

SPATIAL DISTRIBUTION OF $\Delta^{14}\text{C}$ VALUES OF ORGANIC MATTER IN SURFACE SEDIMENTS OFF SARU RIVER IN NORTHERN JAPAN, ONE YEAR AFTER A FLOOD EVENT IN 2006

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ABSTRACT. Dispersion and deposition of terrestrial organic matter by flooding on the inner shelf were studied using C/N ratios, $\delta^{13}\text{C}$, and $\Delta^{14}\text{C}$ values of sedimentary organic matter. Surface sediment samples (top 2 cm) were collected from coastal areas near the Saru River in southwestern Hokkaido, northern Japan, 1 yr after a flood event in 2006. Riverine suspended solids were also collected at a fixed station downstream during 2006–2008. Sandy sediments were located at the front of the river mouth and the western part of the sampling area, with the $\delta^{13}\text{C}$ of organic matter ranging from -23.8‰ to -22.0‰ , $\Delta^{14}\text{C}$ of -655‰ to -388‰ , and an organic carbon/total nitrogen (C/N) ratio of 5.9–7.7. On the other hand, silt and clay sediments were distributed in a restricted area 11–16 km from the river mouth, with lighter $\delta^{13}\text{C}$ (-26.7‰ to -24.1‰) and higher $\Delta^{14}\text{C}$ (-240‰ to -77‰) of organic matter and C/N ratio (7.8–13.3). From end-member analysis, the apparently younger and less degraded organic matter in the silt and clay sediments consists mainly of terrestrial organic matter released by flood events. They remain in the depression, although most flood deposits were moved to deep-sea environments.

INTRODUCTION

Continental shelf sediments play an important role in global geochemical cycles of carbon because they are recognized as the dominant reservoir for organic carbon burial in marine environments (Berner 1982; Hedges and Keil 1995). Coastal and shelf sedimentary organic carbon comprise materials derived from both marine and terrestrial sources. The distribution of terrestrial organic matter on continental margins is tightly linked to the dispersal of river-suspended sediments. It also depends on the characteristics of river systems and the marginal region. The transport of organic matter by flooding is important to elucidate the dynamics and flux of organic matter released from terrestrial environments. Recently, the effects of flooding on the deposition of organic matter have been studied at the continental shelf off the Po River (Miserocchi et al. 2007), Eel River (Blair et al. 2003), and Saru River (Ikehara et al. 2006). Tesi et al. (2008) reported that sedimentary organic matter in the flood deposit was initially dominated by aged, lignin-poor organic matter adsorbed onto the fine materials. About half of the fine-grained organic carbon delivered to the shelf off the Eel River is derived from ancient sedimentary organic carbon found in the watershed (Blair et al. 2003). Ikehara et al. (2006) pointed out that the inner shelf topography controls dispersal and depositional patterns of the flood sediments.

To better elucidate the fate of organic matter after a large flood event, complementary analyses are needed for surface sediments. Various geochemical approaches have been used to define the mixing ratio of marine and terrestrial organic matter, including the C/N ratio, $\delta^{13}\text{C}$, and lignin biomarkers. Radiocarbon analysis is useful for studying the dynamics of soil, river, and marine organic matter on

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timescales of years to decades (Hedges and Keil 1995; Trumbore 2000; Raymond and Bauer 2001) because nuclear weapons testing has injected large quantities of ^{14}C into the atmosphere. The $\Delta^{14}\text{C}$ of particulate organic matter (POM) in rivers shows a great dynamic range of -980‰ to $+227\text{‰}$ and depends on river systems with various watershed conditions. The $\delta^{13}\text{C}$ values and C/N ratios vary among sources (vascular plants, soil organic matter, algae in rivers) and according to the degradation of organic matter in terrestrial environments (e.g. Melillo et al. 1989; Balesdent et al. 1993; Hellings et al. 1999; Onstad et al. 2000; Kendall et al. 2001). On the other hand, marine sedimentary organic matter shows almost constant $\delta^{13}\text{C}$ values and C/N ratios (Miserocchi et al. 1999; Usui et al. 2006). Therefore, simultaneous use of C/N ratios, $\Delta^{14}\text{C}$, and $\delta^{13}\text{C}$ values can provide a convenient and reliable tool for tracing and identifying the fate of bulk POM discharged from rivers (Goñi et al. 1997; Guo et al. 2004; Nagao et al. 2005). Using the ^{14}C isotopic mass balance, the abundances of modern and ancient organic matter can also be constrained quantitatively for marine sediments.

This report describes the spatial distribution of organic matter contents and its character (C/N ratio, $\Delta^{14}\text{C}$, and $\delta^{13}\text{C}$ values) in surface sediments on the continental shelf. We collected surface sediments at a water depth of 19–39 m off the Saru River 1 yr after a severe flood with maximum water discharge of $\sim 3000\text{ m}^3/\text{s}$. We discuss the dispersion and deposition of terrestrial organic matter originating from the Saru River, a small mountainous river in southwestern Hokkaido, Japan.

STUDY AREA

The Saru River flows from the Hidaka Mountains through a forest area ($\sim 90\%$ of watershed) (Figure 1). It is 104-km long with a drainage basin of 1350 km^2 . The annual mean water discharge was measured as $54\text{ m}^3/\text{s}$ at a gauge observatory of Tomikawa during 1972–2007 (Ministry of Land, Infrastructure, Transport and Tourism, Japan 2009). The annual mean precipitation was 1003 mm at Hidakamonbetsu for 1995–2004 (Japan Meteorological Agency 2009). The Saru River has a riverbed gradient of $1/50$ – $1/700$ and is classified as a torrential river. The Saru River Basin is underlain by sedimentary rocks including metamorphic rocks of the Cretaceous period with sedimentary rocks of the Tertiary period distributed in the riversides of the lower reaches (Hokkaido Regional Development Bureau 2007).

Flood Events in the Saru River

In the last 7 yr, the Saru River experienced 2 large floods—in 2003 and 2006—with maximum water discharge greater than $3000\text{ m}^3/\text{s}$. Typhoon No. 10 in 2003, with the typhoon number counting from the beginning of the typhoon season in January, struck the Japanese Islands on August 8–10. It caused extensive flood damage in Kochi, Okayama, Mie, Ehime, and Hokkaido prefectures. Kamei (2006) outlined the flood disaster in the Hidaka area of Hokkaido. This area was subjected to heavy rainfall 3 times during the approach of Typhoon No. 10. Maximum hourly and daily rainfall reached 91 and 307 mm, respectively. The rainwater in the third pulse conducted mud and driftwood into the river, the result of slope failures and debris flow along the upstream parts of the river. The maximum water discharge was $6600\text{ m}^3/\text{s}$ at the Biratori gauge station of the Saru River (Ministry of Land, Infrastructure, Transport and Tourism, Japan 2009).

A large flood also occurred at the Saru River watershed as a result of the stationary front caused by Typhoon No. 10 on 18–19 August 2006. The maximum daily rainfall was $\sim 311\text{ mm}$, similar to the prior flood in 2003. The flooded area in the lower watershed was estimated as $\sim 140\text{ ha}$ (Hokkaido Regional Development Bureau 2006). The maximum water discharge at the Biratori gauge station of the middle Saru River was $2959\text{ m}^3/\text{s}$ (Hokkaido Regional Development Bureau 2006).

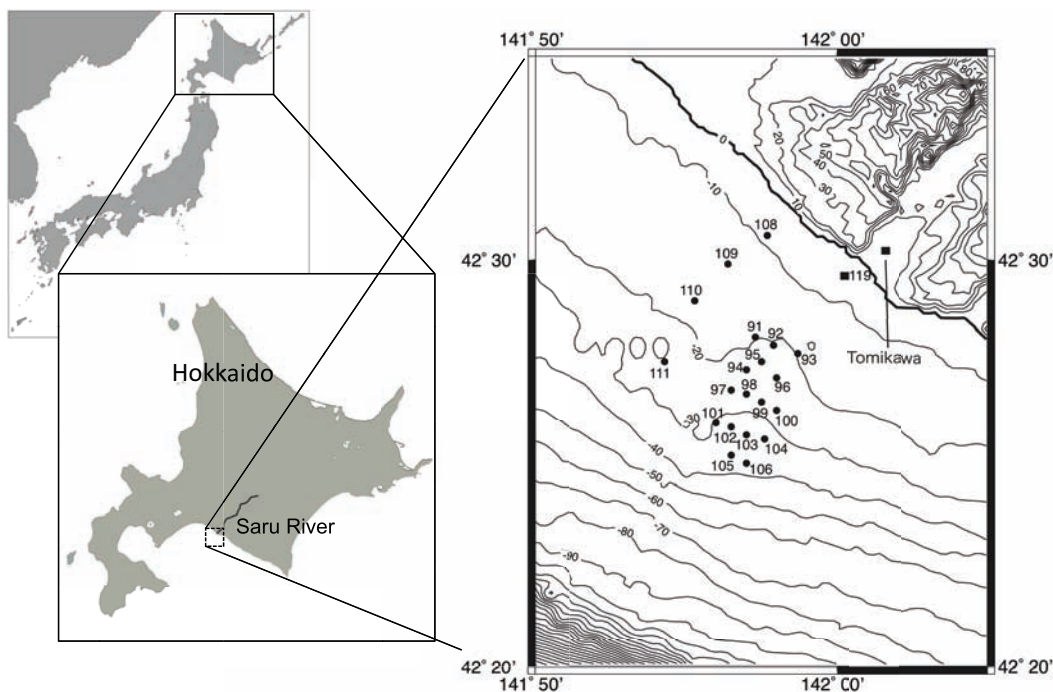


Figure 1 Sampling locations and water depths (m) in the coastal sea near the Saru River in Hokkaido, Japan. Closed circles and squares represent sampling stations for the surface sediments and suspended solids, respectively, used for this study.

MATERIALS AND METHODS

Surface sediments were collected with a K-grab sampler in August 2007, 1 yr after the severe flood in 2006. River water samples were collected at the Tomikawa gauge station, 2.8 km from the river mouth in August 2006 and 2007, and in April 2008. Suspended solid (SS) samples were concentrated from 45–90 L of river water using a single-bowl flow continuous centrifuge technique to obtain a large amount of suspended particulate matter (Nagano et al. 2003). The SS in coastal seawater was concentrated from 103 L of the water sample using continuous centrifugation through a NXXX25 plankton net. We also collected a SS sample from the Saru River water in April 2008 using glass fiber filters (GF/F; Whatman plc.). All samples were freeze-dried and ground in an agate mortar except for the GF/F filter sample.

Aliquots of powdered surface sediments (top 2 cm) and riverine suspended particulate matter were measured for organic carbon and total nitrogen content using a total carbon analyzer (WR-112; Leco Corp.) and/or an elemental analyzer (Flash EA1112; Thermo Scientific) after HCl treatment. The molar ratio of organic carbon/total nitrogen content is expressed as the C/N ratio. The stable carbon isotopic ratio was determined using an elemental analyzer (Flash EA1112; Thermo Scientific) connected to a mass spectrometer (Delta V Plus; Thermo Scientific) via an interface (Conflo III; Thermo Scientific), with $\delta^{13}\text{C}$ notation (‰) versus the VPDB standard. ^{14}C values of organic matter in SS samples were determined using accelerator mass spectrometry (AMS) at the National Institute for Environmental Studies (Tanaka et al. 2000) and the Mutsu Office of the Japan Atomic Energy Agency (Aramaki et al. 2000). ^{14}C values are reported as $\Delta^{14}\text{C}$ corrected for sample $\delta^{13}\text{C}$ (Stuiver and Polach 1977). Absolute errors for $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ analyses are $<0.2\text{‰}$ and $<7\text{‰}$, respectively.

RESULTS AND DISCUSSION

Characteristics of Riverine POM

The particulate organic carbon (POC), C/N molar ratio, and carbon isotope results are presented in Table 1. $\delta^{13}\text{C}$ values of riverine POC are -26.7‰ to -26.4‰ for the summer samples during normal flow conditions, and -25.0‰ to -24.5‰ for the spring samples during higher flow conditions. The $\Delta^{14}\text{C}$ values are -296‰ to -247‰ for the summer and -720‰ for the spring samples. Kao and Liu (1996) reported that the Lanyang His has very low $\Delta^{14}\text{C}$ values (-874‰ to -714‰) of POC. The riverine POC with older age shows the presence of fossil organic matter such as bitumens or kerogen, and/or the entrainment of terrigenous organic matter of long residence times within the drainage basin. On the other hand, POM in coastal seawater off the Saru River has higher $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values than the riverine POC. Turbidity significantly decreased from 114 NTU (nephelometric turbidity unit) for the river water to 16 NTU for the coastal surface water. These results indicate that the riverine POC was diluted by seawater and organic matter that was freshly produced in the sea surface.

Table 1 Organic carbon content, C/N molar ratio, radiocarbon ($\Delta^{14}\text{C}$) and stable carbon isotopic composition ($\delta^{13}\text{C}$) of organic matter from the Saru River, coastal seawater, and surface sediments.^a

Sample	Station	Depth (m)	Sampling date	TOC (wt%)	C/N ratio	$\Delta^{14}\text{C}$ (‰)	$\delta^{13}\text{C}$ (‰)	Type of surface sediment
River SS	Tomikawa	0	2006 July 9	2.03	8.0	-247 ± 2	-26.7	—
		0	2007 May 8	n.m.	n.m.	-720 ± 2	-25.0	—
		0	2007 Aug 7	1.29	9.9	-296 ± 7	-26.4	—
		0	2008 Apr 30	0.65	n.m.	n.m.	-24.5	—
Seawater SS	119	0	2007 Aug 8	2.95	7.2	-121 ± 2	-23.0	—
Surface sediments	91	19	2007 Aug 8	0.72	8.9	n.m.	-24.4	Silt (surface)/coarse sand (0–2 cm)
	93	20	2007 Aug 8	0.08	5.8	-388 ± 3	-22.0	Fine sand, well sorted (0–4 cm)
	94	24	2007 Aug 8	0.13	5.9	-665 ± 1	-23.1	Fine sand, well sorted (0–3 cm)
	95	23	2007 Aug 8	1.68	10.0	-119 ± 3	-24.6	Clay (0–3 cm)
	96	24	2007 Aug 8	1.04	10.9	-168 ± 3	-26.0	Clay (0–6 cm)/fine sand (6–7 cm)
	97	27	2007 Aug 8	0.26	7.7	-476 ± 3	-23.8	Clay (0–1.5 cm)/medium sand (1.5–3.5 cm)
	98	28	2007 Aug 8	0.56	7.8	-240 ± 4	-24.1	Very fine sand (0–1 cm)/silt (1–3 cm)
	99	28	2007 Aug 8	1.26	9.7	-224 ± 4	-24.8	Clay (0–7 cm)
	101	32	2007 Aug 8	0.91	11.0	-136 ± 3	-25.8	Fine sand (0–6.5 cm)
	103	33	2007 Aug 8	0.19	6.2	-490 ± 2	-23.6	Muddy fine sand (surface)/fine sand (0–4 cm)
	104	33	2007 Aug 8	1.87	13.3	-77 ± 6	-26.7	Clay (0–6.5 cm)
105	37	2007 Aug 8	1.45	12.7	-112 ± 3	-25.1	Silt (0–4.5 cm)	
106	39	2007 Aug 8	1.03	12.3	-134 ± 3	-24.8	Sandy silt (0–0.5 cm)/sandy silt (0.5–3 cm)	
108	10	2007 Aug 8	0.25	6.3	-500 ± 2	-23.3	Coarse silt, well sorted (0–5.5 cm)	
109	14	2007 Aug 8	0.09	6.3	-441 ± 2	-22.2	Fine sand (0–3 cm)	
110	17	2007 Aug 8	0.09	4.9	-610 ± 2	-22.1	Fine sand (0–3 cm)	
111	24	2007 Aug 8	0.18	6.2	-496 ± 2	-23.2	Medium sand, granule, shell fragments (0–3.5 cm)	

^an.m.= not measured.

Spatial Distribution of Organic Matter in Surface Sediments

Spatial distributions of TOC, C/N ratio, $\delta^{13}\text{C}$, and $\Delta^{14}\text{C}$ values of surface sediments off the Saru River are shown in Figure 2. The organic carbon content is 0.56–1.87% for silt and clay sediments and 0.09–0.26% for sandy sediments. The C/N molar ratio is 7.8–13.3 for silt and clay sediments and 4.9–7.7 for sandy sediments. The organic materials for the silt and clay sediments and sandy sediments have $\delta^{13}\text{C}$ values of -26.7‰ to -24.5‰ and -23.8‰ to -22.0‰ , respectively. The sedimentary organic matter shows $\Delta^{14}\text{C}$ values between -665‰ and -77‰ . Silt and clay sediments have $\Delta^{14}\text{C}$ values ranging from -240‰ to -77‰ , and sandy sediments, -665‰ to -388‰ . An area with higher values for all parameters is formed as a ring of least 5 km width and length, located 11–16 km from the river mouth. The differences in carbon isotopic composition and C/N ratios are related to the sources as well as the transport and sedimentation processes of POM (Guo et al. 2004; Goñi et al. 2005).

$\Delta^{14}\text{C}$, $\delta^{13}\text{C}$ Values of Surface Sediments on Continental Shelf Areas off Various River Systems

Figure 3 portrays values of $\Delta^{14}\text{C}$ versus $\delta^{13}\text{C}$ of organic matter in surface sediments from estuarine and continental shelf areas off the Saru River, Tokachi River (Nagao et al. 2005), Mackenzie River (Goñi et al. 2005), and Siberian rivers (Guo et al. 2004). The shelf sediments off the Saru River have $\delta^{13}\text{C}$ values of -26.7‰ to -22.0‰ and $\Delta^{14}\text{C}$ of -665‰ to -77‰ . The surface sediments in continental shelf areas off the Tokachi River gave $\delta^{13}\text{C}$ values of -24.5‰ to -21.7‰ and $\Delta^{14}\text{C}$ of -273‰ to -164‰ . The $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values of the sediments near the Siberian Rivers were, respectively, -26.2‰ to -23.8‰ and -805‰ to -279‰ . Land-derived organic carbon accounts for the majority (>60%) of the organic matter present in the Arctic shelf sediments. Although not shown in Figure 3, the shelf flood deposits off the Eel River have $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values of about -25‰ and -600‰ , respectively (Blair et al. 2003). Surface sediments of the prodelta off the Po River have an average $\Delta^{14}\text{C}$ of $-299 \pm 56\text{‰}$ and $\delta^{13}\text{C}$ value of $-24.8 \pm 0.9\text{‰}$ at 2 months after a flood event (Tesi et al. 2008). These results indicate that surface sediments off the Saru River vary widely in $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values for organic matter. The area of $\Delta^{14}\text{C}$ versus $\delta^{13}\text{C}$ of organic matter shown for the sediments off the Saru River differs from those for other shelf sediments, suggesting a linkage of river systems.

Deposition Processes of Terrestrial Organic Matter on the Shelf off the Saru River

Many landslides occurred in 2003 and 2006 due to heavy rain from typhoons. Large amounts of terrigenous sand and mud were supplied to coastal sea areas through rivers (Yamashita 2004; Hokkaido Regional Development Bureau 2006). Based on $\delta^{13}\text{C}$ values of POC, C/N ratio, and contents of freshwater diatoms, Ikehara et al. (2006) determined that mud deposited on the inner shelf off the Saru River, from which the sediment samples were collected in September–October 2005 and July 2006, was of flood origin. Figure 4 portrays the elemental and isotopic compositions of surface sediments off the Saru River collected in August 2007 for this study. Riverine POC values (Table 1) and phytoplankton are also included in Figure 4 and were used to consider quantitative contributions for each end-member. The $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values for temperate phytoplankton used -21‰ and $+59\text{‰}$, respectively (Rau et al. 1982; Wang et al. 1998). The averaged C/N ratio (11.9) for phytoplankton and zooplankton was used to elucidate the maximum range (Wang et al. 1998), although C/N ratios of phytoplankton are 6.6–14 depending on environmental conditions (Mongin et al. 2003; Nieuwerburgh et al. 2004). Figure 4 shows that most sediment samples fell outside the ranges for a mixture of riverine POC and marine end-members. The source for apparent younger sediments might be the floods of 2003 and 2006, when heavy rains caused landslides and flooding in the watershed. The top layer of soil generally shows the presence of bomb carbon, and the decrease in $\Delta^{14}\text{C}$ values with depth in native undisturbed soils might result from new soil organic carbon inputs at the surface and

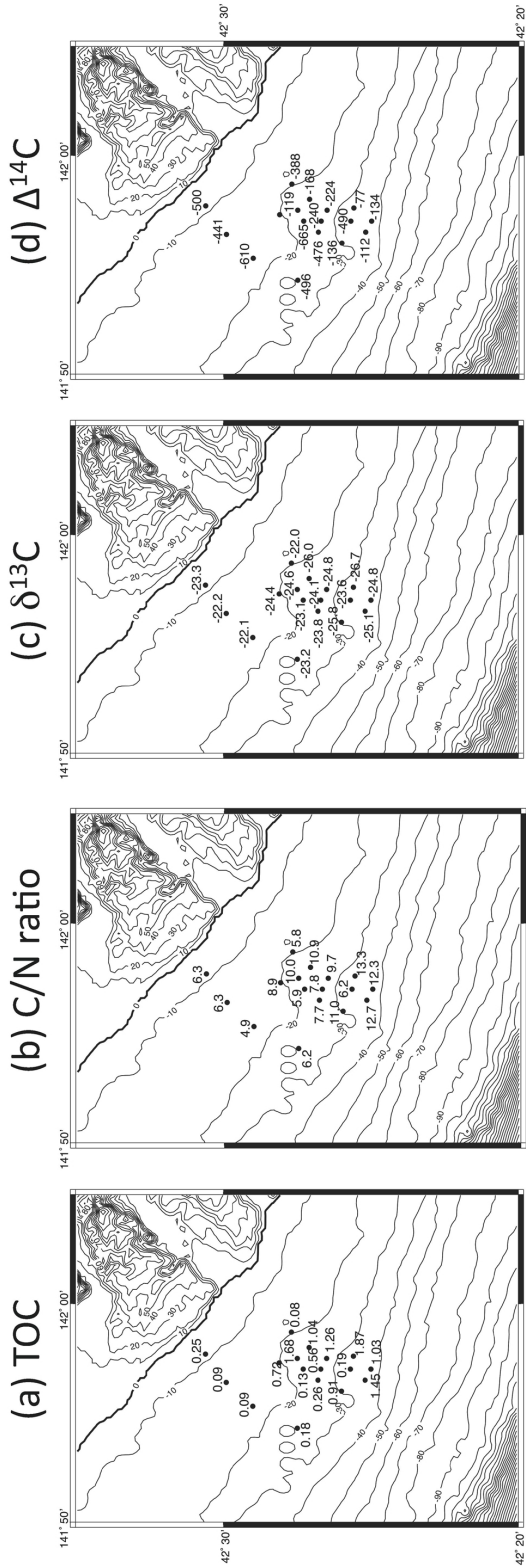


Figure 2 Contour maps of (a) organic carbon content (%), (b) C/N molar ratio, (c) and (d) carbon isotopic compositions ($\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$, respectively) of organic matter from surface sediments off the Saru River. Enclosed circles represent sampling stations.

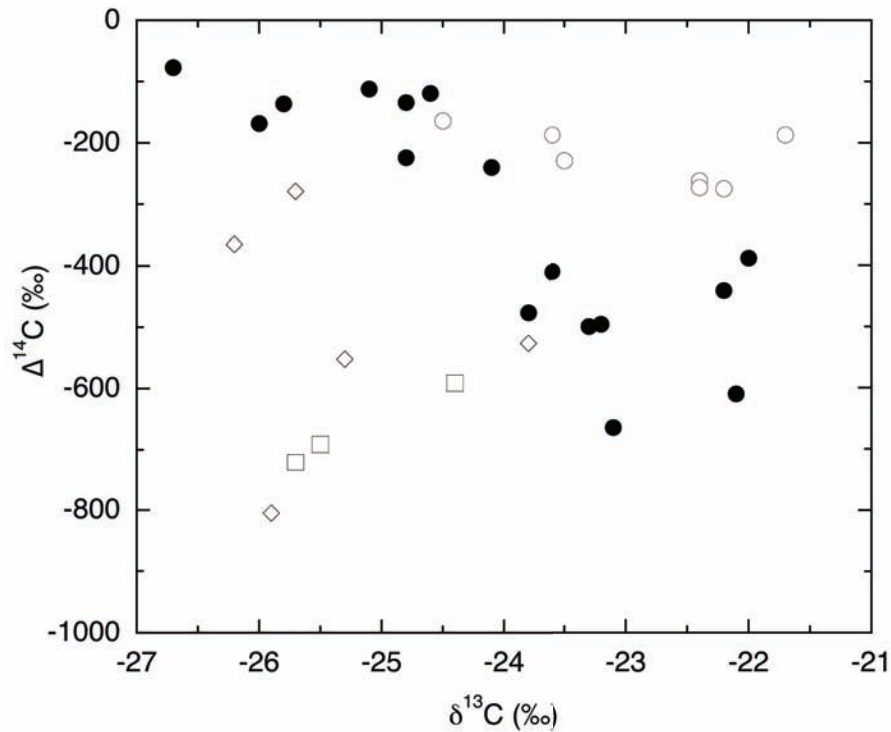


Figure 3 Plot of $\Delta^{14}\text{C}$ versus $\delta^{13}\text{C}$ of the surface sediments from river, estuary and continental shelf off the coast near the Saru River (●Table 1), Tokachi River (○; Nagao et al. 2005), Mackenzie River (□; Goñi et al. 2005) and Siberian Rivers (□; Ob, Yenisey, Khatanga, Lene, and Indigirka rivers from Guo et al. 2004).

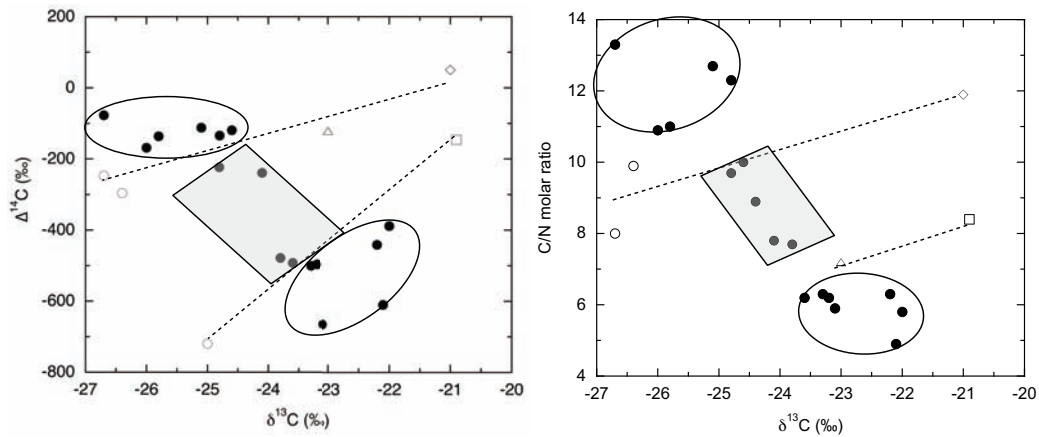


Figure 4 Plots of radiocarbon ($\Delta^{14}\text{C}$) vs. stable carbon isotopic composition ($\delta^{13}\text{C}$) of organic carbon, and $\delta^{13}\text{C}$ vs. C/N ratio in the shelf sediments off the Saru River. Included in this figure are the compositions of plausible sources of organic matter from published data (Wang et al. 1998; Nagao et al. 2005). The symbols are as follows: ●, shelf sediments; ○, Saru River POC; △, coastal sea POC in this study; □, plankton (Rau et al. 1982; Wang et al. 1998); and □, averaged value for the slope sediments off the Tokachi River (Nagao et al. 2005). Shaded areas highlight mixtures of the riverine and marine organic matter.

accumulation of old, resistant components at depth (Trumbore et al. 1995; Trumbore 2000). The downward trend of the C/N ratio and increase of $\delta^{13}\text{C}$ value are found for undisturbed forest soil because of the decomposition of organic matter (Melillo et al. 1989; Balesdent et al. 1993). In the Japanese forest area, organic matter at the 0–16 cm mineral soil layer gave $\delta^{13}\text{C}$ values of -27.3‰ to -25.2‰ , $\Delta^{14}\text{C}$ from -100‰ to $+245\text{‰}$, and a C/N ratio of 20–17 (Yamashita et al. 2004; Koarashi et al. 2005; Liu et al. 2006). Soon after the 2003 flood, mud covered a wide area on the sandy sediment inner shelf (Yamashita 2004). Katayama et al. (2007) reported that mud covered the sandy sediments at a water depth of 23–32 m in a very restricted area, corresponding to the locations of silt and clay sediments in this study. Therefore, the distribution of mud is inconsistent with our hypothesis that the POC with higher $\delta^{13}\text{C}$ and lower $\Delta^{14}\text{C}$ is exported from the surface soil in forest areas in the upper and middle river watersheds.

Kao and Liu (1996) showed that most of the POC associated with high sediment yield is probably fossil carbon originating from kerogen in the bedrock. The sandy sediments with lower $\Delta^{14}\text{C}$ values are located at the northwest part of this study area. $\delta^{13}\text{C}$ values of organic matter in sandy sediments resemble those of coastal POC shown in Table 1. However, their old ages indicate the non-contribution of newly produced marine organic matter and as a result of long residence times. In the Hidaka region, very fine sand to fine sand is widely distributed on the shelf off Hidaka, although silt and clay sediments are distributed on the slope (Katayama 2007). Nevertheless, the sediments deposited by the flood retained the features acquired during flooding 1 yr after 2006 and/or 3 yr from 2003. From a marine geological survey done before the severe flood in 2006, mud-covered sandy sediments in a few areas rest in the depression on the shelf (Katayama et al. 2007).

CONCLUSION

The effects of flood events on the deposition of terrestrial organic matter in coastal environments were studied using C/N ratio, $\Delta^{14}\text{C}$, and $\delta^{13}\text{C}$ data of sedimentary organic matter. Surface sediments off the Saru River, southwestern Hokkaido in northern Japan, were collected from 17 sites using a K-grab sampler in August 2007, a year after a severe flood in 2006. Suspended particles of the Saru River were also collected at a gauge station in the lower region during spring-summer in 2006–2008. $\Delta^{14}\text{C}$ values of organic matter in surface sediments (water depth 10–39 m) range from -665‰ to -77‰ . The silt and clay sediments have $\Delta^{14}\text{C}$ values of -240‰ to -77‰ , but the sandy sediments range from -665‰ to -388‰ . The $\Delta^{14}\text{C}$ values of particulate organic carbon (POC) in the Saru River are -292‰ to -247‰ at normal flow conditions, and -720‰ during the spring snowmelt period. $\delta^{13}\text{C}$ values are -26.7‰ to -24.1‰ for the silt and clay sediments, -23.8‰ to -22.0‰ for the sandy sediments, and -26.7 to -24.5‰ for riverine POC. The C/N molar ratios are 7.8–13.3 for the silt and clay sediments, and 5.7–7.7 for the sandy sediments. Sedimentary organic matter with lighter $\delta^{13}\text{C}$ and higher $\Delta^{14}\text{C}$ and C/N ratio is distributed in the depression; it corresponds to silt and clay sediments. End-member analysis reveals that most sediment samples fell out of the ranges for a mixture of riverine POC and marine end-members. Therefore, these results demonstrate that the terrestrial organic matter was deposited on the inner shelf as mud sediments by flooding of the Saru River, and that it has a negligible contribution of freshly produced organic matter from the sea surface. Terrestrial organic matter derived from the flood events has been preserved in the depression at least 1 yr.

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