

## Characterization ascertained from $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ of particulate organic matter in surface water from a shallow and semi-closed Lake Kiba

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Concentrations of stable and radioactive carbon isotopes of organic matter were determined for suspended solids in surface water from the shallow and semi-closed Lake Kiba, Japan during June 2014 – March 2016. Particulate organic carbon (POC) concentrations were 0.44–5.01 mg L<sup>-1</sup>, and  $\delta^{13}\text{C}$  and  $\Delta^{14}\text{C}$  values were, respectively, –30.3 to –22.8 ‰ and –156 to –33 ‰. The organic matter in suspended solid samples consistently showed depleted  $^{14}\text{C}$ , with an average  $\Delta^{14}\text{C}$  value of  $-81 \pm 37\%$ . The carbon isotopes showed seasonal variation, being higher in summer, and with a positive correlation to POC contents. These results suggest that the POC characteristics are controlled by a mixture of two endmembers, organic matter produced by phytoplankton activity within the lake and the watershed organic matter, under a shallow and semi-closed environment.

### 1. Introduction

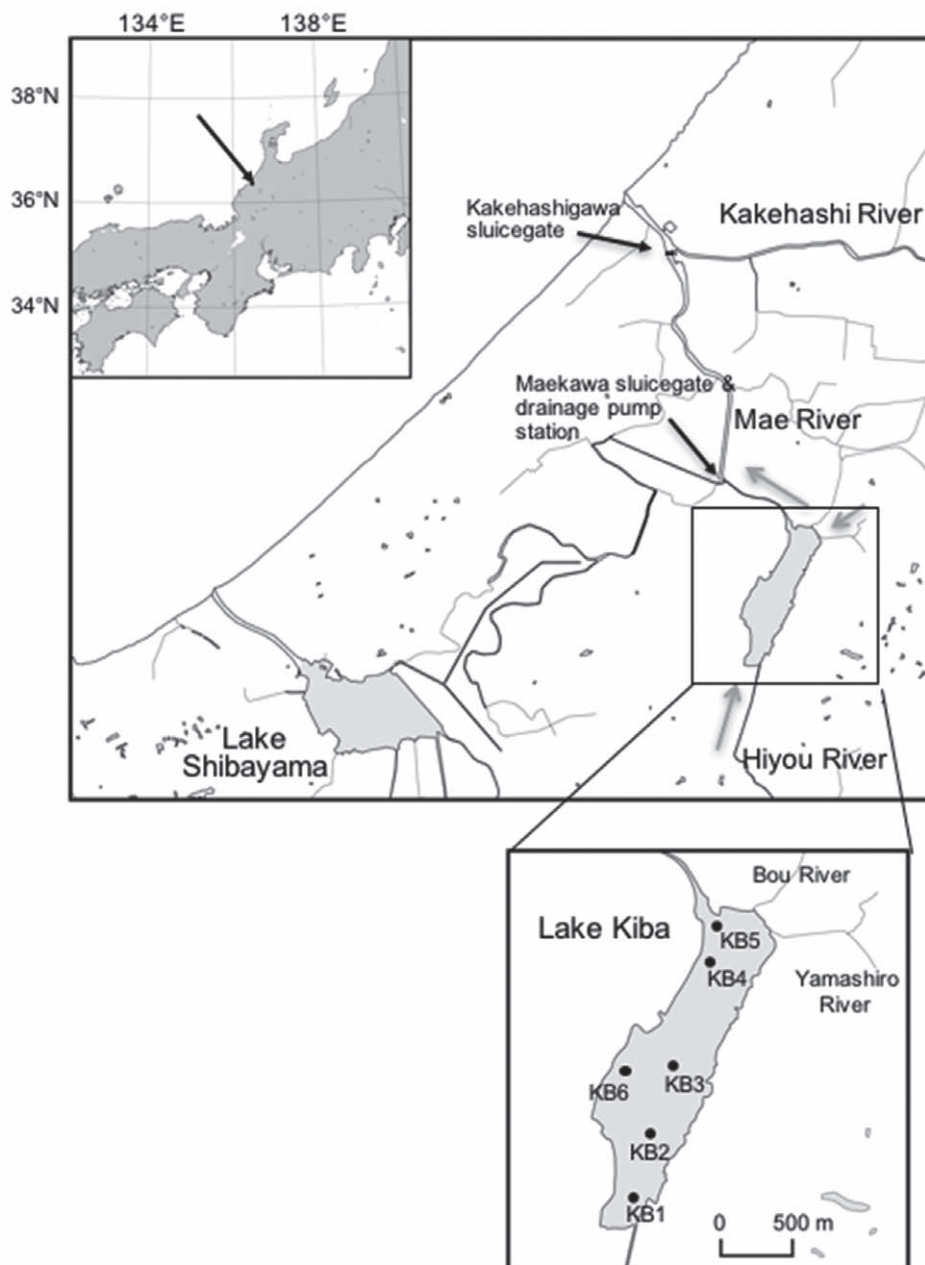
Radiocarbon abundance increased in the natural environment after nuclear weapons testing in the 1950s and 1960s injected large quantities of  $^{14}\text{C}$  into the atmosphere [1,2]. The  $\Delta^{14}\text{C}$  values of organic matter in river suspended particles are –980 to +75‰, but plankton and particulate organic carbon in marine environments have enriched  $^{14}\text{C}$  values of –45 to +110‰ [1-3]. Radiocarbon is useful as a tracer of the dynamics of organic matter in river systems and lake environments because of differences in the contribution of each source with a wide range of  $\Delta^{14}\text{C}$  values [4-6]. The paired  $\Delta^{14}\text{C}$  versus  $\delta^{13}\text{C}$  distributions vary among river systems. The distributions are categorized into three groups based on watershed conditions such as land-use type of wetlands, forest-paddy fields, and permafrost watersheds [7-9]. Riverine particulate organic matter (POM) from wetlands has lower  $\delta^{13}\text{C}$  and higher  $\Delta^{14}\text{C}$  than those of rivers in forests and fluvial plains, which indicates a greater contribution of apparently younger organic matter in wetland river systems. In lake environments, carbon isotopes have been applied to elucidate biogeochemical cycles and freshwater reservoir effect ranges and variations over time [6,10,11]. The phytoplankton  $\Delta^{14}\text{C}$  reflects the  $\Delta^{14}\text{C}$  of dissolved  $\text{CO}_2$ , which might be a mixture of aged and modern carbon [12-14]. Particulate organic carbon (POC) has a wide range of  $\Delta^{14}\text{C}$  extending from –443 to +68‰ in these lakes. Such  $\Delta^{14}\text{C}$  values have been useful in assessing the age, reactivity, and sources of carbon in lake systems. Oguri *et al.* [15] and Ueda *et al.* [16] have demonstrated that radiocarbon behavior in the Lake Obuchi and Lake Hamana ecosystems is influenced strongly by environmental factors.

Lakes play important roles in the carbon cycle through carbon sequestration in sediments and efflux of  $\text{CO}_2$  to the atmosphere [17-20]. Some lake environments in economically

developing countries have sustained significant organic pollution during the last decade because of rapid economic development and increased human populations around watersheds. Organic pollution also has been observed in lake environments at past and present in developed countries. In Japan, water quality survey results for lakes and reservoirs in 2014 indicated that the compliance rate of environmental quality standards for living environmental items is 55.5% for chemical oxygen demand (COD) concentrations [21]. Another water quality problem is the slight increase of COD concentrations in lakes, although total nitrogen, phosphorus, and biochemical oxygen (BOD) concentrations are constant or decreasing with time [22-24]. The increasing annual measurements of COD appear to result from accumulation of refractory organic matter in the lakes. Therefore, the characterization of organic matter is important to elucidate organic matter dynamics, geochemical roles and organic pollution mechanisms in lake systems.

Lake Kiba (Kiba-gata) is located in Komatsu City, Ishikawa Prefecture, Japan (Figure 1) and is classified as a lagoon. The lake has a mean water depth of 1.7 m, area of 1.14 km<sup>2</sup>, and water volume of  $1.7 \times 10^4$  m<sup>3</sup>. Its watershed area is 38 km<sup>2</sup>. In 1932, a sluiceway was constructed at the conjunction of the Kakehashi River to protect against inflow of water from the river during flood events (Figure 1). Lake Kiba was affected by changes in the drainage system and land consolidation around the lagoon during 1954–1969 [25]. One example in that period was construction of the Maekawa sluiceway and drainage pump station at the Mae River to control the Lake Kiba water level. The organic matter flux recorded in the sediment core increased from 1.1 to 2.3 and from 3.9 to 7.5 mg C cm<sup>-2</sup> y<sup>-1</sup>, respectively, during 1903–1974 and 1989–2012, although these values were similar to the flux recorded for 1974–1989 following reclamation [26]. The COD concentration was at a maximum, 11 mg L<sup>-1</sup> in 1990, corresponding to the second worst value seen in Japan at that time. It has remained about double the national standard of this lake class

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**Figure 1.** Sampling sites KB1 to KB6. Water sampling was done at KB3. All six sites were used for surface sediment sampling. Numerical map data are based on Fundamental Geospatial Data provided by the Geospatial Information Authority of Japan, and the National Land Numerical Information provided by the Ministry of Land, Infrastructure, Transport and Tourism, Japan.

(Class A, COD concentration  $< 3 \text{ mg L}^{-1}$  [27]) during the last decade [28], although treatments for water quality improvement have been conducted since 1991.

The objectives of this study were to elucidate the characteristics of POM for the shallow and semi-closed Lake Kiba throughout a year based on monthly observations obtained using data for carbon isotopes,  $\delta^{13}\text{C}$  and  $\Delta^{14}\text{C}$ . The simultaneous use of  $\Delta^{14}\text{C}$  and  $\delta^{13}\text{C}$  values adds a second dimension to observations of carbon cycling in surface aquatic environments [1, 8, 19]. During the sampling period, the fundamental physicochemical parameters, POC concentrations, and carbon isotope concentrations were ascertained. Results of this study are expected to provide basic and useful information to clarify organic pollutants affecting shallow lakes and lagoons.

## 2. Materials and methods

Figure 1 shows sampling sites KB1 to KB6 in Lake Kiba. Water sampling was done at KB3 and all six sites were used

for surface sediment sampling.

Water sampling was conducted monthly during June 2014 – March 2016 at KB3, in the central part of the lake. Collection of the surface water samples was done from a boat using a bucket. During 18 November – 16 December 2016, sampling was also conducted weekly to elucidate changes in water quality during water quality improvement experiments using the Maekawa sluiceway and drainage pump station, as presented in Figure 1 [29]. Detailed results of those experiments will be reported elsewhere.

Water characteristics such as water temperature (WT), pH, conductivity, and turbidity in the surface water were measured using a portable water quality meter (WQC-24; DKK-TOA Corp.). Chlorophyll-a concentration was photometrically quantified using the trichromatic absorption method after extraction of particles using a  $0.45 \mu\text{m}$  membrane filter (Advantec MFS Inc.) with 90% acetone [30]. Samples of chlorophyll-a were collected at a site close to KB3 during monthly research conducted by the Komatsu City Government.

About 60 L of water was taken into polyethylene containers and these were then transferred to the laboratory. Suspended particles in lake water samples were concentrated with a single-bowl continuous-flow centrifuge with a flow rate of 15 L  $\text{h}^{-1}$  [7]. The inside temperature was maintained at 10–20°C to avoid transformation of solids. Solid samples were dried at room temperature or freeze-dried. The suspended solids (SS) concentration was measured using the weight of the suspended solids.

Surface sediments were collected at the six stations (Figure 1) using an Ekman–Birge grab sampler in 2012 and 2013. The sediment samples were freeze-dried and ground in an agate mortar after sieving with a 2-mm mesh size sieve.

Organic carbon contents in suspended solids and sediment samples were determined using an elemental analyzer (NA2500, CE Instrument or Flash EA1112; Thermo Fisher Scientific Inc.). Before analysis of the suspended solids, carbonates were removed by adding 1 M HCl solution. The respective levels of precision of TOC and TN analyses were  $\pm 0.009\%$  and  $\pm 0.003\%$ . The POC concentration is calculated by POC content divided by lake water volume at continuous flow centrifugation. Stable carbon isotopes were assessed for sub-samples of the  $\text{CO}_2$  gas generated during graphite production for radiocarbon analysis using a mass spectrometer (Delta V Plus; Thermo Fisher Scientific Inc.), or were measured using an EA-IRMS analyzer (Delta V coupled to Flash EA1112; Thermo Fisher Scientific Inc.). The  $\delta^{13}\text{C}$  value is expressed as follows.

$$\delta^{13}\text{C} = \left[ \left( \frac{^{13}\text{C}/^{12}\text{C}}{^{13}\text{C}/^{12}\text{C}} \right)_{\text{sample}} / \left( \frac{^{13}\text{C}/^{12}\text{C}}{^{13}\text{C}/^{12}\text{C}} \right)_{\text{standard}} - 1 \right] \times 1000. \quad (1)$$

Here,  $(^{13}\text{C}/^{12}\text{C})_{\text{sample}}$  is the abundance ratio of  $^{13}\text{C}$  to  $^{12}\text{C}$  of the sample.  $(^{13}\text{C}/^{12}\text{C})_{\text{standard}}$  is the ratio of VPDB: ANU-Sucrose ( $\delta^{13}\text{C} = -10.80\%$ ) and L-Alanine ( $\delta^{13}\text{C} = -19.6\%$ ).

$^{14}\text{C}$  measurements were done using an accelerator mass spectrometer at the Beta Analytic Radiocarbon Dating Laboratory. For this study,  $\delta^{14}\text{C}$  is defined as the deviation in parts per thousand from the modern standard (NBS oxalic acid: SRM-4990C):

$$\delta^{14}\text{C} = \left[ \left( \frac{^{14}\text{C}/^{12}\text{C}}{^{14}\text{C}/^{12}\text{C}} \right)_{\text{sample}} / \left( \frac{^{14}\text{C}/^{12}\text{C}}{^{14}\text{C}/^{12}\text{C}} \right)_{\text{standard}} - 1 \right] \times 1000. \quad (2)$$

The  $\Delta^{14}\text{C}$  value is expressed as follows [31]:

$$\Delta^{14}\text{C} = \delta^{14}\text{C} - 2(\delta^{13}\text{C} + 25) (1 + \delta^{14}\text{C}/1000). \quad (3)$$

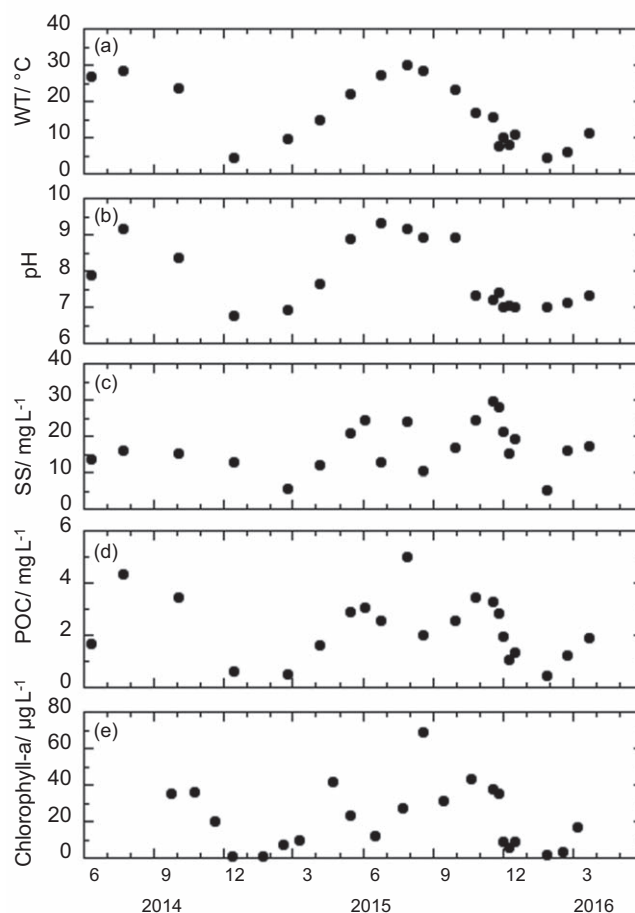
The  $\Delta^{14}\text{C}$  values are normalized to the base value of  $\delta^{13}\text{C} = -25\%$  of standard carbonate (VPDB) to correct for isotopic fractionation of  $^{12}\text{C}$  and  $^{13}\text{C}$ . A positive  $\Delta^{14}\text{C}$  value reflects the presence of  $^{14}\text{C}$  released from nuclear weapon tests in the 1950s and 1960s.

### 3. Results and discussion

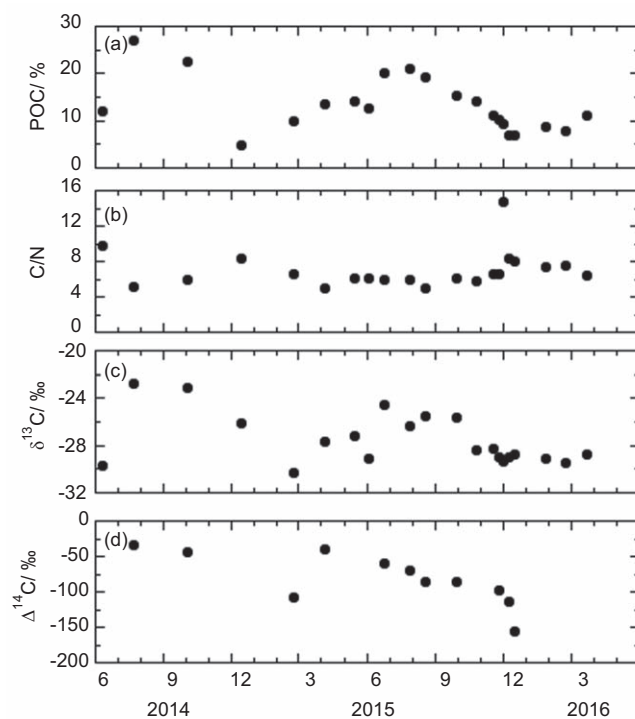
#### Water quality

Water quality data during the sampling period (June 2014 – March 2016) are shown in Figure 2. Water temperature (WT) and pH show seasonal variation. They are higher in summer. Increased pH results from utilization of  $\text{CO}_2$  for phytoplankton activity [32]. Suspended solids (SS) and particulate organic carbon (POC) concentrations are higher in summer (July–August) and lower in winter (December–February). The chlorophyll-a concentration has two peaks in April and August 2015. The POC concentration in the unit of  $\text{mg L}^{-1}$  is correlated with water temperature and pH ( $r = 0.67$ – $0.70$ ,  $p < 0.05$ ), but the POC content in unit of %, as portrayed in Figure 3, shows a stronger positive correlation with water temperature and pH ( $r = 0.87$ ,  $p < 0.0001$ ). This result suggests that the increase in POC concentration is related to phytoplankton

activity within the lake.



**Figure 2.** Variation of water temperature (WT), pH, suspended solids (SS), particulate organic carbon (POC) concentration and chlorophyll-a concentration in surface water from Lake Kiba.



**Figure 3.** Variation of POC content, particulate organic carbon/nitrogen (C/N),  $\delta^{13}\text{C}$  and  $\Delta^{14}\text{C}$  values of POC in surface water from Lake Kiba. Error bars correspond to  $1\sigma$  uncertainty on  $\Delta^{14}\text{C}$  measurements.

*Carbon isotopes of particulate organic matter*

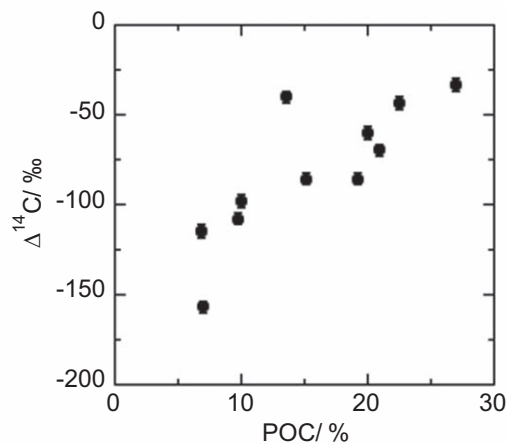
To understand organic matter dynamics through a year, focus was placed on POC content and its characteristics. The POC contents were 4.9–27%, showing a maximum in July–September (Figure 3). The maximum value in 2014 is higher than that of 2015. The C/N ratio varies from 5.0 to 9.8, except for the sample on 2 December 2015. The highest value indicates a larger contribution of soil organic matter and land plant in the watershed [33]. The  $\delta^{13}\text{C}$  value of organic matter in the suspended solids was  $-30.3$  to  $-22.8\%$ . The maximum value in 2014 was higher than that of 2015. These values show similar variation as the POC contents ( $r = 0.80$ ,  $p < 0.0001$ ) do. The  $\delta^{13}\text{C}$  values also correlate with pH of the lake water ( $r = 0.75$ ,  $p < 0.0005$ ). Similar results have been reported for a shallow lake environment [32,34]. The  $\delta^{13}\text{C}$  values of phytoplankton increase because of the reduced isotope fractionation of  $^{12}\text{C}$  and  $^{13}\text{C}$  during photosynthesis, when the phytoplankton biomass increases or the supply of dissolved inorganic carbon is limited [32,35].

The  $\Delta^{14}\text{C}$  value of POC in the Lake Kiba water is more depleted than in the present atmospheric  $\text{CO}_2$ , ca. 30‰ in 2012 [36], exhibiting an average value of  $-81 \pm 37\%$ , although the maximum  $\Delta^{14}\text{C}$  value is observed in summer with higher pH, POC, and chlorophyll-a concentration. The lake water POC has lower  $\Delta^{14}\text{C}$  than inorganic carbon, plankton, and present biota in the river and lake watershed [16,37]. Increases in  $\delta^{13}\text{C}$  and  $\Delta^{14}\text{C}$  values of dissolved organic carbon have been observed in the water of Lake Kasumigaura during spring because of the primary production of phytoplankton, although  $\Delta^{14}\text{C}$ -DOC was more depleted than atmospheric  $\text{CO}_2$  [38]. Lake Kasumigaura is classified as a shallow water lake and it shows depleted  $^{14}\text{C}$  of organic matter, similar to the situation for Lake Kiba. The bulk POC in a large lake, Lake Superior, had a mean  $\Delta^{14}\text{C}$  value of  $+10 \pm 29\%$  ranging from  $-55$  to  $+39\%$ . The  $\Delta^{14}\text{C}$  of dissolved inorganic carbon (DIC) was  $+36 \pm 62\%$  [39]. However, some lake water was reported to have low  $\Delta^{14}\text{C}$  values ( $-443$  to  $-80\%$ ) of POC together with low DIC ( $-718$  to  $-20\%$ ) [13,14]. The results suggest that  $\Delta^{14}\text{C}$  of organic matter in dissolved and particulate phases depends on that of DIC in lake water. Therefore, the depleted  $\Delta^{14}\text{C}$  value of POC in Lake Kiba might be caused by lower  $\Delta^{14}\text{C}$  inorganic compounds in lake water.

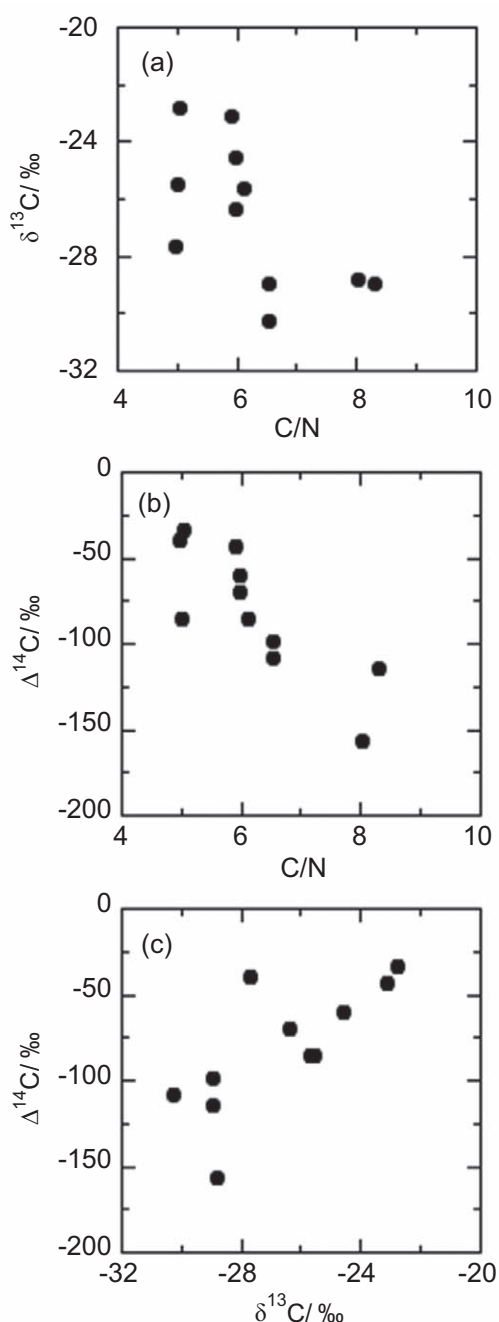
*Factors controlling characteristics of particulate organic matter*

Radiocarbon of POC in lake water is derived from primary production within the lake system, resuspension of sedimentary particulate matter, and inputs from the watershed. To elucidate the contribution of radiocarbon in Lake Kiba, relationships between radiocarbon and characteristics of organic matter must be discussed. Figure 4 presents the relationship between  $\Delta^{14}\text{C}$  values of organic matter and its contents in suspended solids in Lake Kiba. With the increase of POC contents in the suspended solids,  $\Delta^{14}\text{C}$  values increase ( $r = 0.82$ ,  $p < 0.005$ ). Similar variation of chlorophyll-a concentration represents the contribution of phytoplankton activity within the lake.

Figure 5 presents  $\Delta^{14}\text{C}$  values of organic matter in the lake suspended solids, shown as a function of the C/N ratio and  $\delta^{13}\text{C}$  values together with relationship between C/N and  $\delta^{13}\text{C}$  of POC. They have been used as a simple tracer to distinguish inputs of aquatic phytoplankton from soil organic matter. The sedimentary C/N ratio has values of 6 to 9 for freshly deposited organic matter derived from phytoplankton and 20 higher for terrestrial vascular plants [40]. The  $\delta^{13}\text{C}$  of organic matter in lake sediments reveal dynamics of suspended particle organic matter derived from phytoplankton and soil organic matter with land plants [41,42]. As shown in Figure 5(a),  $\delta^{13}\text{C}$  of POC shows a negative correlation with C/N ratio of sus-



**Figure 4.**  $\Delta^{14}\text{C}$  values of POC as a function of POC content in the Lake Kiba surface water. Error bars correspond to  $1\sigma$  uncertainty on  $\Delta^{14}\text{C}$  measurements.



**Figure 5.**  $\delta^{13}\text{C}$  of POC versus C/N ratio (a),  $\Delta^{14}\text{C}$  values of POC as a function of C/N ratio (b) and  $\delta^{13}\text{C}$  value for the POC (c) in the Lake Kiba surface water. Error bars correspond to  $1\sigma$  uncertainty on  $\Delta^{14}\text{C}$  measurements.



pendent solids, indicating the mixture of two endmembers, phytoplankton within the lake and soil organic matter around the lake watershed [43]. A negative correlation is found between the C/N ratio and  $\Delta^{14}\text{C}$  of POC (correlation coefficient of 0.82,  $p < 0.005$ ). Apparently, the higher  $\Delta^{14}\text{C}$  value results from the larger contribution of a newly produced organic matter from phytoplankton activity. The  $\Delta^{14}\text{C}$  of POC shows a positive correlation with  $\delta^{13}\text{C}$  ( $r = 0.73$ ,  $p < 0.01$ ). The  $\delta^{13}\text{C}$  and C/N in winter are within the range of soil organic matter in Ishikawa Prefecture, Japan (C/N ratio of 10.9 to 23.6;  $\delta^{13}\text{C}$  of  $-28.4$  to  $-25.9\%$ ; S. Nagao, unpublished data). The higher  $\Delta^{14}\text{C}$  of POC and the lower C/N ratio suggest an increase in signals of phytoplankton activity in the POC content variation, as presented in Figure 2.

The contribution of surface sedimentary organic matter by resuspension is discussed from C/N and  $\delta^{13}\text{C}$ . Table 1 summarizes carbon isotope data for organic carbon in surface sediments at Lake Kiba. The  $\delta^{13}\text{C}$  values are  $-26.1$  to  $-27.0\%$ , with an average and standard deviation of  $-26.6 \pm 0.3\%$ , and is narrower and higher than the average of surface water POC ( $\delta^{13}\text{C}$  of  $-27.5 \pm 2.1\%$ ). The C/N ratios in surface sediment samples also have higher values (10.6–11.8) than the suspended POC in the lake water, as depicted in Figure 3. A discrepancy exists in  $\delta^{13}\text{C}$  and C/N ratio of organic matter between the suspended solids and surface sediments. Therefore, resuspension of sediment particles from the surface sediment exerts a weak effect on the increase in  $\delta^{13}\text{C}$  and C/N ratio of POC during spring–summer. The  $\delta^{13}\text{C}$  of  $-26.6 \pm 0.3\%$  and C/N ratio of  $11.5 \pm 0.5$  on average from surface sediments might be controlled by selective degradation of newly produced particulate organic matter within the lake [43, 44] and/or a mixture with organic matter transported from the watershed forests and paddy fields.

#### 4. Summary

Simultaneous use of  $\Delta^{14}\text{C}$  and  $\delta^{13}\text{C}$  values adds a second dimension to results of isotopic studies of carbon cycling in surface aquatic environments. This study applied the analysis of carbon isotopes to the lake environment of a shallow and semi-closed lake, Lake Kiba located in Komatsu City, Ishikawa Prefecture, Japan. The field research was conducted during June 2014 to March 2016. Surface water sampling was done at a monitoring station in the central area of Lake Kiba. Suspended solids were collected from these water samples using continuous flow centrifugation. Particulate organic carbon (POC) concentration and its contents were, respectively,  $0.44$ – $5.01$   $\text{mg L}^{-1}$  and  $4.9$ – $27\%$ . Strong positive correlations of POC content were found with its  $\Delta^{14}\text{C}$  ( $-157$  to  $-33\%$ ) and  $\delta^{13}\text{C}$  ( $-30.3$  to  $-22.8\%$ ), and a negative correlation was found with the C/N ratio (5.0–14.7). These results indicated that organic matter in the surface water suspended solids consisted mainly of a mixture of organic matter produced by phytoplankton within the lake and watershed organic matter, although the  $\Delta^{14}\text{C}$  of POC was depleted to a greater degree than present atmospheric  $\text{CO}_2$ .

#### Acknowledgements

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#### References

(1) P. A. Raymond, and J. E. Bauer, *Organic Geochemistry* **32**, 469 (2001).

**TABLE 1: TOC content, C/N ratio and  $\delta^{13}\text{C}$  of organic matter in surface sediments from Lake Kiba.**

Sampling site	Depth (cm)	TOC (%)	C/N	$\delta^{13}\text{C}$ (‰)
St. KB1	0-2	2.86	10.6	-27.0
St. KB2	0-2	5.61	11.2	-26.7
St. KB3	0-2	7.24	11.5	-26.1
St. KB4	0-2	5.27	10.6	-26.5
St. KB5	0-2	4.77	11.1	-26.9
St. KB6	0-1	7.32	11.8	-26.5

- (2) A. P. McNichol, and L. I. Aluwihare, *Chemical Review* **107**, 443 (2007).
- (3) J. I. Hedges, J. R. Ertel, P. D. Quay, P. M. Grootes, J. F. Richey, A. H. Devol, G. W. Farewell, F.W. Schmidt, and E. Salati, *Science* **231**, 1129 (1986).
- (4) C. A. Masieelo, and E. R. M. Druffel, *Global Biogeochemical Cycles* **15**, 407 (2001).
- (5) P. A. Raymond, J. E. Bauer, N. F. Caraco, J. J. Cole, B. Longworth, and S. T. Petsch, *Marine Chemistry* **92**, 353 (2004).
- (6) S. Nagao, H. Kodama, T. Aramaki, N. Fujitake, and K. Yonebayashi, *Nuclear Instrument and Methods in Physical Research Section B* **259**, 552 (2007).
- (7) S. Nagao, T. Aramaki, O. Seki, M. Uchida, and Y. Shibata, *Nuclear Instrument and Methods in Physical Research Section B* **268**, 1098 (2010).
- (8) S. Nagao, T. Usui, M. Yamamoto, M. Minagawa, T. Iwatsuki, and A. Noda, *Chemical Geology* **218**, 63 (2005).
- (9) L. Guo, and R. W. Macdonald, *Global Geochemical Cycles* **20**, GB2011 (2006) doi: 10.1029/2005GB002593.
- (10) M. B. Abbott, and T. W. Stafford Jr., *Quaternary Research* **45**, 300 (1996).
- (11) F. W. Nara, A. Imai, M. Uchida, K. Matsushige, K. Komatsu, N. Kawai, Y. Shibata, K. Amano, H. Mikami, and R. Hanaishi, *Radiocarbon* **52**, 1078 (2010).
- (12) P. K. Zigah, E. C. Minor, and J. P. Werne, *Global Biogeochemical Cycles* **26**, GB1023 (2012) doi:10.1029/2011GB004132.
- (13) P. Albéric, D. Jézéquel, L. Bergonzin, E. Chapron, E. Viollier, M. Massault, and G. Michard, *Radiocarbon* **55**, 1029 (2013).
- (14) E. M. Keaveney, P. J. Reimer, and R. H. Foy, *Radiocarbon* **57**, 407 (2015).
- (15) K. Oguri, E. Matsumoto, A. Hino, H. Kurokura, and K. Okamoto, In *Summaries of Researches Using AMS at Nagoya University (VII)* (Dating and Materials Research Center, Nagoya University, 1996) pp. 184-192.
- (16) S. Ueda, K. Kondo, and J. Inaba, *Radiocarbon* **49**, 161 (2007).
- (17) J. J. Cole, N. F. Caraco, G. W. Kling, and T. K. Kratz, *Science* **265**, 1568 (1994).
- (18) J. J. Cole, N. F. Caraco, G. W. Kling, and T. K. Kratz, *Ecosystems* **10**, 172 (2007).
- (19) G. Gupta, V. Sarma, R. Robin, A. Raman, M. J. Kumar, M. Rakesh, and B. Subramanian, *Biogeochemistry* **87**, 265 (2008).
- (20) A. J. Heathcote, and J. A. Downing, *Ecosystems* **15**, 60 (2012).
- (21) Ministry of the Environment, Water quality data for public waters, <http://www.env.go.jp/water/suiiki/h26/h26-1.pdf> (accessed 30 November 2016).
- (22) National Institute for Environmental Studies, *Ecosystem Management Studies in Lake Towada*. Report of Special Research R-146-'99 (National Institute for Environmental

- Studies, Japan, Tsukuba, 1998) p. 218.
- (23) National Institute for Environmental Studies, *Studies on Mass Balance of Dissolved Organic Matter in Lake and its Functions and Effects on Lacustrine Ecosystems and Water Quality*. Report of Special Research SR-62-2004 (National Institute for Environmental Studies, Japan, Tsukuba, 2004) p. 59.
- (24) K. Hayakawa, and T. Okamoto, In *Lake Biwa: Interactions between Nature and People* edited by H. Kawanabe, M. Nishino and M. Maehata, (Springer, Tokyo, 2012) pp. 441-446.
- (25) Hokuriku Regional Agricultural Administration Office, *Report of Reclamation and Construction Work for Kaga Sanko* (Meiji Publishing Company, Kanazawa, 1970).
- (26) S. Nagao, H. T. Bui, Y. Kawano, T. Suzuki, S. Ochiai, K. Yonebayashi, M. Okazaki, A. Goto, T. Hasegawa, and M. Yamamoto, In *Geomorphology and Society* edited by M. E. Meadows and J.-C. Lin (Springer, Tokyo, 2016) pp. 181-192. doi: 10.1007/978-4-431-56000-s\_11.
- (27) Y. Magara, Classification of water quality standards, Vol. 1, <https://www.eolss.net/Sample-Chapters/C07/E2-19-01-02.pdf>, (accessed on 28 December 2016).
- (28) Komatsu City Government, Research of Natural Environment and Watershed Culture in and around Lake Kiba, [http://www.city.komatsu.lg.jp/secure/15659/kiba\\_lake\\_report.pdf](http://www.city.komatsu.lg.jp/secure/15659/kiba_lake_report.pdf) (accessed 30 November 2016).
- (29) Komatsu City Government, Field demonstration for water quality improvement of Lake Kiba by use of the Maekawa sluiceway, <http://www.city.komatsu.lg.jp/12055.htm> (accessed 30 November 2016).
- (30) Ministry of the Environment, Analytical method for suspended solids concentration, [http://www.env.go.jp/kijun/wt\\_a09.html](http://www.env.go.jp/kijun/wt_a09.html) (accessed 1 December 2016).
- (31) M. Stuiver, and H. A. Polach, *Radiocarbon* **19**, 1127 (1977).
- (32) T. Yoshioka, *Journal of Plankton Research* **19**, 1455 (1997).
- (33) Y. Sampei, and E. Matsumoto, *Geochemical Journal* **35**, 189 (2001).
- (34) H. Doi, E. I. Zuykova, E. Kikuchi, S. Shikano, K. Kanou, N. Yurlova, and E. Yadrenkina, *Hydrobiologia* **571**, 395 (2006).
- (35) B. Fry, *Marine Ecology Progress Series* **134**, 283 (1996).
- (36) Ministry of the Environment, Long-term observation of oxygen and isotopes of carbon dioxide in the atmosphere to evaluate the global budget of carbon dioxide, [https://www.env.go.jp/earth/kenkyuhi/report/pdf/13\\_4\\_2.pdf](https://www.env.go.jp/earth/kenkyuhi/report/pdf/13_4_2.pdf). (accessed 28 December 2016).
- (37) T. Nakamura, S. Kojima, T. Ohta, H. Oda, A. Ikeda, M. Okuno, K. Yokota, Y. Mizutani, and W. Kretschner, *Radiocarbon* **40**, 933 (1998).
- (38) F. Nara, A. Imai, M. Yoneda, K. Matsushige, K. Komura, T. Nagai, Y. Shiha, and T. Watanabe, *Radiocarbon* **49**, 767 (2007).
- (39) P. K. Zigah, E. C. Minor, J. P. Werne, and S. L. McCallister, *Biogeosciences* **9**, 3663 (2012).
- (40) P. A. Meyer, and E. Lavillier-Verges, *Journal of Paleolimnology* **21**, 345 (1999).
- (41) J. C. Finlay, and C. Kendall, In *Stable Isotopes in Ecology and Environmental Science*, Second edition edited by R. Michener and K. Lajtha (Blackwell Publishing Ltd., Malden, 2007) pp. 283-333.
- (42) N. S. Khan, C. H. Vane, and B. P. Horton, In *Handbook of Sea-level Research* edited by I. Shennan, A. J. Long and B. P. Horton (John Wiley & Sons Ltd., New York, 2015) pp. 295-311.
- (43) T. Yoshioka, H. Hayashi, E. Wada, *Japanese Journal of Limnology* **50**, 313 (1989).
- (44) B. J. Eadie, L. M. Jeffrey, *Marine Chemistry* **1**, 199 (1973).