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BACKGROUND CONCENTRATION OF ^{14}C IN AQUATIC SAMPLES FROM BRACKISH LAKE OBUCHI, ROKKASHO, JAPAN, ADJACENT TO NUCLEAR FUEL REPROCESSING FACILITIES

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ABSTRACT. The brackish Lake Obuchi in Rokkasho, Japan, is adjacent to the first Japanese commercial nuclear fuel reprocessing facilities, which are now undergoing performance testing, with commercial operation scheduled to start in 2007. Preparatory surveys were made by measuring the background levels of radiocarbon for water, aquatic biota, and sediment samples using accelerator mass spectrometry (AMS) in order to study the potential effects of ^{14}C released by the plant to the ^{14}C concentration in aquatic samples. Concentrations of ^{14}C in Futamata River in 2004 ranged from 102 ± 0.5 to 109 ± 0.6 pMC (average 106 ± 0.6 pMC), while ^{14}C concentrations in brackish water from Lake Obuchi and in seawater were 89 ± 0.5 to 104 ± 0.4 pMC (average 98 ± 0.5 pMC) and 82 ± 0.6 to 102 ± 0.4 pMC (average 93 ± 0.5 pMC), respectively. The relationship between ^{14}C concentration and salinity showed a negative correlation ($r = 0.68$, $P < 0.01$, $n = 20$). ^{14}C concentration in selected aquatic biota (i.e. fish, benthos, and seagrass) from 2003 to 2004 ranged from 105 ± 0.7 to 107 ± 0.6 pMC and in zooplankton and phytoplankton was 103 ± 2.4 to 105 ± 1.7 pMC. The depth profile of ^{14}C in 3 core sediment samples from Lake Obuchi showed maximum concentrations from 103 ± 0.5 to 106 ± 0.5 pMC at 5–20 cm depth. The vertical profile of ^{14}C concentration in the sediment did not follow global atmospheric ^{14}C fallout. We confirmed that the background level of ^{14}C concentration in aquatic samples in brackish Lake Obuchi before operation of the reprocessing plant was similar to the concentration (~106 pMC) in the recent atmosphere.

INTRODUCTION

Japan's first commercial nuclear fuel reprocessing facilities in Rokkasho Village, Aomori Prefecture (Figure 1), are now undergoing performance testing. The facilities consist of a uranium enrichment plant, a spent-fuel reprocessing plant, a high-level radioactive waste storage plant, and a low-level radioactive waste disposal plant. The reprocessing plant is due to be completed by 2007, while the other 3 plants are already in operation. The small amounts of radioactive materials (^{85}Kr, ^{3}H, ^{14}C, ^{129}I, etc.) to be released from the reprocessing plant will be discharged into the nearby Pacific Ocean from an outlet pipe installed on the sea floor, and into the atmosphere from an exhaust stack. An assessment of the environmental impact of these radioactive substances requires knowing the concentration levels of radionuclides in the natural environment around the reprocessing plant, especially the background level of tritium (^{3}H; half-life = 12.23 yr), radiocarbon (^{14}C; half-life = 5730 yr), and radioiodine (^{129}I; half-life = 1.57×10^{7} yr). ^{3}H released to the atmosphere and ocean from the reprocessing plant was estimated to be 1.9×10^{15} Bq/yr and 1.8×10^{16} Bq/yr, respectively (Japan Nuclear Fuel Limited 2002). The background concentration of ^{3}H in the surrounding environment has been surveyed by Ueda et al. (2006). Moreover, ^{129}I released to the atmosphere and ocean from the reprocessing plant was estimated to be 1.1×10^{10} Bq/yr and 4.3×10^{10} Bq/yr, respectively (Japan Nuclear Fuel Limited 2002). The background concentration of ^{129}I was measured by Aomori Prefecture (2006). ^{14}C (5.2×10^{13} Bq/yr; Japan Nuclear Fuel Limited 2002) will be released from the plant into the atmosphere; however, very little data are available for ^{14}C.

^{14}C discharged into a coastal zone from the nuclear fuel reprocessing plant at Sellafield (United Kingdom) has been reported previously (McCartney at al. 1994; Cook et al. 1997). Cook et al. (1995) reported that ^{14}C levels in plankton and fish around Sellafield were high: 250–270 pMC and 400–500 pMC, respectively. Moreover, reported ^{14}C concentrations and distributions in the vicinity of the nuclear fuel reprocessing plant at La Hague (France) showed that ^{14}C concentrations in the

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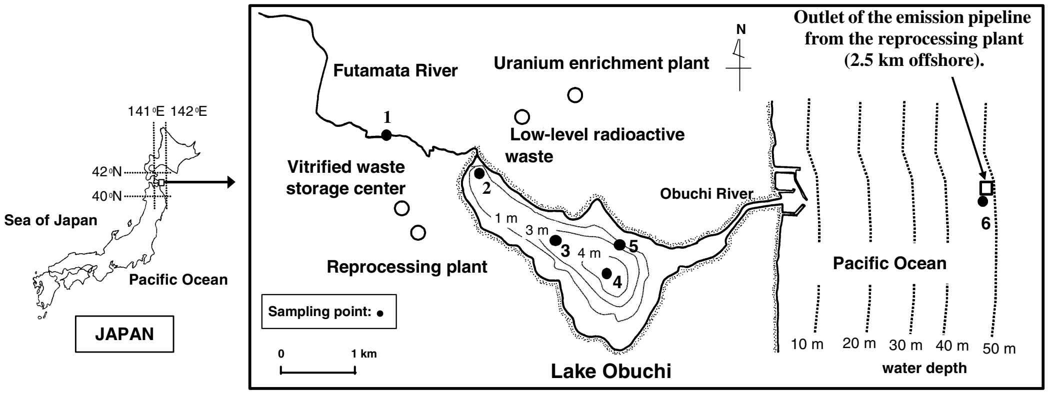

Figure 1 Sampling locations for Futamata River, Lake Obuchi, and the Rokkasho coast, Japan

seaweed and seawater around the plant were 180–270 pMC and 130–400 pMC, respectively (Fontugne et al. 2004; Maro et al. 2004; Fievet et al. 2006).

The aim of this study is to determine the background concentrations of ^{14}C and its distribution in aquatic samples (water, biota, and sediment) in brackish Lake Obuchi, Rokkasho Village, before the nuclear fuel reprocessing plant begins commercial operations.

METHODS

Study Site and Sampling

Table 1 shows the water depth and latitude-longitude for the sampling stations. Lake Obuchi (40°57′N, 141°21′E) is located in northeastern Aomori Prefecture, northern Japan. The Futamata River is the main freshwater river flowing into Lake Obuchi. The lake, which is connected to the Pacific Ocean via the Obuchi River, is small and shallow (3.7 km^2 area; mean depth 2.5 m). It gradually deepens in an easterly direction from the Futamata River, reaching a maximum depth of 4.5 m at Station 3. The salinity of the lake is 10–15 practical salinity units (psu) in the surface layer and 20–30 psu in the bottom layer, and the halocline is formed at depths of 1–3 m (Ueda et al. 2000). Moreover, coastal water samples were collected from the mouth of the Obuchi River to the sea (Station 4) and along the Rokkasho seacoast (Stations 5 and 6).

Table 1 Water depth and latitude-longitude at sampling stations.

Site nr	Water depth (m)	Latitude (N)	Longitude (E)
1	0.7	40°58′21″	141°19′56″
2	1.4	40°58′07″	141°20′20″
3	3.6	40°57′57″	141°21′07″
4	4.3	40°57′00″	141°21′50″
5	1.5	40°57′28″	141°21′44″
6	50	40°57′50″	141°25′30″

Water samples for ^{14}C analysis were collected from Stations 4, 5, 6 using a 2-L Van Dorn water sampler in May, July, September, and November 2004. The samples were taken to the laboratory in 2-L polyethylene bottles. The temperature and salinity of the water samples were measured in situ using a Datasonde-4a (Hydrolab Ltd., Texas, USA).

The biota samples were obtained from the lake in May, July, September, and November 2003 and 2004. The seagrass (*Zostera marina*) was collected by aqualung diving at Station 5. The phytoplankton (0.45–100 µm particle size) samples were collected using a 0.45-µm filter (Millipore) after excluding that which was filtered by a 100-µm net (13XX, Rigo Ltd., Tokyo, Japan). The zooplankton (100–1000 µm particle size) samples were collected using a 100-µm net after excluding that which was filtered by a 1000-µm net (20GG, Rigo Ltd., Tokyo, Japan). Fish samples (*Hypomesus olidus*, *Chaenogobius urotaenia*, *Sebastes schlegeli*, *Hemiramphus sajori*, and *Platichtys stellatus*) were purchased from local fishermen. Benthos samples (Arthropoda [*Crangon affinis*, *Grandidierella japonica*, and *Gnorimosphaeroma ravi*], Mollusca [*Musculus senhausia*], and Annelida [*Prionospio japonicus*]) were collected by a dredge at Station 5.

Sediment cores were collected by aqualung diving using a chloroethylene cylinder (16 cm diameter, 100 cm long) at 3 stations in September 1997. The cores were immediately cut into 5-cm intervals and samples were preserved in plastic containers.

ANALYTICAL TECHNIQUE

Total dissolved inorganic carbon (DIC) and dissolved organic carbon (DOC) concentrations in water samples were measured by the high-temperature catalytic oxidation method (Shimadzu Ltd., TOC-5000A, Kyoto, Japan).

^{14}C activity in the DIC component of water samples was determined following the method of Nakamura et al. (1998). Each sample was opened and filtered in a CO_2-free glove box. Then, 500 mL of each filtrate were transferred into a 500-mL Erlenmeyer flask. This was then acidified, purged with nitrogen, and the CO_2 was trapped out using liquid N_2, or by adding 25 to 100 mL of an ammonical concentrated $SrCl_2$ solution, and then recovering carbon in the form of $SrCO_3$. On completion of the precipitation reaction after a few days, the sample was acidified and purged with nitrogen, and then the CO_2 was transferred to a cold finger surrounded by liquid N_2 and pumped under high vacuum for about 30 min to remove impurities. The graphite target for the accelerator mass spectrometer (AMS) was made from the collected CO_2 with an iron catalyst at 600 °C (Vogel et al. 1984).

The sediment and biota samples were freeze-dried, then digested using 1N HCl (2 hr, 93 °C) in order to remove any carbonate. Samples were combusted at 850 °C with a copper oxide catalyst, and the liberated CO_2 was collected (Minagawa et al. 1984). The graphite target was made from the collected CO_2 with an iron or cobalt powder catalyst at 600 °C. The ^{14}C isotope ratio of the graphite target was analyzed by the Center for Applied Isotope Studies (CAIS) at the University of Georgia, USA, using AMS (NEC Ltd., 1.5SDH-1). Radiocarbon data were reported as $\Delta^{14}C$, with the ‰ deviation from the $^{14}C/^{12}C$ ratio of a standard with the fixed isotopic composition:

$$\Delta^{14}C = [(^{14}C/^{12}C)_{SN} / (^{14}C/^{12}C)_{STD} - 1] \times 1000$$

where $(^{14}C/^{12}C)_{SN}$ is the isotopic abundance ratio of ^{14}C to ^{12}C of the sample normalized to a common $\delta^{13}C$ value of –25‰; and $(^{14}C/^{12}C)_{STD}$ is the absolute international standard ratio, which is selected so that $\Delta^{14}C$ is zero for atmospheric CO_2 in AD 1950. The absolute international standard ratio is defined as 0.7459 times of the $^{14}C/^{12}C$ ratio, in 1950, of NBS oxalic acid (SRM 4990) standard normalized to $\delta^{13}C = -25‰$ (Stuiver and Polach 1977). The normalization to $\delta^{13}C = -25‰$ in the sample is for correcting isotopic fraction during fixation of carbon from atmospheric CO_2 into objective materials (Stuiver 1983). Positive $\Delta^{14}C$ values indicate the presence of carbon labeled with ^{14}C produced by atmospheric nuclear weapons testing and accidents, causing activities higher than the 1950 atmospheric level. The $\Delta^{14}C$ measurements had a total uncertainty (counting statistics and laboratory reproducibility) of ±5‰ (1 σ). ^{14}C was corrected by $\delta^{13}C$ results of –25‰, and expressed in percent modern carbon (pMC) as defined by Stuiver and Polach (1977). Typical analytical errors were ±0.5 pMC (1 σ) for ^{14}C. A 100-pMC value is roughly equivalent to 226 Bq/kg C, in the year 1950. ^{14}C ages of the sediment samples were calculated according to the method of Nakamura and Nakai (1988).

For measuring the stable carbon isotopic ratio ($\delta^{13}C$), we used an isotope ratio mass spectrometer (IRMS; Finnigan Ltd., MAT-252, Bremen, Germany) and an elemental analyzer (CE-instruments Ltd., NC-2500, Milan, Italy). The $\delta^{13}C$ values were expressed in terms of ‰ deviation:

$$\delta^{13}C = [(^{13}C/^{12}C)_{\text{Sample}} / (^{13}C/^{12}C)_{\text{PDB}} - 1] \times 1000$$

where $(^{13}C/^{12}C)_{\text{Sample}}$ is the abundance ratio of ^{13}C to ^{12}C of the sample, and $(^{13}C/^{12}C)_{\text{PDB}}$ is the ratio of Pee Dee Belemnite (PDB) to the original standard. $\delta^{13}C$ analysis precision was within ±0.1‰.

RESULTS AND DISCUSSION

Water Samples

Table 2 shows the analytical results of DIC, DOC, ^{14}C, $\delta^{13}C$, water temperature, and salinity measurements in water samples from Futamata River (Station 1), Lake Obuchi (Station 4), and the Rokkasho coast (Station 6). DIC concentrations in freshwater samples of Futamata River ranged from 5.9 to 9.4 mg/L, with an average of 7.9 mg/L, while DIC concentrations in water samples from brackish Lake Obuchi were 13.7–21.0 mg/L. Mean values of DIC at depths of 1 m and 3 m were 18 and 20 mg/L, respectively. DIC concentrations in seawater from the Rokkasho coast (21.9–24.7 mg/L) were higher than those in other station samples. In contrast, DOC concentrations in freshwater and seawater (0.6–0.9 mg/L) were lower than those in brackish lake water (1.4–2.1 mg/L). The mean values of $\delta^{13}C$ in freshwater, brackish water, and seawater samples were –12, –6, and –4‰, respectively. $\delta^{13}C$ tended to increase as salinity increased.

Concentrations of ^{14}C in water samples from Futamata River ranged from 102 ± 0.5 to 109 ± 0.6 pMC (average 105 ± 4 pMC). ^{14}C concentrations in Lake Obuchi water samples collected at depths of 1 m and 3 m were 94 ± 0.5 to 104 ± 0.5 pMC (average 100 ± 4 pMC) and 89 ± 0.5 to 101 ± 0.4 pMC (average 97 ± 5 pMC), respectively. For the Rokkasho coast, ^{14}C concentrations in water samples were low, ranging from 83 ± 0.6 to 102 ± 0.4 pMC (average 93 ± 8 pMC). The relationship between ^{14}C concentration and salinity showed a negative correlation ($r = 0.68$, $P < 0.01$, $n = 20$) (Figure 2). Moreover, ^{14}C concentrations in water samples from Futamata River were similar to the ^{14}C level (~106 pMC) in the atmosphere; however, the ^{14}C concentrations in brackish water samples were lower than the atmospheric concentration, showing a dilution effect of seawater.

Aquatic Biota Samples

Table 3 shows the analytical results of TOC, ^{14}C, and $\delta^{13}C$ in aquatic biota samples collected at Station 5 in Lake Obuchi. The TOC concentrations in the fish group ranged from 45 to 52 mg/g, the highest concentration among the biota (Figure 3). Samples of benthos, seagrass (*Zostera marina*), zooplankton (100–1000 μm particle size), and phytoplankton (0.45–100 μm particle size) were 37 ± 5, 36 ± 3, 37 ± 13, and 19 ± 3 mg/g, respectively. TOC concentrations in a lower-trophic biological stage (phytoplankton) in biota tended to be lower than that in higher-trophic stages (fish and benthos). The $\delta^{13}C$ concentrations in the fish, benthos, and plankton groups were –17, –17, and –23, respectively. The $\delta^{13}C$ concentration in biota tended to be higher as the high-trophic stages dominated the ecosystem, except for the seagrass group.

Table 2 Results of water temperature, salinity, dissolved inorganic carbon (DIC), dissolved organic carbon (DOC), δ^{13}C, and ^{14}C (pMC and Δ) measurements in water samples from Futamata River, Lake Obuchi, and the Rokkasho coast.

	Month (2004)	Station 1	Station 4		Station 6	
		0.5 m	1 m	3 m	1 m	40 m
Water temp. (° C) ±1 σ	May	12.7	13.9	13.3	11.0	10.2
	July	13.9	20.6	17.5	15.1	14.0
	September	14.1	20.1	20.6	19.6	18.2
	November	7.6	5.4	6.9	16.0	16.5
	Average	*12.1 ± 3.0*	*15.0 ± 7.1*	*14.6 ± 5.9*	*15.4 ± 3.5*	*14.7 ± 3.5*
Salinity (psu)[a] ±1 σ	May	0.1	17.0	21.0	32.0	32.0
	July	0.1	26.0	29.0	32.0	32.0
	September	0.1	27.0	28.0	33.0	33.0
	November	0.1	14.8	26.7	32.0	32.0
	Average	*0.1 ± 0.0*	*21.2 ± 6.2*	*26.2 ± 3.6*	*32.3 ± 0.5*	*32.3 ± 0.5*
DIC (mg/L) ±1 σ	May	7.3	18.1	18.9	22.5	23.0
	July	9.0	19.3	20.8	21.9	24.7
	September	9.4	21.0	20.7	24.6	22.1
	November	5.9	13.7	19.6	22.5	24.6
	Average	*7.9 ± 1.6*	*18.0 ± 3.1*	*20.0 ± 0.9*	*22.9 ± 1.2*	*23.6 ± 1.3*
DOC (mg/L) ±1 σ	May	1.0	1.6	1.8	0.9	1.0
	July	0.9	1.9	1.8	0.9	0.7
	September	0.1	2.1	1.8	0.7	0.9
	November	0.6	1.9	1.4	1.1	1.2
	Average	*0.6 ± 0.4*	*1.9 ± 0.2*	*1.7 ± 0.2*	*0.9 ± 0.2*	*0.9 ± 0.2*
δ^{13}C (‰) ±1 σ	May	–6.1	–4.1	–6.7	–2.4	–6.3
	July	–14.5	–5.5	–5.6	–2.6	–2.9
	September	–14.4	–6.8	–8.3	–3.9	–5.7
	November	–14.4	–9.3	–6.5	–7.8	–2.3
	Average	*–12.4 ± 4.2*	*–6.4 ± 2.2*	*–6.8 ± 1.1*	*–4.2 ± 2.5*	*–4.3 ± 2.0*
^{14}C (pMC) ±1 σ	May	99.5 ± 0.5	104.2 ± 0.5	100.6 ± 0.4	102.3 ± 0.4	100.2 ± 0.5
	July	107.7 ± 0.7	100.1 ± 0.5	100.5 ± 0.4	100.3 ± 0.5	97.1 ± 0.5
	September	105.0 ± 0.7	93.9 ± 0.5	89.2 ± 0.5	88.5 ± 0.5	86.3 ± 0.5
	November	109.0 ± 0.6	102.2 ± 0.6	96.0 ± 0.6	83.3 ± 0.6	82.6 ± 0.5
	Average	*105.3 ± 4.2*	*100.1 ± 4.4*	*96.6 ± 5.4*	*93.6 ± 9.2*	*91.5 ± 8.4*
Δ^{14}C (‰) ±1 σ	May	–11.1 ± 4.7	36.1 ± 4.7	0.0 ± 4.2	27.3 ± 4.4	–3.7 ± 4.8
	July	70.8 ± 6.6	–5.2 ± 4.5	–0.7 ± 4.3	–3.0 ± 5.4	–35.4 ± 5.2
	September	51.0 ± 6.8	–66.4 ± 5.0	–113.6 ± 4.7	–120.2 ± 5.1	–142.3 ± 5.0
	November	90.0 ± 6.4	15.5 ± 6.1	–45.7 ± 6.0	–171.8 ± 5.6	–199.1 ± 5.4
	Average	*50.2 ± 43.8*	*–5.0 ± 44.3*	*–40.0 ± 53.5*	*–66.9 ± 94.5*	*–95.1 ± 91.2*

[a] psu = practical salinity units.

The ^{14}C concentrations in the fish, benthos, seagrass, and zooplankton groups were 106 ± 0.7, 105 ± 0.6, 106 ± 0.7, and 106 ± 1.7 pMC, respectively. However, the ^{14}C in the phytoplankton group was lower (103 ± 2.4 pMC). The salinity at the time when the phytoplankton was collected was 15 ± 5

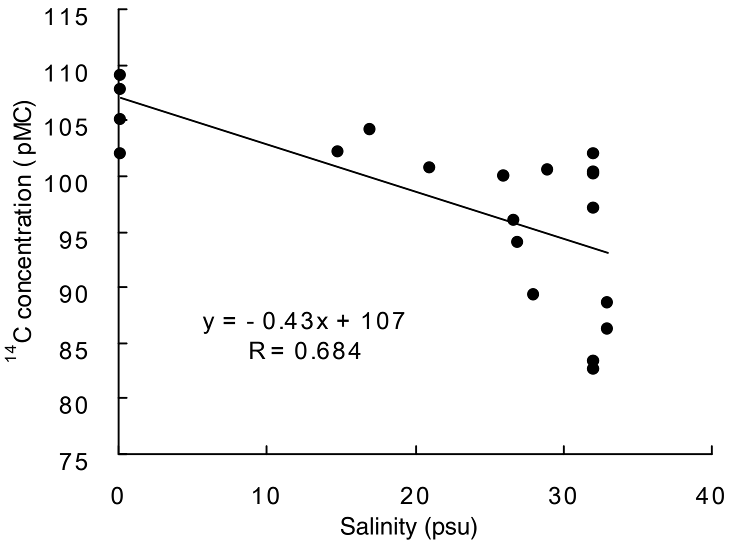

Figure 2 Relationship between ^{14}C activity and salinity in the water samples

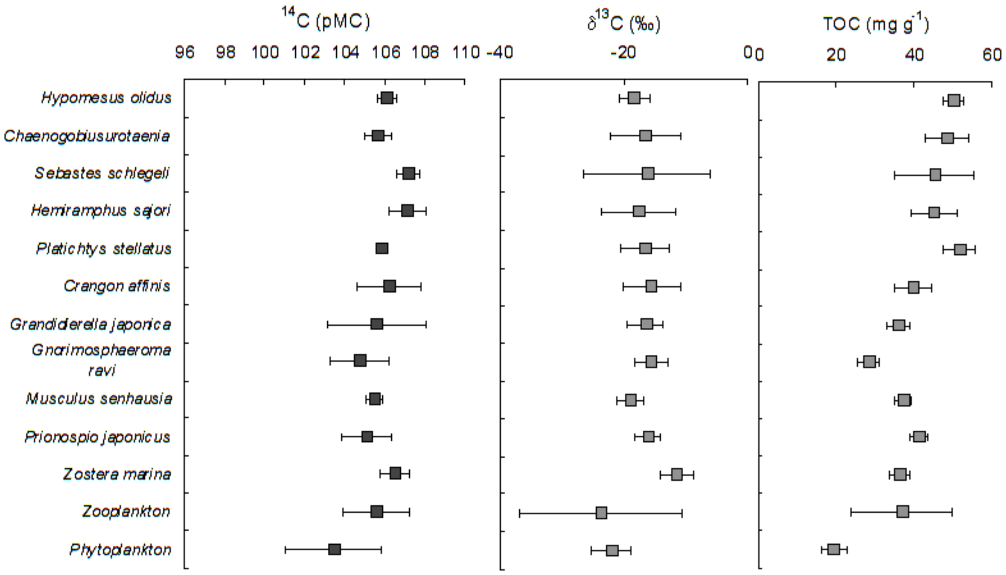

Figure 3 ^{14}C activity, δ^{13}C, and TOC (total organic carbon) in aquatic biota from Lake Obuchi

psu, and the ^{14}C concentration in brackish water samples collected at this time was low (average 100 ± 4 pMC). The ^{14}C concentration in the phytoplankton was lower due to direct uptake of brackish water, followed by photosynthesis of the phytoplankton.

As described above, the ^{14}C concentration in biota (fish, benthos, seagrass, and zooplankton) of the lake was 105 pMC, very similar to the concentration of ^{14}C in the atmosphere. The difference between the lower- and higher-trophic biological stages was negligible.

Table 3 Analytical results of total organic carbon (TOC), δ^{13}C, and ^{14}C (pMC and Δ) in aquatic biota samples from Lake Obuchi.

		TOC	δ^{13}C	^{14}C		
Group	Creature	(mg/g) ±1 σ	(‰) ±1 σ	pMC(%) ±1 σ	Δ^{14}C(‰) ±1 σ	*n*
Fish	*Hypomesus olidus*	50.1 ± 2.5	–18.3 ± 0.7	106.1 ± 0.5	61.1 ± 4.7	6
	Chaenogobius urotaenia	48.6 ± 5.7	–16.6 ± 1.3	105.7 ± 0.7	56.6 ± 6.5	6
	Sebastes schlegeli	45.3 ± 10	–16.3 ± 0.3	107.2 ± 0.6	71.6 ± 5.9	2
	Hemiramphus sajori	45.3 ± 5.9	–17.6 ± 0.3	107.1 ± 0.9	71.3 ± 9.1	2
	Platichtys stellatus	51.8 ± 4.0	–16.7 ± 0.1	105.8 ± 0.1	58.4 ± 1.3	2
	Group average	*48.2 ± 2.9*	*–17.1 ± 0.8*	*106.4 ± 0.7*	*63.8 ± 7.2*	
Benthos	*Crangon affinis*	39.8 ± 4.7	–15.6 ± 0.7	106.2 ± 1.6	62.1 ± 15.8	6
	Grandidierella japonica	35.9 ± 2.9	–16.5 ± 1.9	105.6 ± 2.4	56.1 ± 24.4	6
	Gnorimosphaeroma sp.	28.4 ± 2.7	–15.6 ± 1.1	104.7 ± 1.4	47.5 ± 14.5	5
	Musculus senhausia	37.4 ± 2.2	–19.0 ± 5.8	105.5 ± 0.4	50.9 ± 12.6	4
	Prionospio japonicus	41.3 ± 2.1	–16.1 ± 4.4	105.1 ± 1.3	67.5 ± 13.1	3
	Group average	*36.6 ± 5.0*	*–16.6 ± 1.4*	*105.4 ± 0.6*	*56.8 ± 8.2*	
Seagrass	*Zostera marina*	36.4 ± 2.7	–11.4 ± 1.0	106.5 ± 0.7	65.5 ± 7.5	6
Plankton	Zooplankton (100–1000 µm)	36.9 ± 13	–23.7 ± 3.4	105.6 ± 1.7	55.9 ± 16.5	6
	Phytoplankton (0.45–100 µm)	19.3 ± 3.3	–22.1 ± 1.8	103.5 ± 2.4	34.8 ± 23.7	6

Sediment Samples

Table 4 shows the analytical results of TOC, δ^{13}C, ^{14}C, ^{137}Cs, and ^{210}Pb$_{\text{excess}}$ in core sediment samples at Stations 2, 3, and 4 from Lake Obuchi. The vertical distributions of TOC in sediments from the 3 stations tended to be lower when going deeper into the sediment; TOC in the sediments from Stations 2, 3, and 4 ranged from 35–46, 34–55, and 32–53 mg/g, respectively (Figure 4), and the amount of organic substance in surface sediment (0–5 cm depth) from Stations 3 (55 mg/g) and 4 (average 53 mg/g) was slightly greater than that from Station 2 (46 mg/g).

Mean δ^{13}C values in sediment samples from Stations 2, 3, and 4 ranged from –24.9 to –21.5, –21.7 to –21.2, and –21.3 to –20.5‰, respectively (Figure 4). Though the δ^{13}C concentrations in the sediments of Stations 3 and 4 were nearly equal from the upper to lower layers, the δ^{13}C concentrations at Station 2 tended to be higher when going deeper into the sediment. The mean values of ^{14}C in sediment samples from Stations 2, 3, and 4 ranged from 92–104, 91–104, and 92–106 pMC, respectively. The core sediment depths with the maximum ^{14}C activities at Stations 2, 3, and 4 were 15–20, 10–15, and 10–15 cm, respectively. Observed ^{14}C activities were similar to that in the atmosphere; however, though atmospheric ^{14}C peaked around 1963 (~180 pMC) due to atmospheric nuclear weapons testing (Nydal and Lovseth 1983; UNSCEAR 2000; Yamada et al. 2005), the maximum concentration of ^{14}C in the sediments was ~106 pMC, considerably lower than the atmospheric ^{14}C concentration in the past. The ^{14}C concentration in the Lake Obuchi water samples was low (96–100 pMC on average), as was the ^{14}C concentration of phytoplankton (103 ± 2 pMC on average). The dominant species of phytoplankton in this lake was *Skeletonema costatum* of Bacillariophyceae (Ueda et al. 2004a), and the optimal condition of salinity for its multiplication is >20 psu (Kondo et al. 2004). Thus, the particulate matter that settled into the bottom sediment would be phytoplankton, which has the lowest ^{14}C concentration throughout the year.

Table 4 Analytical results of total organic carbon (TOC), $\delta^{13}C$, ^{14}C (pMC and Δ), and the ^{14}C age in core sediment samples from Lake Obuchi.

		TOC	$\delta^{13}C$	^{14}C		Age of ^{14}C
Site	Depth (cm)	(mg/g) ±1 σ	(‰) ±1 σ	pMC(%) ±1 σ	$\Delta^{14}C$(‰) ±1 σ	BP ±1 σ
Station 2	0–5	46.4	–24.9	101.4 ± 0.5	1.3 ± 5.0	—
	5–10	44.6	–24.1	103.1 ± 0.5	18.1 ± 5.1	—
	10–15	42.8	–23.9	102.5 ± 0.5	12.2 ± 5.1	—
	15–20	39.8	–22.9	103.7 ± 0.5	24.0 ± 4.8	—
	20–25	43.4	–22.3	101.9 ± 0.5	6.3 ± 4.9	—
	25–30	40.8	–22.0	94.8 ± 0.5	–63.8 ± 4.7	430 ± 40
	30–35	38.6	–22.1	92.1 ± 0.5	–90.4 ± 4.6	660 ± 40
	35–40	35.5	–22.0	93.0 ± 0.5	–81.3 ± 4.6	580 ± 40
	40–45	35.0	–21.5	93.2 ± 0.5	–80.2 ± 4.6	570 ± 40
	45–50	35.4	–22.2	92.2 ± 0.5	–89.3 ± 4.6	650 ± 40
	Average	*40.2 ± 4.1*	*–22.8 ± 1.1*	*97.8 ± 5.1*	*–34.3 ± 50.1*	
Station 3	0–5	55.3	–21.2	102.2 ± 0.4	21.6 ± 3.5	—
	5–10	52.9	–21.3	102.6 ± 0.4	25.9 ± 3.7	—
	10–15	50.9	–21.2	103.6 ± 0.4	35.9 ± 3.8	—
	15–20	49.0	–21.4	98.6 ± 0.4	–14.4 ± 3.6	175 ± 29
	20–25	43.3	–21.6	93.1 ± 0.3	–68.6 ± 3.4	626 ± 29
	25–30	40.9	–21.8	93.2 ± 0.3	–67.7 ± 3.4	615 ± 29
	30–35	38.7	–21.8	92.0 ± 0.3	–80.5 ± 3.3	725 ± 29
	35–40	36.5	–21.7	91.8 ± 0.3	–82.1 ± 3.4	742 ± 30
	40–45	33.8	–21.6	92.1 ± 0.4	–79.3 ± 4.1	719 ± 36
	45–50	34.0	–21.5	91.5 ± 0.3	–85.1 ± 3.3	771 ± 29
	50–55	34.0	–21.7	91.3 ± 0.3	–87.0 ± 3.4	784 ± 30
	Average	*42.7 ± 8.1*	*–21.5 ± 0.2*	*95.0 ± 5.0*	*–50.3 ± 50.2*	
Station 4	0–5	52.5	–20.5	104.1 ± 0.5	19.1 ± 5.2	—
	5–10	48.4	–20.6	106.0 ± 0.5	37.9 ± 5.3	—
	10–15	48.1	–20.8	102.4 ± 0.5	2.3 ± 5.1	—
	15–20	47.9	–21.0	98.2 ± 0.5	–39.2 ± 4.8	220 ± 40
	20–25	38.7	–21.3	93.5 ± 0.5	–83.6 ± 4.6	600 ± 40
	25–30	37.4	–21.3	93.3 ± 0.5	–85.8 ± 4.6	620 ± 40
	30–35	31.9	–21.2	92.7 ± 0.5	–91.5 ± 4.6	670 ± 40
	35–40	31.8	–21.3	93.5 ± 0.5	–83.6 ± 4.6	600 ± 40
	40–45	33.5	–21.2	92.6 ± 0.5	–92.6 ± 4.6	680 ± 40
	45–50	33.5	–21.2	92.3 ± 0.5	–95.0 ± 4.6	700 ± 40
	Average	*40.4 ± 8.0*	*–21.0 ± 0.3*	*96.9 ± 5.4*	*–51.2 ± 52.1*	

In vertical profiles, modern carbon (post-AD 1950) was found in sediments at Station 2 at 0–25 cm depth, with ^{14}C ages from 430 ± 40 to 650 ± 40 BP (±1 σ). The modern carbon in sediments at Stations 3 and 4 was found at 0–15 cm depth, with ^{14}C ages from 175 ± 29 to 784 ± 30 BP. The sedimentation rates from the core sediments were calculated using the ^{210}Pb dating method (Ueda et al. 2004b). The vertical profiles of $^{210}Pb_{excess}$ activity in sediments from Stations 3 and 4 exponentially decreased from the surface to the bottom layer; sedimentation rates at Stations 3 and 4 were estimated as 0.11 ± 0.04 and 0.10 ± 0.02 g cm^{-2} yr^{-1}, respectively. From the ^{210}Pb dating results, the

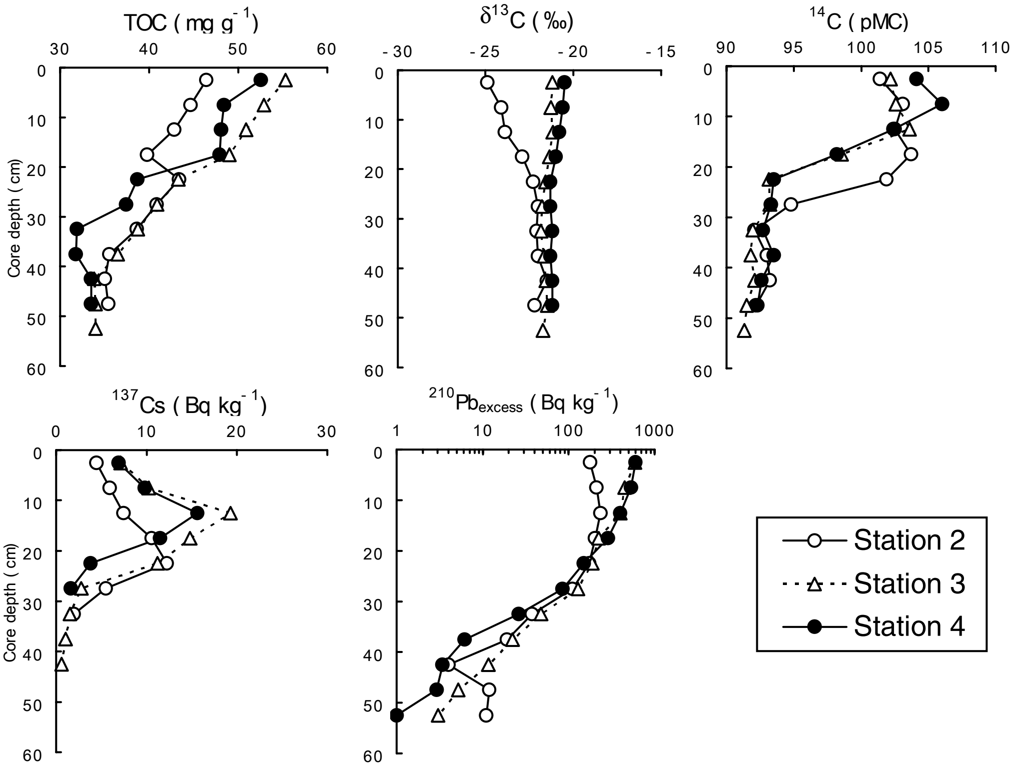

Figure 4 Vertical profiles of total organic carbon (TOC), $\delta^{13}C$, ^{14}C, ^{137}Cs and $^{210}Pb_{excess}$ in core sediment samples at Stations 2, 3, and 4 from Lake Obuchi. Profile data of ^{137}Cs and $^{210}Pb_{excess}$ are from Ueda et al. (2004b).

ages of the 15–20-cm layer in the sediments from Stations 3 and 4 were estimated as 130 ± 40 and 140 ± 30 BP, respectively. Compared with the results of ^{14}C dating (175–220 BP), the ^{210}Pb dating results tended to be about 50 yr younger.

A maximum depth of ^{14}C in sediments was compared with a peak for ^{137}Cs (Ueda et al. 2004b). Although atmospheric fallout of ^{14}C and ^{137}Cs peaked in 1963–1964, the ^{14}C peak in sediment was slightly later than the ^{137}Cs peak (Bergan 2002). This probably reflects the different sedimentation rates of each radionuclide (McGeehin et al. 2004). Almost no cesium that accumulated in the sediment was eluted to lake water (Ueda et al. 1999), but carbon was easily eluted from sediment to lake water by decomposition of organic matter in the sediment. Differences in vertical profiles of ^{14}C and ^{137}Cs seem to be strongly dependent on the dissolution characteristics of both radionuclides.

CONCLUSION

The concentration levels and horizontal distribution of ^{14}C in aquatic samples from brackish Lake Obuchi in Rokkasho Village were obtained before the nuclear fuel reprocessing facilities commenced operations. Concentrations of ^{14}C in freshwater, brackish water, and seawater samples in 2004 had ranges of 102 ± 0.5 to 109 ± 0.6 (average 105 ± 4 pMC); 89 ± 0.5 to 104 ± 0.4 (average 98 ± 0.5 pMC); and 82 ± 0.6 to 102 ± 0.4 (average 93 ± 0.5 pMC), respectively. The relationship between ^{14}C concentration and salinity showed a negative correlation ($r = 0.68$, $P < 0.01$, $n = 20$). ^{14}C concentration in water samples from Futamata River was similar to the ^{14}C level (~106 pMC) in the atmosphere; however, the ^{14}C concentration in brackish water samples was lower than that of the atmosphere due to a dilution by seawater.

Concentration of ^{14}C in aquatic biota (fish, benthos, and seagrass) from 2003 to 2004 ranged from 105 ± 0.7 to 107 ± 0.6 pMC, while values of 103 ± 2.4 to 105 ± 1.7 pMC were observed for zooplankton and phytoplankton. The ^{14}C concentration in aquatic biota was ~105 pMC, and the difference among biota was small.

The vertical profile of ^{14}C in 3 core sediment samples collected from this lake in 1997 showed maximum concentrations of ^{14}C from 103 ± 0.5 to 106 ± 0.5 pMC at depths of 5 to 20 cm. The profile of ^{14}C in the sediment was different from the variation in the atmospheric ^{14}C fallout. We confirmed that the background level of ^{14}C concentration in aquatic samples in brackish Lake Obuchi before operation of the reprocessing plant was similar to the concentration level in the atmosphere.

^{14}C may enter this lake from the ocean via tides, from the land via rivers, and/or from the atmosphere via fallout. The behavior of ^{14}C is strongly influenced by environmental factors in the lake ecosystem. This study will contribute to evaluations of the surrounding environment regarding discharge of radioactive nuclides from the nuclear fuel cycle facilities after commercial operation begins.

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