan. The <sup>13</sup>C/<sup>12</sup>C ratios of organic carbon were measured with a Finnigan Delta S mass spectrometer after combustion of the sediment samples. A combustion temperature of 800°C was used to guarantee complete oxidation of both reactive and resistant forms of organic matter. NBS standards were regularly and frequently used to calibrate the instruments. Results are reported relative to the PDB standard.

#### Results and discussion

#### The Lake Biwa sediment record

Accumulation of organic matter appears to have continued with little change over the past 15 kyr in Lake Biwa sediments. Organic carbon concentrations remain generally between 1 and 2.5% throughout the sediment record (Fig. 2). These values are typical of sediments of large oligotrophic lakes. C/N ratios vary over the narrow range of ca 6–10. Several samples at ca 9 m have ratios between 10 and 12; these samples also have enhanced carbon concentrations. Nakai and Koyama (1987) measured the C/N ratios of 7–8 in samples of mixed plankton filtered from the waters of Lake Biwa and of over 30 in plants from the surrounding land areas. The correspondence

between the lake sediment C/N ratios and those of the plankton samples indicates that the dominant source of organic matter to the lake sediments has been phytoplankton throughout the cored sediment record. A small increase in organic carbon concentrations is evident in sediments shallower than 10 m in the core (Fig. 2). An increase in burial of aquatic organic matter is consistent with the enhanced productivity and sedimentation rates in postglacial times inferred by Meyers et al. (1993).

Unlike the total organic carbon and the C/N ratios, organic carbon isotopic compositions show a major change within this core. A shift of ca -4‰ occurs between sediments deposited ca 15 kyr and ca 3.6 kyr (Fig. 2). Sediments shallower than 6 m have  $\delta^{13}$ C values between -25 and - 26‰. These isotopic ratios are within the ranges of -25 to -28% for mixed plankton samples and -25 to -30% for land plants measured by Nakai and Koyama (1987). The similarity between the isotopic compositions of aquatic and terrigenous organic matter means that  $\delta^{13}$ C values cannot be used to identify organic matter sources in the modern Lake Biwa system. There are to be two excursions towards lighter values—one between near 9 m and another near 10 m—which punctuate the pattern of heavier isotopic values in sediments deeper than 7.5 m. These excursions correspond to increases in C/N ratios at these sediment depths (Fig. 2).

Pollen assemblage data from Horie et al. (1971)

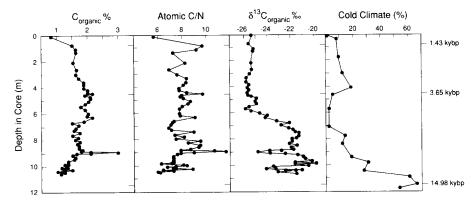


Fig. 2. Organic carbon concentrations, atomic C/N ratios, and organic  $\delta^{13}$ C values of samples from 12 m piston core of Lake Biwa sediment. Climate record is inferred from pollen contents of lake sediments (Horie, 1991). Organic carbon <sup>14</sup>C ages determined by Horie et al. (1971) are given.

show a shift from regionally cold climate to a more temperate climate in sediments from near the bottom of this core (Fig. 2). The pollen data confirm that the piston core recovered part of the sediment record of the latest Pleistocene.

#### Isotopic fractionation in aquatic organic carbon

Organic matter synthesized by aquatic organisms can provide an indirect record of fluctuations in atmospheric CO<sub>2</sub> concentrations. Biota selectively incorporate 12C during formation of organic matter, and the amount of discrimination against <sup>13</sup>C depends on the concentration of dissolved CO<sub>2</sub> available to phytoplankton (cf. O'Leary, 1981; Freeman and Hayes, 1992). If dissolved CO<sub>2</sub> is in equilibrium with atmospheric CO<sub>2</sub>, then the carbon isotope ratios of sedimentary organic matter can provide a generally reliable proxy record of change in atmospheric pCO<sub>2</sub> (Rau et al., 1991). If the concentration increases, then the dynamics of photosynthetic carbon fixation should enhance the difference between the 13C contents of dissolved CO2 and those of organic carbon (Popp et al., 1989; Freeman and Hayes, 1992), that is, towards more negative  $\delta^{13}$ C values in the organic carbon.

A number of factors can complicate the isotopic record of  $pCO_2$  change which may be preserved in sedimentary organic matter. When aquatic productivity is high, for example, preferential biotic uptake of  $^{12}C$  can shift the isotopic ratio of dissolved  $CO_2$  towards heavier values, that is, less negative  $\delta^{13}C$  values (McKenzie, 1985; Hollander and McKenzie, 1991). Recycling of isotopically light land-derived organic carbon in coastal marine and lacustrine waters can create an isotopic shift to lighter values in algal organic matter. Finally, selective diagenesis of isotopically heavy components of organic matter has been shown to cause a shift of as much as +2% in sedimentary organic matter (Spiker and Hatcher, 1984).

Two marine examples of the organic carbon isotope record in sediments suggest that natural systems largely avoid the potential complications that might confuse the paleo-pCO<sub>2</sub> record. Sediments from DSDP Site 619 in the Pygmy Basin (northern Gulf of Mexico), for example,

record a  $\delta^{13}$ C source change from predominantly marine-derived organic matter in the present and the previous interglacial times to a larger proportion of land-derived organic matter in the last glacial period (Jasper and Gagosian, 1989). The carbon isotopic composition of methyl ketones, which are synthesized only by marine phytoplankton and can be isolated from sedimented organic matter, nonetheless shows a clear record of concomitant change in atmospheric CO<sub>2</sub> content. A shift of -5% from glacial to modern times occurs, and this corresponds to an increase of about 90 ppm in pCO<sub>2</sub> (Jasper and Hayes, 1990). The carbon isotope contents of bulk organic matter present in sapropels in the eastern Mediterranean Sea display almost identical shifts as found in the methyl ketones, ca -4%, between glacial and interglacial sediments deposited over the past 440 kyr (Fontugne and Calvert, 1992). Changes in atmospheric CO<sub>2</sub> concentrations correlate well with these fluctuations and are probably principal causes, although changes in isotopic composition of CO<sub>2</sub>(aq) due to regional freshwater inputs or temporary upwelling may have possibly been involved.

Two lacustrine examples of isotope records similarly indicate that sedimentary organic carbon retains a record of atmospheric  $pCO_2$ . Sediments from Meerfelder Maar in Germany (Brown et al., 1991) and a lake in Sweden (Haakanson, 1986) exhibit shifts of about -8% in organic carbon  $\delta^{13}$ C values at the Pleistocene/Holocene boundary. The glacial-postglacial transition has been accompanied by important changes in land vegetation in these two examples; consequently, the possibility of changes in the source of organic matter has hindered conclusive interpretation of the isotopic shifts. Unlike these areas, where tundra was replaced by more recent forests, Honshu has been forested continually since the Pliocene (Fuji, 1988).

#### Interpretation of the Lake Biwa isotopic record

Several possible paleoenvironmental factors may participate in creating the glacial-postglacial shift found in the carbon isotopic composition of organic matter in Lake Biwa sediments: Effects of changes in the sources of organic matter to Lake Biwa sediments

It is possible that the proportions of aquatic and land-derived organic matter changed over the glacial-postglacial transition in Lake Biwa. Fuji (1988) notes that interglacial periods have been wetter than glacial periods, resulting in greater amounts of land runoff which would wash more land-plant debris into the lake. Land plants typically have higher C/N ratios than found in plankton, and this diagnostic difference has been confirmed for Lake Biwa biota (Nakai and Koyama, 1987). A general correlation between higher concentrations of organic carbon and higher C/N ratios is found (Fig. 3), suggesting that contributions of land-derived organic matter have indeed added to lake-derived material. Peak concentrations of organic carbon correlate with peak C/N values (Fig. 2), yet the C/N ratios remain well below 20, the threshold value diagnostic of landderived organic matter. Greater runoff would also wash in more nutrients and thereby enhance aquatic productivity. Meyers et al. (1993) conclude from grain size and diatom assemblage data that both sedimentation rates and aquatic productivity rates increase during interglacial periods. Unlike

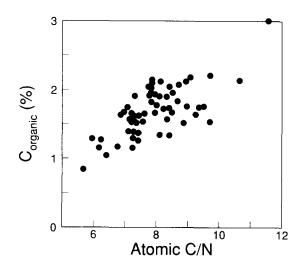


Fig. 3. Relationship between organic carbon concentrations and atomic C/N ratios of samples from 12 m piston core of Lake Biwa sediment. Positive correlation implies that variable contributions of land-derived organic matter have augmented the principal, algal source of organic matter to sediments.

in ocean margin locations where a source change is evident in the carbon isotopic record (cf. Jasper and Gagosian, 1989; Jasper and Hayes, 1990), a change in the aquatic/land ratio would be concealed in Lake Biwa. Nakai and Koyama (1987) have shown that the isotopic compositions of modern plankton (-25 to -28%) and of land plants (-25 to -30%) are indistinguishable. The lack of major change in organic matter C/N ratios (Figs. 2 and 3) can be used, however, to conclude that a large source change has not occurred and that phytoplankton have been the principal source of the sedimentary organic matter throughout the 12 m core record.

#### Effects of changes in aquatic productivity

Fluctuations in lake productivity are capable of creating variations in the carbon isotopic composition of aquatic organic matter (e.g. McKenzie, 1986; Hollander and McKenzie, 1991). During periods of enhanced productivity, CO<sub>2</sub>(aq) becomes depleted, and phytoplankton discriminate less towards selective uptake of <sup>12</sup>C. If the 4‰ shift shown in Fig. 2 were caused by a change in aquatic productivity, then rates were higher during the last glacial and diminished into postglacial times. This is contrary to the diatom evidence which indicates that postglacial productivity has been greater than during glacial times (Mori and Horie, 1984). It remains possible, however, that the change to a more productive phytoplankton assemblage has had a species effect on the isotopic composition of aquatic organic matter.

## Effects of diagenesis on the isotopic content of sediment organic matter

Selective diagenesis has been suggested as a possible cause of change in the isotopic composition of sedimentary organic matter. A diagenetic shift of -2% in organic matter has been found in the sapropelic sediments of Mangrove Lake, Bermuda (Spiker and Hatcher, 1984). The organic carbon content of these sediments is over 20%, very unlike the concentrations in Lake Biwa (Fig. 2). The Mangrove Lake isotopic shift is created by the existence of large proportions of

diagenetically sensitive, isotopically heavy amino compounds and sugars which can be selectively destroyed to shift the bulk organic matter isotopic composition. These compounds are abundant only in sediments having extraordinarily well preserved organic matter and not in more typical lacustrine sediments like those of Lake Biwa. The preserved glacial—interglacial isotopic shifts documented by Jasper and Hayes (1990) and Fontugne and Calvert (1992) in marine sediments show that diagenesis does not significantly alter the isotopic composition of bulk organic matter in sediments having typically low (<2-3%) concentrations of organic carbon.

## Effects of changes in the availability of dissolved $CO_2$

Several processes modify the availability of dissolved CO<sub>2</sub> in lakes and thereby impact the isotopic composition of aquatic organic matter. First, periods of high productivity and associated depletion of dissolved CO<sub>2</sub> in the photic zone can lead to organic matter less depleted in <sup>13</sup>C than normal (e.g. Hollander and McKenzie, 1991). Second, enhanced mixing or overturn of lake waters can return isotopically light CO<sub>2</sub> derived from oxidation of organic matter to the photic zone for reutilization by phytoplankton, thereby producing organic matter which is even more isotopically lighter (McKenzie, 1985). Third, cooler waters accommodate higher concentrations of dissolved CO<sub>2</sub>, permitting phytoplankton to incorporate <sup>12</sup>C more selectively than in warmer waters (Rau et al., 1989) and therefore to synthesize isotopically lighter organic matter. Finally, glacial-interglacial changes in atmospheric pCO<sub>2</sub> are recorded in the carbonate and organic carbon isotopic compositions of marine and probably lacustrine organic matter (Jasper and Hayes, 1990; Fontugne and Calvert, 1992). Aquatic plants are not able to discriminate so strongly towards 12C during glacial times of lowered pCO<sub>2</sub> and consequent limited availability of dissolved CO<sub>2</sub>.

Both the temperature change and the productivity change can be ruled out as causes of the Lake Biwa carbon isotope shift to lighter  $\delta^{13}$ C values in recent sediments. The effects of these two

factors would cause a shift to heavier, not lighter, modern values. Enhanced mixing and overturn of lake waters would have a one-time impact but then simply establish more rapid equilibration with atmospheric CO<sub>2</sub>; this factor consequently cannot be responsible for the sustained isotopic change.

#### The answer-change in atmospheric pCO<sub>2</sub>

The most likely cause for the carbon isotopic shift present in Lake Biwa sediments is the glacial postglacial change in atmospheric pCO<sub>2</sub> documented in occluded air bubbles by Neftel et al. (1982) and Barnola et al. (1987). The decrease in atmospheric CO2 would lead to lowered equilibrium concentrations of CO<sub>2</sub> dissolved in lake waters and diminished discrimination by phytoplankton towards 12C. As determined by Jasper and Hayes (1990) for organic matter synthesized by marine plankton, the amount of diminished discrimination would be ca 5%, which is approximately the amount of change found in the Lake Biwa isotope record. The isotopic composition of organic matter in Lake Biwa sediments consequently appears to preserve a record of the change in atmospheric pCO2 which occurred at the Pleistocene-Holocene transition.

The lower  $pCO_2$  of the glacial-age atmosphere would affect isotopic discrimination by C3 plants utilizing CO<sub>2</sub> dissolved in water but not those taking up gaseous CO<sub>2</sub>. Organic matter produced by land plants therefore would not exhibit the shift in isotopic composition that would be evident in aquatic organic matter. The excursions to lighter isotopic values at ca 9 m and 10 m in the Lake Biwa record (Fig. 2) correspond to peaks in organic carbon concentration and in C/N ratios. The isotopic composition of Pleistocene land plants was undoubtedly the same as modern plants growing around Lake Biwa, ca -25 to -30% (Nakai and Koyama, 1987), whereas the composition of aquatic plants was probably heavier. Sediments at the 9 and 10 m horizons evidently record two brief intervals of increased wash-in of land-derived organic matter to Lake Biwa near the end of the latest glacial period.

The temporal difference between appearance of the changes in organic matter  $\delta^{13}$ C values and in

proportion of cold climate pollen in this core suggests that the climate of Japan began to warm before the atmospheric  $pCO_2$  began to increase (ca 10 kyr: Barnola et al., 1987). Information from nearby areas of the north Pacific support this inference. In particular, Chinzei et al. (1987) conclude from microfossil data that the warm and northward flowing Kuroshio Current had migrated northward to flow past southern Honshu by 13 kyr. The return of this current from its glacial, more southward location evidently ameliorated the regional climate around Lake Biwa prior to 10 kyr.

#### **Summary and conclusions**

The 1967 12-m piston core of Lake Biwa sediment provides a high-resolution record of climate change and of paleolimnologic history for Japan dating to ca 15 kyr. The principal source of organic matter to the sediments of Lake Biwa over the core record is phytoplankton production. Several brief episodes of greater-than-normal delivery of land-derived organic matter in the latest Pleistocene are preserved as excursions in the organic C/N and carbon isotopic ratios. These episodes may possibly record interludes of wetter climate, but they are more likely to represent tectonically induced turbidity flows or slumps of nearshore sediment. The elemental and isotopic excursions are especially important in confirming that the carbon isotopic content of late Pleistocene algal organic matter differs from that of modern plankton.

A shift of ca -4% occurs in the organic carbon  $\delta^{13}C$  of sediments deposited between radiocarbon ages of 15 kyr and 3.6 kyr. The most plausible explanation for this shift is the documented change in atmospheric  $pCO_2$  from 200 ppm at the last glacial maximum to 280 ppm throughout most of the Holocene. The diminished availability of dissolved  $CO_2$  is reflected in the heavier isotopic composition of Pleistocene algal organic matter. A similar shift in the isotopic content of algal organic matter has been reported from several marine areas and has been postulated to occur in lakes. The Lake Biwa record confirms that freshwater algae mimic the marine algal response to changes in atmospheric  $CO_2$  concentrations.

The shift from heavier late Pleistocene to lighter Holocene carbon isotope values can be used to identify the Pleistocene–Holocene boundary in the sediments of Lake Biwa. Pollen contents of these sediments indicate that climate in the Lake Biwa region began to ameliorate before 10 kyr, probably because warm-water conditions returned to nearby areas of the North Pacific Ocean and the Sea of Japan in the late Pleistocene.

Our successful investigation of the use of organic carbon isotopic ratios in Lake Biwa sediments as a measure of past concentrations of atmospheric CO<sub>2</sub> opens opportunities for future studies in this and other lakes. In particular, the 250 m deep T bed of the 1982 drilled core in Lake Biwa is an apparently uninterrupted sequence of deep lake sediments which records the past 0.5 myr of depositional history (Meyers et al., 1993). A highresolution record of changes in atmospheric pCO<sub>2</sub> over the last five glacial-postglacial cycles should be present in the organic matter contents of this cored sedimentary sequence. Similar highresolution records of regional and global climate change should be available from the sediments of other ancient lakes.

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