Lake water dissolved inorganic carbon dynamics revealed from monthly measurements of radiocarbon in the Fuji Five Lakes, Japan

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Lakes are sensitive recorders of anthropogenic activities, as human society often develops in their vicinity. Lake sediments thus have been widely used to reconstruct the history of environmental changes in the past, anthropogenic, or otherwise, and radiocarbon dating provides chronological control of the samples. However, specific values of radiocarbon in different carbon reservoirs due to the different pathways of radiocarbon from the upper atmosphere to the lake, called the radiocarbon reservoir age, is always difficult to evaluate because of dynamic processes in and around lakes. There are few systematic studies on radiocarbon reservoir ages for lakes owing to the complex radiocarbon transfer processes for lakes. Here, we investigate lake waters of the Fuji Five Lakes with monthly monitoring of the radiocarbon reservoir effects. Radiocarbon from dissolved inorganic carbon (DIC) for groundwater and river water is also measured, with resulting concentrations ($^{14}$C) at their lowest at Lake Kawaguchi in August 2018 ($–122.4 \pm 3.2$‰), and at their highest at Lake Motosu in January 2019 ($–22.4 \pm 2.5$‰), despite a distance of 25 km. However, winter values in both lakes show similar trends of rising $^{14}$C (about 2‰). Our lake water DIC $^{14}$C results are compared to previously published records obtained from sediments in Lake Motosu and Lake Kawaguchi. These suggest that total organic carbon and compound-specific radiocarbon found in sediments are heavily influenced by summer blooms of aquatic organisms that fix DIC in water. Thus, future studies to conduct similar analyses at the various lakes would be able to provide further insights into the carbon cycle around inland water, namely understanding the nature of radiocarbon reservoir ages.

Keywords: Radiocarbon reservoir effect, Radiocarbon, Lake water, Groundwater, Lake sediments

Introduction
Reconstructing past terrestrial environments is vital for a comprehensive understanding of the Anthropocene. Various techniques have been employed to this end, including the utilization of speleothems and tree-ring archives (e.g., Sakashita et al., 2017; Cheng et al., 2020), but lake sediments are unique as they are distributed widely in many parts of the world. In some lakes, annual varve counting techniques can be used to establish sediment chronology and quantify the timing of past environmental changes (e.g., Bronk Ramsey et al., 2012), but the majority of lake sediment studies rely on radiocarbon dating (e.g., Nakamura et al., 2016; Yamamoto et al., 2018; Ghazoui et al., 2019; Safaierad et al., 2020). Radiocarbon ($^{14}$C) measurements require careful consideration of the local radiocarbon reservoir effects (e.g., Jull et al., 2013; Yokoyama et al., 2019a). The radiocarbon reservoir effect is a phenomenon in which the $^{14}$C ages of water and sediments have an older $^{14}$C age than the ambient atmospheric radiocarbon ages due to the exchange of $^{14}$C-depleted carbon stored in soil or host rock (Asough et al., 2010). Although freshwater ecosystems fix carbon from the atmosphere, dissolved carbon can also enter the lake water via groundwater. If the dissolved inorganic carbon (DIC) is derived from eroded carbonaceous bedrock, it may have very low amounts of $^{14}$C and lead to a reservoir age in the catchment or basin area (Broecker and Walton, 1959).

Since radiocarbon reservoir ages reflect hydrosphere dynamics, there are many studies that consider changes in seawater radiocarbon in the ocean with time (e.g., Hirabayashi et al., 2017, 2019; Ota et al., 2019; Servettaz et al.,...
However, few systematic lake water reservoir age studies have been reported. The radiocarbon reservoir effect of lakes has been reported in many areas, with some areas having a sediment radiocarbon reservoir age of more than 1,500 years (e.g., Nakamura et al., 2016, Schneider et al., 2019). One source of atmospheric $^{14}$C to consider is bomb-derived $^{14}$C produced in the atmosphere during the nuclear weapons tests in the 1950s. “Bomb-$^{14}$C” increased rapidly during the decade of weapons testing (Broecker and Walton, 1959), then decreased due to exchange between the atmosphere and other carbon reservoir such as the oceans and biosphere (Sweeney et al., 2007). Subsequently, usage of fossil fuels has been increasingly important to reduce atmospheric $^{14}$C level (Suess effect; Suess, 1955). In addition to atmospheric $^{14}$C, groundwater and/or river water may be the source of lake $^{14}$C (Brady et al., 2009). Yu et al. (2007) constructed a mass balance model considering variations in $^{14}$C due to changes in atmospheric, groundwater, and river water inflow or outflow. It has been suggested that the contribution of $^{14}$C from these sources may vary due to environmental changes including anthropogenic forcing, such as the use of fossil fuels (Graven, 2015; Blattmann et al., 2018). However, few studies mention the time scale of the variation of radiocarbon reservoirs in lake water. Therefore, systematic sampling and analysis are needed to estimate reliable radiocarbon reservoir ages of lake water, including measurements on groundwater and river water.

The Fuji Five Lakes were selected for this study in order to monitor DIC radiocarbon fluctuations in lake water. Their proximity to each other allows us to collect water monthly from each of the lakes. The lakes are situated within 40 km from the flank of Mount Fuji. It is a unique research setting to understand lake dynamics because the size and depths of each lake are different (Table 1). Water stored in the Fuji Five Lakes is sourced from precipitation, groundwater, and surface water. Geological and hydrological surveys were conducted in the mid-1940s and 1960s for groundwater resource development (Hamano, 1976), and the regional hydrology was studied by Hayashi and Tsuboi (2005), reporting the source distribution of groundwater by utilizing hydrogen and oxygen isotope ratios. In recent years, chlorofluorocarbons (CFCs) and tritium measurements have been used to estimate groundwater ages (Asai and Tsujimura, 2010). However, the degree of groundwater contribution has not been determined due to limitations in the earlier studies (e.g., Ochiai, 1995).

In this study, DIC radiocarbon was measured monthly for the Fuji Five Lakes. The results are discussed in regard to local hydrological dynamics. Previously measured sediment total organic carbon (TOC) and compound-specific radiocarbon data were compared to DIC radiocarbon data for Lake Motosu and Lake Kawaguchi in order to examine local reservoir ages in the context of paleo environmental studies using lake sediment.

Materials and methods

The Fuji Five Lakes are dammed lakes formed by the volcanic activity of Mt. Fuji. They are surrounded to the north by the Misaka and Tanzawa Mountains and to the south by Mt. Fuji (Takeuchi et al., 1995). The groundwater stream boundary 50 m underground was found by a survey conducted previously at altitudes of 900 m to 1,000 m at the foot of Mt. Fuji (Hamano, 1976). Most of the lake water in the Fuji Five Lakes is considered to be derived from the Mikasa or Tanzawa Mountains, and groundwater from Mt. Fuji is thought to rarely flow into the lake (Hamano, 1976). The section below 50 m is an impermeable layer, and groundwater has been shown to move along the buried fossil valley through the permeable upper layer (Ogata et al., 2014; Yamamoto et al., 2017a).

The map, water level, elevation, and water depth of the Fuji Five Lakes are shown in Figure 1 and Table 1. Lakes Motosu, Shoji, and Sai are situated at around 900 m above sea level, whereas Lake Kawaguchi, on the east side of Lake Sai, is 831 m above sea level. Lake Yamanaka is the most eastern lake in the five lakes area and has an altitude of 981 m. There are no rivers that constantly discharge to or from the lakes, other than for Lakes Kawaguchi and Yamanaka. However, lake water is artificially released from Lake Motosu to the Fuji River, from Lake Sai to Lake Kawaguchi, and from Lake Kawaguchi to the Katsura River.

Lakes Motosu, Sai, and Shoji are located to the northwest of Mount Fuji (Figure 1). Lava flows cover the southern side of the Lakes Sai and Shoji from the surface to 135 m in depth (Chiba et al., 2010), and at the eastern side of Lake Motosu, a lava flow of 167 m thick was reported (Koshimizu et al., 2007). This characterizes the high under-water flow velocity (Ochiai, 1995). The other 3 sides are surrounded by the steep Misaka mountains of 1,000 m to 1,800 m, and there is no constant river inflow or outflow. Hamada et al. (2012) conducted a yearlong vertical survey of water temperature and water quality, noting that precipitation dominated the water balance.

Lake Kawaguchi is the lowest of the Fuji Five Lakes at an altitude of 831 m, with the southern shore of Lake Kawaguchi covered by lava from Mount Fuji (Takada et al., 2016). The other 3 sides are surrounded by Misaka Mountains with an altitude of 1,400 m to 1,700 m. The inflowing water from Rivers Tera and Oku from the Misaka Mountains is considered to be mainly underground water (Yoshizawa and Mochizuki, 2005; Yamamoto et al., 2017a). Groundwater resources have been investigated by Hamano (1976), and the outflow from the lake is suggested to be due to the underground valley structure in the southwestern part of Lake Kawaguchi (Yoshimura and Kawada, 1942; Kanno et al., 1986; Hayashi and Tsuboi, 2005; Ogata and Kobayashi, 2015; Yamamoto et al., 2017b). Mt. Fuji lava does not contain rocks such as limestone that reduce $^{13}$C, but calcareous rock layers have been reported in the Misaka Mountains to the northern side of the Fuji Five Lakes (Matsuda, 2007).

To the southwest of Lake Yamanaka, sediments of gently sloping volcanic foot fans from Mt. Fuji overlap, and from the north to the east, the Tanzawa mountains with an altitude of 1,200 m to 1,400 m of mountains. The Tanzawa Mountains around Lake Yamanaka are classified as impervious layers. The groundwater level of the Ichinosuna and Ohori rivers that inflow in the lake is higher.
than that of the western parts of the lake (Susuki and Taba, 1994).

**Sampling**

Lake water was collected each month from June 2018 to April 2019 at the surface (0.5 m) of each lake for $\Delta^{14}$C measurements (Figure 2b). A 250-ml glass bottle shaded with aluminum was used for water sampling, and 56 lake water samples were collected. Various other water samples were also analyzed to compare $\Delta^{14}$C values, including groundwater (wells: $n = 11$) and river water ($n = 6$). For Lake Shoji, we were unable to collect lake water samples.

**Table 1.** Summary of sizes, depth, altitude, and retention time of Fuji Five Lakes (Geospatial Information Authority of Japan and Yamanashi Prefecture, 1993). DOI: https://doi.org/10.1525/elementa.2020.00149.t1

<table>
<thead>
<tr>
<th>Lake</th>
<th>Motosu</th>
<th>Shoji</th>
<th>Sai</th>
<th>Kawaguchi</th>
<th>Yamanaka</th>
</tr>
</thead>
<tbody>
<tr>
<td>Area (km$^2$)</td>
<td>4.7</td>
<td>0.5</td>
<td>2.1</td>
<td>5.7</td>
<td>6.8</td>
</tr>
<tr>
<td>Altitude (m)</td>
<td>900</td>
<td>900</td>
<td>900</td>
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<td>981</td>
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<tr>
<td>Maximum depth (m)</td>
<td>121.6</td>
<td>15.2</td>
<td>71.7</td>
<td>14.0</td>
<td>13.3</td>
</tr>
<tr>
<td>Average depth (m)</td>
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<td>3.7</td>
<td>34.8</td>
<td>9.8</td>
<td>9.2</td>
</tr>
<tr>
<td>Retention time (year)</td>
<td>7.9</td>
<td>0.11</td>
<td>2.3</td>
<td>0.37</td>
<td>0.52</td>
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</table>
because we did not receive sampling approval in time, but we hope to measure lake water from Lake Shoji in the future. Groundwater was collected from drinking water wells in Fujikawaguchiko town and Narusawa and Yamanakako village. We chose sampling sites for each lake from at least 2 locations to identify the sources of lake water, namely, one for the Mt. Fuji side and the other close to the Misaka Mountains. River water was collected in the autumn when the average flow rate was at a high. Water samples from the Tera and Oku Rivers in the Misaka Mountains around Lake Kawaguchi were collected in October and November 2019. For Lake Yamanaka, river water from the Ohori River and Ichinosuna River were collected in November and December 2019.

Method
A few drops of mercurous chloride (HgCl₂) were added to each sample to suppress biological activity after water
collection. The samples were processed using the bubbling method (McNichol et al., 1994); 4 ml of phosphoric acid were added to 250 ml of lake water, and bubbling was carried out for 15 min in a glass tube filled with 60 mPa of helium gas to strip CO2 liberated from DIC. During this process, water vapor is released along with the CO2 gas, so 2 cryogenic traps containing ethanol cooled to –120°C were used to remove the water vapor. The CO2 was captured in a liquid nitrogen trap (Servettaz et al., 2019). The CO2 collected was processed under the protocol described in Yokoyama et al. (2007): CO2 was injected into a quartz tube together with 4 mg of iron powder pre-reduced at 450°C and graphitized by heating at 630°C for 6 h. The samples were measured using a single-stage accelerator mass spectrometer at the University of Tokyo Atmosphere and Ocean Research Institute (Yokoyama et al., 2019b).

Throughout this study, we report Δ14C using Equation 1, where x is the year of the sample measurement, and F is the fraction modern carbon as was defined in a previous study (Stuiver and Polach 1977).

\[
\Delta^{14}C = \left( \frac{F(1990-x)}{F(8267)} - 1 \right) \times 1000 \text{‰,} \tag{1}
\]

where radiocarbon age and F can be written as follows (Donahue et al., 1990):

\[
14C \text{ age} = -8033 \ln F. \tag{2}
\]

The radiocarbon reservoir age (R) expresses the difference in the age of the lake DIC and the contemporary atmosphere, in radiocarbon years. R can be written as follows:

\[
R = |\text{radiocarbon age}|_{\text{lake DIC}} - |\text{radiocarbon age}|_{\text{atmosphere}}. \tag{3}
\]

Substituting Equations 1 and 3, the following relationship between R and F can be obtained:

\[
R = -8033 \cdot \ln F_{\text{lake}} = (-8033 \cdot \ln F_{\text{atm}}) \approx -8033[\ln F_{\text{lake}} - \ln F_{\text{atm}}] \tag{4}
\]

\[
R = -8033 \cdot \ln \left( \frac{F_{\text{lake}}}{F_{\text{atm}}} \right) = 8033 \cdot \ln \left( \frac{F_{\text{atm}}}{F_{\text{lake}}} \right). \tag{4}
\]

Solving Equation 1 for F, and substituting into Equation 4, we get a relationship between R and Δ14C:

\[
R = 8033 \cdot \ln \left( \frac{(\Delta^{14}C_{\text{atm}}/1000) + 1}{(\Delta^{14}C_{\text{lake}}/1000) + 1} \right). \tag{5}
\]

Thus, the 14C offset between the lake water and the ambient atmosphere is presented in the current study, and the younger (smaller) the Δ14C provides the older (larger) the radiocarbon reservoir ages (e.g., Yokoyama et al., 2000; Hirabayashi et al., 2019; Fukuyo et al., 2020).

Results

Radiocarbon measurement in lake water

The results of radiocarbon measurements are listed in Table 2 and Figure 2. The Δ14C in Lake Motosu was at its highest value in January (–22.4 ± 2.5‰), and the lowest was recorded in June (–53.8 ± 4.3‰). Winter average values were calculated to be around –20‰, but the values from summer to autumn, that is, between July and November, showed average values of around –30‰. It was not possible to measure the radiocarbon content of the lake water sample obtained in December from Lake Motosu due to the extremely small concentration of CO2 gas in the sample.

The results of Lake Sai show the lowest Δ14C values in June (–47.3 ± 5.9‰), with peak values in September (–25.3 ± 2.3‰). The values fall between –30‰ and –45‰ after October. When comparing the values between the Lake Sai and the Lake Motosu, similar Δ14C values in both lakes are observed from June to November, but the Lake Sai values drop after December by as much as 10‰.

The Δ14C trend at Lake Kawaguchi follows a similar trajectory as that of Lake Motosu, namely lower values in summer and higher values in winter. However, actual values are distinctly lower than the other lakes, where Δ14C observed at a site in the center of the lake is –122.4 ± 3.2‰ in August and –87.8 ± 3.6‰ in January.

The Δ14C values in Lake Yamanaka were below –60‰ in August and October, which is about 20‰ less than the value in September (–36.8 ± 5.3‰). The Δ14C fluctuated more than 20‰ per month from August to October.

Radiocarbon values of groundwater (wells) and river water

Table 2 shows the radiocarbon results for 11 groundwater (wells) and 6 river water samples. Groundwater sampling was conducted from October to November 2019, whereas 2 separate sampling campaigns (October and November 2019) were made for the Tera River and the Oku River. At least 2 ground water sampling sites were designed to collect waters for each lake, considering the incoming water routes from either the northern mountain ranges (e.g., Misaka Mountains) or southern mountain (i.e., Mt. Fuji). In the case of Lake Motosu, the groundwater from the Mt. Fuji side (YAUT-05815) had extremely low Δ14C (–246.0 ± 1.9‰). The groundwater Δ14C values from Koan (on the Misaka Mountains side: YAUT-050826) were measured at –7.1 ± 2.2‰, which was higher than that of surface lake water (Figure 2). For Lake Shoji, groundwater Δ14C was as low as –165.9 ± 2.0‰ from Shojiaoki, on the Mt. Fuji side (YAUT-050813), with higher values measured from Shojikyoson (–45.5 ± 2.3‰: YAUT-050812), on the Misaka Mountains side. At Lake Sai, Δ14C values of –38.9 ± 2.5‰ were obtained from Aoki (on the Mt. Fuji side: YAUT-050809), while Δ14C values in Shinhonsawa were 9.8 ± 2.3‰ (on the Misaka Mountains side: YAUT-050811), which is equivalent to, or higher, than the corresponding lake water Δ14C value.

Results from 2 other lakes, Lake Kawaguchi and Lake Yamanaka, show different features than the other 3 lakes described above. At Okune, located on the southern (Mt. Fuji) side of Lake Kawaguchi, Δ14C values of –74.6 ± 2.5‰ were obtained, whereas values of –62.2 ± 3.1‰ were measured from the sample collected from Goto (YAUT-050805). The values from Wakahiko, located further
Table 2. Radiocarbon results of lake water, groundwater, and river water dissolved inorganic carbon radiocarbon. DOI: https://doi.org/10.1525/elementa.2020.00149.t2

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<tr>
<th>Lab. No.</th>
<th>Location</th>
<th>Date</th>
<th>Δ¹⁴C (‰)</th>
<th>Error (±)</th>
<th>¹⁴C Age (yBP)</th>
<th>Error (±)</th>
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(continued)
north on the Misaka mountains side are considerably higher, with Δ14C values of –256.1 ± 1.9‰ (YAUT-050806). The 2 rivers flowing into Lake Kawaguchi, namely the Tera and Oku Rivers, had water Δ14C values of –106.3 ± 2.3‰ to –94.7 ± 2.5‰ and –48.3 ± 2.2‰ to –45.2 ± 2.2‰, respectively.

At Lake Yamanaka, Δ14C values of 4.0 ± 2.6‰ were obtained (on the Mt. Fuji side: YAUT-050803), whereas the value obtained from Hirano was –53.6 ± 2.9‰ (northern side: YAUT-050802). They are similar to the Δ14C values of 2 river waters flowing into the lake: Ichinosuna and Ohori of –6.5 ± 2.2‰ and –137.4 ± 2.0‰, respectively.

**Discussion**

**Factors controlling the radiocarbon reservoir ages of lake waters**

Systematic Δ14C analyses and reservoir ages for Fuji Five Lakes show seasonal variations depend on their setting. As all the lakes are located within 40 km of the northern flank of Mt. Fuji, climate conditions including precipitations and evaporations are similar for the Fuji Five Lakes. Monthly precipitation is preeminent in the summer months, with the maximum in September 2018. However, the value of lake water Δ14C did not show a significant change in September, except for Lake Yamanaka. Thus, the specific hydrological dynamics of each lake are likely responsible for the fluctuations in Δ14C.

The results can be categorized into 2 groups: The first includes lakes that show seasonal variations in Δ14C, such as Lake Motosu and Lake Kawaguchi. Their Δ14C values are relatively low (i.e., older reservoir ages) in summer but high (i.e., younger reservoir age) in winter. The second group, such as Lake Sai and Lake Yamanaka, does not show clear seasonality.

Two possible causes can explain the Δ14C fluctuations (i.e., reservoir ages) for these lakes. The first is that the decrease in Δ14C is due to the changes in retention time of each lake. However, since a previous study revealed that...
the retention time of lake water in Lake Motosu is typically 7.9 years and the retention of each lake is significantly different (Table 1), it is unlikely that seasonal $\Delta^{14}C$ fluctuations are due to retention time. If there is an effect of retention time, the longer the retention time, the lower the $\Delta^{14}C$ value. However, Lake Motosu has the longest retention time of 7.9 years, yet the $\Delta^{14}C$ value in Lake Motosu is the highest of the five lakes.

The second possible cause could be changes in contributions of waters with low $\Delta^{14}C$ flowing into the lake. Since the groundwater $\Delta^{14}C$ values around the Fuji Five Lakes are sufficiently low compared to the ambient atmospheric values, it is likely that groundwater supplies water with lower $\Delta^{14}C$ to the lake waters. In Lakes Motosu and Kawaguchi, $\Delta^{14}C$ tended to increase during winter (Figure 2, Table 2), while no seasonal $\Delta^{14}C$ variation was observed at Lakes Sai and Yamanaka. In the following section, hydrological dynamics changes are discussed in the view of the mixtures of low $\Delta^{14}C$ water that enter each lake.

At Lake Motosu, the water is heated from the surface, which leads to the development of a thermocline during spring and summer. In contrast, the lake waters do not stratify during winter because of the U-shaped lake basin with a maximum depth of 121.6 m. Hamada et al. (2012) suggested the possibility of meteoric water accumulation at the surface layer of the lake during the stratified period based on seasonal changes from electric conductivity measurements. However, the lake water $\Delta^{14}C$ showed relatively lower values from spring to autumn compared to winter, suggesting contribution from groundwater with low $\Delta^{14}C$. Although the groundwater from the Misaka Mountains side (YAUT-050826) has high $\Delta^{14}C$ in October 2018, the data may not be indicative of average values of the groundwater because the water was sampled just after a typhoon passed through. It could be explained by higher hydrostatic pressure caused by precipitation during summer to autumn and snowmelt in spring (Figure 3).

The $\Delta^{14}C$ value during the summer at the eastern site (Funatsu) is around $-122\%$, which is lower than the site where water is collected at the center of Lake Kawaguchi (Central site: $\Delta^{14}C$ is around $-100\%$). Groundwater samples collected from Wakahiko, located further north and situated on the Misaka mountains side, have $\Delta^{14}C$ of $-256.1 \pm 1.9\%$ (YAUT-050806). The water temperature of Funatsu is higher than that of the lake center in summer (Horiuchi et al., 1992), suggesting that the subsurface water or groundwater influx is more significant during summer (July–October).

Unlike the above lakes, no seasonal variation was observed at Lakes Sai and Yamanaka. The $\Delta^{14}C$ increase of around 20% in September in Lake Yamanaka is attributed to the increase in precipitation due to a typhoon (Typhoon #21), which directly hit the region on September 4 and 5, 2018. The typhoon resulted in 200 mm of precipitation, causing the surface lake water $\Delta^{14}C$ to rise. This typhoon also caused lake level to rise by up to +24 cm. An increased amount of low $\Delta^{14}C$ groundwater flowed into the lake due to an increase in groundwater pressure during October 2018, or lake water was diluted by precipitation around September 2018. The DIC $\Delta^{14}C$ value in lake waters is considered to be a function of groundwater, precipitation, and carbon retained in the lake water. A combination of these factors causes $\Delta^{14}C$ fluctuations in the lake waters. In this paragraph, we conducted a 2-box model simulation to estimate relative contributions of groundwater and precipitation for the Fuji Five Lakes surface waters (Figure 4). Previous studies have suggested that the main sources of water for the lakes are either groundwater or
precipitation; however, groundwater is abundant in Lake Yamanaka (see Hamada and Kitagawa, 2010; Hamada et al., 2012). In fact, 3 lakes: Lake Motosu, Lake Shoji, and Lake Sai, have no rivers that constantly discharge to or from the lakes. Although Tera and Oku Rivers are flowing into Lake Kawaguchi, and Ichinosuna and Ohori Rivers flow into Lake Yamanaka, we cannot confirm the effects of river water due to the lack of flow rate data. Although the case mentioned above exists only under limited conditions, semiquantitative analysis of the hydrology of the lakes is useful in evaluating the mechanisms for understanding Δ14C changes in the lake surface water. Newly

Figure 4. Results of groundwater 14C contribution (%) in the Fuji Five Lakes compared to actual Δ14C values. Percentage of groundwater 14C in lake water 14C (gray bar), lake water DIC 14C (black: Motosu, red: Sai, yellow: Kawaguchi Funatsu, gray: Kawaguchi Center, and blue: Yamanaka). DOI: https://doi.org/10.1525/elementa.2020.00149.f4
obtained groundwater Δ¹⁴C values by this study are employed, and an atmospheric Δ¹⁴C value of each month ranged from 7.0 ± 1.9‰ to −9.98 ± 1.5‰ (ICOS RI, 2019) at station Saclay was used in the following equation. The calculation formula is described below.

\[
\Delta^{14}C_{\text{lake}} = (1 - f) \times \Delta^{14}C_{\text{atm}} + (f) \times \Delta^{14}C_{\text{groundwater}}.
\]

Here, \( f \) is the proportion (%) of groundwater \( \Delta^{14}C \) in the lake, whereas \( \Delta^{14}C_{\text{lake}} \) DIC is the lake water \( \Delta^{14}C \), and \( \Delta^{14}C_{\text{atm}} \) is the atmospheric \( \Delta^{14}C \) value observed in 2018. Groundwater also originates from precipitation, but \( \Delta^{14}C \) in groundwater decreases before reaching the lake because it is isolated from the atmosphere (e.g., Bente, 2013). Although the lake water in the Fuji Five Lakes is considered to be mainly derived from the Mikasa and Tanzawa Mountains (Hayashi, 2020), \( \Delta^{14}C_{\text{groundwater}} \) DIC values obtained from wells around the Fuji Five Lakes are employed for Lakes Motosu and Sai where the \( \Delta^{14}C_{\text{groundwater}} \) DIC values of the Misaka Mountain are higher than the lake DIC \( \Delta^{14}C \) values. The values obtained from the Misaka Mountains side (i.e., Northern Sites) are used to calculate the proportion of groundwater \( \Delta^{14}C \) for Lakes Kawaguchi and Yamanaka. The proportion of groundwater contribution to DIC\(^{14}C\) for the surface lake water in Lake Motosu is about 20%. This is consistent with the chemical measurements of waters reported in Hamada et al. (2012). Water in Lake Kawaguchi is largely influenced by groundwater, as the proportion is approximately 50%. For the Lakes Sai and Yamanaka, more than 80% of the lake water is derived from groundwater DI\(^{14}C\), including months when lake water \( \Delta^{14}C \) values are smaller than groundwater DI\(^{14}C\) value of each lake. Since the groundwater collected in this study is very limited in this area, further investigation is needed to clarify the origin of the lake water.

**Implications for reservoir ages of Lake Motosu and Lake Kawaguchi**

Previous studies collected sediment cores from Lake Motosu to study past environmental changes in the region (Lamair et al., 2018, 2019; Obrochta et al., 2018). Obrochta et al. (2018) measured radiocarbon in TOC from sediments, and they reported a Δ14C surface value of Δ14C of −41 ± 2.5‰. Our observed lake water Δ14C in Lake Motosu from this study between July and November is similar with values around −30‰. This suggests that the carbon produced in the lake due to carbon fixation during spring to summer by phytoplankton living in the lake surface water is the dominant source of carbon for the sediments in the lake. As a result, it is considered that the DIC in the lake water from spring to summer is comparable to the TOC Δ14C of the sediments. Thus, the Δ14C of lake water DIC in spring to summer can be utilized to estimate the reservoir age of the sediments.

We also compared lake water DIC Δ14C with compound-specific Δ14C reported for Lake Kawaguchi sediments (Yamamoto et al., 2020). Compound-specific Δ14C extracted from fatty acids from Lake Kawaguchi sediments were reported for each fraction. The results show that C16 fatty acid Δ14C (−124 ± 6‰), chlorophyll a Δ14C (−133 ± 6‰), and lake water DIC Δ14C (−117 ± 2‰) are consistent with each other for the samples collected in June 2017. Our study revealed that the seasonal magnitude of Δ14C changes was around 35% from August 2018 (−122.4 ± 3.2‰) to March 2019 (−88.7 ± 2.4‰); thus, an increase in Δ14C could have occurred between the summer and winter in 2017 when Yamamoto et al. (2020) collected samples. If so, the value of DIC Δ14C observed from the current study is close to the values of C24 fatty acid (Δ14C = −77 ± 6‰) or C28 fatty acid (Δ14C = −75 ± 7‰). Both C24 and C28 fatty acids are considered to be derived from higher plants, soil-derived compounds (Gagosian et al., 1981; Simonneit 1977), and/or aquatic macrophytes (Ficken et al., 2000). Hence, the Δ14C of these fatty acids could be partly affected by additional aquatic sources in addition to terrestrial plant materials.

Further systematic studies are needed to clarify the relationship between lake water and organic matter produced by living organisms. To elucidate the origin of organic matter, δ13C measurements would be able to provide clear pathways of carbon transported from lake water to organic matter.

**Conclusions**

Monthly Δ14C measurements on the Fuji Five Lakes surface water DIC revealed the following:

1. Δ14C values of Lake Motosu and Lake Kawaguchi decreased in summer, as compared to winter, whereas relatively low Δ14C values were observed in Lake Sai. Δ14C values decrease in June–August and October at Lake Yamanaka.

2. Reservoir ages of sediment may reflect lake water DIC in spring to summer. Lake water DIC is likely to be fixed as organic matter and supplied to sediments due to the expansion of aquatic organisms that are active from spring to summer.

3. Seasonal variations of lake water Δ14C are mainly driven by groundwater amount and also suggest that Δ14C of lake seasonality was shown to be useful for improving the accuracy of lake hydrology and radiocarbon dating.

**Data accessibility statement**

The following datasets were generated:

- Sizes, depth, altitude, and retention time of Fuji Five Lakes: Geospatial Information Authority of Japan (https://www.gsi.go.jp/), and Yamanashi
References


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