



# Molecular and stable carbon isotopic compositions of saturated fatty acids within one sedimentary profile in the Shenhu, northern South China Sea: Source implications



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## ABSTRACT

This study examined the distributions and stable carbon isotopic compositions of saturated fatty acids (SaFAs) in one 300 cm long sedimentary profile, which was named as Site4B in Shenhu, northern South China Sea. The concentrations of total SaFAs in sediments ranged from 1.80 to 10.16  $\mu\text{g/g}$  ( $\mu\text{g FA/g}$  dry sediment) and showed an even-over-odd predominance in the carbon chain of  $\text{C}_{12}$  to  $\text{C}_{32}$ , mostly with  $n\text{-C}_{16}$  and  $n\text{-C}_{18}$  being the two major components. The short-chain fatty acids (ScFAs;  $n\text{-C}_{12}$  to  $n\text{-C}_{18}$ ) mainly from marine microorganisms had average  $\delta^{13}\text{C}$  values of  $-26.7\text{‰}$  to  $-28.2\text{‰}$ , whereas some terrigenous-sourced long-chain fatty acids (LcFAs;  $n\text{-C}_{21}$  to  $n\text{-C}_{32}$ ) had average  $\delta^{13}\text{C}$  values of  $-29.6\text{‰}$  to  $-34.1\text{‰}$ . The other LcFAs ( $n\text{-C}_{24}$  &  $n\text{-C}_{26}$  ~  $n\text{-C}_{28}$ ; average  $\delta^{13}\text{C}$  values are  $-26.1\text{‰}$  to  $-28.0\text{‰}$ ) as well as  $n\text{-C}_{19}$  and  $n\text{-C}_{20}$  SaFAs (average  $\delta^{13}\text{C}$  values are  $-29.1\text{‰}$  and  $-29.3\text{‰}$ , respectively) showed a mixed signal of carbon isotope compositions.

The relative bioproductivity calculation (marine *vs.* terrigenous) demonstrated that most of organic carbon accumulation throughout the sedimentary profile was contributed by marine organism. The high marine productivity in Shenhu, South China Sea may be related to the hydrocarbon seepage which evidenced by diapiric structures. Interestingly, there is a severe fluctuation of terrigenous inputs around the depth of 97 cm below the seafloor (bsf), probably resulting from the influence of the Dansgaard-Oeschger events and the Younger Dryas event as revealed by  $^{14}\text{C}$  age measurements.

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## 1. Introduction

Fatty acids (FAs) are one of the major components in modern marine sediments. It is well known that the saturated short-chain FAs (ScFAs) were typically to be from vascular plants, algae, bacteria, and other sources (Cranwell, 1982; Kattner et al., 1983; Zegouagh et al., 1996), while the saturated long-chain FAs (LcFAs) are the major components found in leaf waxes, suberin, and cutin in terrestrial higher plants (Kolattukudy, 1980). Therefore, the occurrence of LcFAs in modern marine sediments was usually considered to be an indicative of terrestrial input (Simoneit, 1978; Brassell, 1993). Prahl et al. (1989) used LcFAs to estimate the contributions of organic matter from terrigenous input in one sedimentary rock core. Bourbonniere and Meyers (1996) defined a new parameter  $\text{TAR}_{\text{FA}}$  as  $[(\text{C}_{24} + \text{C}_{26} + \text{C}_{28})/(\text{C}_{12} + \text{C}_{14} + \text{C}_{16})]$  to

estimate the relative contribution of terrigenous and marine inputs to total FA (TFA) concentration. With an introduction of carbon preference index (CPI) concept, Naraoka and Ishiwatari (2000) developed this parameter as  $2\sum(\text{even } \text{C}_{20} \text{ to } \text{C}_{28})/[\sum(\text{odd } \text{C}_{19} \text{ to } \text{C}_{27}) + \sum(\text{odd } \text{C}_{21} \text{ to } \text{C}_{29})]$  to estimate terrigenous input into marine sediments. Although there are numerous publications on the estimation of terrigenous input to marine sediments using concentrations of LcFAs,  $\text{TAR}_{\text{FA}}$ , CPI etc., it is still problematic of the methods used due to the multiple source of LcFAs (e.g. diatoms, bacteria, etc.) (Naraoka and Ishiwatari, 2000; Volkman et al., 1980; Viso et al., 1993).

Compound-specific stable carbon isotopic analysis has proven to be a powerful tool to determine the source of organic compounds in sediments. In terms of the sources of FAs in sediments, there should be a big difference of  $\delta^{13}\text{C}$  values from those of various types of presursors because the  $\delta^{13}\text{C}$  of organic compound is mainly determined by carbon source and isotopic fractionation during the synthesis and metabolic activity of organic matter (Fang et al., 1993; Abrajano et al., 1994; Parker, 1964; Monson and

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Hayes, 1982). The carbon isotope fractionation between FAs and substrates is greater than 20‰ when a substrate is an organic matter with low molecular weight (e.g. methane, methanol, acetic acid), but fractionation is less than 5‰ when microorganisms show a preference for complex mixed substrates. Different metabolic pathways also cause varying carbon fractionation levels. For example, aerobic heterotrophic metabolism (e.g. that displayed by *Shewanella putrefaciens*) can cause a carbon fractionation of approximately −3‰, and anaerobic heterotrophic metabolism can cause a carbon fractionation of −16‰. The carbon isotope fractionation of FAs during autotrophic metabolism with CO<sub>2(g)</sub> as the carbon source (e.g. *planctomycetes*, specifically *anammox* bacteria) can reach −42‰. Under *Thiobacillus* metabolism, carbon isotope fractionation may reach −19‰ (Balkwill et al., 1997; Tunlid and White, 1992; Hill et al., 2000; Frostegard and Baath, 1996; Joergensen and Potthoff, 2005; Sakamoto et al., 2004; Baath and Anderson, 2003). These results provide the base for the use of FAs δ<sup>13</sup>C to identify their biological sources (e.g. terrestrial plants and marine microorganisms), as well as source dynamics with palaeoenvironment change (Duan et al., 1997; Goni et al., 1997; Street-Perrott et al., 1997).

Although Deuser (1971) suggested that the primary productivity of marine organisms is the main contribution to marine organic matter, and terrigenous input accounts for 1% of total input, terrigenous input plays an important role in continental margin, especially estuary and delta (Wang and Chen, 1988). In this study, profiles of saturate fatty acid (SaFA) and their δ<sup>13</sup>C compositions were determined in one 300 cm long sedimentary core collected from continental slope of the Shenhu area, northern South China Sea. The aim of this study is to, (1) examine the source of SaFAs using FAs δ<sup>13</sup>C, further to calculate proportion of terrigenous fraction of LcFAs and its productivity; (2) investigate the possible influence of paleoclimate and regional geological structure evolution on the terrigenous and marine productivity.

## 2. Samples and experimental

### 2.1. Sample collection

Guangzhou Marine Geological Survey, Chinese Ministry of Land and Resources collected sediments with a large gravity core on the

r/v GMGS Ocean IV during a cruise from May to June, 2009. This core is located on the Baiyun Sag of the Pearl River Mouth Basin, northeast of the continental slope of Shenhu (20°08.4374'N, 116°31.0455'E; Fig. 1) and was defined as Site4B. The core is 3 m long collected at a water depth of 970 m. Sediments from 0 to 95 cm depth are unconsolidated, low-viscosity, yellowish-gray and fine-grained sands, and others were all sticky clay silt with layer structure. Water contents in sediments from 0 to 95 cm depth were very high and decreased with increasing depth (Fig. 1; Xie et al., 2012).

The sediments were immediately wrapped with tin foil paper and shipped to lab on ice packs, then were archived at −20 °C for molecular geochemistry analyses. Samples were freeze dried at −50 °C, ground to 80 mesh with an agate mortar and pestle, and then stored at −20 °C until analysis.

### 2.2. FAs extracts, separation and measurements

A method similar to that of Duan et al. (1996) was used to extract FAs. About 10 g to 20 g of powdered samples were extracted with dichloromethane and methanol (9:1) for 72 h in a Soxhlet apparatus. Daturic acid (NuchekPrep, Elysian, MN, USA) was added as an internal standard. The extracted organic matter was saponified with a KOH–methanol solution, followed by separation of neutral lipids, acidification and filtration for FAs recovery. The boron trifluoride–methanol was used to convert the FAs into FA methyl esters (FAMES) prior to gas chromatography–mass spectrometry (GC/MS) and gas chromatography–isotope ratio–mass spectrometry (GC/ir/MS) analyses.

GC–MS analysis of FAMES was performed on a Thermo Scientific DSQ II mass spectrometry coupled to a Thermo Scientific Trace gas chromatography (Zhang et al., 2011). Chromatographic separation was achieved with a 60 m × 0.32 mm ID fused silica capillary column coated with a 0.25 mm film of J & W DB-1MS stationary phase. The oven temperature started at 60 °C, and was then programmed from 60 to 110 °C at 30 °C/min, 110 to 220 °C at a rate of 2 °C/min, and 220 to 315 °C at a rate of 10 °C/min, followed by a 20 min hold. Helium was used as the carrier gas, with a flow rate of 1.1 ml/min. The transfer line temperature was 280 °C, and the ion source temperature was 250 °C. The ion source was operated in the electron impact (EI) mode at 70 eV. Full scan and the selected ion monitoring mode were simultaneously adopted. The

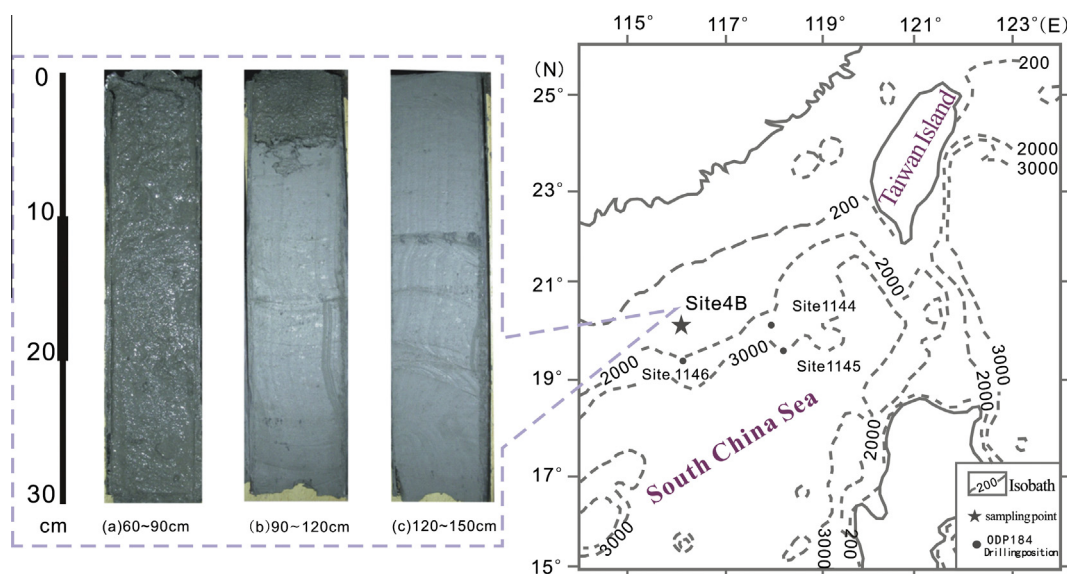


Fig. 1. Geographic location of Site4B in Shenhu and surface characteristics of the profile in the study area.

FAMES were identified on the basis of their mass spectra and relative retention times, as well as by comparison with published data.

The stable carbon isotopic compositions of the individual FAMES were analyzed using an Agilent 6890N gas chromatography interfaced to a GV IsoPrime 100 system. The gas chromatography was equipped with a 30 m × 0.25 mm ID fused silica capillary column coated with a 0.25 μm film of DB-5MS. The column led directly to the combustion interface. The oven temperature program was initiated at 100 °C, then by heating rates of 20 °C/min to 160 °C/min, 1.5 °C/min to 220 °C/min, 10 °C/min to 295 °C/min, and followed by a 20 min hold. Helium was used as the carrier gas with a flow rate of 1.5 ml/min. The samples were injected in splitless mode. The combustion furnace was operated at 800 °C and loaded with CuO and Pt wires as the oxidant and catalyst, respectively. For calibration, a CO<sub>2</sub> reference gas (calibrated against Charcoal Black, a national calibrated standard, with a value of −22.43‰, as indicated in the VPDB standard) was automatically introduced into the isotope ratio mass spectrometer in a series of pulses positioned before and after array peaks of interest. The isotope values were calibrated against the reference gas and are reported in the usual delta notation relative to VPDB.

Reported δ<sup>13</sup>C values represent averages of 2 to 4 analyses, with normally <±0.5‰ standard deviation, corrected for the contribution from the methylating reagent (methanol) by

$$\delta^{13}\text{C}_{\text{FA}} = [(\text{C}_{n+1})\delta^{13}\text{C}_{\text{FAME}} - \delta^{13}\text{C}_{\text{MeOH}}]/\text{C}_n,$$

where δ<sup>13</sup>C<sub>FA</sub> is the δ<sup>13</sup>C of an FA, C<sub>n</sub> is the number of carbons in the FA, δ<sup>13</sup>C<sub>FAME</sub> is the δ<sup>13</sup>C of the methylated FA, and δ<sup>13</sup>C<sub>MeOH</sub> is the δ<sup>13</sup>C of the methanol used for methylation reaction.

The FAs were identified on the basis of their mass spectra and relative retention times, as well as by comparison with published data (Duan et al., 1996; Elvert et al., 2003). FA nomenclature is in the form “A:BωC,” where “A” denotes the total number of carbons, “B” is the number of double bonds, and “C” refers to the distance of the closest site of unsaturation from the aliphatic end of a molecule. The suffixes “c” for *cis* and “t” for *trans* refer to geometric isomers. The prefixes “I,” “a,” “cyc,” “me,” and “br” refer to iso and anteiso methyl branching, cyclopropyl branching, mid-chain methyl branching, and undetermined methyl branch position, respectively.

### 3. Results

#### 3.1. The concentration and distribution of fatty acids in sediments

The concentrations of total SaFA (TSaFAs) in sediments ranged from 1.80 μg/g to 10.16 μg/g, with mostly from 2 μg/g to 6 μg/g (Table 1). There is a significant change on the concentrations of TSaFAs within the core profile. The TSaFAs contents in sediments demonstrated a quick decrease with increasing depth in the first 0–80 cm bsf, then followed by an increasing trend with a severe fluctuation in the section of 80–168 cm bsf. The TSaFAs contents in sediments almost kept stable (4–6 μg/g) in the section of 168–300 cm bsf (Fig. 2).

The carbon chains of the SaFAs in sediments ranged from C<sub>12</sub> to C<sub>32</sub> with an even-over-odd predominance (Fig. 3). The ScFAs are the main group of total SaFAs with enrichments in n-C<sub>16</sub> and n-C<sub>18</sub>, accounting for 24.72–57.65% and 8.27–28.42%, respectively, followed by the n-C<sub>14</sub> and n-C<sub>15</sub> as the second most common components, at most, 5.47% (75 cm bsf) and 3.18% (273 cm bsf), respectively in the profile. Although the relative abundance of LcFAs is normally lower than that of the ScFAs, the n-C<sub>22</sub> and n-C<sub>25</sub> could reach maximum values of 4.82% and 8.39%, respectively in some of the samples (e.g. those collected at 85 and 102 cm bsf).

#### 3.2. Stable carbon isotope of individual saturated fatty acid

Compared to the LcFAs, the δ<sup>13</sup>C values of the ScFAs are relatively enriched in <sup>13</sup>C in studied profile. The δ<sup>13</sup>C values of n-C<sub>14</sub>, n-C<sub>15</sub>, and n-C<sub>16</sub> showed small changes throughout the profile, from −25.1‰ to −27.9‰, −25.2‰ to −27.9‰, and −26.1‰ to −29.1‰, respectively, with a magnitude of less than 3‰ difference across the profile. This is exactly the same as δ<sup>13</sup>C value of n-C<sub>18</sub> in sediments from above 90 cm bsf (less than 3.3‰), although there was a dramatic change below 90 cm bsf, and the δ<sup>13</sup>C value of n-C<sub>18</sub> ranged from −22.4‰ to −29.6‰ (Table 2A).

The δ<sup>13</sup>C values of the LcFAs across the profile demonstrated visibly dynamics, especially from the sediments below 90 cm bsf. The stable carbon isotopic compositions of n-C<sub>24</sub>, n-C<sub>26</sub>, n-C<sub>27</sub> and n-C<sub>28</sub> in sediments above 90 cm bsf were in the range of −25.0‰ to −29.8‰, −25.1‰ to −27.1‰, −21.2‰ to −28.6‰ and −26.9‰ to −29.0‰, respectively, showing relatively positive characteristic. While the rest LcFAs showed more negative carbon isotope values, and had δ<sup>13</sup>C values from −30.9‰ to −33.5‰ (n-C<sub>21</sub>), −30.4‰ to −32.7‰ (n-C<sub>22</sub>), −29.7‰ to −33.3‰ (n-C<sub>23</sub>), −33.5‰ to −35.3‰ (n-C<sub>25</sub>), −28.3‰ to −33.1‰ (n-C<sub>29</sub>), −29.4‰ to −31.4‰ (n-C<sub>30</sub>), and −28.4‰ to −32.0‰ (n-C<sub>32</sub>), respectively (Tables 2A and 2B).

The δ<sup>13</sup>C values of n-C<sub>19</sub> and n-C<sub>20</sub> exhibited visible fluctuations throughout the profile, but similarly as small changes in δ<sup>13</sup>C trends above 90 cm bsf with differences only by 2.6‰ and 2.5‰, respectively (Table 2A).

Interestingly, the variation in δ<sup>13</sup>C of mostly individual SaFA above 90 cm bsf is significantly lower than 3‰. For example, n-C<sub>14</sub> from −25.1‰ to −27.9‰, n-C<sub>26</sub> from −25.1‰ to −27.1‰, n-C<sub>19</sub> from −28.2‰ to −30.8‰ as well as n-C<sub>20</sub> from −28.3‰ to −30.8‰. The results are probably an indicative of stable source supply in this part of sediments.

### 4. Discussion

#### 4.1. Relationship between total organic carbon and total saturated fatty acids

The amount of total organic carbon (TOC) ranged from 0.195% (99 cm bsf) to 0.614% (153 cm bsf), most of the samples in the range of 0.25–0.4% (Fig. 2, Table 1). The relatively low TOC values compared with other marine sediments (Magen et al., 2010; Seki et al., 2006) and estuary sediments (Bianchi et al., 2007; Cowie et al., 2009) may be due to the relatively lower terrigenous organic matter input than estuary sediments (Zhu et al., 2005) and relatively low marine productivity with other marine sediments (Babu et al., 1999; Isla et al., 2002).

Previous studies suggested that the TSaFA compositions in marine sediments strongly depended on locations where the sediments deposited and depositional environment (Harvey, 1994; Wakeham et al. 1997). The concentrations of TSaFAs in sediments investigated in the current work ranged from 1.80 μg/g (80 cm bsf) to 10.16 μg/g (97 cm bsf), which matched well with previous reported data for marine sediments (less than 23.8 μg/g) (Ohkouchi, 1995), and the TSaFAs concentrations reached its maximum level at 97 cm bsf (Fig. 2, Table 1). The TSaFAs/TOCs range from 0.52 mg/g (80 cm bsf) to 3.25 mg/g (97 cm bsf) also with its maximum level at 97 cm bsf (Fig. 2, Table 1).

In terms of the TSaFAs concentrations or TSaFAs/TOC ratio in sediments from the Shenhu area, its accumulation of organic carbon was significantly lower than that in the Equatorial Pacific (5°N to 12°S) or northern Pacific (48°N to 35°N) where productivity are much higher (Ohkouchi, 1995). The strong fluctuations of the TSaFAs through the profile may be attributed to the balance between

**Table 1**  
Parameters of the profile.

Depth (cm bsf)	1	2	3	4	5	6 (%)	7 (%)	8	9	10
30.00–35.00	0.491	5.01	1.02	2.58	0.11	11.34	88.66	7.8	0.57	4.44
35.00–40.00	0.489	5.59	1.14	1.9	0.13	12.58	87.42	7	0.70	4.89
40.00–45.00	0.574	6.12	1.07	2.32	0.18	12.88	87.12	6.8	0.79	5.33
45.00–50.00	0.595	5.67	0.95	1.82	0.14	9.05	90.95	10.1	0.51	5.16
50.00–55.00	0.448	3.63	0.81	2.84	0.28	12.40	87.60	7.1	0.45	3.18
55.00–60.00	0.429	4.58	1.07	3.15	0.15	10.93	89.07	8.1	0.50	4.08
65.00–70.00	0.261	2.66	1.02	0.95	0.17	16.15	83.85	5.2	0.43	2.23
70.00–75.00	0.338	3.15	0.93	1.84	0.12	17.67	82.33	4.7	0.56	2.59
75.00–80.00	0.346	1.8	0.52	2.46	0.05	10.32	89.68	8.7	0.19	1.61
80.00–85.00	0.282	3.41	1.21	1.26	0.23	25.64	74.36	2.9	0.87	2.54
85.00–90.00	0.495	4.39	0.89	1.41	0.26	17.60	82.40	4.7	0.77	3.62
90.00–95.00	0.409	4.98	1.22	2.13	0.23	10.24	89.76	8.8	0.51	4.47
95.00–97.00	0.312	10.16	3.25	8.44	0.06	3.35	96.65	28.8	0.34	9.82
97.00–99.00	0.195	3.69	1.89	5.19	0.17	6.78	93.22	13.7	0.25	3.44
99.00–102.00	0.209	3.33	1.59	1.23	0.21	17.53	82.47	4.7	0.58	2.75
102.00–105.00	0.269	4.67	1.74	7.86	0.1	3.33	96.67	29	0.16	4.51
105.00–108.00	0.287	5.34	1.86	1.91	0.06	5.51	94.49	17.1	0.29	5.05
108.00–111.00	0.277	6.09	2.2	2.38	0.07	5.52	94.48	17.1	0.34	5.75
111.00–114.00	0.277	5.94	2.14	2.4	0.15	6.96	93.04	13.4	0.41	5.53
114.00–117.00	0.203	4.2	2.07	3.46	0.12	8.21	91.79	11.2	0.34	3.86
117.00–120.00	0.274	5.39	1.97	2.95	0.09	7.48	92.52	12.4	0.40	4.99
120.00–123.00	0.356	4.21	1.18	2.78	0.1	7.29	92.71	12.7	0.31	3.90
123.00–126.00	0.415	8.53	2.06	2.31	0.06	4.66	95.34	20.5	0.40	8.13
126.00–129.00	0.33	4.44	1.34	3.44	0.1	6.48	93.52	14.4	0.29	4.15
129.00–132.00	0.299	4.29	1.43	2.82	0.09	7.43	92.57	12.5	0.32	3.97
132.00–135.00	0.347	9.62	2.77	3.81	0.05	3.23	96.77	30	0.31	9.31
135.00–138.00	0.393	7.65	1.95	2.85	0.06	6.06	93.94	15.5	0.46	7.19
138.00–141.00	0.29	3.18	1.1	2.49	0.07	5.65	94.35	16.7	0.18	3.00
141.00–144.00	0.277	6	2.16	2.08	0.07	7.63	92.37	12.1	0.46	5.54
144.00–147.00	0.279	4.71	1.69	2.21	0.06	6.22	93.78	15.1	0.29	4.42
147.00–150.00	0.324	7.41	2.29	2.57	0.12	6.05	93.95	15.5	0.45	6.96
150.00–153.0	0.614	6.68	1.09	2.66	0.07	5.45	94.55	17.4	0.36	6.32
153.00–156.00	0.273	4.32	1.58	1.9	0.07	8.15	91.85	11.3	0.35	3.97
156.00–159.00	0.303	5.18	1.71	2.32	0.14	10.25	89.75	8.8	0.53	4.65
159.00–162.00	0.295	3.19	1.08	2.43	0.1	5.89	94.11	16	0.19	3.00
162.00–165.00	0.288	3.85	1.34	2.52	0.12	9.74	90.26	9.3	0.37	3.48
165.00–168.00	0.371	6.43	1.73	2.08	0.08	5.82	94.18	16.2	0.37	6.06
168.00–171.00	0.342	3.9	1.14	2.52	0.08	7.26	92.74	12.8	0.28	3.62
171.00–174.00	0.274	4.34	1.58	2.11	0.05	5.72	94.28	16.5	0.25	4.09
174.00–177.00	0.273	3.49	1.28	2.11	0.07	6.34	93.66	14.8	0.22	3.27
177.00–180.00	0.285	2.47	0.87	1.65	0.09	7.41	92.59	12.5	0.18	2.29
180.00–183.00	0.306	4.38	1.43	1.61	0.06	5.30	94.70	17.9	0.23	4.15
183.00–186.00	0.24	3.05	1.27	1.7	0.07	6.68	93.32	14	0.20	2.85
186.00–189.00	0.284	3.68	1.29	2.98	0.06	3.65	96.35	26.4	0.13	3.55
189.00–192.00	0.344	3.3	0.96	1.01	0.04	6.01	93.99	15.6	0.20	3.10
192.00–195.00	0.358	3.65	1.02	1.07	0.04	6.95	93.05	13.4	0.25	3.40
198.00–201.00	0.33	3.24	0.98	1.28	0.05	6.41	93.59	14.6	0.21	3.03
201.00–204.00	0.297	2.77	0.93	2.7	0.03	2.56	97.44	38.1	0.07	2.70
204.00–207.00	0.28	3.75	1.34	1.36	0.05	6.90	93.10	13.5	0.26	3.49
207.00–210.00	0.288	4.37	1.52	4.43	0.03	2.34	97.66	41.7	0.10	4.27
210.00–213.00	0.359	4.26	1.19	1.49	0.06	5.61	94.39	16.8	0.24	4.02
213.00–216.00	0.29	2.6	0.9	2.01	0.06	5.22	94.78	18.2	0.14	2.46
216.00–219.00	0.275	3.97	1.44	1.53	0.04	6.19	93.81	15.1	0.25	3.72
219.00–222.00	0.292	4.52	1.55	3.64	0.02	2.14	97.86	45.8	0.10	4.42
222.00–225.00	0.293	1.91	0.65	1.21	0.11	11.35	88.65	7.8	0.22	1.69
225.00–228.00	0.334	4.39	1.31	1.53	0.03	4.07	95.93	23.6	0.18	4.21
228.00–231.00	0.288	2.71	0.94	1.82	0.08	10.35	89.65	8.7	0.28	2.43
231.00–234.00	0.295	2.62	0.89	3.44	0.05	6.28	93.72	14.9	0.16	2.46
234.00–237.00	0.289	3.65	1.26	3.24	0.03	3.15	96.85	30.8	0.11	3.54
237.00–240.00	0.283	3.7	1.31	3.15	0.04	2.50	97.50	39	0.09	3.61
240.00–243.00	0.338	4.4	1.3	2.45	0.03	2.19	97.81	44.8	0.10	4.30
243.00–246.00	0.278	4.45	1.6	2.16	0.02	2.16	97.84	45.2	0.10	4.35
246.00–249.00	0.269	4.28	1.59	1.56	0.03	2.58	97.42	37.7	0.11	4.17
249.00–252.00	0.262	3.6	1.37	2.73	0.02	2.30	97.70	42.6	0.08	3.52
252.00–255.00	0.271	4.19	1.55	3.55	0.01	2.04	97.96	48.1	0.09	4.10
255.00–258.00	0.256	3.58	1.4	3.92	0.03	1.91	98.09	51.4	0.07	3.51
258.00–261.00	0.283	2.89	1.02	3.55	0.02	2.41	97.59	40.5	0.07	2.82
261.00–264.00	0.284	4.26	1.5	2.37	0.02	2.50	97.50	39	0.11	4.15
264.00–267.00	0.278	3.86	1.39	2.78	0.03	3.31	96.69	29.2	0.13	3.73
267.00–270.00	0.292	4.47	1.53	0.92	0.05	8.17	91.83	11.2	0.37	4.10
270.00–273.00	0.283	5.16	1.82	2.75	0.04	3.35	96.65	28.8	0.17	4.99
279.00–282.00	0.283	5.02	1.77	3.68	0.02	2.40	97.60	40.7	0.12	4.90
282.00–285.00	0.306	5.83	1.9	3.61	0.01	1.69	98.31	58.2	0.10	5.73
285.00–288.00	0.309	4.99	1.61	3.19	0.02	2.03	97.97	48.3	0.10	4.89

(continued on next page)

Table 1 (continued)

Depth (cm bsf)	1	2	3	4	5	6 (%)	7 (%)	8	9	10
288.00–291.00	0.294	4.83	1.64	2.72	0.01	2.98	97.02	32.6	0.14	4.69
291.00–294.00	0.293	4.51	1.54	3.17	0.01	2.34	97.66	41.8	0.11	4.40
294.00–297.00	0.28	5.64	2.01	0.62	0.01	7.88	92.12	11.7	0.44	5.20
297.00–300.00	0.288	4.36	1.51	1.59	0.01	2.92	97.08	33.2	0.13	4.23

1: Carbon%; 2: TSaFAs ( $\mu\text{g/g}$ ); 3: TSaFA/TOC ( $\text{mg/g}$ ); 4: CPI; 5:  $\text{TAR}_{\text{FA}}$ ; 6: The relative terrigenous input; 7: The relative marine input; 8: The marine input/terrigenous input; 9: the absolute terrigenous input; 10: the absolute marine input.

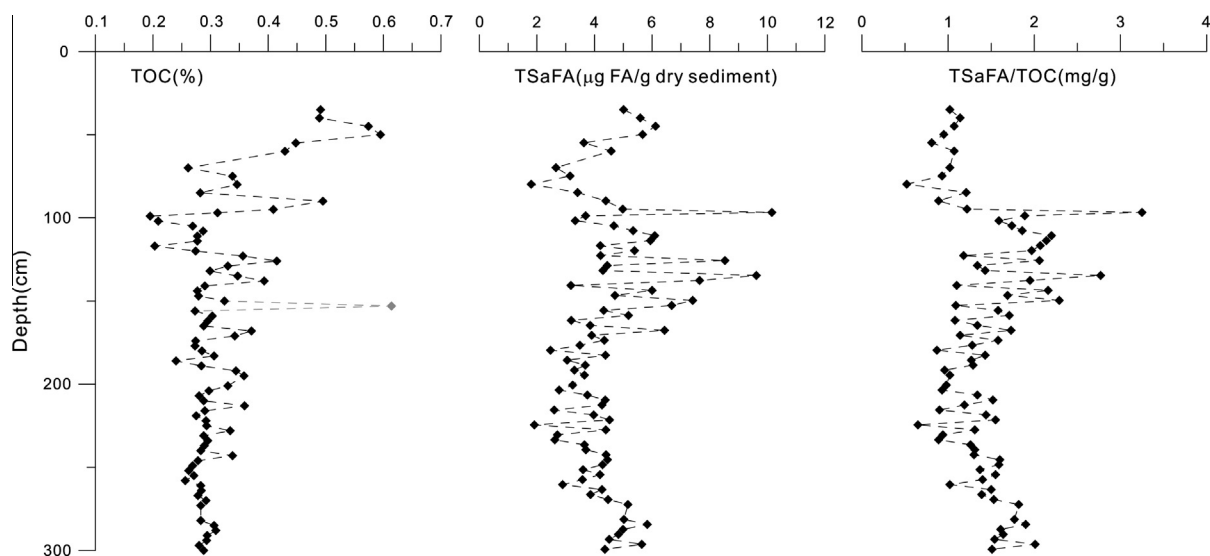


Fig. 2. Distribution of TOC, TSaFAs, and TSaFAs/TOCs in the profile of Site4B.

the rate of organic inputs and the rate of mineral depositions. The sediment of Shenhu likely originated from organic sources that differ from those reported for vent marine sediments (e.g. Yamanaka and Sakata, 2004), which consist of hydrothermal precipitates and minerals with pelagic clay; such composition indicates that the TSaFA compositions in vent marine sediments are several times higher than those in the non-vent sediment of the Shenhu area (Hedrick et al., 1992).

As showed in Fig. 4, the TOCs and TSaFAs displayed good correlation ( $R^2 = 0.7$ ) above 97 cm bsf. Moreover, there was a better correlation ( $R^2 = 0.86$ ) between the TOCs and TSaFAs above 90 cm bsf but poor correlation ( $R^2 = 0.1$ ) below 97 cm bsf. This is in contrast with the traditional understanding, which typically had a weak positive correlation between the TOCs and TSaFAs (Wakeham et al., 1997). Therefore, the good correlation between the TOCs and TSaFAs above 97 cm bsf was probably an indicative of the excellent balance between the rates of marine production and sedimentation throughout the water column.

The relatively high TOC and low TSaFAs above 97 cm bsf can be ascribed to the relatively high terrigenous organic matter input in the sediment of Site4B (Tsugaru et al., 1991) (Fig. 6). Similarly, the relatively low TOC values and high TSaFAs below 97 cm bsf were probably characterized by a relatively high marine production in the sediments and a minor contribution from terrigenous matter (Fig. 6). The relatively low TSaFAs/TOCs suggested that the deposition area is located in a non-eutrophic region (Wakeham et al., 1997). Such deposition can be interpreted as a result of the selective consumption of FAs through biogeochemical degradation in the marine water column. This consumption leads to the significantly low TSaFAs/TOCs in the non-vent sediment relative to eutrophic biomass.

#### 4.2. *n*-weighted average $\delta^{13}\text{C}$ values of saturated fatty acids

The prerequisite for the *n*-weight average  $\delta^{13}\text{C}$  calculation of saturated fatty acids is that the organic source must be kept stable. Xie et al. (2012) found that there was a significant change of the water content of the sedimentary profile studied at 95 cm bsf. The sediments were humid above this depth, and then changed to dry and solid below 95 cm bsf (Fig. 1). On the other hand, Elemental sulfur species analysis showed that sulfate content is almost 100% above 95 cm bsf, suggesting that there was only slight change in respect to the oxic/reducing conditions (GuoDong Zheng, personal communication). An excellent correlation between the TOCs and TSaFAs in sediments above 97 cm (especially above 90 cm) bsf and the similar distribution of stable carbon isotopes of SaFAs among compounds identified in sediments above 90 cm bsf strongly supported its relative uniform for the organic source at this part of the profile (Tables 2A and 2B). Normally, there are multiple complex sources for the TOCs and TSaFAs due to the changeable environments, resulting in a weak correlation between the TOCs and TSaFAs (Wakeham et al., 1997). The maximum difference of carbon isotopes across the lipid compounds in given sample here was usually lower than 3.7‰. These values are within 7‰ of the  $\delta^{13}\text{C}$  differences among lipid compounds in the marine plants and marsh plants examined by Canuel et al. (1997), and within 8‰ of the  $\delta^{13}\text{C}$  differences among alkanes in the higher plant leaf waxes investigated by Rielley et al. (1991).

Geological and geochemical integrations suggested that the sedimentary environment above 90 cm bsf in Site4B is undisturbed, indicating that neither a huge destruction of geological processes nor unexpectedly substantial organic matter input has been found elsewhere. The lack of disturbance means that all of

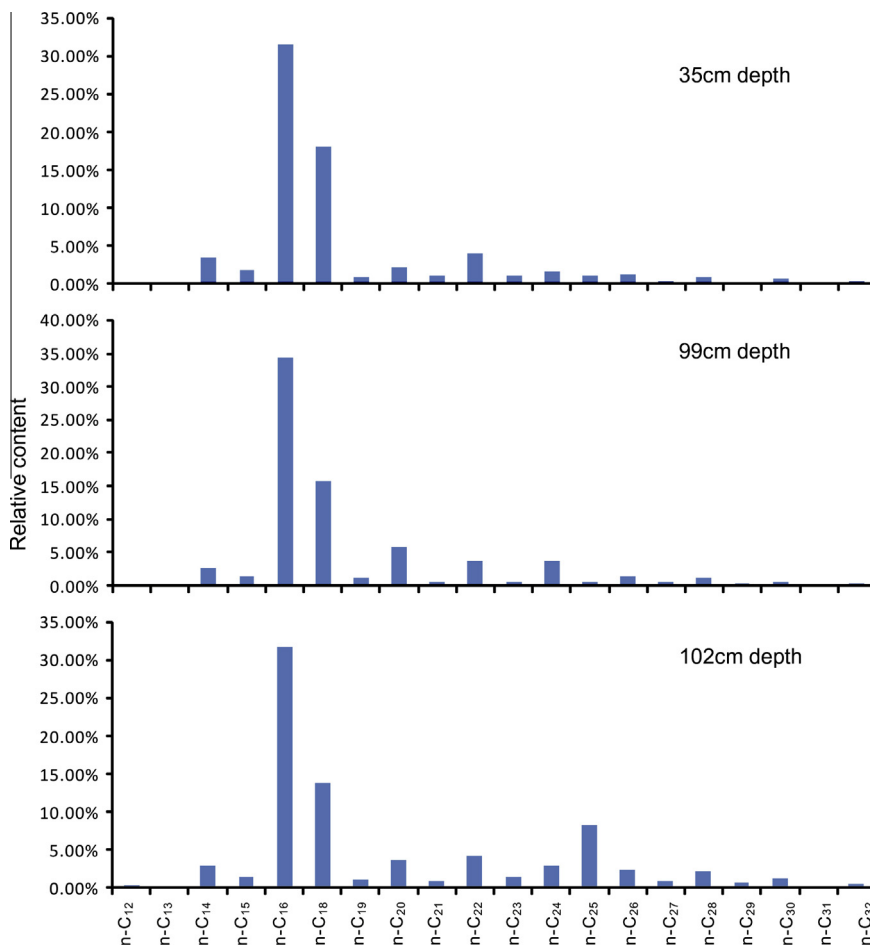


Fig. 3. Distribution of SaFAs at 35, 99, and 102 cm bsf in the profile.

the samples collected from the area above 90 cm bsf can be combined into one huge sample that corresponds to the sediment found at 35–90 cm bsf. Thus, the  $\delta^{13}\text{C}$  of each FA above 90 cm bsf can be replaced by an  $n$ -weighted average  $\delta^{13}\text{C}$  of the total values of each FA above 90 cm bsf. These  $n$ -weighted  $\delta^{13}\text{C}$  of the FAs can be used to determine probable sources from a macroscopic perspective. Thereafter, the equation for  $n$ -weighted average  $\delta^{13}\text{C}$  is formulated as follows:

$$\delta^{13}\text{C} = \frac{\sum(\text{TOC}_m \times \delta^{13}\text{C}_n)}{\sum\text{TOC}_m},$$

where  $\delta^{13}\text{C}$  represents the  $n$ -weighted average of each SaFA,  $n$  represents the carbon number of an SaFA (in this study,  $n = 14\text{--}32$ ),  $m$  denotes the depth of the profile above 90 cm bsf (in this study,  $m = 35$  to 90 cm bsf),  $\text{TOC}_m$  is the TOC of a specific depth above 90 cm bsf,  $\sum\text{TOC}_m$  denotes the total TOC from 35 to 90 cm bsf in the profile, and  $\delta^{13}\text{C}_n$  represents the  $\delta^{13}\text{C}$  values of the SaFAs with carbon numbers 14–32.

As printed in Fig. 5, the  $n$ -weighted average  $\delta^{13}\text{C}$  values of the SaFAs were distributed in two zones above 90 cm bsf. The  $\delta^{13}\text{C}$  values of the ScFAs and some of the LcFAs (n-C<sub>24</sub>, n-C<sub>26</sub>, n-C<sub>27</sub>, and n-C<sub>28</sub>) were relatively enriched in  $^{13}\text{C}$  with a carbon isotopic range from  $-26.1\text{‰}$  to  $-28.2\text{‰}$ , and other LcFAs were depleted in  $^{13}\text{C}$  with  $\delta^{13}\text{C}$  values from  $-29.6\text{‰}$  to  $-34.1\text{‰}$ . Interestingly, the n-C<sub>25</sub> had a more negative stable carbon isotopic composition compared to other LcFAs. The components of n-C<sub>19</sub> and n-C<sub>20</sub> showed similar carbon isotope values, that is,  $-29.1\text{‰}$  and  $-29.3\text{‰}$ , respectively.

### 4.3. Probable sources of saturated fatty acids

#### 4.3.1. Probable sources of short-chain fatty acids

The FAs in modern marine sediments may originate from marine microorganisms, plankton, and terrigenous sources. ScFAs are prevalent in sediments and may be derived from vascular plants, algae, bacteria, and other sources (Cranwell, 1982; Kattner et al., 1983; Zegouagh et al., 1996). The differences in  $\delta^{13}\text{C}$  among the ScFAs in sediments from the studied area may be controlled by: (1) uncertain organic matter input from the Pearl River Estuary, (2) bacterial contributions, and (3) different carbon sources and the special biological synthesis pathway of ScFAs. Gong and Hollander (1997) claimed that n-C<sub>15</sub> ( $-27\text{‰}$ ) counts as conclusive evidence for a bacterial origin, that is, generally, autotrophic bacteria that use chemical energy. In terms of the stable carbon isotopic compositions of ScFAs in this study, with  $n$ -weighted average  $\delta^{13}\text{C}$  values of  $-26.7\text{‰}$  to  $-28.2\text{‰}$ , it should be possible for these compounds with a bacterial origin because of the presence of heavy carbon sources in pore water or the high  $\text{CO}_2$  concentrations in the sedimentary environment (a condition suitable for the development of autotrophic bacteria that use chemical energy), resulting in a relative enrichment of  $^{13}\text{C}$  of ScFAs. The small differences (maximum of less than  $2\text{‰}$ ; Fig. 5) in  $\delta^{13}\text{C}$  among the ScFAs indicated intense cycling of organic matter caused by strong bacterial activity in the sedimentary history of the study area (Gong and Hollander, 1997). Also, these ScFAs were resynthesized by microbial activity in the deposition process rather than derived from



213.00–216.00	0.02	N.D.	0.01	N.D.	N.D.	N.D.	0.05	N.D.	1.71	-29.1	0.43	-28.7	0.01	-28.2	0.05	-30.1	0.02	-31.3	0.09	-31.4
216.00–219.00	N.D.	N.D.	0.01	N.D.	N.D.	N.D.	0.08	-26.4	2.62	-29.1	0.67	-29.4	0.02	-27.9	0.07	-31.4	0.03	-32.5	0.16	-33.2
219.00–222.00	0.02	N.D.	0.01	N.D.	N.D.	N.D.	0.18	-26.2	3.12	-28.2	0.85	-27.1	0.02	-24.8	0.07	-29.5	0.02	-31.3	0.11	-32.1
222.00–225.00	0.01	N.D.	0.01	N.D.	0.08	-27.5	0.04	-27	0.88	-28.4	0.42	-27.2	0.01	-26.4	0.05	-29.5	0.02	-32.1	0.09	-32.2
225.00–228.00	0.05	N.D.	0.02	N.D.	N.D.	N.D.	0.13	-26.5	2.98	-28.8	0.73	-28.2	0.02	N.D.	0.06	-30.4	0.03	N.D.	0.11	-31.9
228.00–231.00	N.D.	N.D.	0	N.D.	0.09	N.D.	0.05	-27.9	1.48	-28.7	0.56	-27.7	0.02	N.D.	0.07	-30	0.03	N.D.	0.13	-32.2
231.00–234.00	N.D.	N.D.	0	N.D.	0.15	-27.3	0.06	-26.7	1.6	-28.3	0.47	-27.4	0.02	N.D.	0.05	-29.8	0.02	N.D.	0.12	-30.8
234.00–237.00	0.01	N.D.	N.D.	N.D.	N.D.	N.D.	0.11	N.D.	2.53	-28.7	0.63	-28.4	0.02	N.D.	0.07	-30.9	0.03	N.D.	0.11	-30.9
237.00–240.00	0.01	N.D.	N.D.	N.D.	N.D.	N.D.	0.14	N.D.	2.39	-28.3	0.78	-27.3	0.02	N.D.	0.08	-30.7	0.02	N.D.	0.11	-30.9
240.00–243.00	0.01	N.D.	0.01	N.D.	N.D.	N.D.	0.19	-25.3	2.75	-27.5	1.04	-25.2	0.03	N.D.	0.08	-28.7	0.02	-29.9	0.09	-30.3
243.00–246.00	0.01	N.D.	0.01	N.D.	N.D.	N.D.	0.17	-25.8	3.23	-28.5	0.74	-29	0.02	-23.3	0.06	-31.2	0.02	N.D.	0.08	-30.5
246.00–249.00	0.02	N.D.	0.01	N.D.	N.D.	N.D.	0.18	-26.3	2.99	-28.2	0.61	-28.8	0.03	-23.5	0.08	-31.6	0.03	-30.9	0.1	-31
249.00–252.00	0.01	N.D.	0.01	N.D.	N.D.	N.D.	0.14	N.D.	2.59	N.D.	0.57	N.D.	0.02	N.D.	0.06	N.D.	0.02	N.D.	0.08	N.D.
252.00–255.00	0.01	N.D.	0.01	N.D.	N.D.	N.D.	0.16	-26.2	3.1	-28.7	0.67	-29.3	0.02	N.D.	0.06	-31.4	0.02	-31.8	0.08	-31.8
255.00–258.00	0.01	N.D.	0.01	N.D.	N.D.	N.D.	0.14	-26.5	2.57	-28.5	0.6	-29.2	0.02	N.D.	0.06	-30.5	0.02	-31.6	0.07	-30.8
258.00–261.00	0.01	N.D.	0.01	N.D.	N.D.	N.D.	0.1	-26.2	2.07	-28.7	0.5	-29.1	0.02	N.D.	0.05	N.D.	0.01	N.D.	0.06	-31.5
261.00–264.00	0.01	N.D.	0.01	N.D.	N.D.	N.D.	0.16	-26.7	3	-28.5	0.72	-28.9	0.04	N.D.	0.08	-30.8	0.03	-29.5	0.1	-29.8
264.00–267.00	0.01	N.D.	0.01	N.D.	N.D.	N.D.	0.13	-26.4	2.69	-28.7	0.67	-29.2	0.03	N.D.	0.07	-31.1	0.03	-30.7	0.09	-30.9
267.00–270.00	0.02	N.D.	0.01	N.D.	N.D.	N.D.	0.13	-27.7	2.86	-28.9	0.72	-28.4	0.02	-28	0.07	-31.4	0.02	N.D.	0.09	-31.5
270.00–273.00	0.05	N.D.	0.03	N.D.	N.D.	N.D.	0.21	-27.3	3.73	-28.6	0.68	-28.6	0.03	-27.5	0.06	-31.4	0.03	-30.8	0.09	-32.1
279.00–282.00	0.04	N.D.	0.02	N.D.	N.D.	N.D.	0.19	-27.7	3.62	-28.5	0.84	-28.4	0.02	N.D.	0.06	-30.2	0.02	-31.4	0.11	-31
282.00–285.00	0.06	N.D.	0.02	N.D.	N.D.	N.D.	0.23	-27.7	4.33	-28.5	0.92	-28.1	0.02	-26.5	0.07	-31.2	0.03	-31.6	0.1	-31.5
285.00–288.00	0.06	N.D.	0.02	N.D.	N.D.	N.D.	0.19	-27	3.64	-28.5	0.74	-28.5	0.02	-27.6	0.06	-30.6	0.03	-31.5	0.1	-31.4
288.00–291.00	0.03	N.D.	0.02	N.D.	N.D.	N.D.	0.16	-26.6	3.68	-28.8	0.6	-29.4	0.03	N.D.	0.05	-31.4	0.03	N.D.	0.14	-31.7
291.00–294.00	0.02	N.D.	0.01	N.D.	N.D.	N.D.	0.17	N.D.	3.44	-28.9	0.62	-29.4	0.02	N.D.	0.05	-31.5	0.02	N.D.	0.08	-31.4
294.00–297.00	0.03	N.D.	0.01	N.D.	N.D.	N.D.	0.14	N.D.	4.02	-29	0.74	-29.6	0.03	-32.3	0.07	-32.1	0.03	-31.6	0.14	-31.8
297.00–300.00	0.03	N.D.	0.01	N.D.	N.D.	N.D.	0.18	-26.5	3.15	-28.7	0.63	-29.1	0.03	N.D.	0.06	-30.7	0.03	N.D.	0.08	-31.1

Note: N.D. means not detected; unit of  $\delta^{13}\text{C}$  is ‰;  $\mu\text{g/g}$  means  $\mu\text{g FA/g}$  dry sediment.

\* n-Weighted average:  $\delta^{13}\text{C} = \frac{\sum(\text{TOC}_m \times \delta^{13}\text{C}_n)}{\sum\text{TOC}_m}$ , n means the number of carbons in the fatty acid, m means the depth of the samples from 35 to 90 cm bsf in the profile.





Table 2B (continued)

Depth (cm)	n-C <sub>23</sub>		n-C <sub>24</sub>		n-C <sub>25</sub>		n-C <sub>26</sub>		n-C <sub>27</sub>		n-C <sub>28</sub>		n-C <sub>29</sub>		n-C <sub>30</sub>		n-C <sub>32</sub>	
	μg/g	δ <sup>13</sup> C	μg/g	δ <sup>13</sup> C	μg/g	δ <sup>13</sup> C	μg/g	δ <sup>13</sup> C	μg/g	δ <sup>13</sup> C	μg/g	δ <sup>13</sup> C	μg/g	δ <sup>13</sup> C	μg/g	δ <sup>13</sup> C	μg/g	δ <sup>13</sup> C
279.00–282.00	0.03	–22	0.03	–30.5	N.D.	N.D.	0.04	N.D.	0	N.D.	N.D.	–27.8	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
282.00–285.00	0.02	–21.4	0.03	–31.2	N.D.	N.D.	0	–23.5	0	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
285.00–288.00	0.03	N.D.	0.04	–30.7	N.D.	–32.4	0.03	N.D.	0.01	N.D.	0.01	–29.7	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
288.00–291.00	0.04	N.D.	0.02	–31.3	0.01	N.D.	0.03	N.D.	0	N.D.	N.D.	–29.4	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
291.00–294.00	0.02	–23.8	0.02	–30.7	0.01	N.D.	0.01	N.D.	0	N.D.	0.01	–29	N.D.	–29.7	0.01	–31.2	N.D.	–32.5
294.00–297.00	0.02	–23.3	0.04	–33.5	0.36	N.D.	0.01	N.D.	0	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	–32.7	N.D.	N.D.
297.00–300.00	N.D.	–23.1	0.03	–31	0.03	N.D.	0.01	–25.1	0.04	N.D.	N.D.	–29.3	0.03	N.D.	0.01	–31.7	N.D.	–32.5

Note: N.D. means not detected; unit of δ<sup>13</sup>C is ‰; μg/g means μg FA/g dry sediment.

\*n-Weighted average: δ<sup>13</sup>C = Σ(TOC<sub>m</sub> × δ<sup>13</sup>C<sub>n</sub>) / ΣTOC<sub>m</sub>, n means the number of carbons in the fatty acid, m means the depth of the samples from 35 to 90 cm bsf in the profile.

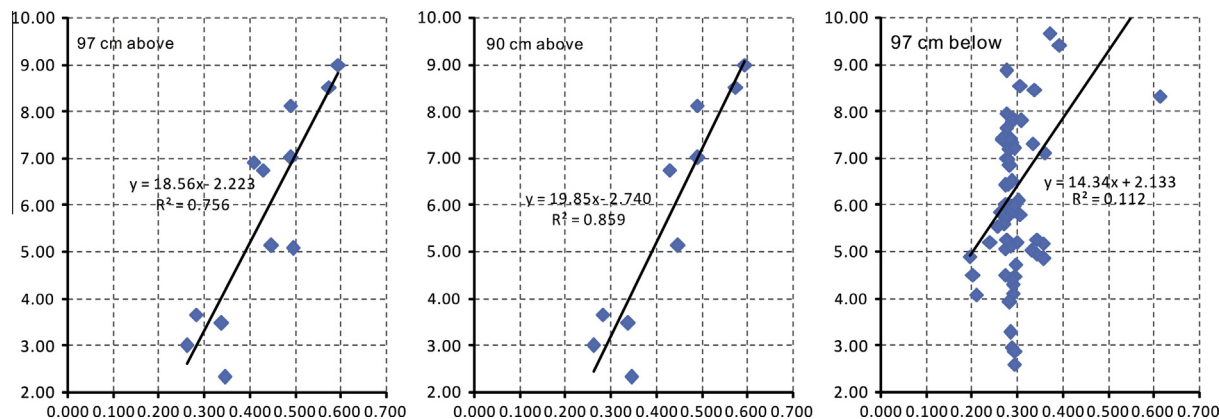


Fig. 4. Correlation between TSaFAs and TOCs in the profile.

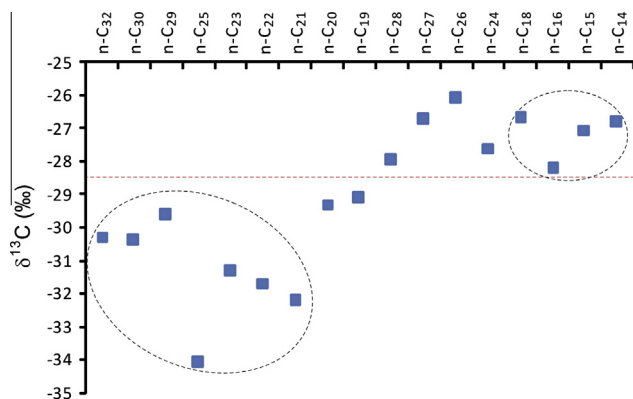


Fig. 5. Distribution of the n-weighted average δ<sup>13</sup>C above 90 cm bsf in the profile.

different carbon sources from the marine column or uncertain organic matter input from the Pearl River Estuary.

#### 4.3.2. Probable sources of long-chain fatty acids

The differences between the n-weighted average δ<sup>13</sup>C values of some LcFAs (except n-C<sub>24</sub>, n-C<sub>26</sub>–n-C<sub>28</sub>; –29.6‰ to –34.1‰) and ScFAs (–26.7‰ to –28.2‰) may be attributed to two factors: (1) the carbon isotope fractionation of the elongation of the FAs during biosynthesis (Fang et al., 1993; Monson and Hayes, 1982) and (2) the presence of different biological sources. The first factor can be disregarded because the differences in δ<sup>13</sup>C caused by changes in the length of carbon chains are less than 18‰, as discussed by Tunlid and White (1992); moreover, the δ<sup>13</sup>C differences that result from the elongation of FAs are confined to a limited number of higher plants (Ratnayake et al., 2005). As for the second factor,

these LcFAs may be derived from higher plants (Kvenvolden, 1967; Matsuda and Koyama, 1977; Duan et al., 1995). As suggested by Volkman et al. (1980), the bacteria in sediments differ from LcFA sources because bacteria seldom synthesize LcFAs. By contrast, Naraoka and Ishiwatari (2000) showed that LcFAs originate not only from terrestrial plants but also from marine diatoms. There is large body of literatures showed that the even-over-odd predominance distribution of LcFAs is partially caused by marine diatoms, and that this distribution is not a completely terrestrial input signal (e.g. Volkman et al., 1980; Viso et al., 1993). Nevertheless, the LcFAs that originate from marine algae are infrequently detected in marine sediments. This contradictory phenomenon may be explained by the fact that limited marine organisms can produce LcFAs characterized by even-over-odd predominance distribution; the absence of the usual phytoplankton markers in the sediments (e.g. 20:5ω3 or 22:6ω3) also excludes the possibility that marine algae are LcFA sources in this study. The long-chain n-alkanes in marine sediments are exclusively derived from terrestrial organic matter and biologically synthesized via LcFAs from terrestrial higher plants (Ratnayake et al., 2005). Therefore, the existence of long-chain n-alkanes in the sediment of Site4B (unpublished data) reflects that the LcFAs are, at least, partially a result of terrestrial input. Furthermore, the CPI values of the Site4B sediment range from 0.62 to 8.44, with most of the values being less than 4 (Table 1). These CPIs are significantly less than those of leaf waxes from terrigenous higher plants (>10) (Simoneit, 1978; Collister et al., 1994). Given this backdrop, Naraoka and Ishiwatari (2000) argued that very low CPI values with positive δ<sup>13</sup>C characteristics (–25‰ to –28‰) may indicate that the aforementioned LcFAs originated from a mixed marine and terrigenous source. On the other hand, the n-weighted average δ<sup>13</sup>C values of n-C<sub>24</sub>, n-C<sub>26</sub>, n-C<sub>27</sub>, and n-C<sub>28</sub> above 90 cm bsf ranged from –26.1‰ to –28.0‰ (Table 2B). These values are highly similar to the δ<sup>13</sup>C of the ScFAs (Fig. 5), suggesting that these four LcFAs

may have the same marine microorganism origins as the ScFAs, although the terrigenous input cannot be completely excluded. Thereafter, n-C<sub>24</sub>, n-C<sub>26</sub>, n-C<sub>27</sub>, and n-C<sub>28</sub>, as well as the others LcFAs, particularly those with  $\delta^{13}\text{C}$  values similar to those of ScFAs, may also reflect mixed terrestrial higher plant contributions and marine input, as suggested by Naraoka and Ishiwatari (2000).

#### 4.4. Estimation of terrigenous contribution from the saturated fatty acids in Site4B

Although the n-C<sub>24</sub>, n-C<sub>26</sub>, n-C<sub>27</sub>, and n-C<sub>28</sub> LcFAs were clearly determined as originating from mixed input using their stable carbon isotopes, the proportion of terrigenous input remains unclear. Although the low TAR<sub>FA</sub> values (maximum, 0.28; Table 1) showed the small terrigenous input in the TFAs in the sediments, it is still unclear how much the contribution of organic matter from terrigenous is. Here the SaFAs in the sediment were assumed to have maintained their inherited carbon isotopic characteristics. In such a way, the terrigenous input versus the in situ marine input of the SaFAs in the sediment can be calculated using following equation (defined by Naraoka and Ishiwatari, 2000).

$$\delta_{\text{mean}} = (1 - f) \times \delta t + f \times \delta m$$

where  $f$  is the proportion of marine input,  $\delta_{\text{mean}}$  denotes the known  $n$ -weighted average  $\delta^{13}\text{C}$  of the n-C<sub>24</sub>, n-C<sub>26</sub>, n-C<sub>27</sub>, and n-C<sub>28</sub> LcFAs above 90 cm bsf,  $\delta t$  is  $\delta^{13}\text{C}$  that is thought to originate from terrigenous input, and  $\delta m$  is  $\delta^{13}\text{C}$  that is believed to originate from marine input.

The differences in  $\delta^{13}\text{C}$  of the ScFAs, particularly those of n-C<sub>14</sub> to n-C<sub>16</sub> ( $n$ -weighted average  $\delta^{13}\text{C}$  ranges from  $-26.8\text{‰}$  to  $-28.2\text{‰}$ ), were less than  $3\text{‰}$ , similar to those reported by Gong and Hollander (1997). In conclusion, the similar  $\delta^{13}\text{C}$  values ( $-27.2 \pm 1\text{‰}$ ) indicated that n-C<sub>14</sub> to n-C<sub>16</sub> originated from in situ microorganisms, and that the average  $-27.2\text{‰}$  can be regarded as the characteristic  $\delta^{13}\text{C}$  of marine input. The average  $\delta^{13}\text{C}$  of the LcFAs from terrestrial higher plants was  $-31.4\text{‰}$ , similar to the  $\delta^{13}\text{C}$  ( $-31.0\text{‰}$  to  $30.7\text{‰}$ ) of the LcFAs in Nansha studied by Duan

et al. (1997), suggesting a source of land plants of the FAs in Site4B. Therefore,  $-31.4\text{‰}$  was considered as the diagnostic  $\delta^{13}\text{C}$  of the terrigenous input to the Shenhu sediments.

Mathematical calculation showed that the proportion of the terrigenous inputs of n-C<sub>24</sub> and n-C<sub>28</sub> account for 1% and 1.2%, respectively above 90 cm bsf in the profile. The more positive  $\delta^{13}\text{C}$  values of n-C<sub>26</sub> and n-C<sub>27</sub> may reflect exclusive marine microorganism input because marine LcFAs originated not only from terrestrial plants but also from marine bacteria (Naraoka and Ishiwatari, 2000). The terrigenous input relative to the marine input was very low in the sediments, amounting to only 9.05% to 25.64%; the highest concentration in the profile was  $0.87 \mu\text{g/g}$  and the lowest was  $0.19 \mu\text{g/g}$  (Fig. 6, Table 1). Below 90 cm bsf, the sedimentary environment had extreme changes, the source of LcFAs was uncertain and varied, and the  $\delta^{13}\text{C}$  values of the LcFAs were frequently altered. Thus, the proportion of land source input cannot be simply calculated by the formula defined by Naraoka and Ishiwatari (2000) with the representative  $\delta^{13}\text{C}$  of FAs.

The terrigenous contribution of four LcFAs (n-C<sub>24</sub>, n-C<sub>26</sub>, n-C<sub>27</sub>, and n-C<sub>28</sub>) above 90 cm bsf was significantly small (maximum of 1.2%). Together with isotope data, the n-C<sub>19</sub>, n-C<sub>20</sub>, n-C<sub>24</sub>, n-C<sub>26</sub>, n-C<sub>27</sub>, and n-C<sub>28</sub> LcFAs in sediments below 97 cm bsf were considered to be marine microorganism origin, whereas the n-C<sub>21</sub> to n-C<sub>23</sub>, n-C<sub>25</sub>, and n-C<sub>29</sub> to n-C<sub>32</sub> LcFAs to be terrigenous origin. Thus, the proportion of terrigenous input below 97 cm bsf was also minimal, ranging from 1.69% to 17.53%. Generally, the terrigenous input within the profile demonstrated a decreasing trend (Fig. 6, Table 1). The ratio of marine input to terrigenous input was 2.9 to 58.18 (Table 1), indicating that the FAs in the sediment were primarily derived from in situ marine organic matter.

The terrigenous input was significantly higher in the sediments above 90 cm bsf than that of sediments below 97 cm bsf, and also there was a distinct distribution pattern of the relative contributions from terrestrial organic matter and in situ marine organic matter in the sediments below 97 cm bsf. The open sea of Shenhu enabled terrigenous input to be transported in two ways: by flow and by wind. Gagosian et al. (1981, 1987) suggested that

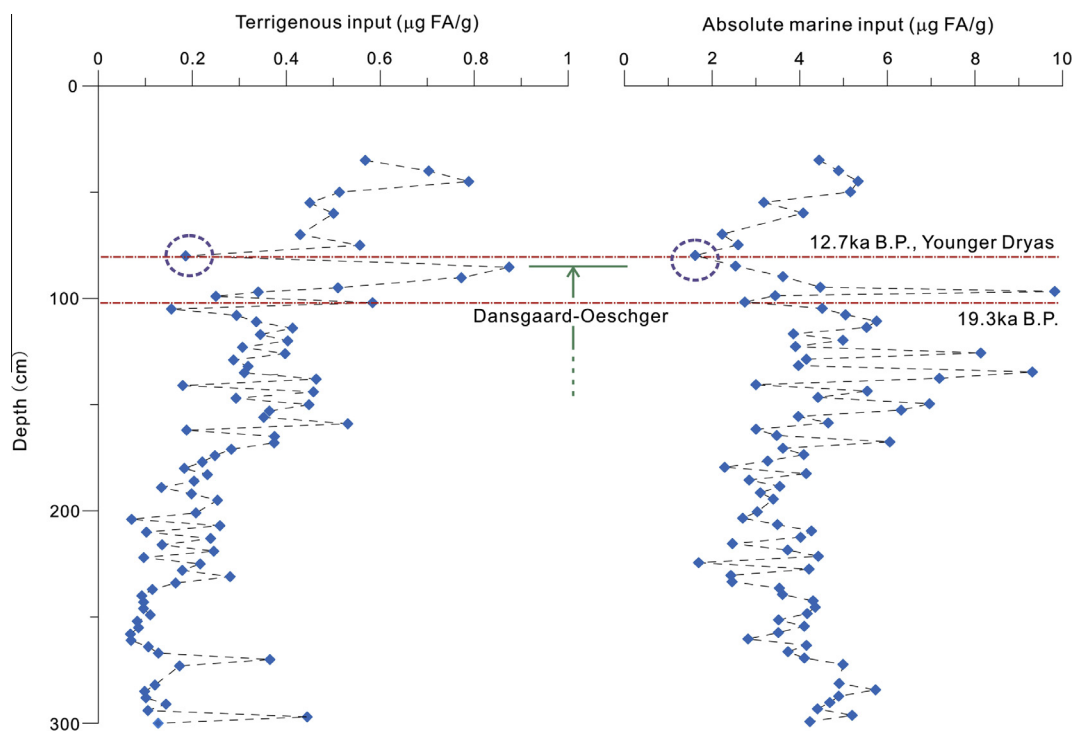


Fig. 6. Distribution of the terrigenous and marine inputs in the profile.

terrigenous organic matter spread by wind accounts for a larger proportion than terrigenous organic matter transported by flow. By contrast, [Simoneit et al. \(1977\)](#) detected LcFAs in the sediments of the Black Sea, where the main terrigenous input is transported only through the water. [Wang et al. \(1999\)](#) studied 10 sediment cores and 40 core-top samples from the South China Sea and found that the terrigenous input in the region was transported through the water. [Hu et al. \(2003\)](#) also inferred that the terrigenous input into core 17,962 from the Nansha Sea area was transported by flow. If the terrigenous input transported to the Shenhu area was dominated by eolian rather than river and ocean currents, the concentration and distribution of the land input above 90 cm would not be completely different from that of below 97 cm. Therefore, the cycled changes in the terrigenous input below 97 cm bsf could be caused primarily by the influence of a monsoon climate system in the South China Sea. In winter, wind blows from the northeast and terrigenous input was transported in the direction of the Shenhu sea area, which exhibits a high terrestrial input feature. In summer, southwest wind blew from the Shenhu sea area to the Pearl River Estuary, and terrestrial input reflected a lower value. Moreover, the frequently and abruptly changing of terrigenous input below 97 cm bsf (102 cm bsf, 19.3 ka B.P., as cited in Qi Lin and JiaSheng Wang, unpublished data) may be related to Dansgaard–Oeschger events (since 35.0 ka B.P.) (Fig. 6)—fluctuations that were confirmed by climatic records ([Heinrich, 1988](#); [Bond et al., 1992](#); [Grimm et al., 1993](#)). Such events were characterized by rapid warming, slow cooling, and extremely rapid cooling in a short cycle during the last glacial period to the interglacial period. Above 90 cm bsf, wind-transported terrigenous input became an insignificant factor, and the increasing terrigenous input may reflect elevated riverine transport on the coast. The terrigenous input with a minimum value characteristic at 80 cm bsf (12.7 ka B.P., as cited in Qi Lin and JiaSheng Wang, unpublished data) (Fig. 6) corresponded to the conditions of the last cold period, that is, the Younger Dryas event. During this period, the marine input also showed a minimum value above 90 cm and when the climate warmed to enable organisms to live, either the terrigenous input transported through the water from the Pearl River Estuary or the in situ marine input showed an obviously increasing trend relative to the Younger Dryas event (Fig. 6).

#### 4.5. Indication of marine productivity in Site4B

The FAs from the marine sources account for 74.36–98.31% of the studied profile (Table 1). This high proportion of marine source input indicated that the FAs in Site4B were primarily derived from marine input and that these FAs, especially the ScFAs, were biosynthesized in the marine environment. Therefore, these ScFAs can be a representative of marine productivity. Some of the LcFAs ( $n\text{-C}_{24}$ ,  $n\text{-C}_{26}$ ,  $n\text{-C}_{27}$ , and  $n\text{-C}_{28}$ ) originated from the microorganisms in the sediments, and their carbon sources depended on the organic matter produced from the original marine source. The concentrations of these LcFAs were also determined by marine productivity. The marine input increased below 97 cm bsf (Fig. 7) and was generally greater than 90% (Table 1), suggesting that terrigenous input in the sediments was rare. Furthermore, below 97 cm bsf, the marine input and marine productivity fluctuations were more likely caused by changes in the marine environment rather than by interferences from the rare terrigenous input transported by wind. Another possibly important factor is the hydrocarbon seepage in this region caused by the diapiric structures, as indicated in either the research on alkanes in the profile (unpublished data) or seismic profile data ([Wu et al., 2009](#)) (Fig. 8). This geological structure also changed the marine ecosystems and seeping hydrocarbon benefits the proliferation of numerous marine microorganisms, thereby increasing marine productivity and causing the deposition of microorganism

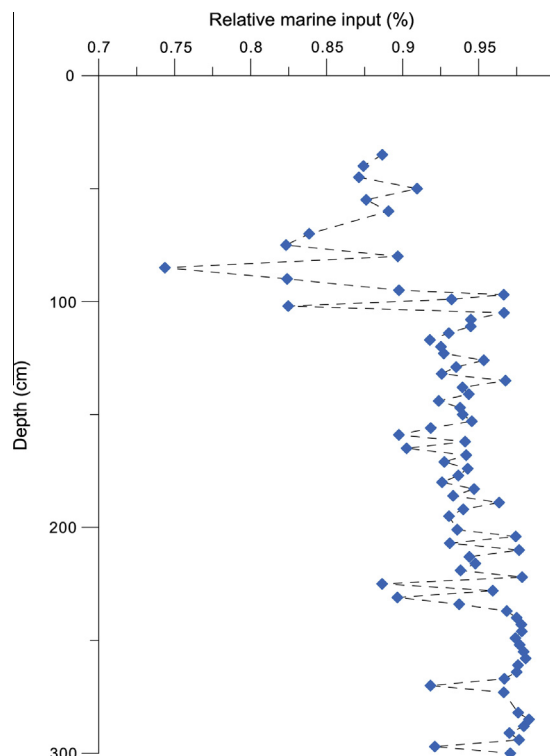


Fig. 7. Distribution of the relative marine inputs in the profile.

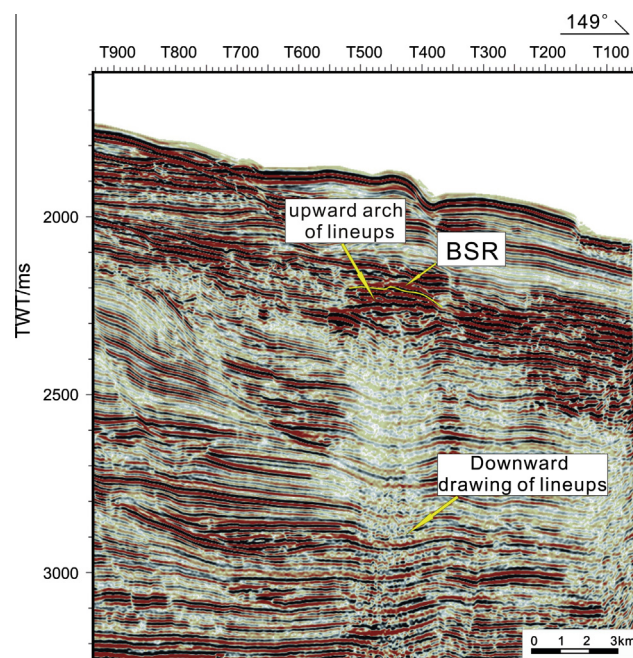


Fig. 8. Seismic profile showing the diapiric structure and its VAMP feature around Site 4B in Pearl River Mouth Basin (from [Wu et al., 2009](#)).

communities into the sediment. This process, in turn, causes massive death and accumulation of marine biomass at 97 cm bsf.

## 5. Conclusion

In this study, the SaFA contents and their  $\delta^{13}\text{C}$  characteristics of the surface sediment of Site4B in the Shenhu, northern South China Sea were investigated. The results suggested that:

- (1) The TSaFAs in Site4B ranged from 1.80  $\mu\text{g/g}$  to 10.16  $\mu\text{g/g}$ , indicating common organic matter accumulation in the sediment of the Shenhu Sea area. The TSaFAs/TOCs were between 0.52 and 3.25  $\text{mg/g}$  in the Site4B profile—values that coincide with other similar latitude studies, and also it showed that deposition occurred in the non-eutrophic zone.
- (2) The ScFAs in the sediments mainly originated from marine microorganisms, whereas most of the LcFAs may have originated from terrigenous plants. Few parts of the LcFAs may be derived from a mixture of marine and terrigenous inputs.
- (3) The terrestrial input demonstrated a boundary around 97 cm bsf in the Site4B sediments. Below 97 cm bsf, organic source input may be related to the marine monsoon climate and transported by wind, whereas above 90 cm bsf, input was transported mainly through predominant current. The minimum values of the marine and terrigenous inputs at 80 cm bsf were related to the Younger Dryas event.
- (4) The marine input was far greater than the terrestrial input in the studied profile, indicating a high marine productivity level; reaching its maximum at 97 cm bsf, an occurrence presumably related to the hydrocarbon seepage caused by diapiric structures.

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