

Chapter 13

Recent Eutrophication and Environmental Changes in the Catchment Inferred from Geochemical Properties of Lake Onuma Sediments in Japan

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Abstract This study investigated the continuous record of eutrophication in Lake Onuma based on the geochemical properties of two lake sediment cores obtained from the deepest part of the lake in 2011. Based on a tuff layer deposited during the eruption of Mt. Komagatake, and on the correlation between fluctuations in $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values, two sediment cores, ON11-2-2 and ON11-6, were dated to the 1920s and 1890s, respectively. The $\delta^{13}\text{C}$ value and C/N ratio for the lake sediments show values within the ranges for planktonic material and river sediment, suggesting

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that the lake sediment is a mixture of these sources and that their mixture ratio was almost constant since the 1920s. On the other hand, the $\delta^{15}\text{N}$ of two cores show a similar trend with increasing $\delta^{15}\text{N}$ from the 1950s–1960s to the present time. It is attributed to the increase in the $\delta^{15}\text{N}$ value of planktonic material reflecting anthropogenic nitrogen inflow to the lake.

Keywords Eutrophication • Lake sediment • Organic matter • $\delta^{13}\text{C}$ • $\delta^{15}\text{N}$

13.1 Introduction

Lakes are important resources for drinking water, irrigation, and hydroelectric power supply. Lake eutrophication is one of the serious environmental pollution problems (e.g., Smith et al. 1999) damaging to these lake water resources. Eutrophication has become a serious environmental problem in various lakes, such as Lake Nansihu, China (Liu et al. 2010), Lake Dianchi, China (Xiong et al. 2010), Myall Lake, Australia (Drew et al. 2008), and is sometimes caused by the inflow of anthropogenic nutrients.

Lake Onuma, a volcanic dammed lake on the southern Hokkaido Island in Japan, faces the progression of eutrophication resulting from water pollution during the past few decades. The chemical oxygen demand (COD), which reflects the concentration of dissolved organic matter, was relatively low (0.54–2.04 mg/L) during the 1930s in lake water (Yoshizumi et al. 1972). However, it increased to 2–4 mg/L in the 1970s (Yoshizumi et al. 1972; Imada et al. 1983) and remained stable at 3–5 mg/L in the 2000s, which is higher than the environmental quality standard for lake water (Hokkaido 2013). If eutrophication continues to progress, it may influence not only water quality but also fishery productivity, lake scenery, and tourism resources at the lake.

The cause of eutrophication in Lake Onuma is postulated to be anthropogenic nutrient inflow. The major nitrogen source may be livestock-derived nitrogen from the rivers (Tanaka 2005). To investigate the mechanism of eutrophication progression and to allow for accurate future predictions, reconstruction of the continuous record of past environmental change in lake-catchment is needed. The geochemical properties, total organic carbon (TOC), total nitrogen (TN), and carbon and nitrogen isotope ratios of lake sediments have been widely used to reconstruct long-term and recent environmental changes (e.g., Meyers and Ishiwatari 1993; Enters et al. 2006). The nitrogen isotope ratio, $\delta^{15}\text{N}$, can be used to evaluate eutrophication in rivers, lakes, and coastal areas (e.g., Valiela et al. 2000; Kohzu 2006; Liu et al. 2010). Therefore, this study aims to reveal the detailed continuous record of the progression of eutrophication based on the geochemical properties of Lake Onuma sediments.

13.2 Samples and Methods

Lake Onuma is located in the southern part of Hokkaido Island in Japan (Fig. 13.1). Lake Onuma is a dammed lake formed by the volcanic eruption of Mt. Komagatake in 1640 (Yoshimoto et al. 2007). This lake consists of the eastern basin (Onuma) and the western basin (Konuma) connected by a narrow channel. Water and catchment areas are about 8.9 km² and 173 km², respectively (Tanaka 2005). Average and maximum depths are 4.7 and 12.9 m. Three major rivers (Shukunobe, Ikusa, and Karima Rivers) flow into the Onuma basin. There was an outflow river in the eastern end of the Onuma basin before 1961. The outlet was closed and a waterway tunnel for a hydroelectric power plant was constructed in the southern part of the Konuma basin in 1961 (Fig. 13.1).

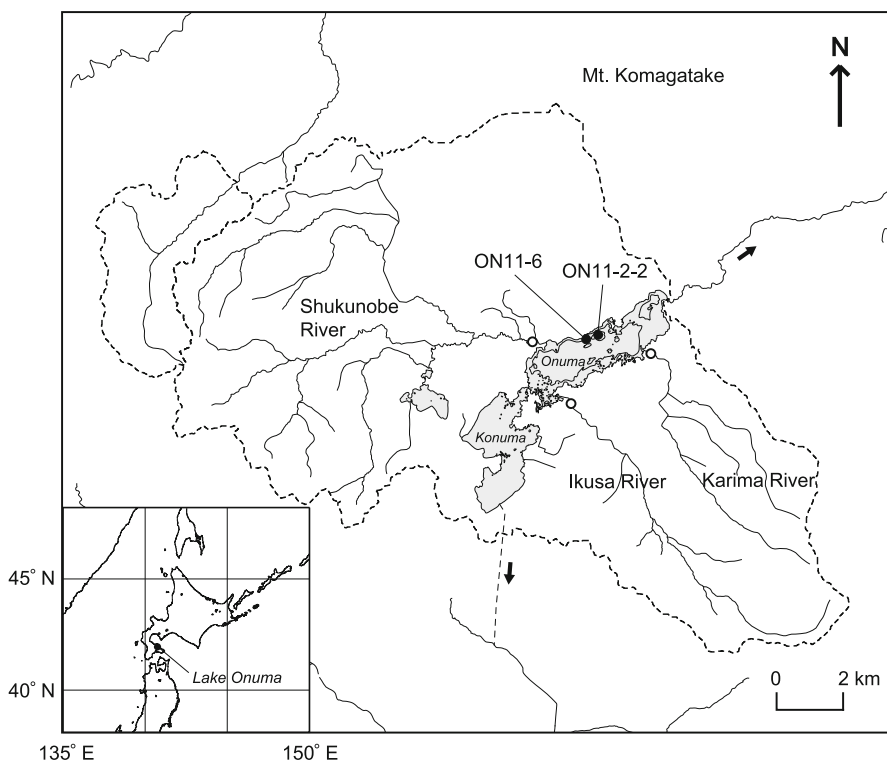


Fig. 13.1 Location and topographic maps of Lake Onuma. The *dashed line* indicates the catchment area of Lake Onuma. *Closed and open circles* show the sampling points of lake sediment cores and river sediments, respectively. The numerical map data were based on Fundamental Geospatial Data provided by the Geospatial Information Authority of Japan, and National Land Numerical Information provided by the Ministry of Land, Infrastructure, Transport and Tourism, Japan

Two sediment cores (ON11-2-2, 68 cm; ON11-6, 45 cm) were obtained around the deepest part of the Onuma basin with a gravity core sampler (Satake-type, RIGO, Japan) on September 20, 2011 (closed circles in Fig. 13.1). These sediment cores were sliced into 1-cm-interval subsamples. Planktonic material and lake water samples were collected from the sampling point of ON11-2-2 on June 5 and 6, 2012. Riverbed sediment and water samples were also collected at the main river mouths on June 6, 2012 (opened circles in Fig. 13.1). Water samples were filtered using a glass fiber filter for the dissolved organic carbon (DOC) and dissolved nitrogen (DN) measurements.

Freeze-dried sediment samples were grounded and treated with 1 M HCl to remove inorganic carbon for analyses of total organic carbon (TOC), total nitrogen (TN), and stable carbon and nitrogen isotope ratios. TOC and TN contents were measured with the elemental analyzer (2400 Series II, PerkinElmer, USA). The precision of TOC and TN analyses were $\pm 0.009\%$ and $\pm 0.003\%$, respectively. The stable carbon and nitrogen isotope ratios were analyzed with three types of mass spectrometers (IsoPrime EA, GV Instruments, UK; DLTApplus and DELTA V Advantage, Thermo Fisher Scientific Inc., USA). Stable carbon and nitrogen isotope ratios are shown as $\delta^{13}\text{C}$ values relative to VPDB and $\delta^{15}\text{N}$ values relative to atmospheric N_2 as follows:

$$\delta^{13}\text{C}, \delta^{15}\text{N} = (R_{\text{sample}}/R_{\text{standard}} - 1) \times 1,000 \quad (13.1)$$

where R_{sample} and R_{standard} are the $^{13}\text{C}/^{12}\text{C}$ or $^{15}\text{N}/^{14}\text{N}$ atomic ratios of the sample and international standard, respectively. The reference materials USGS40 ($\delta^{13}\text{C}_{\text{VPDB}} = -26.39$, $\delta^{15}\text{N}_{\text{air}} = -4.52$), L-Alanine ($\delta^{13}\text{C}_{\text{VPDB}} = -19.6$, $\delta^{15}\text{N}_{\text{air}} = 1.6$), ANU-sucrose ($\delta^{13}\text{C}_{\text{VPDB}} = -10.80$), IAEA-N1 ($\delta^{15}\text{N}_{\text{air}} = 0.4$), and IAEA-N2 ($\delta^{15}\text{N}_{\text{air}} = 20.3$) were used to calibrate the measurements. The precision of the $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ analyses were $\pm 0.18\text{‰}$ and $\pm 0.31\text{‰}$, respectively. The dissolved organic carbon (DOC) and dissolved nitrogen (DN) concentrations of lake and river water samples were analyzed with a TOC analyzer (TOC-V_{SCN}, Shimadzu, Japan).

The ^{210}Pb and ^{137}Cs concentrations were measured to estimate the age of the cores. The powdered samples were sealed into plastic bags (5.0×3.5 cm). After establishing the radioactive equilibrium between ^{222}Rn and ^{214}Pb (about 1 month), the activity concentration of ^{210}Pb (46.5 keV), ^{214}Pb (352 keV), and ^{137}Cs (661.6 keV) were determined by gamma-ray spectrometry using a Ge detector (LO-AX-51370-20, ORTEC, USA). The activity of excess ^{210}Pb ($^{210}\text{Pb}_{\text{ex}}$) was estimated by subtracting the activity of ^{214}Pb from that of ^{210}Pb .

The numerical map data were based on Fundamental Geospatial Data provided by the Geospatial Information Authority of Japan, and National Land Numerical Information provided by the Ministry of Land, Infrastructure, Transport and Tourism, Japan.

13.3 Results

13.3.1 Chemical Properties of Lake Sediments and Lake Water Samples

Figure 13.2a–d show the vertical changes in TOC and TN contents, C/N atomic ratio, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values of ON11-2-2. The C/N ratios, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values of the sediment samples represent the TOC/TN ratio, $\delta^{13}\text{C}$ value of organic matter, and $\delta^{15}\text{N}$ value of total nitrogen in the following discussion. The TOC and TN contents of sediment decrease with depth from 9 % to 3 %, and from 0.9 % to 0.4 %, respectively. The C/N ratio of sediment ranges from 9 to 11 and this range corresponds to the value of algae (Meyers 1994), suggesting that the sediment of Lake Onuma is largely influenced by autochthonous organic matter. The $\delta^{13}\text{C}$ value exhibits a relatively small variation around -38‰ . On the other hand, the $\delta^{15}\text{N}$ value continuously increases upward through the core, from 2 to 7 ‰.

Figure 13.2e, f show the vertical changes in $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values through ON11-6. The $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values of the upper 30 cm show a similar trend to that of ON11-2-2, with $\delta^{15}\text{N}$ values increasing upward through the core. This result suggests that the bottom of ON11-2-2 corresponds to the $\sim 30\text{-cm}$ -deep layer of ON11-6. On the other hand, the lower 30 cm section exhibits a different trend. The $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values synchronously decrease upward through the core.

Table 13.1 shows the C/N ratio, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values of planktonic material and river sediment. The $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values of river sediment range from -26.3 to -28.3 and from 3.1 to 3.9, respectively, which represent the organic matter from the catchment. On the other hand, the $\delta^{15}\text{N}$ value of planktonic material in Lake

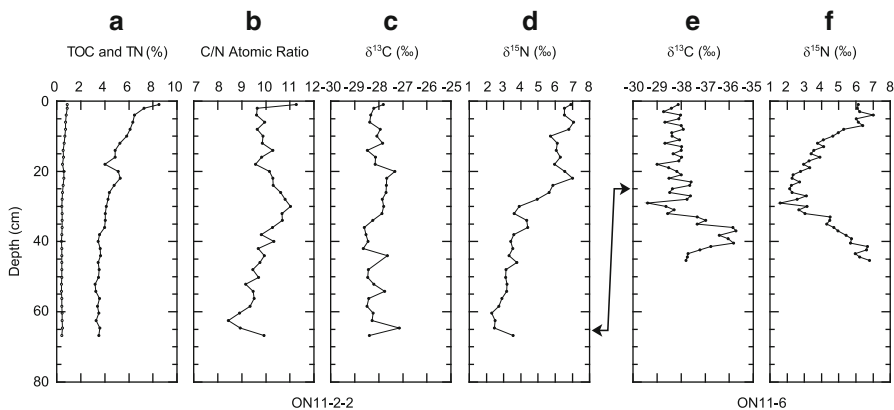


Fig. 13.2 Vertical changes in (a) TOC and TN contents, (b) C/N atomic ratio, (c) carbon isotope ratio $\delta^{13}\text{C}$, (d) nitrogen isotope ratio $\delta^{15}\text{N}$ for the ON11-2-2 core. Vertical changes in (e) carbon isotope ratio $\delta^{13}\text{C}$, and (f) nitrogen isotope ratio $\delta^{15}\text{N}$ for the ON11-6 core

Table 13.1 C/N ratio, $\delta^{13}\text{C}$, and $\delta^{15}\text{N}$ of planktonic material and river sediment

Sample	C/N atomic ratio	$\delta^{13}\text{C}$ (‰)	$\delta^{15}\text{N}$ (‰)	DOC (mgC/L)	DN (mgN/L)
ON11-2-2					
Plankton	6.12	-31.72	7.53		
Lake water				1.0240	0.4435
Ikusa River					
Sediment	11.48	-26.32	3.85		
River water				1.6780	0.6720
Karima River					
Sediment	15.35	-26.40	3.28		
River water				0.8875	1.4070
Shukunobe River					
Sediment	13.58	-28.27	3.09		
River water				1.1835	0.6270

Onuma is higher (7.5‰) than that of river sediment. The $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values of the lake sediments are within the range for planktonic material and river sediment, suggesting that the sediment is a mixture of these sources. The DOC concentrations of river and lake water exhibit the same range (0.8875–1.6780 mgC/L). The DN concentration of river water ranges between 0.6720 and 1.4070 mgN/L, and is higher than that of lake water (0.4435 mgN/L), suggesting that the river is one of the major sources of dissolved nitrogen in Lake Onuma.

13.3.2 Age of the Sediment Cores

Figure 13.3 shows the vertical changes in water content of ON11-2-2 and ON11-6. Water content of the ON11-6 core was analyzed by Itono et al. (2015, this volume). A layer with low water content, corresponding to the tuff layer, is observed at a depth of 28–30 cm in ON11-6. This tuff layer can be correlated to the 1929 volcanic deposit (Ko-a) of Mt. Komagatake (Yoshimoto et al. 2007). On the other hand, the tuff layer could not be found in ON11-2-2, based on water content fluctuation and observation, implying that the bottom of the core is younger than 1929.

The age of the cores were also estimated based on the ^{210}Pb (Krishnaswamy et al. 1971; Appleby and Oldfield 1978) and ^{137}Cs methods (Ritchie and McHenry 1990). Figure 13.4a, b indicate the vertical changes in the activity concentrations of $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs for ON11-2-2 and ON11-6, shown as a function of mass depth. The regression curves show the fitting results of the constant initial concentration (CIC) model (Pennington et al. 1973; Appleby and Oldfield 1983) as a first estimation.

The sedimentation rate of ON11-2-2 was estimated at 0.0924 g/cm²/year. Based on this sedimentation rate, the bottom of ON11-2-2 was dated to 1908. However, this result does not correspond to the ages of the tuff layer and to those estimated using the ^{137}Cs method. The ^{137}Cs fluctuation first appears and peaks at 30 cm

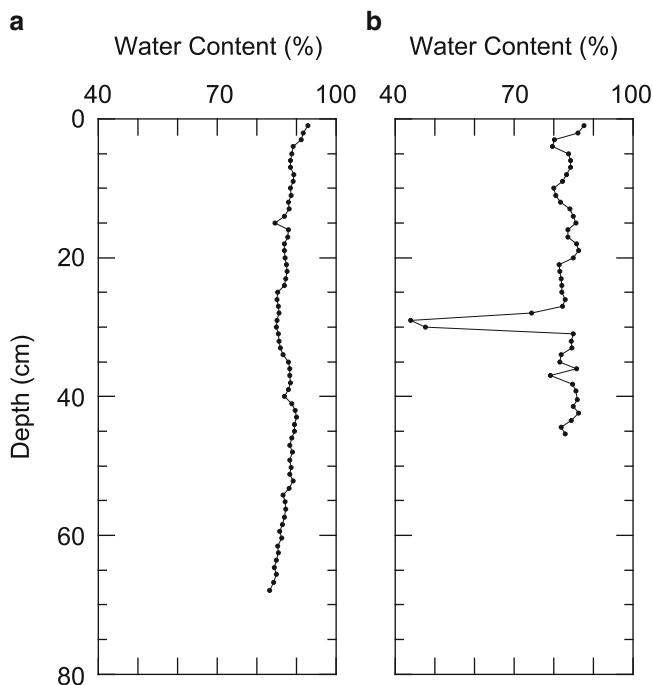


Fig. 13.3 Vertical changes in water content for the (a) ON11-2-2 and (b) ON11-6 cores

(4.1 g/cm²) and 26 cm (3.4 g/cm²), corresponding to the beginning of the ¹³⁷Cs global fallout in 1954 (Ritchie and McHenry 1990), and to the fallout peak of ¹³⁷Cs in 1963 in Japan (Katsuragi 1983; Katsuragi and Aoyama 1986; Igarashi et al. 1996). However, the ²¹⁰Pb ages for these depths are younger than those of ¹³⁷Cs (1963 and 1974, respectively). These discrepancies may result from artificial disturbance of sediment layer and/or dilution effect of ²¹⁰Pb_{ex} by biogenic silica. Fishery operations (smelt fishing using dragnets) were performed around the deepest area of the lake every year (Onuma Fisheries Cooperative Association, personal communication), possibly disturbing the surface sediment layer and causing the mismatches between tuff layer, ¹³⁷Cs and ²¹⁰Pb ages. Additionally, the dilution of catchment-derived ²¹⁰Pb_{ex} by autochthonous materials such as biogenic silica may affect the ²¹⁰Pb age. Mineral content from the catchment of the ON11-2-1 core, which was obtained at the same location as ON11-2-2, exhibit a significant variation from 20 to 60 % (Itono et al. 2015, this volume), reflecting the change in biogenic silica content. Therefore, the CIC model, assuming a constant initial ²¹⁰Pb_{ex} concentration on the sediment surface, may be influenced by a change in biogenic silica productivity. Because of this problem, the ²¹⁰Pb dating result was not used to establish the age model of the core.

For ON11-6, the sedimentation rate was estimated at 0.0782 g/cm²/year. The age of the middle section of the tuff layer (29 cm deep) was estimated to 1926.

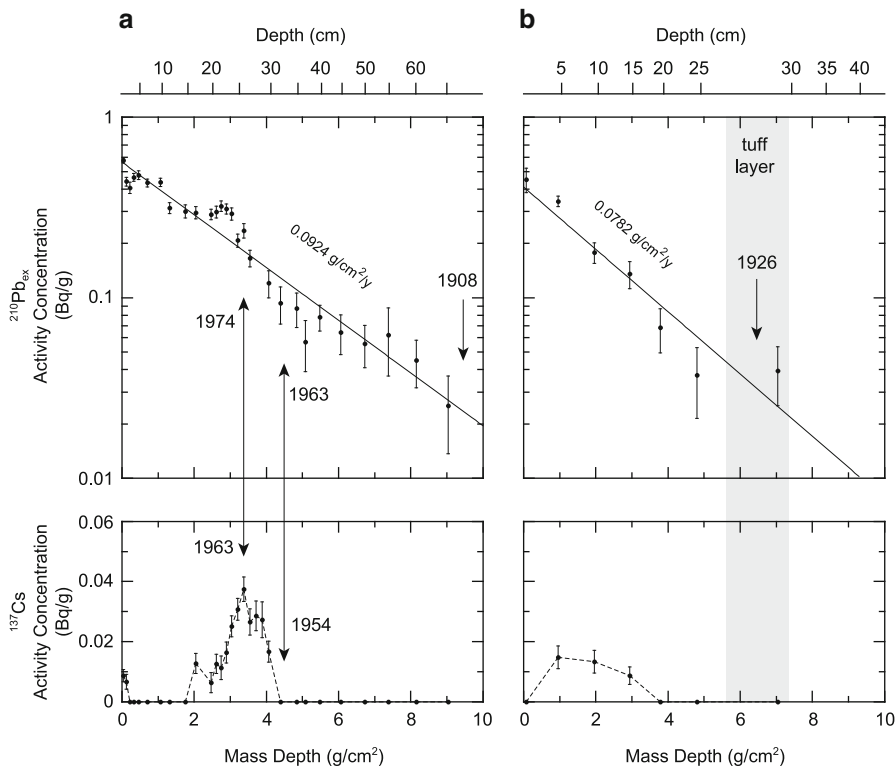


Fig. 13.4 Vertical changes in activity concentrations of excess $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs for the (a) ON11-2-2 and (b) ON11-6 cores

This result supports the hypothesis that the tuff layer in ON11-6 corresponds to Ko-a deposit and that the disturbance by fishery operation and the dilution effect of autochthonous materials are negligible in this point. Assuming that the sedimentation rate of the section below the tuff layer is similar as that of the upper layer, the bottom of ON11-6 was dated to the 1890s.

The correlation between fluctuations in $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values of ON11-2-2 and ON11-6 suggest that the bottom of ON11-2-2 can be correlated to the section just above the tuff layer (about 25 cm deep) of ON11-6. The bottom of ON11-2-2 was dated to the 1920s.

13.4 Discussion

Figure 13.5a shows the $\delta^{13}\text{C}$ -C/N plot of lake sediment of ON11-2-2 and expected organic matter sources (autochthonous planktonic material and river sediment from catchment) in Lake Onuma. The values for the lake sediments are within the range for planktonic material and river sediment, suggesting that the organic matter

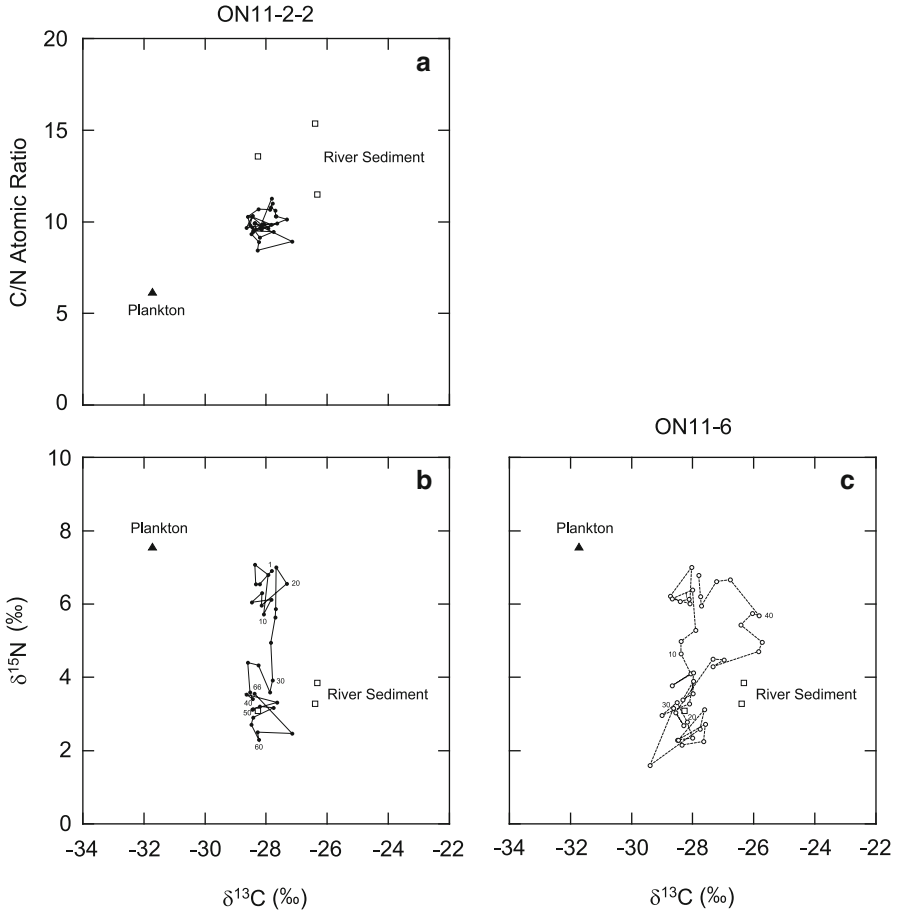


Fig. 13.5 (a) $\delta^{13}\text{C}$ -C/N ratio and (b) $\delta^{13}\text{C}$ - $\delta^{15}\text{N}$ plots of ON11-2-2 sediment, planktonic material, and river sediment. (c) The $\delta^{13}\text{C}$ - $\delta^{15}\text{N}$ plots of ON11-6 sediment. Circles indicate sediment samples. Triangle and square indicate planktonic material and river sediment, respectively

contained within the lake sediment is a mixture of these organic matter sources, and that their mixture ratio has been almost constant since the 1920s.

On the other hand, the $\delta^{15}\text{N}$ value of the sediment has increased, although the organic sources are almost constant (Fig. 13.5b, c). Additionally, the $\delta^{15}\text{N}$ value of planktonic material is higher than that of river sediment. These results indicate that the increase of the sediment $\delta^{15}\text{N}$ value is attributed to the increase of the $\delta^{15}\text{N}$ value of plankton. Because the $\delta^{15}\text{N}$ value of phytoplankton is mainly determined by the isotopic composition of inorganic nutrients (e.g., Fogel et al. 1992), the $\delta^{15}\text{N}$ value of plankton reflects that of dissolved inorganic nitrogen (DIN) in the lake.

The $\delta^{15}\text{N}$ value of DIN derived from livestock and sewage water is larger than that of natural sources such as precipitation (Heaton 1986; Fogg et al. 1998).

It has been reported that the $\delta^{15}\text{N}$ value of organic matter becomes larger in watersheds with significant anthropogenic nitrogen loading (e.g., Valiela et al. 2000; Carmichael et al. 2004). Therefore, increasing anthropogenic nitrogen inflow has promoted the eutrophication of Lake Onuma and its influence has been recorded in the $\delta^{15}\text{N}$ fluctuation of the lake sediment. The dissolved nitrogen concentration of river water is higher than that of lake water (Table 13.1). These results also support the hypothesis that nitrogen loading from the river is the primary cause of eutrophication. The major nitrogen source may be the livestock-derived nitrogen, which is estimated at 50 % of nitrogen discharge from the main three rivers in 2000 (Tanaka 2005). The sediment records indicate that the contribution of livestock waste water to the lake has rapidly increased since the 1950s–1960s, and its influence still continues in present time.

In the section below the tuff layer in ON11-6 (Figs. 13.2e, f and 13.5c), $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values synchronously increased downward through the core. This variation seems to differ from that of the post-tuff section where only the $\delta^{15}\text{N}$ value changed. This change in the isotopic composition of the sediment may be attributed to the shift of organic matter source and/or post-deposition diagenesis. Volcanic eruption and wildfire cause the clearance and the shift in the composition of vegetation in the catchment. Catchment vegetation disturbance affects the transport of soil and particulate organic matter (Beschta 1978; Bormann et al. 1974; Miller 1984), and their influences are recorded in the $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values of the lake sediments (Lane et al. 2004; Routh et al. 2007). The post-deposition diagenesis also changes the isotopic composition of the sediment. The synchronous increase in $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values can be interpreted as the result of diagenesis effect. Longer sediment record may provide more detailed information on vegetation changes and/or geochemical conditions in the lake.

13.5 Conclusions

This study aims to reconstruct the continuous record of lake-catchment environmental changes to evaluate the progression of eutrophication in Lake Onuma. The study of the sediment record of lake sediment geochemical properties leads to the following conclusions.

The ages estimated with the ^{210}Pb method does not correspond to the ages of the tuff layer and ^{137}Cs fallout peak in the ON11-2-2 core. These results may be attributed to the artificial disturbance of the sediment layer and/or to the dilution effect of $^{210}\text{Pb}_{\text{ex}}$ by biogenic silica. ON11-6 exhibits a tuff layer corresponding to the 1929 volcanic eruption. Correlation between fluctuations in $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values of the ON11-2-2 and ON11-6 cores suggest that the bottom of ON11-2-2 is dated to the 1920s.

The $\delta^{13}\text{C}$ value and C/N ratio for lake sediments are within the ranges indicative of planktonic material and river sediment, suggesting that organic matter of lake

sediment is a mixture of these sources and that their mixture ratio has been almost constant since the 1920s. The $\delta^{15}\text{N}$ values of two cores show a similar trend of increasing $\delta^{15}\text{N}$ values since the 1950s–1960s. It indicates that the contribution of livestock waste water to the lake has rapidly increased since the 1950s–1960s, and that its influence continues in present time. In the section below the tuff layer, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values synchronously increased downward through the core. This fluctuation may be attributed to the shift of organic matter sources and/or to post-deposition diagenesis.

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