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# Distribution of terrigenous lipids in marine sediments off northeastern Taiwan

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## Abstract

Surface sediments on the continental margin off northeastern Taiwan have been analyzed for terrigenous lipids including *n*-alkanes, *n*-fatty alcohols, and sterols. Marine input to the sediments is particularly low based on the average *n*-C<sub>17</sub>/*n*-C<sub>29</sub> alkane and *n*-C<sub>16</sub>/*n*-C<sub>28</sub> fatty alcohol ratios,  $0.15 \pm 0.13$  and  $0.13 \pm 0.06$ , respectively; this may be due to the fact that marine lipids are more prone to degradation than terrestrial ones. The study area has the highest plant wax *n*-alkane contribution (average carbon preference index  $3.9 \pm 1.2$ ) among the coastal marine areas surrounding Taiwan; lateral particle transport from the southern East China Sea shelf and river runoff from the east Taiwan coast are considered to be the major contributors. The distributions of plant wax *n*-alkane and *n*-alkanol concentrations normalized to total organic carbon (TOC) in the study area generally show maximum values on the upper slope of the southernmost Okinawa Trough, but not for phytosterols. Linear regression of TOC versus plant wax *n*-alkane concentrations show a weak relationship ( $r = 0.64$ ,  $p = 0.001$ ), and an even weaker relationship ( $r = 0.42$ ,  $p = 0.05$ ) between TOC and plant wax *n*-fatty alcohol concentrations is found. This could be attributed to several factors: (1) a complex input (not a point source) of terrigenous organic matter to the study area, (2) TOC also including marine organic matter, (3) temporal variations in river flow due to flooding, and (4) different rates of degradation for TOC and individual biomarkers. However, in spite of those factors, TOC and phytosterol concentrations are positively linearly correlated ( $r = 0.85$ ,  $p < 0.001$ ), implying that the dilution of phytosterols in terrigenous organic carbon with marine organic carbon with or without the phytosterols follows a nearly constant ratio, which is remarkable. In addition, the predominant source of diploptene in the sediments does not appear to be of higher plant origin.

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## 1. Introduction

Hydrocarbon distribution as indicators of source matter identification has been an important and useful tool in organic geochemistry studies. In general, biomarkers for higher plants can be found

in most continental shelf sediments. For instance, *n*-alkanes in marine sediments that show an odd carbon number predominance in the *n*-C<sub>25</sub>—*n*-C<sub>35</sub> region indicate a contribution from land sources (Gearing et al., 1976; Farrington and Tripp, 1977; Keizer et al., 1978; Jeng, 1984). Higher plant waxes tend to contain mainly longer-chain (>C<sub>22</sub>) saturated alcohols (Eglinton and Hamilton, 1963, 1967; Kolattukudy, 1970; Tulloch, 1976).

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Therefore, longer-chain alcohols also have been used commonly as higher-plant markers in sediments (Cranwell, 1984; Volkman et al., 1987). However, in areas of higher productivity, terrigenous biomarkers are less abundant than marine biomarkers. As an example, Smith et al. (1983) reported that a marine sediment taken from the Peru continental shelf showed a larger contribution from marine sources (probably mainly from phytoplankton and bacteria) and little terrigenous influence. Campesterol, stigmasterol and  $\beta$ -sitosterol are the three common phytosterols generally found in epicuticular waxes of vascular plants (Scheuer, 1973; Weete, 1976; Wannigama et al., 1981), and are normally considered to be higher-plant input to sediments (Gagosian et al., 1983; Shaw and Johns, 1985). However,  $C_{29}$  sterols also could originate from marine organisms such as certain microalgae and perhaps cyanobacteria (Volkman, 1986).

Huang and Meinschein (1976) have shown that the % composition of cholesterol increases as the % composition of  $\beta$ -sitosterol decreases in the sample sequence of river inlet sediments—bay sediments—Gulf sediments—bay and Gulf plankton. Calculations based on the sterol ratios gave the percent of the sterols in the Gulf sediments derived from terrigenous sources. Lee et al. (1979) observed a decrease in sterol contents seaward in the sediments collected (farther east of the SEEP sampling locations) in the Atlantic Ocean, and concluded that part of the sterols in the slope sediments originate from terrigenous debris.

For the transport of organic matter from the land to the sea, organic geochemical analyses of benthic sediments from a Northern Queensland (Australia) coastal transect indicated that most terrestrial organic material is confined to near-shore sediments (less than 10 km offshore) (Currie and Johns, 1989). For the transport on the continental margin, Venkatesan et al. (1987) examined the lipid fraction in six sediment cores collected from the Atlantic shelf, slope and the rise areas, and found that the concentrations of organic compound classes decreased from the shelf through the slope to the rise, and estimated that about 50% of the shelf organic matter is exported to the slope. Similar results were also

obtained for sediments from the continental shelf south of New England (Venkatesan et al., 1988). In general, the terrestrial component of the organic matter in unconsolidated sediments decreases progressively offshore, but this is not so on the Scotian margin (Canada) where organic matter with the greatest marine input is found in inshore basins (Pocklington et al., 1991).

Lin et al. (1992) investigated the organic carbon content in the continental margin sediments off northeastern Taiwan and found that lowest organic carbon concentrations were found in the shelf sediments while high concentrations were observed in the upper continental slope sediments. Chen et al. (1992) studying the composition and texture of surface sediments off northeastern Taiwan concluded that in the Okinawa Trough the sediments beneath the path of the main Kuroshio flow are composed of non-biogenic, fined-grain mud.

The circulation patterns of the seas off northeastern Taiwan have been given elsewhere (Hsu et al., 1998). Briefly, the Kuroshio Current flows northward along the east coast of Taiwan. Its subsurface water shoals up off the northeast coast and turns northeastward along the East China Sea continental slope. The inputs of terrigenous organic matter to this area are rather complex—from the northwest and south together with the runoff of the Lan-yang River. In order to examine the influence of high river flow and the Kuroshio Current on the lipid and organic carbon distributions in this area, *n*-alkanes, *n*-fatty alcohols and sterols in the sediments were analyzed to determine terrestrial lipid distribution and contribution to this area and its relation to organic carbon, which can provide important information for understanding carbon budget.

## 2. Materials and methods

Samples were collected on the continental margin off northeastern Taiwan using a box corer on cruises ORI-417 (April 24–May 1, 1995) and ORI-456 (July 6–12, 1996). A total of 23 box core sediments were collected, and sample locations are given in Table 1. Surface sediments (top 3–5 cm

Table 1  
Sampling locations off northeastern Taiwan

Station	Latitude (N)	Longitude (E)	Water depth (m)
<i>ORI-417</i>			
5	24°50.18'	122°49.08'	1522
8	25°00.97'	122°30.84'	1438
9	24°59.44'	122°40.24'	1465
10	25°00.17'	122°50.16'	1518
16	25°11.35'	122°30.88'	894
20	25°01.59'	122°18.03'	1096
21	24°45.32'	122°22.57'	1115
27	25°46.13'	123°07.08'	118
34	24°39.91'	122°30.08'	462
36	24°30.57'	122°20.7'	542
37	24°30.18'	122°12.22'	404
42	25°31.06'	123°03.77'	805
43	25°18.59'	123°00.51'	1360
44	25°15.12'	122°52.10'	1465
45	25°06.15'	122°48.25'	1526
46	24°50.09'	123°00.12'	1665
<i>ORI-456</i>			
27	25°33.88'	122°36.72'	887
33	25°13.94'	122°34.96'	911
34	25°22.28'	122°25.15'	451
35	25°31.86'	122°14.98'	117
37	25°11.93'	122°07.99'	156
39	24°49.93'	122°08.04'	612
K	25°30.97'	122°50.98'	1211

from subcores) were individually analyzed. Sediment samples were freeze-dried. After adding internal standards (*n*-C<sub>24</sub>D<sub>50</sub> and 1-heptadecanol), sediment samples were extracted with a mixture of benzene and methanol (1:1, v/v) for 24 h in a Soxhlet apparatus. The lipid extract was then saponified by reflux for 3 h with 0.5 N KOH solution in methanol. The non-saponifiable lipids were isolated by hexane extraction four times and concentrated using N<sub>2</sub> gas. Aliphatic hydrocarbons and the fatty alcohols and sterols containing fraction were isolated from the neutral lipids by silica gel (deactivated with 5% H<sub>2</sub>O) column chromatography using hexane and a mixture of dichloromethane/methanol (4/1, v/v), respectively. The lipids between the two fractions were eluted with hexane/dichloromethane (2/3, v/v) and discarded. The isolated alcohols and sterols were taken to dryness, redissolved in benzene, and derivatized with N,O-bis(trimethylsilyl)acetamide. For gas chromatography (GC) analysis, an HP

5890 gas chromatograph equipped with a split/splitless injector and a flame ionization detector (FID) was used. An SGE (Australia) OCI-5 cool on-column injector also was fitted in the gas chromatograph for quantitation. Separation of aliphatic hydrocarbons was achieved on an SPB-1 capillary column (30 m × 0.25 mm i.d.), and that of alkanols/sterols (as TMS ethers) was accomplished by another SPB-1 column (25 m × 0.25 mm i.d.). Oven-temperature programming was 45–90°C at 15°C/min and 90–270°C at 3°C/min for analyzing aliphatic hydrocarbons, and 45–90°C at 15°C/min and 90–250°C at 3°C/min, 30 min at 250°C for analyzing alkanols/sterols. Identification was made with co-injection of authentic standards and gas chromatography–mass spectrometry (GC–MS). The GC–MS analyses were performed with an HP 6890 GC (HP-1MS crosslinked methyl siloxane column, 30 m × 0.25 mm i.d.) interfaced directly to an HP 5973 quadrupole mass selective detector (electron impact, electron energy 70 eV, scanned from 50 to 550 D). Based on replicate analyses by GC–FID, the analytical precision of lipids was calculated to be 2–8%.

Organic carbon was analyzed using infrared determination of CO<sub>2</sub> evolved from high temperature (1400°C) combustion with a LECO SC-444 carbon/sulfur analyzer. Approximately 0.25 g of dry sediment was pre-acidified with ~2 ml of 6 N HCl to remove carbonate carbon. After drying on a hot-plate for 8 h at ~60°C, the sample was combusted in a LECO carbon analyzer to measure its organic carbon content. The LECO carbon standard (502-062) was used for calibration. The average analytical precision was 0.88% (1 s.d.).

### 3. Results and discussion

#### 3.1. Terrestrial lipid contribution and distribution

Gas chromatograms of aliphatic hydrocarbons in the study area show a minor unresolved complex mixture (UCM) (Fig. 1), suggesting some input from petroleum or biodegradation sources (Brassell and Eglinton, 1980) or from eroded hydrocarbons. In addition, it is noted that the lower molecular weight region (<C<sub>23</sub>) has a CPI

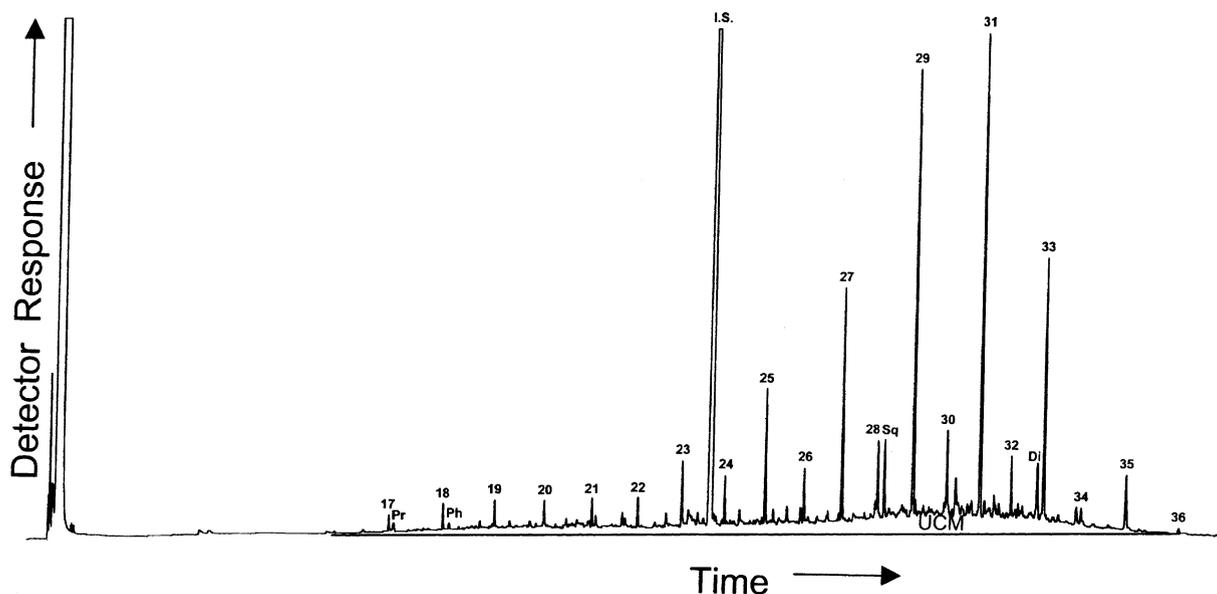


Fig. 1. Gas chromatogram of aliphatic hydrocarbons from station 456-37. Numbers above the peaks refer to carbon number of *n*-alkanes; Pr = pristane; Ph = phytane; Sq = squalene; Di = diploptene; UCM = unresolved complex mixture.

of 0.99, possibly indicating a contribution from fossil fuels. As shown in Fig. 2, mass chromatograms of characteristic ions  $m/e$  85, 191, and 217 illustrate the distribution of alkanes, triterpanes and steranes, respectively. For pentacyclic triterpanes ( $m/e$  191), besides diploptene, two compounds cannot be unequivocally identified from mass spectra that are similar to those of lupane and oleanane. Sterane ( $m/e$  217) characteristic ions are of very low intensity, and no steranes were found. In addition, generally there were no low-molecular-weight ( $\leq 3$  aromatic rings) polycyclic aromatic hydrocarbons (PAHs) along with their alkylated homologues found in the sediments, nor were high-molecular-weight ( $\geq 4$  aromatic rings) parental (non-alkylated) PAH compounds found. This result indicates probably lack of fossil fuels and their combustion products in the sediments. However, there was one predominant PAH—perylene found (Fig. 3; Table 2), which is considered to be derived from diagenesis (Venkatesan, 1988). For instance, a down-core increase of perylene concentrations (191, 311, 388, 514, 629, 542 ng/g, each section being 4 cm) was observed in a box core taken from station 417-20, reflecting transformation from unknown natural precursors

(Louda and Baker, 1984). From the above results, it can be concluded that the study area is considered to have minimal contamination by hydrocarbons related to petroleum.

The sediments have predominantly *n*-C<sub>25</sub>, *n*-C<sub>27</sub>, *n*-C<sub>29</sub>, *n*-C<sub>31</sub> and *n*-C<sub>33</sub> paraffins, typical of terrestrial plant waxes (Eglinton and Hamilton, 1967), and a unimodal distribution having a maximum at C<sub>31</sub> (the abundance order  $n\text{-C}_{25} < n\text{-C}_{27} < n\text{-C}_{29} < n\text{-C}_{31}$ ). In comparison, the *n*-alkane profiles of the Changjiang estuary sediments exhibit a bimodal distribution with the first maximum at *n*-C<sub>31</sub> and the second maximum at *n*-C<sub>17</sub> or other lower molecular weight ( $< n\text{-C}_{23}$ ) *n*-alkanes (Bigot et al., 1990). The common feature of these two areas is the *n*-C<sub>31</sub> maximum. The major *n*-alkanes found in marine phytoplankton (Blumer et al., 1971), benthic algae (Youngblood et al., 1971), and pelagic *Sargassum* (Burns and Teal, 1973) are *n*-C<sub>15</sub> and *n*-C<sub>17</sub>, which are present in only small amounts in the sedimentary *n*-alkanes of this area. The ratio of  $n\text{-C}_{17}/n\text{-C}_{29}$  was employed by Venkatesan et al. (1987) as a parameter to indicate marine vs. terrigenous input of *n*-alkanes; this ratio in the 23 samples examined has a wide range of 0.00–0.31 with a mean of

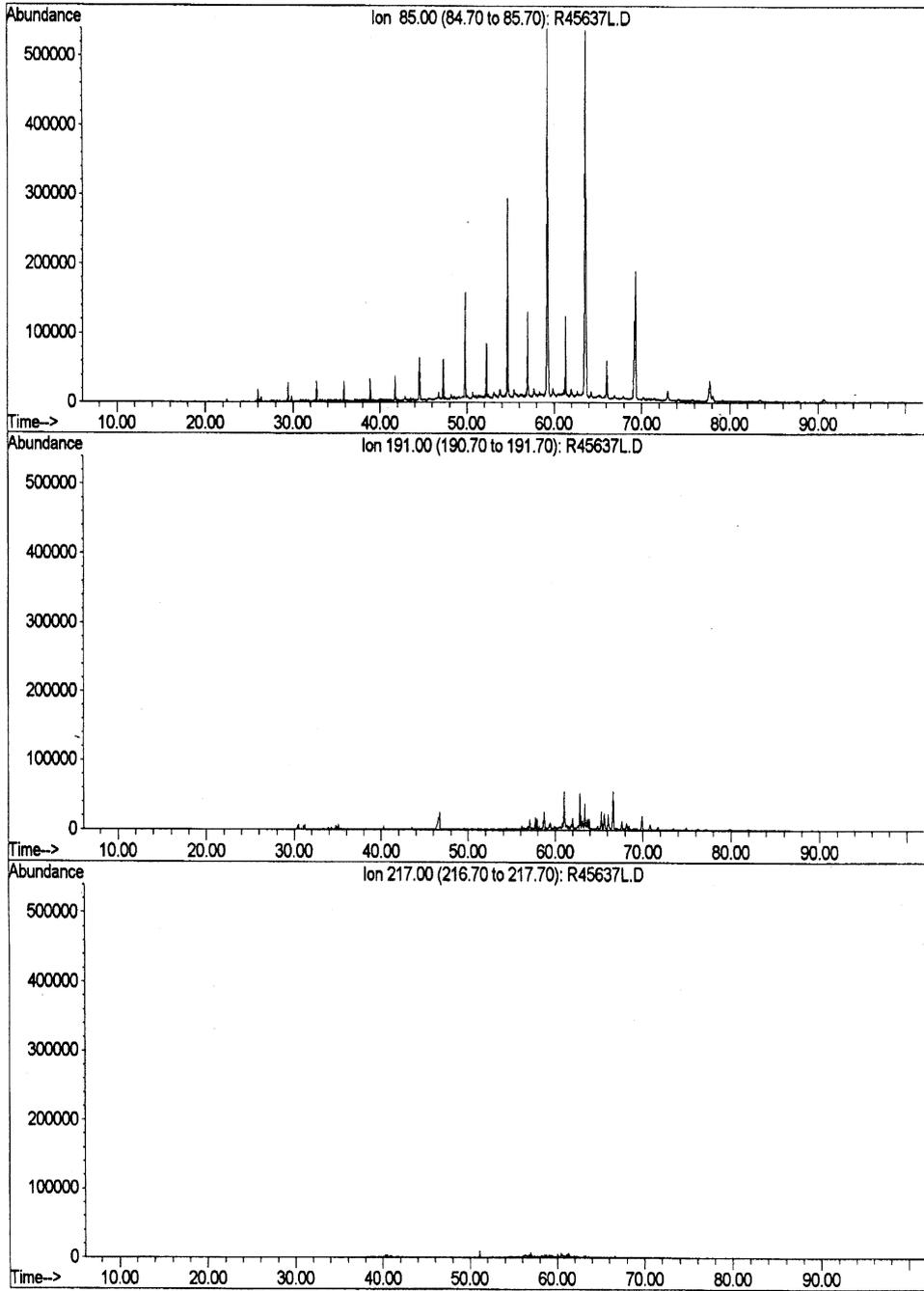


Fig. 2. Specific ion plots for  $m/e$  85 (alkanes), 191 (triterpanes), and 217 (steranes) from the GC-MS analysis of the aliphatic fraction from station 456-37.

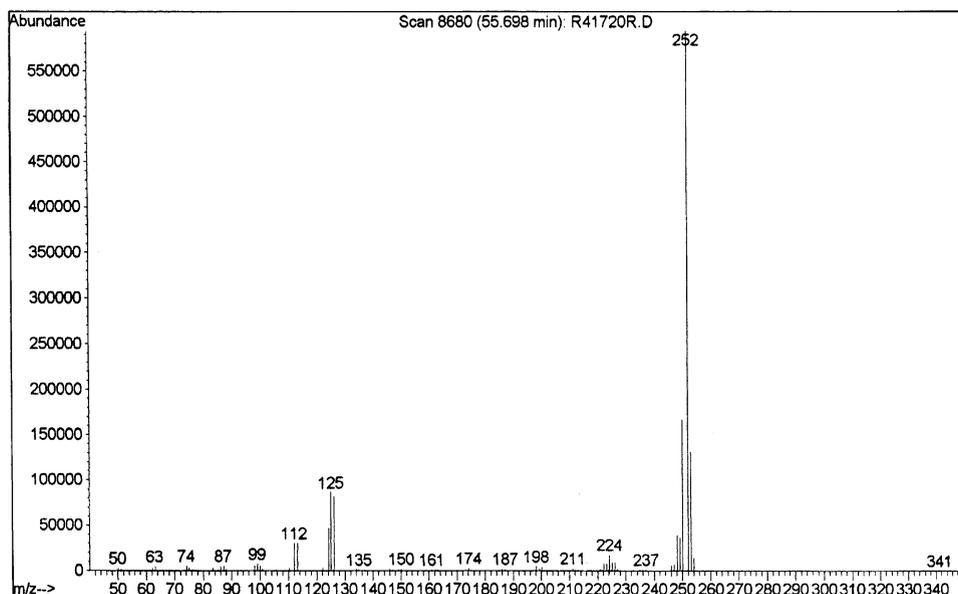


Fig. 3. Electron impact mass spectrum of perylene from station 417-20.

$0.15 \pm 0.13$  (Table 2). Two surficial sediments with a low level of contamination from the Changjiang estuary and adjacent East China Sea have  $n\text{-C}_{17}/n\text{-C}_{29}$  ratios of 0.20 and 0.60 (calculated from histograms given by Bigot et al., 1990). The ratio in the suspended particles from surface waters of the Changjiang has an average of  $0.33 \pm 0.18$  ( $n=13$ , calculated from  $n$ -alkane concentrations given by Sicre et al., 1993). Surface sediments (0–10 cm) on the Amazon shelf have a mean  $n\text{-C}_{17}/n\text{-C}_{29}$  ratio of 0.38 (six values being 0.23, 0.33, 0.37, 0.39, 0.41 and 0.57 calculated from histograms given by Elias and Cardoso, 1996). Two sediment core sections (20–40 and 40–60 cm) from the Bering Sea have the  $n\text{-C}_{17}/n\text{-C}_{29}$  ratio of 0.26 and 0.59 (calculated from the peak heights of GC traces given by Kennicutt et al., 1991). One surface sediment from the Bellingshausen Sea, Antarctica has a  $n\text{-C}_{17}/n\text{-C}_{29}$  ratio of 0.80 (calculated from histogram given by Cripps, 1995). The ratio for sediments from southeast Florida is 6.68 (reef track), 10.26 (Hawk Channel), 2.54 (nearshore Atlantic), 0.46 (bay), and 4.15 (ABC channel) (calculated from data given by Snedaker et al., 1995). The top 1 cm sediment from the middle Black Sea has a  $n\text{-C}_{17}/n\text{-C}_{29}$  ratio of 0.09 (calculated from data given by Wakeham, 1996).

For our samples, the ratio is apparently at the low end; this can probably be attributed to (1) the predominance of the terrestrial  $n$ -alkane contribution to the coastal marine sediments, and (2) preferential degradation of marine-derived  $n$ -alkanes relative to terrigenous  $n$ -alkanes (Prah et al., 1980; Meyers et al., 1984).

To compare with other marine sediments surrounding Taiwan, another parameter is used. Land plant  $n$ -alkane contribution to marine sediments is often expressed as carbon preference index, (CPI). Compared with the CPIs of northern and southern Taiwan Strait surface sediments and eastern Taiwan shelf (ca. 1–20 km in width) sediments, 2.0, 1.7 and 2.3, respectively (Jeng, 1978, 1979, 1984), the average CPI value of the study area is relatively high,  $3.88 \pm 1.16$  (Table 2) indicating a strong terrestrial  $n$ -alkane influence. A similar index for higher plant wax contribution to marine sediments is odd–even predominance (OEP) proposed by Scalan and Smith (1970) as a means of reducing the mathematical limitations of the CPI. The Mackenzie River is regarded as providing a homogeneous, well-defined terrigenous input of  $n$ -alkanes to the Mackenzie shelf (Yunker et al., 1991). Five OEP  $\text{C}_{29}$  ratios for Mackenzie shelf (Beaufort Sea) sediments are 2.20,

Table 2  
Hydrocarbon data

Sample	TOC (10 <sup>-2</sup> g/g)	<i>n</i> -C <sub>17</sub> (ng/g)	<i>n</i> -C <sub>29</sub> (ng/g)	<i>n</i> -C <sub>17</sub> / <i>n</i> -C <sub>29</sub>	∑ <i>n</i> -C <sub>25–33</sub> (ng/g)	(∑ <i>n</i> -C <sub>25–33</sub> / TOC) × 10 <sup>-7</sup>	CPI <sup>a</sup> & OEP <sup>b</sup>	Diploptene (ng/g)	Perylene (ng/g)
<i>ORI-417</i>									
5(0–5 cm)	1.01	95	467	0.20	1820	1800	2.61/2.62	1140	n.d. <sup>c</sup>
8(0–3 cm)	1.05	108	509	0.21	1970	1880	2.54/2.40	2020	n.d.
9(0–3 cm)	1.09	113	439	0.26	1760	1610	2.49/2.27	518	151
10(0–3 cm)	1.00	10	439	0.02	1690	1690	2.60/2.50	530	n.d.
16(0–5 cm)	0.82	n.d.	394	—	1640	2000	3.47/3.35	169	157
20(0–4 cm)	0.63	32	691	0.05	2450	3880	6.06/5.27	42	191
21(0–4 cm)	0.77	222	418	0.53	1630	2110	2.81/2.76	433	118
27	0.16	1	57	0.02	208	1300	5.49/5.55	10	31
34(0–4 cm)	0.69	32	731	0.04	2620	3800	5.24/5.21	29	740
36(0–4 cm)	0.42	51	502	0.01	1870	4450	5.76/5.69	37	124
37(0–4 cm)	0.29	17	91	0.19	358	1230	3.17/3.46	9	n.d.
42(0–4 cm)	0.41	51	154	0.33	581	1420	3.82/4.24	32	24
43(0–4 cm)	0.73	8	288	0.03	1090	1500	3.69/3.50	182	86
44(0–4 cm)	0.84	84	378	0.24	1460	1940	3.34/3.27	303	102
45	0.87	58	380	0.15	1470	1690	3.23/3.16	529	155
46(0–4 cm)	0.79	59	335	0.18	1300	1650	3.83/3.86	533	204
<i>ORI-456</i>									
27	0.74	27	784	0.03	2150	1910	5.17/4.69	92	n.d.
33(0–4 cm)	0.38	n.d.	177	—	552	1450	4.17/3.90	70	n.d.
34(0–4 cm)	0.74	n.d.	827	—	3000	4050	5.91/5.54	74	n.d.
35	0.14	3	43	0.07	135	964	4.13/4.24	15	n.d.
37	0.36	11	341	0.03	998	2770	3.19/3.03	37	n.d.
39(0–4 cm)	0.71	59	477	0.12	1140	1610	3.19/3.14	176	n.d.
K(0–4 cm)	0.60	62	252	0.25	763	1270	3.22/2.95	205	n.d.
Average				0.15			3.88/3.77		
s.d.(1σ)				0.13			1.16/1.09		

$$^a \text{Carbon preference index} = \frac{1}{2} \left( \frac{nC_{25} + nC_{27} + nC_{29} + nC_{31} + nC_{33}}{nC_{24} + nC_{26} + nC_{28} + nC_{30} + nC_{32}} + \frac{nC_{25} + nC_{27} + nC_{29} + nC_{31} + nC_{33}}{nC_{26} + nC_{28} + nC_{30} + nC_{32} + nC_{34}} \right)$$

$$^b \text{odd even predominance } n\text{-}C_{29} \text{ ratio} = (n\text{-}C_{27} + 6n\text{-}C_{29} + n\text{-}C_{31}) / (4n\text{-}C_{28} + 4n\text{-}C_{30})$$

<sup>c</sup> n.d. = not determined.

2.18, 2.11, 3.14 and 4.28 (Yunker et al., 1993), with a mean of 2.78. The average of our OEP C<sub>29</sub> ratios is 3.77 ± 1.09 (Table 2). The difference could be due to stronger terrigenous *n*-alkane influence in the study area or difference in *n*-alkane maximum—C<sub>31</sub> for the study area and C<sub>29</sub> for the Mackenzie shelf.

Several factors can account for this interesting result of high land plant *n*-alkane inputs to the study area. The most important one is that shorter-chain alkanes, such as *n*-C<sub>17</sub>, are more prone to degradation than longer-chain *n*-alkanes.

Second, a part of the study area is a region with comparatively high sedimentation rates around Taiwan (Hung and Chung, 1994; Chung and Chang, 1995); this tends to create a reducing environment, especially on the upper slope (e.g., mottled and filamented authigenic pyrite has been found by Chen et al., 1995), which is favorable for preserving organic matter. Third, suspended particulates in runoff of other rivers on the eastern coast carried by the Kuroshio Current presumably tend to be deposited in the southern part of the study area.

Prior to discussing the distribution of lipid data, it is advantageous to know the nearby river flow and predominant ocean currents. The inputs of terrigenous organic matter to the study area are rather complex—from the northwest and south together with the runoff of the Lan-yang River (Fig. 4). A small alongshore flow just off north Taiwan may carry materials from the southern East China Sea and Taiwan Strait (west of Taiwan) to this area (arrow 1). Contribution of materials from the Lan-yang River runoff is apparent (arrow 2). Another source is other river runoffs from eastern Taiwan carried by the Kuroshio Current to the study area (arrow 3). Their respective contributions of terrigenous lipids

to this area will be compared in the following discussion.

In coastal regions receiving predominantly detrital input of organic material essentially from a point source, plant wax *n*-alkane concentrations normalized to TOC show a general decrease with distance offshore, e.g., Washington State, USA (Prahl and Carpenter, 1984). The trend can be attributed to dilution of the biomarkers in terrestrial organic carbon, with an increasing proportion of marine organic carbon containing none of the terrestrial *n*-alkanes. The spatial distribution of land plant derived *n*-alkanes (sum of *n*-C<sub>25</sub>, *n*-C<sub>27</sub>, *n*-C<sub>29</sub>, *n*-C<sub>31</sub> and *n*-C<sub>33</sub>) normalized to TOC (to eliminate the grain size effect) is given

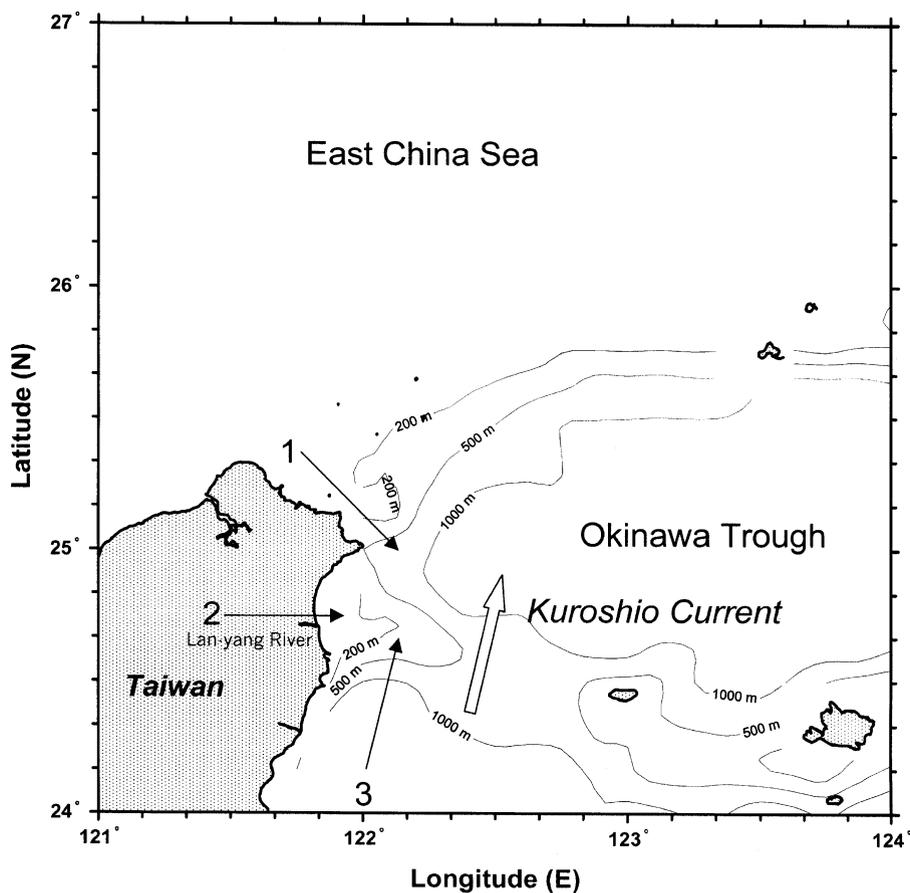


Fig. 4. Map showing river inputs and ocean currents in the study area. Arrow 1: A small alongshore flow just off north Taiwan carrying materials from the southern East China Sea and Taiwan Strait (west of Taiwan) to the study area. Arrow 2: Direct inputs of terrigenous materials from the Lan-yang River. Arrow 3: Other river runoffs from eastern Taiwan carried by the Kuroshio Current to the study area.

in Fig. 5. The major distribution features are that (1) the westernmost four stations in the study area have low values, (2) the four stations with highest values are on the upper slope (shown in solid squares) near Taiwan, and (3) the stations on the lower slope show low values. The distribution suggests a major terrestrial contribution of *n*-alkanes to the area from Taiwan. This is supported by the fact that the immediate shelf north of the study area has no net deposition. For instance, we have collected six other surface sediments (located from 120°10'E to 123°E and from 25°40'N to 25°50'N) from the immediate shelf area and have found that the samples contain shell debris and have low concentrations of mud (0.03–0.77%).

Further, the Keelung shelf offshore northern Taiwan exhibits a rocky bottom with sparsely scattered sandy sediments. These facts suggest that the adjacent shelf has little sediment for export to the study area. However, three sediment-trap moorings deployed in the canyon located between the two highest values of plant wax *n*-alkanes normalized to TOC stations (Fig. 5) in the north have been shown to have high apparent mass fluxes, 5–72 gm<sup>-2</sup>d<sup>-1</sup> (Hung and Chung, 1998). The particle flux could result from a combined flow of an anticyclonic eddy (whose center located at 122°35'E, 25°20'N, details given by Tang et al., 1999) and a southeastward filament (a small alongshore flow) occasionally appearing some-

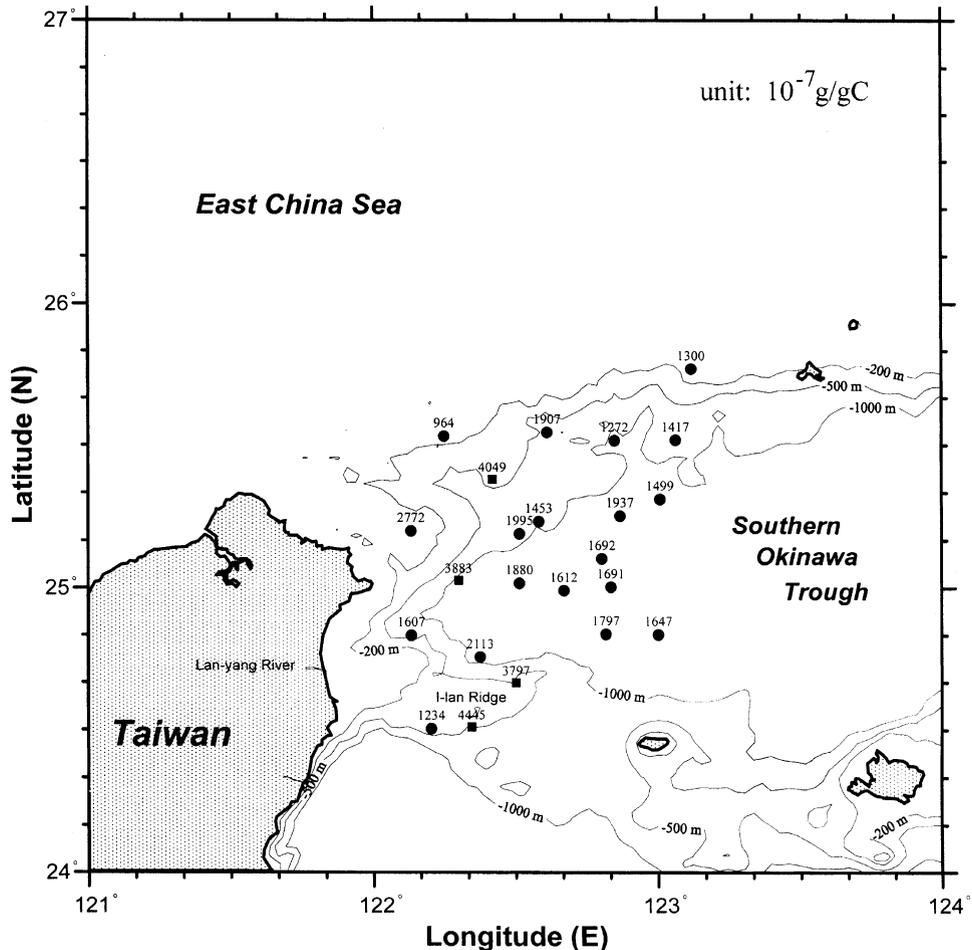


Fig. 5. Spatial distribution of plant wax *n*-alkanes ( $n\text{-C}_{25} + n\text{-C}_{27} + n\text{-C}_{29} + n\text{-C}_{31} + n\text{-C}_{33}$ ) normalized to TOC.

where between the eddy and northern Taiwan (Chern et al., 1990). This suggests that the major mode of sediment transport is suspended particle flow from the southern East China Sea continental shelf. Another possible contribution to this area (particularly the two highest stations in the south) could be the deposition of particles carried by the northbound Kuroshio Current from the south.

For better illustration of terrigenous *n*-alkane distribution, CPI is used because it is a non-mass dependent and dimensionless quantity. It is used to indicate the degree of diagenesis of straight-chain geolipids, and is a numerical representation of how much of the original biological chain-length specificity is preserved in geological lipids

(Meyers and Ishiwatari, 1995). Since all samples examined are surface sediment, the difference in CPIs should reflect more different input sources than diagenetic change. As shown in Fig. 6, it is noted that three northernmost stations exhibit high CPIs, which are not seen in Fig. 5. Sample 456-37 (at the northwest corner of the study area), located on the Keelung shelf, has been investigated for grain size effect on CPI. The unfractionated sample has a CPI value of 3.19, and the <30 mesh fraction has a CPI value of 5.30. This is not in accord with the general observation that CPIs decrease as particle-size decreases (Thompson and Eglinton, 1978; Meyers and Ishiwatari, 1995). It is thought that the larger particle size (>30 mesh)

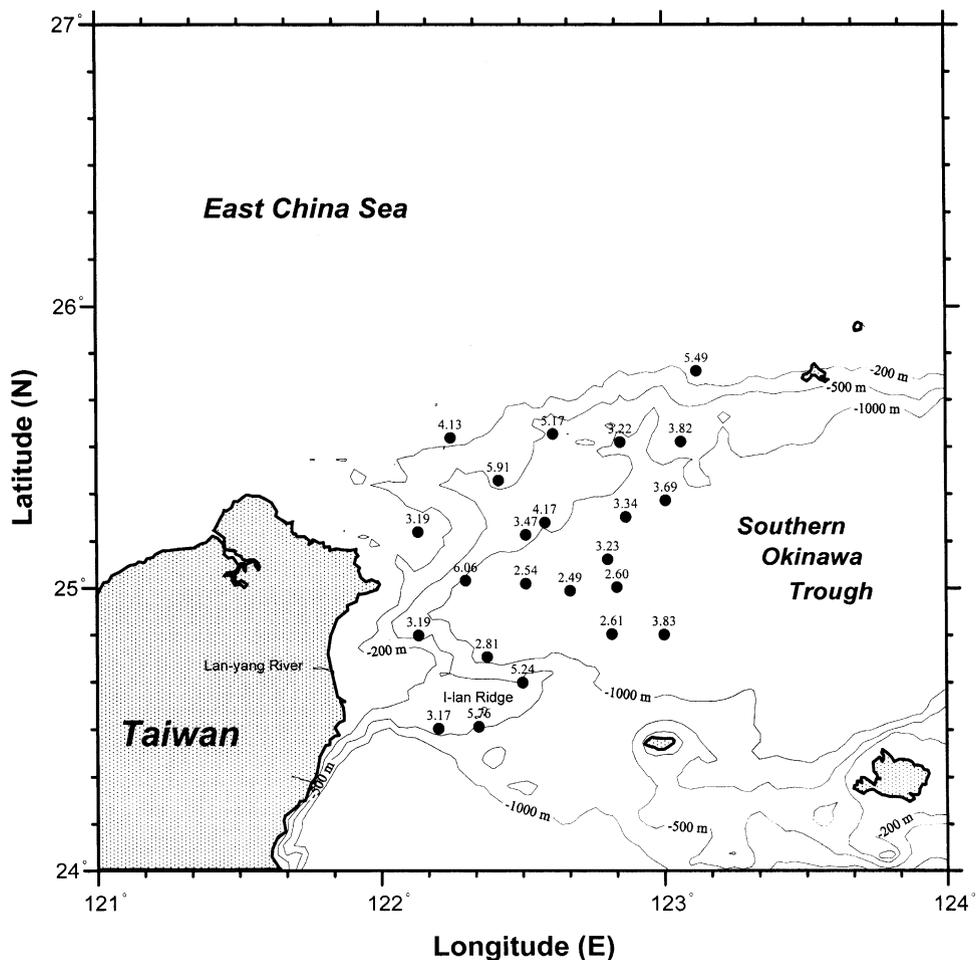


Fig. 6. Spatial distribution of carbon preference index ( $CPI_{25-33}$ ) of *n*-alkanes.

fraction is made up of eroded older (relict) sediment containing degraded lipids with low CPIs and that the smaller particle size (<30 mesh) fraction contains recent sediment with high CPIs. One important feature is that the three stations nearest to Taiwan exhibit low CPIs (3.19, 3.19 and 3.17) and that the highest CPIs are farther from shore. This does not seem to be reasonable if the sediment source is Taiwan.

Geographically, the Lan-yang River, which discharges its sediment (ca. 8.0 Mt/yr, Water Resources Bureau, 1998) directly into the study area, plays a major role in the distribution of terrigenous lipids in the study area, and the CPI signature of its suspended sediments is expected to reflect that of marine sediments. Three samples of total suspended matter from the river have CPIs of 1.38 (Fig. 7), 1.43 and 1.46, which are lower than those of the marine sediments. The result is consistent with a recent report that modern carbon in the POC discharged from the main channel of the river was <30% (Kao and Liu, 1996). In other words, the particulates in the river water are mainly old material, which tends to have lower CPIs. This leads us to consider that, on the two-end member assumption, the marine sediments examined are a mixture of two sources—high CPI sediment from either northwest or south of the study area and low CPI sediment from the nearby Lan-yang River. Those samples closest to Taiwan would have much higher CPIs if there were no input from the Lan-yang River. However, comparatively lower CPIs for the lower slope sediments can be attributed to contribution of old organic carbon from the Lan-yang River.

Terrestrial hydrocarbons (for example, the plant wax alkanes and retene) in Washington shelf sediments have been found to be highly concentrated in the sand-sized, low-density fraction (particles <1.9 g/ml) of bulk sediments (Prah and Carpenter, 1983). However, the sediments off northeastern Taiwan have somewhat high concentrations of plant wax alkanes, but have comparatively low sand contents except some stations located in the north. For instance, two samples taken from this area (lower slope) have sand contents of 2.2% and 2.5% (Jeng and Chen, 1995). Deploying ten moorings for sediment traps in the

northern half of our study area, Chung and Hung (2000) found that the major components of the trap samples in the canyon were silt and sand, but those on the slope were mainly silt. This difference can be attributed to different depositional environments. The low sand percentage could be due to the effect of the Kuroshio Current which turns northeastward in the study area. Coarse suspended particles from the East China Sea shelf are blocked by the Kuroshio and not deposited on the immediate shelf off north Taiwan, and fine particles cross the shelf and settle on the slope of the southern Okinawa Trough (Chen et al., 1992).

The distribution of *n*-fatty alcohols shows a general distribution pattern maximizing at C<sub>22</sub> (for the samples on the lower slope in particular, Fig. 8) or >C<sub>22</sub>. Cranwell (1981) suggests that decomposer organisms may be the source of the *n*-C<sub>22</sub> alcohol present in the sediments; however, Jeng and Chen (1995), studying the grain size effect, show that bacteria do not seem to be the source of the C<sub>22</sub> alcohol. Recent study indicates that the C<sub>22</sub> alcohol can be contributed from freshwater and marine sources (Volkman et al., 1998; and references therein). Lan-yang River suspended matter has an *n*-alkanol distribution with a maximum at C<sub>22</sub> (Fig. 7), which may play a role in the marine sediments. The low-molecular-weight alkanols (*n*-C<sub>14</sub>–*n*-C<sub>22</sub>), which may be formed by hydrolysis of esterified alcohols derived from a wide variety of organisms, such as zooplankton (Boon and de Leeuw, 1979), are considered to be from marine sources. The major alcohols of zooplankton are C<sub>16:0</sub>, C<sub>20:1</sub>, and C<sub>22:1</sub> (Kattner and Krause, 1989). Higher plants generally have their *n*-alkanol distribution >C<sub>22</sub> (Kolattukudy, 1970; Wannigama et al., 1981). Linear regression between *n*-C<sub>22</sub>OH and other biomarker *n*-alkanols (Table 4) shows that the relationship is better correlated for C<sub>20</sub>, C<sub>24</sub>, C<sub>26</sub>, and C<sub>28</sub> than for C<sub>14</sub>, C<sub>16</sub>, and C<sub>18</sub> *n*-alkanols. This result appears to indicate that the sediments have received more *n*-C<sub>22</sub>OH from terrigenous sources than from marine sources. In addition, Lan-yang River suspended matter also contains a high concentration of phytol (Fig. 7), which may contribute to the marine sediments. Linear regression of phytol against other biomarker *n*-alkanols

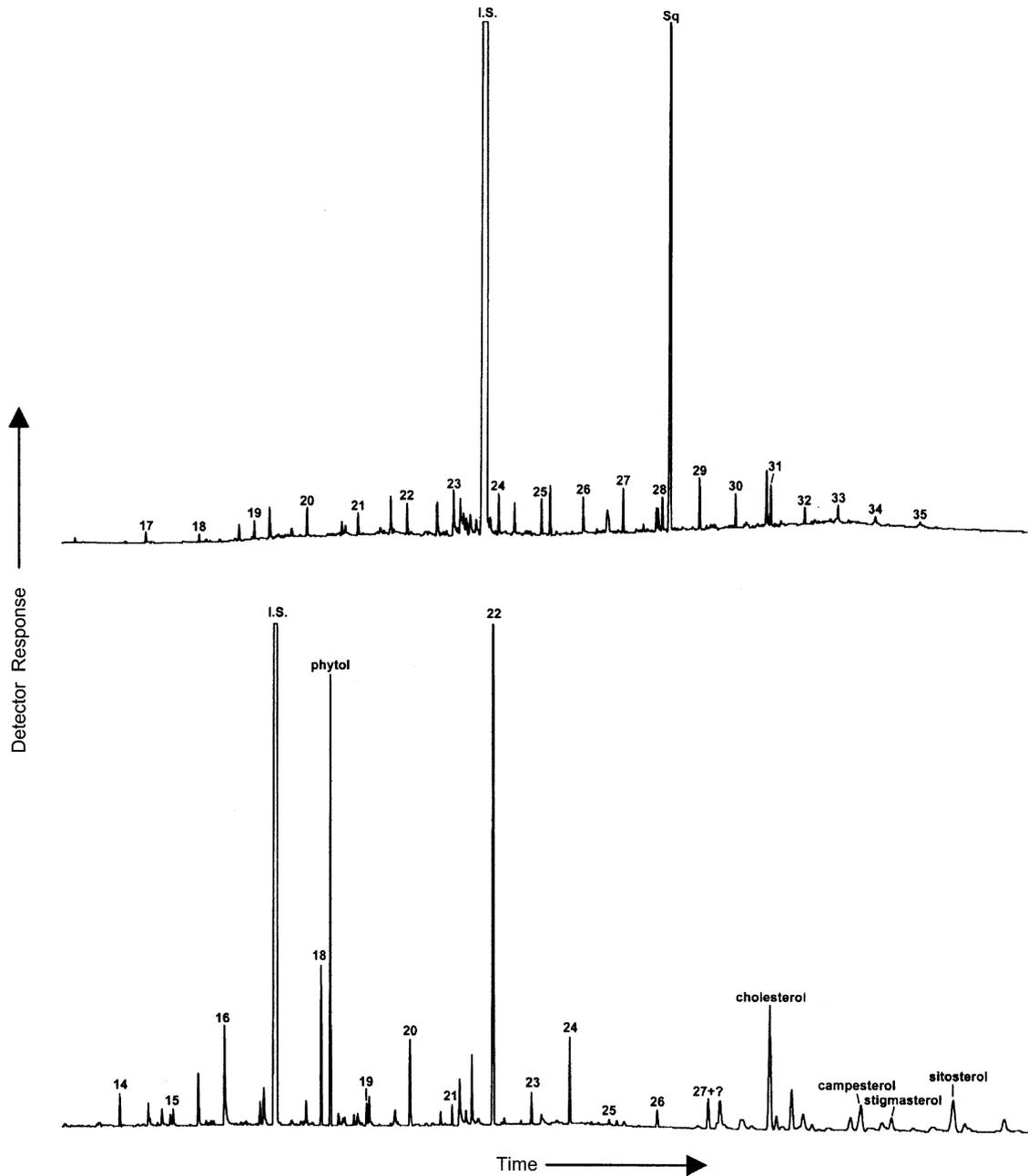


Fig. 7. (upper) Gas chromatogram of the aliphatic fraction from the Lan-yang River suspended matter. Numbers above peaks refer to carbon number of  $n$ -alkanes; Sq = squalene; I.S. = internal standard ( $n\text{-C}_{24}\text{D}_{50}$ ).  $\text{CPI}_{25-33} = 1.38$ . (lower) Gas chromatogram of the alkanol/sterol fraction from the Lan-yang River suspended matter. Numbers above peaks refer to carbon number of  $n$ -alkanol; I.S. = internal standard ( $n\text{-C}_{17}\text{OH}$ ).

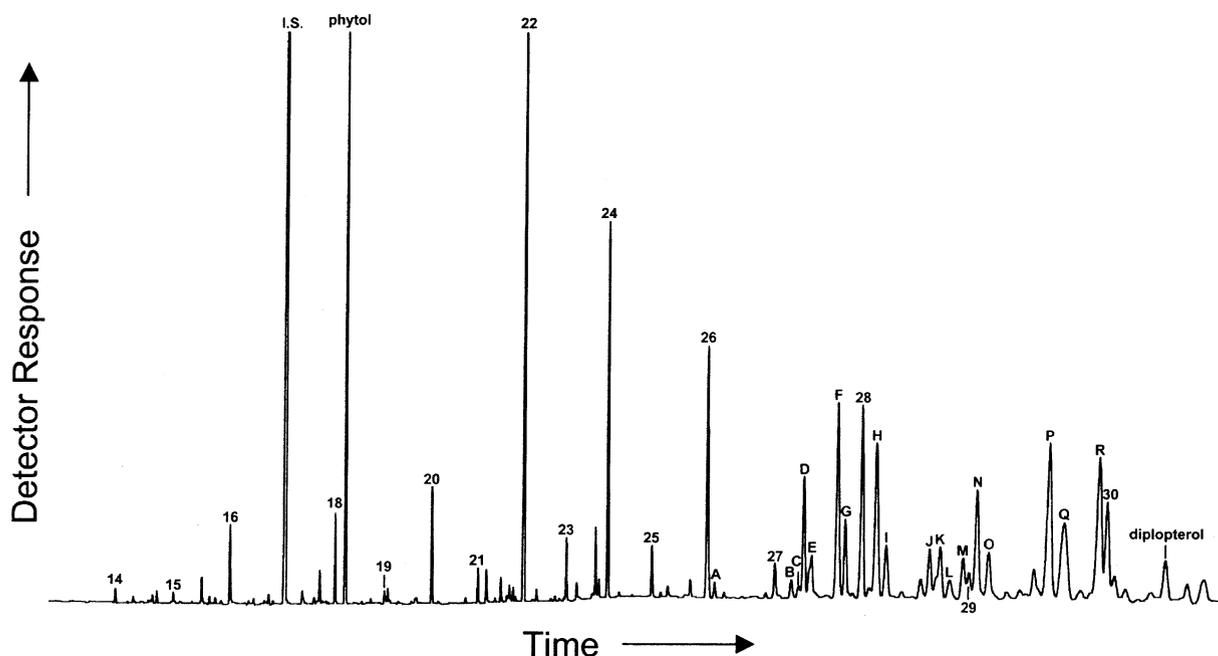


Fig. 8. Gas chromatogram of the alkanol/sterol fraction for sample 417-45. Numbers above peaks refer to carbon number of *n*-alkanols; I.S. = internal standard (*n*-C<sub>17</sub>OH). Letters above peaks refer to sterols: (A) 24-nor-5 $\alpha$ -cholest-22E-en-3 $\beta$ -ol, (B) 27-nor-24-methylcholesta-5,22E-dien-3 $\beta$ -ol, (C) 27-nor-24-methyl-5 $\alpha$ -cholest-22E-dien-3 $\beta$ -ol, (D) cholesta-5,22E-dien-3 $\beta$ -ol (22-dehydrocholesterol), (E) 5 $\alpha$ -cholest-22E-en-3 $\beta$ -ol, (F) cholest-5-en-3 $\beta$ -ol (cholesterol), (G) 5 $\alpha$ -cholestan-3 $\beta$ -ol (cholestanol), (H) 24-methylcholesta-5,22E-dien-3 $\beta$ -ol (diatomsterol), (I) 24-methyl-5 $\alpha$ -cholest-22E-en-3 $\beta$ -ol, (J) 24-methylcholesta-5,24(28)-dien-3 $\beta$ -ol (24-methylenecholesterol), (K) 24-methylcholest-5-en-3 $\beta$ -ol (campesterol) + 24-methyl-5 $\alpha$ -cholest-24(28)-en-3 $\beta$ -ol(?), (L) 24-methyl-5 $\alpha$ -cholestan-3 $\beta$ -ol, (M) 23,24-dimethylcholesta-5,22E-dien-3 $\beta$ -ol, (N) 24-ethylcholesta-5,22E-dien-3 $\beta$ -ol (stigmasterol), (O) 24-ethyl-5 $\alpha$ -cholest-22E-en-3 $\beta$ -ol, (P) 24-ethylcholest-5-en-3 $\beta$ -ol (sitosterol), (Q) 24-ethyl-5 $\alpha$ -cholestan-3 $\beta$ -ol + unknown, and (R) 4 $\alpha$ ,23,24-trimethyl-5 $\alpha$ -cholest-22E-en-3 $\beta$ -ol (dinosterol). Dipl. = diplopterol.

(Table 5) shows that phytol is better correlated with C<sub>14</sub>, C<sub>16</sub>, C<sub>18</sub>, and C<sub>20</sub> than with C<sub>24</sub>, C<sub>26</sub>, and C<sub>28</sub> *n*-alkanols, indicating that the sedimentary phytol receives more contribution from marine sources.

The ratio of *n*-C<sub>16</sub>OH to *n*-C<sub>28</sub>OH was used by Venkatesan et al. (1987) as an index to show marine vs. terrigenous input of *n*-alkanols to the sediments. The ratios of the sediments range from 0.03 to 0.24 (with a single outlier of 3.70) with a mean of  $0.13 \pm 0.06$  (Table 3), indicating the predominance of higher plant *n*-alkanols in the area, which is in agreement with the *n*-alkane distribution. To show the spatial distribution of terrestrial *n*-alkanols in the area, the sum of *n*-C<sub>24</sub>OH + *n*-C<sub>26</sub>OH + *n*-C<sub>28</sub>OH normalized to TOC was calculated (Table 3) and shown in Fig. 9. The spatial distribution pattern of *n*-fatty alcohols

matches well with that of *n*-alkanes with respect to the occurrence of high values (shown in solid squares).

Three phytosterols—campesterol, stigmasterol, and  $\beta$ -sitosterol—are generally found in epicuticular waxes of vascular plants (Scheuer, 1973; Weete, 1976). They are a major part of the sterol fraction in the sediments analyzed (Fig. 8) and account for, on average, 24.3% of the total sterols (Table 6). The spatial distribution of the phytosterol concentrations normalized to TOC (Fig. 10) is totally different from those of *n*-alkanes and *n*-alkanols. The four stations with the highest values for *n*-alkanes and *n*-alkanols do not exhibit the highest values for phytosterols. Instead, the highest value spot is located just off northeastern Taiwan. It is seen from Fig. 10 that phytosterols are randomly distributed. The cause of this

Table 3  
Fatty alcohol data

Sample	TOC (10 <sup>-2</sup> g/g)	Phytol (ng/g)	<i>n</i> -C <sub>16</sub> OH (ng/g)	<i>n</i> -C <sub>22</sub> OH (ng/g)	<i>n</i> -C <sub>28</sub> OH (ng/g)	<i>n</i> -C <sub>16</sub> OH/ <i>n</i> -C <sub>28</sub> OH	Σ <i>n</i> -C <sub>24,26,28</sub> (ng/g)	(Σ <i>n</i> -C <sub>24,26,28</sub> / TOC) × 10 <sup>-7</sup>
<i>ORI-417</i>								
5(0–5 cm)	1.01	981	96	695	682	0.14	1770	1750
8(0–3 cm)	1.05	1740	215	880	881	0.24	2220	2120
9(0–3 cm)	1.09	1100	139	489	632	0.22	1650	1520
10(0–3 cm)	1.00	1110	148	863	701	0.21	1810	1810
16(0–5 cm)	0.82	370	88	437	529	0.17	1420	1740
20(0–4 cm)	0.63	350	72	903	1560	0.05	3940	6250
21(0–4 cm)	0.77	1370	94	842	595	0.16	1590	2060
27	0.16	66	15	194	154	0.10	350	2190
34(0–4 cm)	0.69	204	59	857	1800	0.03	4380	6340
36(0–4 cm)	0.42	65	46	758	1040	0.04	2740	6510
37(0–4 cm)	0.29	34	30	374	162	0.19	378	1300
42(0–4 cm)	0.41	138	34	480	259	0.13	652	1590
43(0–4 cm)	0.73	431	53	694	418	0.13	1120	1530
44(0–4 cm)	0.84	941	81	894	618	0.13	1630	1940
45	0.87	863	72	750	553	0.13	1460	1680
46(0–4 cm)	0.79	450	76	750	562	0.14	1460	1850
<i>ORI-456</i>								
27	0.74	861	62	739	1120	0.06	2930	3960
33(0–4 cm)	0.38	250	38	461	279	0.14	771	2030
34(0–4 cm)	0.74	291	51	740	1540	0.03	4140	5590
35	0.14	47	17	180	91	0.19	235	1680
37	0.36	36	185	89	50	3.70	148	411
39(0–4 cm)	0.71	888	77	433	673	0.11	1510	2130
K(0–4 cm)	0.60	451	40	599	367	0.11	966	1610
Average						0.13		
s.d.(1σ)						0.06 <sup>a</sup>		

<sup>a</sup> Exclusive of 456–37.

Table 4  
Linear regressions between *n*-C<sub>22</sub>OH and other biomarker *n*-alkanols for the 23 marine sediments

Regression	<i>r</i>	<i>p</i>
<i>n</i> -C <sub>22</sub> OH = 4.77 <i>n</i> -C <sub>14</sub> OH + 517	0.24	0.26
<i>n</i> -C <sub>22</sub> OH = 0.890 <i>n</i> -C <sub>16</sub> OH + 544	0.18	0.40
<i>n</i> -C <sub>22</sub> OH = 4.65 <i>n</i> -C <sub>18</sub> OH + 263	0.58	0.004
<i>n</i> -C <sub>22</sub> OH = 4.45 <i>n</i> -C <sub>20</sub> OH + 131	0.84	< 0.001
<i>n</i> -C <sub>22</sub> OH = 0.561 <i>n</i> -C <sub>24</sub> OH + 344	0.74	< 0.001
<i>n</i> -C <sub>22</sub> OH = 0.411 <i>n</i> -C <sub>26</sub> OH + 382	0.69	< 0.001
<i>n</i> -C <sub>22</sub> OH = 0.360 <i>n</i> -C <sub>28</sub> OH + 374	0.70	< 0.001

*r* = correlation coefficient, *p* = significance value.

Table 5  
Linear regressions of phytol against other biomarker *n*-alkanols for the 23 marine sediments

Regression	<i>r</i>	<i>P</i>
Phytol = 24.6 <i>n</i> -C <sub>14</sub> OH + 74.1	0.63	0.001
Phytol = 6.05 <i>n</i> -C <sub>16</sub> OH + 97.2	0.64	0.001
Phytol = 11.6 <i>n</i> -C <sub>18</sub> OH - 311	0.73	< 0.001
Phytol = 7.60 <i>n</i> -C <sub>20</sub> OH - 256	0.73	< 0.001
Phytol = 1.07 <i>n</i> -C <sub>22</sub> OH - 90.2	0.54	0.007
Phytol = 0.266 <i>n</i> -C <sub>24</sub> OH + 440	0.18	0.42
Phytol = 0.180 <i>n</i> -C <sub>26</sub> OH + 466	0.15	0.48
Phytol = 0.162 <i>n</i> -C <sub>28</sub> OH + 460	0.16	0.47

*r* = correlation coefficient, *p* = significance value.

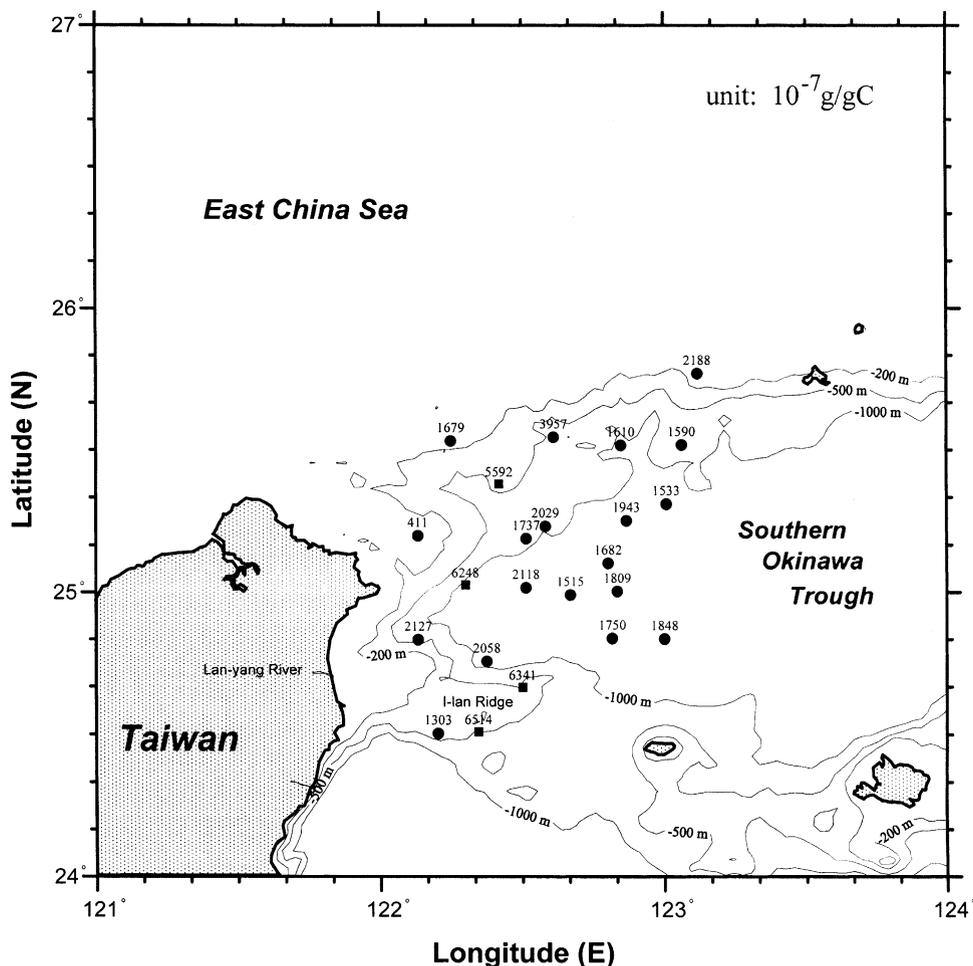


Fig. 9. Spatial distribution of plant wax *n*-fatty alcohols ( $n\text{-C}_{24}\text{OH} + n\text{-C}_{26}\text{OH} + n\text{-C}_{28}\text{OH}$ ) normalized to TOC.

difference is unclear. Part of the reason might be that the three phytosterols originally thought to be terrigenous markers also could be produced by marine organisms (Volkman, 1986).

### 3.2. Correlation between TOC and terrigenous lipid biomarkers

Linear regression of TOC versus concentrations of land plant wax *n*-alkanes (sum of  $n\text{-C}_{25}$ ,  $n\text{-C}_{27}$ ,  $n\text{-C}_{29}$ ,  $n\text{-C}_{31}$  and  $n\text{-C}_{33}$ ) in sediments is expected to give a strong positive linear correlation if a point source exists in a region of interest, such as that for the Columbia River drainage basin (Hedges and Prahl, 1993). The relationship for the sediments in

the study area is positive but weak ( $r = 0.64$ ,  $p = 0.001$ ). Linear regression of TOC against concentrations of land plant wax *n*-alkanols (sum of  $n\text{-C}_{24}\text{OH}$ ,  $n\text{-C}_{26}\text{OH}$  and  $n\text{-C}_{28}\text{OH}$ ) also shows positive linear correlation ( $r = 0.42$ ,  $p = 0.048$ ), which is weaker. This result could be attributed to several factors: (1) a complex input (not a point source) of terrigenous organic matter to the study area, (2) sediment receiving a blend of eroded and recent lipids (discussed earlier), (3) TOC also including marine organic matter, (4) temporal variations in river flow due to flooding, and (5) different rates of degradation for TOC and individual biomarkers. However, there is a strong positive linear relationship between TOC and the

Table 6  
Sterol and diplopterol data

Sample	TOC (g/100 g)	Campesterol (ng/g)	Stigmasterol (ng/g)	Sitosterol (ng/g)	Total <sup>a</sup> Phytosterols (ng/g)	(Total phytosterols/ TOC) × 10 <sup>-7</sup>	Total sterols (ng/g)	Diplopterol (ng/g)
<i>ORI-417</i>								
5(0–5 cm)	1.01	257	432	1000	1690	1670	6270	3050
8(0–3 cm)	1.05	416	630	1260	2360	2200	9340	2960
9(0–3 cm)	1.09	398	587	1080	2060	1890	7870	401
10(0–3 cm)	1.00	287	479	1010	1780	1780	6640	892
16(0–5 cm)	0.82	313	455	772	1540	1880	6190	79
20(0–4 cm)	0.63	116	79	282	477	757	2200	55
21(0–4 cm)	0.77	214	377	727	1320	1710	5540	492
27	0.16	25	13	32	70	438	449	26
34(0–4 cm)	0.69	112	101	362	575	833	2450	85
36(0–4 cm)	0.42	84	117	332	533	1270	2130	51
37(0–4 cm)	0.29	36	33	92	161	555	869	13
42(0–4 cm)	0.41	62	118	246	426	1040	1520	38
43(0–4 cm)	0.73	94	190	390	674	923	2780	130
44(0–4 cm)	0.84	191	346	669	1210	1440	4960	232
45	0.87	213	344	685	1240	1430	4860	376
46(0–4 cm)	0.79	117	197	502	816	1030	3320	786
<i>ORI-456</i>								
27	0.74	220	191	544	955	1290	4550	90
33(0–4 cm)	0.38	86	111	403	501	1320	1810	71
34(0–4 cm)	0.74	178	178	434	790	1070	3220	108
35	0.14	15	22	44	81	579	371	12
37	0.36	107	162	392	661	1840	2610	38
39(0–4 cm)	0.71	495	502	1100	2090	2950	8180	187
K(0–4 cm)	0.60	113	174	399	686	1140	2660	128

<sup>a</sup> Phytosterols = campesterol + stigmasterol + sitosterol.

concentration of phytosterols (sum of campesterol, stigmasterol and sitosterol),  $r = 0.85$  ( $p < 0.001$ ). The strong positive correlation could mean that the mixing or dilution of phytosterols in terrestrial organic carbon with marine organic carbon with or without the phytosterols follows a nearly constant ratio, which is especially remarkable for the study area with a complex input.

### 3.3. Source of diploptene

Both diploptene (hop-22(29)-ene, Fig. 1) and diplopterol (hopan-22-ol, Fig. 8) were found in the sediments; their EI mass spectra are given in Fig. 11. Diploptene is derived from terrestrial higher plants and is also formed by bacteria. If it is derived from higher plants, a strong correlation

between diploptene and terrestrial higher plant *n*-alkanes has been demonstrated (Prahl et al., 1992). A plot of diploptene versus  $\Sigma C_{25-33}$  *n*-alkanes (odd carbon only) for our samples shows scatter (Fig. 12), indicating no correlation. The diploptene in the study area is not predominantly derived from terrestrial higher plants, but from bacteria. It is also known that diplopterol is present in many bacteria and that concentrations up to 1600  $\mu\text{g/g}$  dry wt have been found in some methylotrophs, although small amounts of diplopterol also have been found in some cyanobacteria, mosses and ferns (Rohmer et al., 1984). Moreover, diplopterol plotted against diploptene exhibits strong positive linear correlation ( $r = 0.93$ ,  $p < 0.001$ , Fig. 12). This result suggests that the diploptene in the study area may be mainly derived from diplopterol

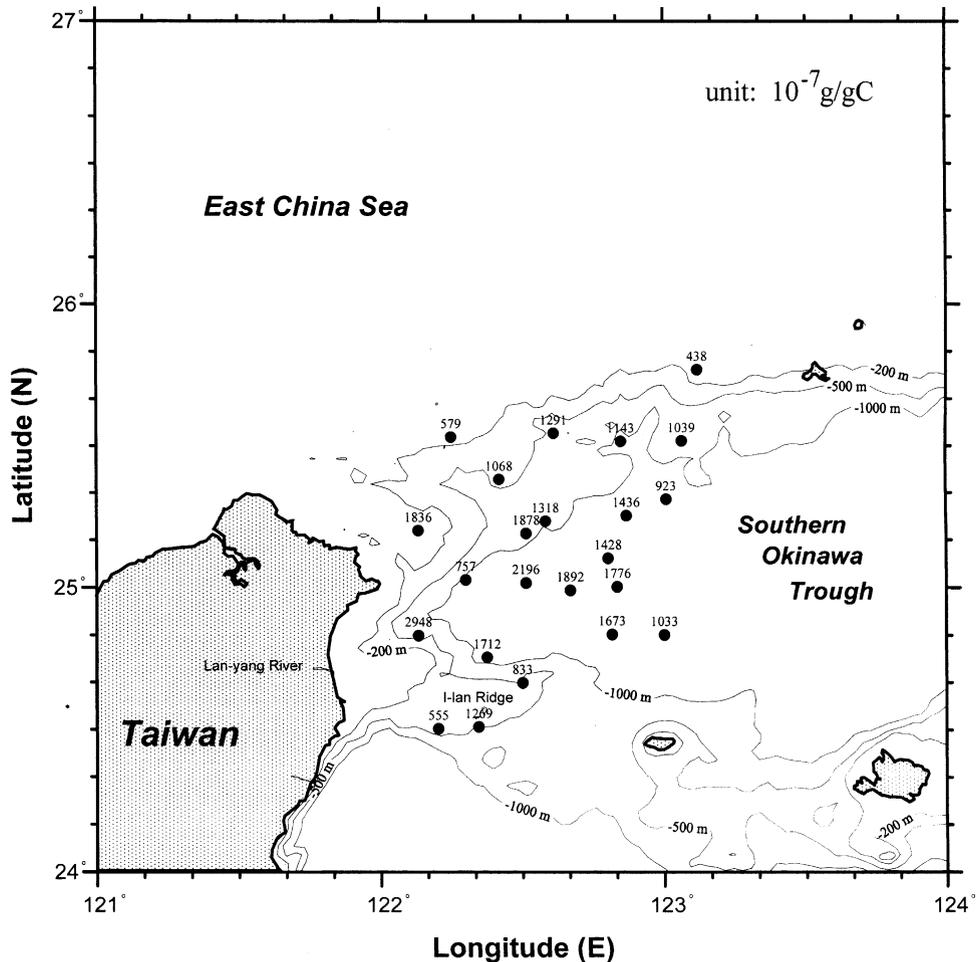


Fig. 10. Spatial distribution of phytosterols normalized to TOC.

through dehydration in early diagenesis. In addition, many active submarine hydrothermal activities in the study area have been observed in the southern Okinawa Trough (Lee et al., 1998). Furthermore, an extraordinarily high concentration (23 730 nl/l) of dissolved methane in the bottom water (depth 1440 m) was found at 25°04.18'N, 122°35.20'E (KEEP-MASS, 1992) although other high levels of dissolved methane in the bottom waters were also found in the study area (Chen, 1994). Subsequent collection of bottom water (within 2 m of sea bottom) and sediment simultaneously using a specially-designed sampler for methane study was made closest to this site. Abnormally high to high dissolved

methane concentrations in bottom water were observed at stations P (25°03.12'N, 122°34.23'E; water depth 1283 m), Q (25°04.37'N, 122°35.32'E; water depth 1395 m) and R (25°04.88'N, 122°40.07'E; water depth 1380 m)—11 100, 1500 and 320 nl/l, respectively (Chen, 1994). As shown in Fig. 13, relatively high diploptene and diplopterol concentrations in sediments also were obtained at stations Q (5440 and 7550 ng/g, respectively) and R (1380 and 842 ng/g, respectively). If stations Q and R are included in Fig. 12, the regression equation is diplopterol =  $-38.7 + 1.40$  diploptene ( $r = 0.97$ ,  $p < 0.001$ ). This leads us to consider some relation between methane, diplopterol and diploptene. As a

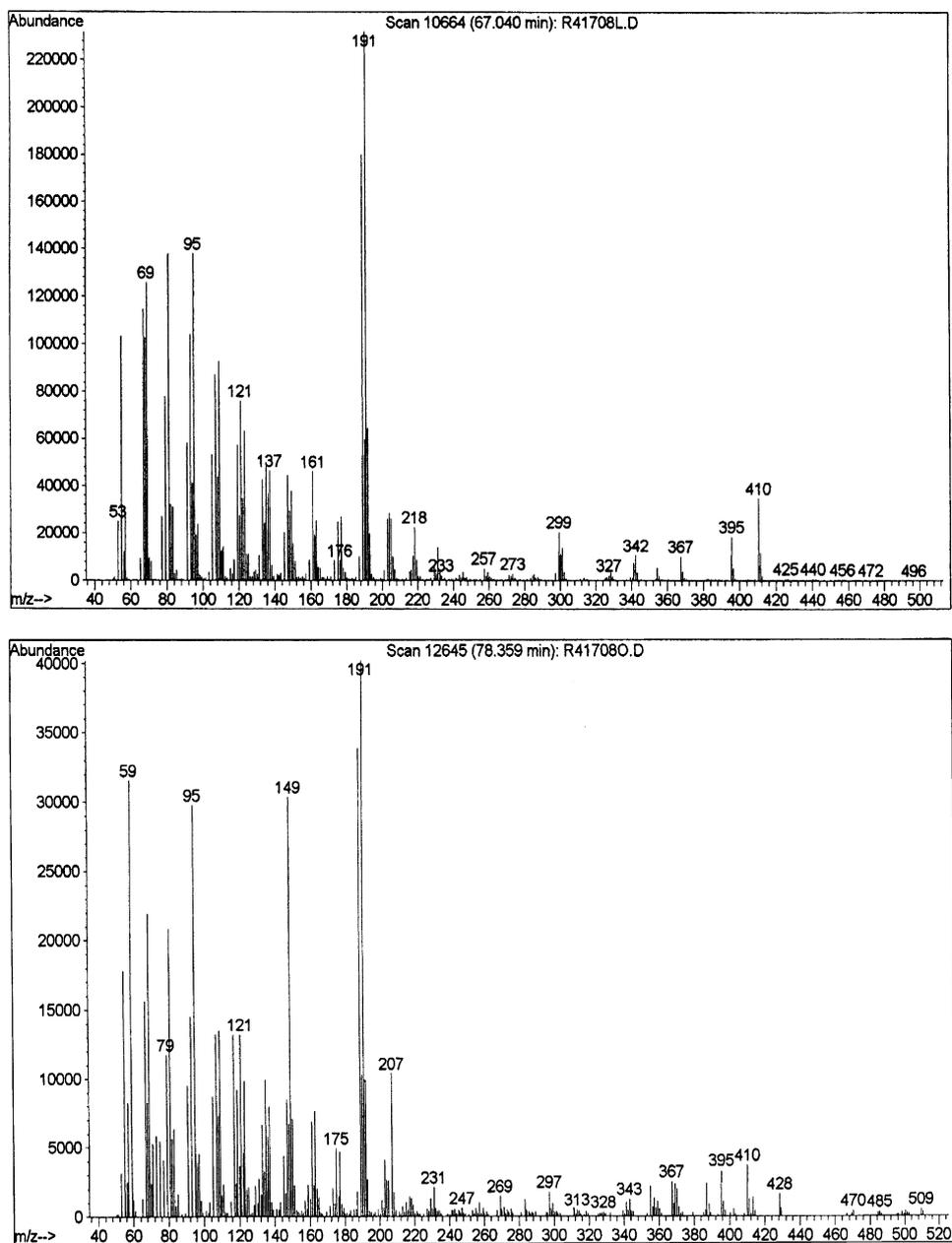


Fig. 11. Electron impact mass spectra of diploptene (upper) and diplopterol (lower) from station 417-8. Note that the present silylation procedure using BSA alone did not silylate diplopterol, and the spectrum shown here represents its free form.

consequence, it is speculated that diplopterol in the sediments could be synthesized by methylo-trophic bacteria using methane as their carbon source.

#### 4. Conclusions

Compared with other coastal areas, the sediments off northeastern Taiwan contain comparatively

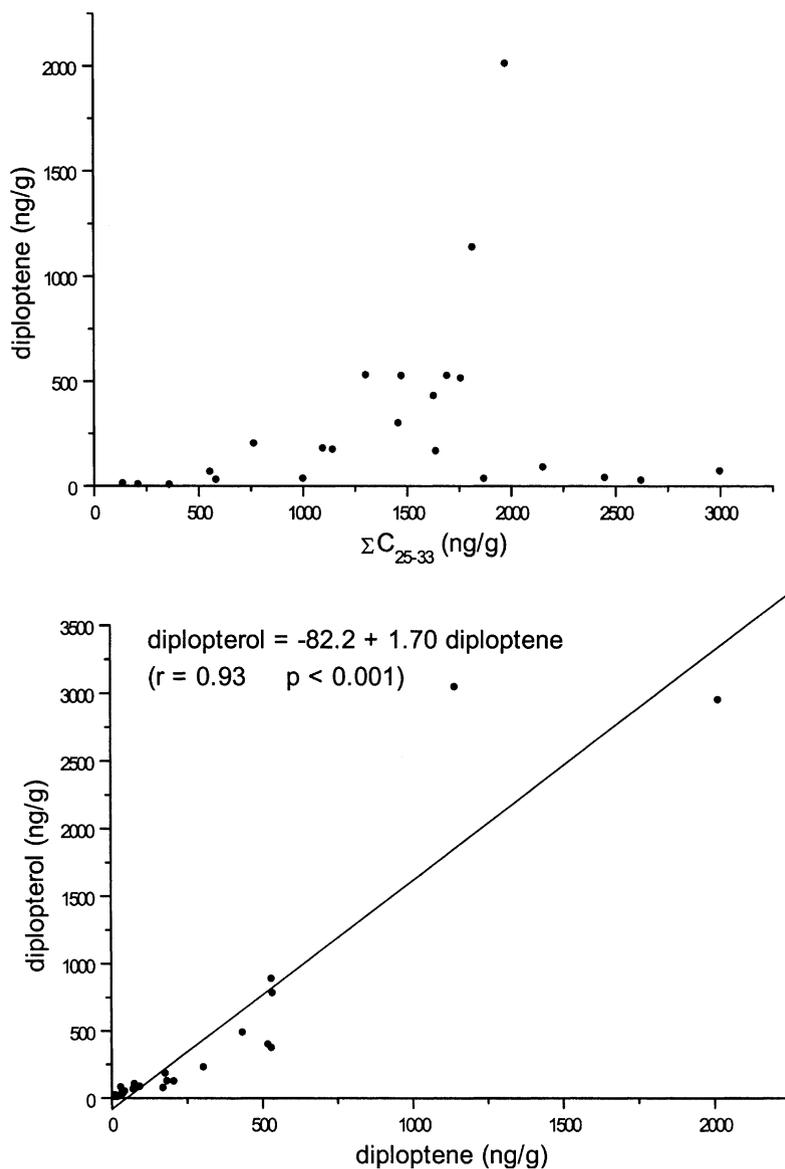


Fig. 12. (upper) Plot of diplotene versus  $\text{CPI}_{25-33}$  of *n*-alkanes and (lower) plot of diplopterol versus diplotene.

high proportions of terrigenous *n*-alkanes and *n*-alkanols. The highest concentrations of plant waxes were found on the upper slope of the southernmost Okinawa Trough near Taiwan. The major terrigenous inputs to this area are particle fluxes from the southern East China Sea shelf and river runoff from the east Taiwan coast. However, contribution from the Lan-yang River

input is less important. TOC and phytosterol concentrations are positively correlated ( $r = 0.85$ ,  $p < 0.001$ ).

In general, the present results show that the *n*-alkane and *n*-alkanol concentrations from marine sources are smaller than those from terrigenous sources. Therefore, biased conclusions may be obtained if only *n*-alkanes and *n*-alkanols are

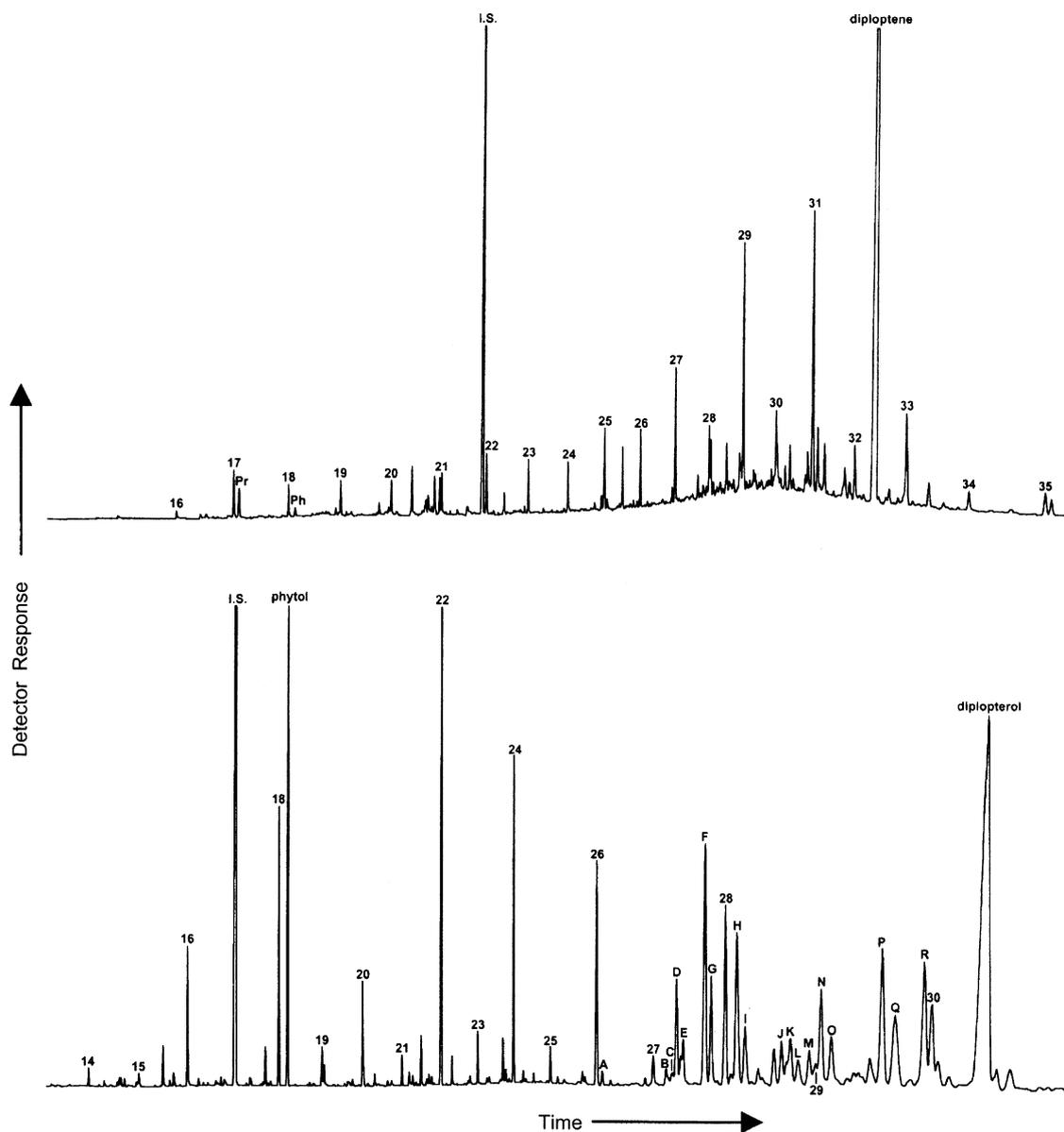


Fig. 13. Chromatograms containing diptoptene (upper) and diptoptol (lower) from station Q with high concentration of dissolved methane in overlying bottom water. Note that the retention time of diptoptene differs from that of Fig. 1 since the analysis was made earlier using an SE-30 column. Numbers above peaks refer to carbon number of *n*-alkanes (upper) and *n*-alkanol (upper). Letters above peaks refer to sterols; their identifications as in Fig. 8.

considered. In effect, our data do indicate that some marine-derived lipids outweigh terrigenous ones such as phytol and sterols. The observed lower concentrations of marine lipids in the sediments are attributed to the fact that terrige-

nous lipids are often preferentially preserved due to sequestration and that marine lipids are more prone to degradation in the marine environment especially those compounds derived from plankton.

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