COMPOUND-SPECIFIC RADIOCARBON AGES OF FATTY ACIDS IN MARINE SEDIMENTS FROM THE WESTERN NORTH PACIFIC

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ABSTRACT. Compound-specific radiocarbon analysis of five fatty-acid biomarkers was conducted for marine sediments collected from the western North Pacific. The fatty acids (C_{12} to C_{34}) showed a typical bimodal distribution pattern with two maxima at C_{16} and C_{26}. Their carbon isotopic compositions ranged from –25.1‰ (C_{16}) to –31.8‰ (C_{28}), suggesting that they derived from terrestrial higher plants and marine organisms. A large variations of ^{14}C ages were found among the fatty acids detected in the same sedimentary horizon of the core, ranging from 530 BP (C_{18}) to 3250 BP (C_{28}). The results of ^{14}C analysis of fatty acids could be divided into two groups, i.e., lower molecular weight (LMW) fatty acids (C_{16}, C_{18}) derived from marine organisms and higher molecular weight (HMW) fatty acids (C_{24}, C_{26}, C_{28}) derived from terrestrial higher plants. The HMW fatty acids showed older ages, ranging from 2550 BP (C_{24}) to 3250 BP (C_{28}), than LMW fatty acids (530 BP [C_{18}] to 1,820 years BP [C_{16}]). On the other hand, bulk-phase total organic matter (TOM) showed the age of 2260 BP that is between those two groups, suggesting that it was likely a mixture of organic matter derived from marine and terrestrial sources. The compound specific ^{14}C ages and $\delta^{13}C$ data of sedimentary fatty acids presented here could provide useful information to decipher the fate and transport process of terrestrial organic matter to marine sediments.

INTRODUCTION

It is important to study glacial-interglacial changes in North Pacific circulation and hydrography because the Pacific Ocean is the end-member of the modern ocean circulation regime, and the circulation may have been different during previous climate states (Keigwin 1998). Such records as deciphering the interaction between the ocean and the atmosphere during late quaternary environment have been collected from the analysis of sediment cores with higher sedimentation rate, which in turn need more accurate dating methods for each sediment layer. Today we understand that a foraminifera-based chronology is best. However, this method requires a good preservation condition of calcium carbonate in the sediments. In the North Pacific regions it is often difficult to get sufficient amounts of planktonic foraminifera from sediment cores for accelerator mass spectrometry (AMS) analysis. Therefore, an alternative chronology tool is needed for sediment analysis in the Pacific Ocean.

The recently developed technique of compound-specific radiocarbon analysis (CSRA) has been proposed as an alternative dating tool (Domack et al. 1999; Eglinton et al. 2000). This technique has been achieved using a preparative capillary gas chromatography (PCGC) system (Eglinton et al. 1996; Uchida et al. 2000) and microscale ^{14}C analysis (Pearson et al. 1998). The application of this technique to the marine sediments with varieties of different sedimentation conditions is in progress and is presenting some technical challenges (Eglinton et al. 1997; Uchida et al. 2000; Pearson et al. 2000a, 2000b). On the other hand, ^{14}C ages of total organic carbon (TOC) in the marine sediments have been considered to be less reliable for dating because of the reworking of organic material, such as humic detritus, and uncertainties of ratios of contribution from marine and terrestrial sources (e.g. Benoit et al. 1979; Jones and Gagnon 1994). There are several reports showing that TOC in marine sediment could be derived from multiple, isotopically heterogeneous sources (e.g. Hedges and Parker 1976; Eglinton et al. 1997; Uchida et al. 2000). The development of the CSRA technique to

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the marine sediments, therefore, will enable us to construct compound-based sediment chronologies such as alkenones and other organic compounds as proxies, which are applicable to most oceanographic regions (Domack et al. 1999; Eglinton et al. 2000). Recently, Pearson et al. (2000) reported that $\Delta^{14}C$ of some sterols derived from phytoplankton production in varved sediments were in good agreement with the $\Delta^{14}C$ of surface water dissolved inorganic carbon (DIC).

In this study the CSRA was conducted on the fatty acids isolated from hemi-pelagic sediments in the western North Pacific. Fatty acids are synthesized as primary products through elongation of acetyl-CoA and are one of the important and major components in living organisms (e.g. Hitchcock and Nichols 1971; Bradshaw and Eglinton 1993). They also play a variety of roles, such as membrane structure (phospholipids) and energy storage compounds (long-chain alkyl esters or wax esters and triacylglycerols) and are ubiquitous in marine sediments (Parker 1962, 1964; Volkman et al. 1980). The compound-specific ^{14}C data of fatty acids coupled to compound specific stable carbon isotope data are also discussed in terms of origins and transport process to marine sediments.

METHODS

The sediment sample was collected by a multiple-core sampler at a water depth of 1536 m on the continental margin in the western North Pacific (40°29′N, 142°59′E; Figure 1) during the MR00-K01 cruise of JAMSTEC R/V *Mirai* (Harada et al. 2000). The major lithology of the sediment core is dark olive-colored diatomaceous mud. The length is 26 cm.

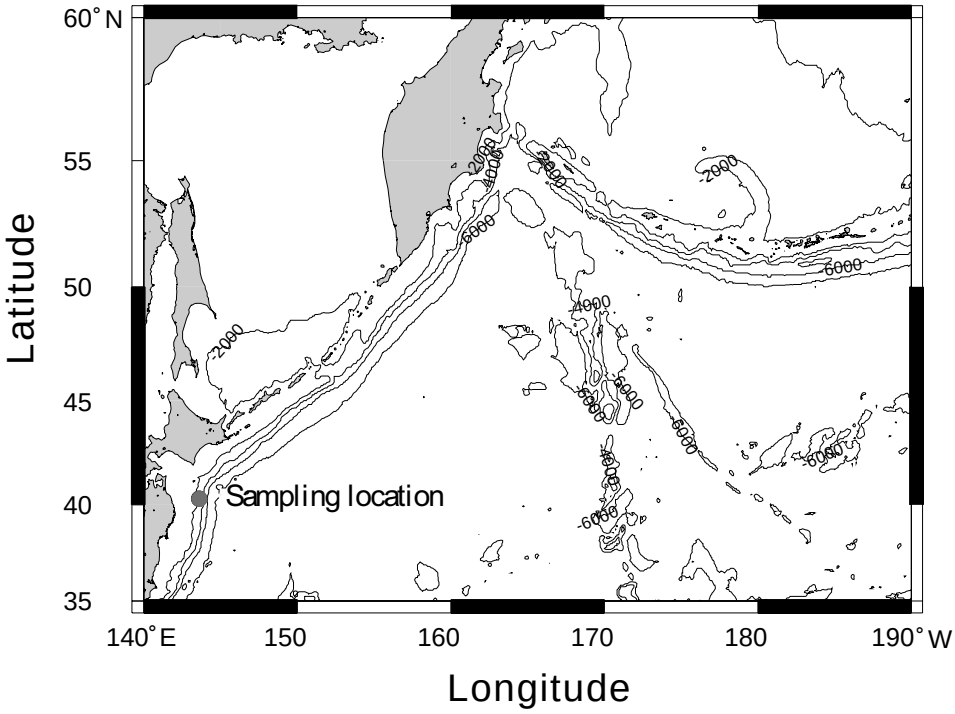

Figure 1 Sampling location of multiple cores. Samples were collected at a water depth of 1536 m on the continental margin in the western North Pacific.

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TOC contents ranged from approximately 2.5% in the top to 1.0% in the bottom layers. The sedimentation rate was calculated as 2.8 cm/ka from slope of ^{14}C ages of bulk-phase organic matter (Uchida et al. unpublished data).

Sediment section from 12 cm (1700 BP calculated based on the above sedimentation rate) to 15 cm (2770 BP), which corresponds to the time range of approximately 1070 years, was used for CSRA. The content of individual fatty acids was also determined. A freeze-dried and homogenized sediment samples (300 g) for CSRA was extracted with a large Soxhlet apparatus with CH_2Cl_2/MeOH (99:1 v/v) (Eglinton et al. 1996). Separation of fractions to several compounds types was conducted according to Kawamura et al. (1995). The total extracts were then saponified with 0.5M KOH/methanol for 2 hr under reflux. Neutral lipids were separated by extraction with CH_2Cl_2/*n*-hexane (10:1), whereas acidic lipids were extracted with CH_2Cl_2 after the remaining solution was acidified to below pH 2. The neutral fraction was further separated into four sub-fractions on a silica gel column. The acidic lipids were derivatized to methyl esters with 14% BF_3/methanol. The methyl esters were separated into three sub-fractions on the silica gel column by stepwise elution. Fatty acid methyl esters (FAMEs) were eluted with *n*-hexane/CH_2Cl_2 (1:2). To determine the compound concentration and yield in a series of lipid extraction, 15-methyl hexadecanoic acid and 19-methyl octadecanoic acid were used as internal and external standards, respectively.

Each fraction of fatty acids preparatively isolated by PCGC was analyzed by GC-FID and GC/MS to determine their amounts and purities. Stable carbon isotope ratios of isolated compounds were determined by isotope ratio monitoring gas chromatography/mass spectrometer (GC/IRMS), consisted of an HP6890GC and a Finnigan MAT252 mass spectrometer. We also determined the carbon isotopic compositions of bulk phase organic matter by combustion in the sealed quartz tube with CuO/Ag, 850 °C for 4 hr. Stable carbon isotope ratios are calculated relative to the NBS-19.

The PCGC systems used here is composed of an HP 6890 GC with FID, a cooled injection system (CIS, Gerstel, Germany), a zero-dead-volume effluent splitter, and a cryogenic preparative collection device (PFC, Gerstel). The PFC device consists of an eight-port zero-dead-volume valve in a heated interface (320 °C) and six 10-μL glass traps and a 100-μL waste glass trap supported in cooled units (−5 °C) with circulation of ethyleneglycol cooled by an electric cooler.

The injection volume was approximately 10 μL in *n*-hexane per injection, which corresponds to the total amounts of 1–5 μg C. The injection port (CIS, Gerstel) was set at temperature programmed from 40 °C at a rate of 12.0 °C/min to 350 °C (hold time: 10 min) at a rate of 12.0 °C/min. Individual compounds were separated on a 60-m megabore (0.53 mm inner diameter) fused silica capillary column coated with a cross-bonded methyl silicone phase (R_{TX-1}, RESTEK; film thickness 0.5 μm). The GC oven temperature was programmed from 50 °C (hold time: 1 min) to 120 °C at a rate of 10 °C/min and to 320 °C at a rate of 5 °C/min (hold time: 10 min). Run time was about 60 min. Helium was used as carrier gas with a flow rate of 5 mL/min.

Prior to compound-specific ^{14}C analysis, we investigated the reproducibility of replicate injection and purity of target compounds on PCGC. The capillary gas chromatogram of FAMEs is also shown in Figure 2. Standard deviations of retention times for the target compounds from the 51 consecutive PCGC runs ranged from 0.02 min for $C_{15:0}$ fatty acid to 0.07 min for $C_{28:0}$ fatty acid. The reliability of isolation of target compounds by PCGC was checked. HRGC chromatograms of target compounds before and after isolation are shown in Figure 2 and it is seen that the isolations were successful.

After PCGC isolation, the trapped components were recovered from the U-tubes by addition of CH_2Cl_2 (1 mL) and transferred to a 2-mL glass vial. An aliquot (50 μL) was used to determine the

purity, yields, and stable carbon isotope analysis. For combustion, trapped compounds were transferred to quartz tubes (10 cm, 6 mm outer diameter) using CH_2Cl_2 and the solvent was removed under a stream of high purity helium (99.999%). Then CuO, Ag, and Cu were added to the quartz tubes and combusted at 850 °C. As a precaution to remove the residual solvent from the quartz tubes, the tubes were evacuated to 10^{-6} Torr while immersed in a dry ice/EtOH. Preparation of graphite targets for 1-mg order samples was conducted according to the batch preparation method (Kitagawa et al. 1993). Graphitization of small amounts of carbon, less than 100 μgC, was made using technique modified in NIES-TERRA on the basis of the microscale ^{14}C analysis developed at NOSAMS (Pearson et al. 1998; Uchida et al. forthcoming). ^{14}C analysis was conducted at the AMS facility of the National Institute for Environmental Studies (NIES-TERRA; Kume et al. 1997; Tanaka et al. 2000).

Correction of $\Delta^{14}C$ ages by subtracting contribution from methyl group derivatized on fatty acids prior to chromatographic separation was made by using a simple isotopic mass balance equation by measuring $\Delta^{14}C_{MeOH}$ (−995‰) of the derivative reagent (BF_3/MeOH). The equation of isotopic mass balance is as follows

$$C_n \cdot \Delta^{14}C_{free} = (C_n+1) \cdot \Delta^{14}C_{ester} \cdot 1 \cdot \Delta^{14}C_{MeOH} \quad (1)$$

$\Delta^{14}C_{free}$ is real age and C_n represents carbon number of derivatived compounds.

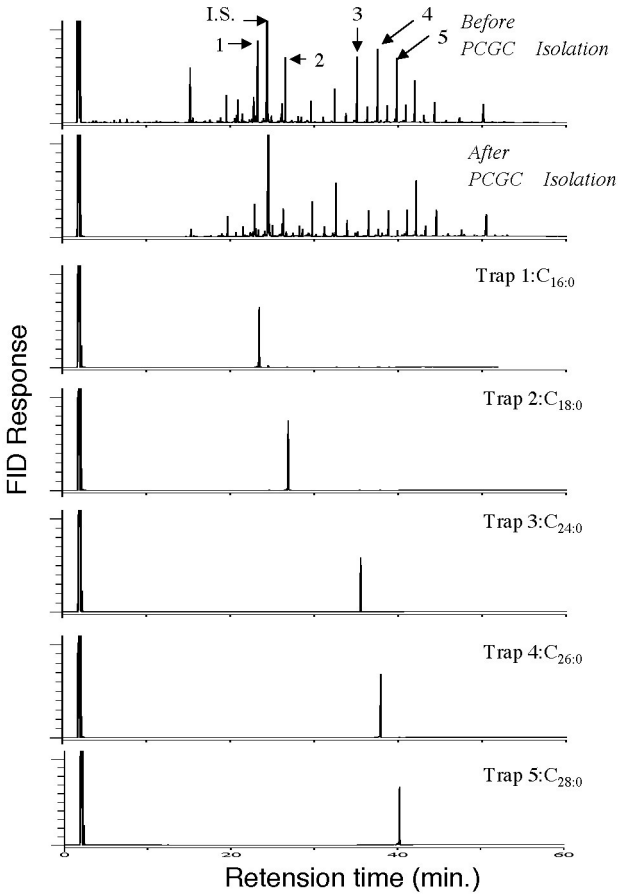

Figure 2 HRGC chromatograms of fatty acid methyl esters (FAMEs). The upper two chromatograms show GC traces before and after PCGC isolations. Numbers of the peaks represent compounds isolated by PCGC.
1: $C_{16:0}$, 2: $C_{18:0}$, 3: $C_{24:0}$, 4: $C_{26:0}$, 5: $C_{28:0}$. Chromatograms of trapped compounds after PCGC separations show successful isolations of target compounds with enough purity.

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RESULTS AND DISCUSSION

Table 1 shows the abundance of *n*-fatty acids in the sediment. Straight-chain saturated C_{12}-C_{34} fatty acids were detected and showed a bimodal pattern with maxima at C_{16} and C_{26} and a predominance of even-carbon number. Monounsaturated fatty acids ($C_{16:1}$, $C_{18:1}$, $C_{18:2}$) and branched-fatty acids ($C_{15:0}$), which originate from plankton and bacteria, respectively, are also detected in the sediments. The distribution pattern was similar to that of samples collected from pelagic sediments in the Pacific Ocean (Kawamura 1995; Ohkouchi et al. 1997). Moreover, similar distribution pattern has also been reported in the marine aerosol samples from the western Pacific Ocean (Kawamura 1995), suggesting that substantial portions of HMW fatty acids are derived from air-to-sea deposition of aerosol particles. The abundance (7 μg/g-dry weight) of C_{23}-C_{34} fatty acids in the sediments was, however, about twice higher than that of pelagic sediments (Ohkouchi et al. 1997). This may suggest that contribution of organic matter derived from continental landmass nearby in our study site, about 100 km off the coast of northern Honshu Island, Japan, is also negligible.

Table 1 Concentrations of individual fatty acids in the western North Pacific sediments (Sediment layers of 12–15 cm).

Carbon nr	Concentrations (μg/g dry weight)	Carbon nr	Concentrations (μg/g dry weight)
12	0.09	24	1.34
13	0.05	25	0.27
14	0.48	26	1.68
15	0.16	27	0.39
16	1.90	28	1.36
17	0.11	29	0.29
18	1.32	30	0.82
19	0.11	31	0.13
20	0.38	32	0.41
21	0.10	33	0.05
22	0.60	34	0.12
23	0.16		

Table 2 shows the $\delta^{13}C$ values of *n*-fatty acids and bulk-phase organic matter in the sediment. The $\delta^{13}C$ values of *n*-fatty acids ranged from –31.8‰ (C_{28}) to –25.1‰ (C_{16}). The $\delta^{13}C$ values of C_{16}–C_{18} fatty acids (–25.1‰ to –27.1‰) were also consistent with those of C_{16} and C_{18} fatty acids in marine algal lipids (–25.0‰ to –20.5‰) and in planktonic foraminifera lipids (–25.0‰ to –20.8‰) (Riebsell et al. 2001; Uhle et al. 1997).

The result of isotopically lighter C_{18} fatty acids compared to C_{16} fatty acids by 2.0‰ was significantly different from the other studies of plants, bacteria, mytilids, and mussels (e.g. Abrajano et al. 1994; Monson and Hayes 1982; Fang et al. 1993; Ballentine et al. 1996), whose $\delta^{13}C$ values of fatty acids showed consistent enrichment in accordance with the increase of carbon number. On the other hand, $\delta^{13}C$ values for C_{16} and C_{18} fatty acids in terrestrial higher plants ranged from –36.0‰ to –35.1‰ (Ballentine et al. 1998). The contribution of terrestrial source fatty acids to the LMW fatty acids fraction of the present sediments is thought to be low. The $\delta^{13}C$ values of C_{16}–C_{18} fatty acids were lighter than that of TOC (–20.9‰), which was consistent with those reported in previous studies on the marine sediments (Naraoka et al. 1995; Ishiwatari et al. 1997). On the other hand, TOC had heavier $\delta^{13}C$ values than that of C_{24}–C_{28} fatty acids (–28.2‰ to –31.8‰) by 6‰ to 11‰. These results are

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Table 2 AMS ^{14}C ages of fatty acids, foraminiferas, and bulk-phase OM in the western North Pacific sediment layers of 12–15 cm.

Target compounds	Inferred source	$\delta^{13}C$[a] (‰)	Yields[b] (μgC)	$\Delta^{14}C$[c] (‰)	^{14}C age[d] (BP)	Error (BP)
C16:0	Marine	–25.1	265	–198	1828	128
C18:0	Marine	–27.1	220	–62	530	60
C24:0	Terrestrial	–30.6	158	–268	2550	70
C26:0	Terrestrial	–28.2	262	–296	2900	210
C28:0	Terrestrial	–31.8	233	–328	3250	370
Benthic Foraminifera[e]		–1.2			2520	40
Bulk-phase OM[e]		–20.9			2260	70

[a]Isotope ratio is relative to the PDB standard material and is corrected by measuring isotope ratio of derivative reagent (MeOH; –29.3‰)
[b]Determined after PCGC isolation
[c]^{14}C concentration corrected for the presence of derivative carbon (MeOH; –995‰)
[d]^{14}C age (BP) reported using the Libby half-life of 5568 yr
[e]Collected in the layer for compound-specific ^{14}C analysis

consistent with the previous study in pelagic sediments in the western North Pacific, Japan (Naraoka et al. 1995). The $\delta^{13}C$ value of C_{24} fatty acid of terrestrial higher plant was reported to be –33.8‰ (Ballentine et al. 1998). Thus, the HMW fatty acids in the present sediments seem to be derived mainly from terrestrial higher plants.

Table 2 also lists conventional ^{14}C ages and $\Delta^{14}C$ values of *n*-fatty acids as methyl ester (FAMEs) in the layers of 12–15 cm. The ^{14}C ages of bulk-phase organic matter and benthic foraminifera were also dated as 2260 BP and 2520 BP, respectively. Interestingly, individual fatty acids showed large variances in ^{14}C ages, from about 530 BP (C_{18}) to 3250 BP (C_{28}), even among the same sedimentary horizon. The results of CSRA are likely separated into two groups, i.e., LMW fatty acids (C_{16}, C_{18}) and HMW fatty acids (C_{24}, C_{26}, C_{28}). The age difference between the two groups may reflect differences of transport processes and their origins as inferred from compound specific stable isotope data.

HMW fatty acids (C_{24}, C_{26}, and C_{28}) derived from plant leaf waxes are dated from 2550 BP (C_{24}) to 3250 BP (C_{28}) with the average of 2890 BP. The ages of HMW fatty acids showed tendencies to have older ages with an increase in carbon numbers. HMW fatty acids might experience a long residence time in reservoirs such as soil, river, and lake sediments before reaching the sediments. The major transport processes of terrigenous organic matter to the marine sediments are likely both fallout of aerosol particles and river runoff from the land (e.g. Gagosian et al. 1982; Kawamura 1995; Hedges et al. 1997). A recent study on the ^{14}C dating of particulate organic carbon (POC) in river discharging to the Ocean showed that the river is a source of predominantly old terrestrial POC to the Ocean (Kao and Liu 1996; Raymond and Bauer 2001). The result of the present study seems to be consistent with these findings.

Bulk-phase organic carbon age in the same layer was dated as 2260 BP and was between ages of the LMW and HMW fatty acids, supporting the idea that it was really mixture of organic matter derived from both marine and terrestrial sources.

On the other hand, the age of C_{18} fatty acids (530 BP) was much younger than that of C_{16} (1830 BP). Although $\delta^{13}C$ values of these fatty acids showed those of marine organisms as mentioned above, the

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cause of age difference between both fatty acids was unknown from small data sets on the present study. Influence of bioturbation might be not negligible because macro benthos such as starfish and molluscan extensively occupied the study site (Fujita and Ohta 1989) and thick mixed layer (about 10 cm) was recognized from the vertical profiles of bulk organic matter ^{14}C ages (Uchida et al. unpublished data). We should require a further study using other compounds derived from phytoplanktonic production such as alkenones molecules and dinosterol in the present sediments.

From compound-specific ^{14}C analysis, we found a significant age difference between LMW- and HMW-fatty acids in the same horizon in the marine sediment. These age differences of individual fatty acids may reflect the different sources (marine and terrestrial) of organic matter to the marine sediments, and CSRA together with compound specific stable isotope data will be a powerful tool to study detailed transport, sedimentation process of organic materials in marine environment.

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