



PERGAMON

Applied Geochemistry 16 (2001) 95–108

**Applied  
Geochemistry**

www.elsevier.com/locate/apgeochem

# Comparative study of sterols in shelf and slope sediments off northeastern Taiwan

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Received 24 September 1998; accepted 13 January 2000

Editorial handling by B.R.T. Simoneit

## Abstract

Bottom sediments and suspended matter from the shelf and slope areas off northeastern Taiwan were analyzed for sterol, *n*-alkanol and *n*-alkane compositions. The  $\Sigma(\text{algal sterols/cholesterol})$  ratios (mean  $\pm 1\sigma$ ) were  $1.00 \pm 0.31$  ( $n = 7$ ) for the shelf sediments,  $3.29 \pm 0.61$  ( $n = 7$ ) for the slope sediments, and  $1.54 \pm 0.19$  ( $n = 3$ ) for the suspended matter. The much higher proportion of cholesterol in the shelf sediments is most likely derived from mollusks in the relict sediments. An additional sterol source for the shelf sediments probably is suspended matter in the water column although the shelf has no apparent sedimentation. Results from phytol and *n*-alkanols suggest recent inputs of lipids from the water column to the shelf. The stanol/stenol ratios are lower for the older shelf sediments than for the younger slope sediments. © 2000 Elsevier Science Ltd. All rights reserved.

## 1. Introduction

The seafloor off northeastern Taiwan is characterized by a shelf-slope-trough complex with a rather sharp NW–SE gradient (Fig. 1). As the northbound Kuroshio Current impinges on the slope, it turns northeastward and causes a cyclonic eddy and upwelling (Tang et al., 1999). The combined effects of topography and water circulation on material exchange and biological productivity in this region have drawn much attention in recent years (Chen Lee, 1992; Chen et al., 1995; Chung and Wu, 1995; Shieh et al., 1996).

Sediments from the shelf are composed mainly of shell fragments and light brown sands, which are pri-

marily remnants of relic beaches of the last glaciation when the sea level was low. The slope sediments consist of fine-grained mud of non-biogenic origin (Chen et al., 1992). Studies have shown that the shelf area has no apparent sedimentation in the Holocene, whereas the slope area has apparent sedimentation rates ranging from 0.10 to 0.52 cm a<sup>-1</sup> (e.g., Chen et al., 1995; Chung and Chang, 1995). Thus, these two types of material represent two sharply different sedimentary environments.

Sterols and their derivatives are important geochemical biomarkers. The distribution of sterols found in biota and in sediments, as well as some of the diagenetic modifications affecting this compound class has been reviewed by Mackenzie et al. (1982), de Leeuw and Baas (1986), and Volkman (1986). Nishimura and Koyama (1977) compared algae and vascular plants from Lake Suwa (Japan) and the area around the lake, and showed that cholesterol is the dominant algal

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sterol, whereas  $\beta$ -sitosterol is the major sterol in emergent water plants and in watershed plants. Mermoud et al. (1985) noted that epicholestanol is present in surficial sections of a core from the anoxic, deep part of Lake Lemman (Switzerland) but absent from oxygenated sediments in other parts of the lake and in older parts of the core. In Motte Lake (France), sediments throughout a 6-m core contain gorgostanol, believed to be derived from the microbial reduction of gorgosterol, which is probably produced by the dinoflagellate phytoplankton abundant in this lake (Wuenschel et al., 1987). Rieley et al. (1991) reported that leaf waxes contain  $C_{28}$  and  $C_{29}$  sterols but lack  $C_{27}$  sterol. The use of  $\beta$ -sitosterol as a biomarker specifically for pulp-mill effluent is given by Leeming and Nichols (1998). Numerous studies use coprostanol as a tool for tracing sewage pollution (O'Leary et al., 1999; and references therein). Lee et al. (1977) studied sterol diagenesis in recent sediments from Buzzards Bay (USA) and found that extractable sterols decreased in concentration while bound sterols increased in concentration with

depth. From this result, they inferred that a transformation between extractable and bound sterols appeared to be occurring over the entire core. In the anoxic Tan-shui Estuary (Taiwan), Jeng and Han (1996) showed that extractable  $\Sigma$ coprostanol/TOC and cholesterol/TOC increased with depth, but extractable cholesterol showed the opposite trend. Both bound  $\Sigma$ coprostanol/TOC and cholesterol/TOC also displayed a decreasing trend with depth downcore, which might be ascribed to assimilation by bacteria and/or chemical decomposition.

The shelf area off northeastern Taiwan is uncommon in that the old (relict) sediments are on the surface of the sea bottom in the oxic environment. This oxic shelf is presumably unfavorable for the preservation of old organic compounds since their degradation is generally more rapid under oxic conditions than under anoxic conditions (Hulthe et al., 1998). As a result, the shelf sediments are expected to contain some residual lipids derived from mollusks and/or other recent lipids from the water column (very little from sediment transport

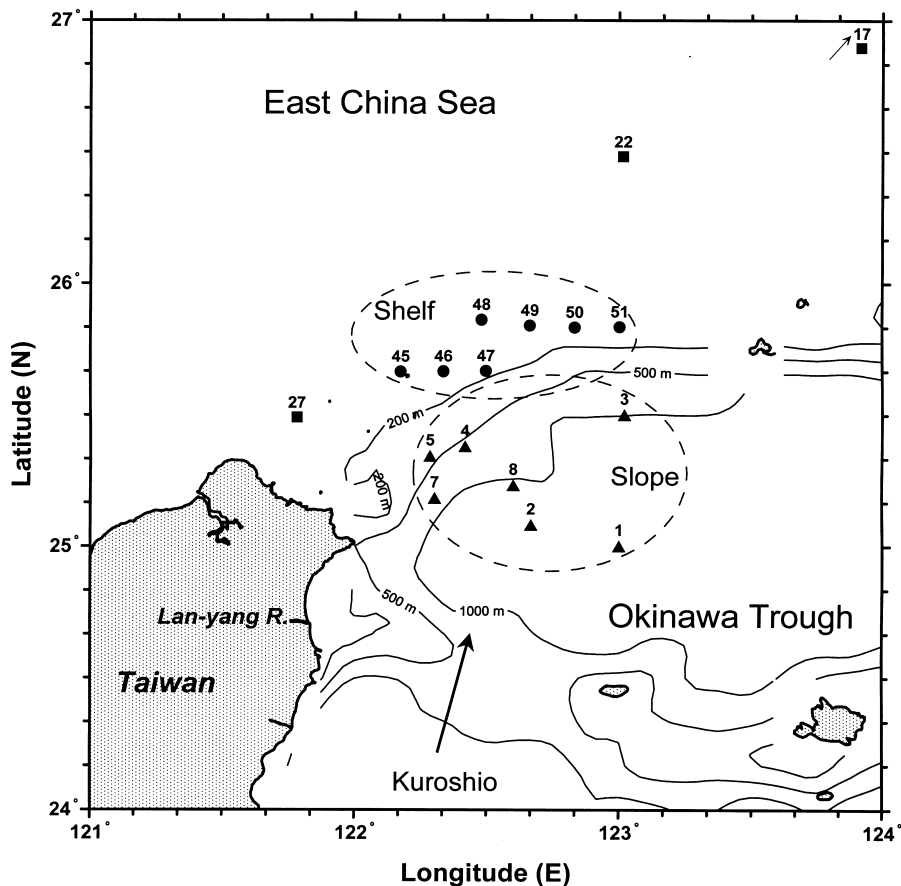


Fig. 1. Map of the sampling sites for shelf sediments (●), slope sediments (▲) and surface water samples (■). Note that station 17 is off the map, located at 124°30.21'E, 28°00.02'N. Also note that the contours are simplified greatly.

due to no apparent sedimentation). The purpose of this work is to compare the sterol, *n*-alkanol and *n*-alkane compositions of the shelf and slope sediments in this region and gain some insights into the sources, pathways and diagenesis of these compounds. Emphasis is placed on the source and distribution of sterols in the shelf sediments because they contain relic shell fragments and debris, and because cholesterol is usually the predominant sterol of most mollusks and other shell-containing animals (Ballantine et al., 1983; and references therein).

## 2. Methodology

### 2.1. Sampling

Box cores were taken from the shelf and the slope areas off northeastern Taiwan on board the R/V *Ocean Researcher I* (Fig. 1). Sediment samples from the shelf area were wet-sieved using a 30 mesh stainless steel screen to separate shell fragments and coarse debris, followed by a 230 mesh screen (63  $\mu\text{m}$ ) to collect the mud fraction for sterol analysis. The relative proportions of the 3 size fractions (>30 mesh, 30–230 mesh, and <230 mesh) in some samples are shown in Table 1. In general, the mud fraction accounts for <1% of the total mass. Samples from the slope area were mostly muddy, and no further size fractionation was made except sample 7 which was passed through a 30 mesh stainless steel screen due to its poor sorting.

Besides bottom sediments, 3 surface (1 m below the sea surface) water samples (50–60 l each) were collected from the shelf area and processed in situ. The water samples were pressure filtered through pre-cleaned (batch washed with acetone and dichloromethane several times) and pre-weighed 142 mm, 0.45  $\mu\text{m}$  Whatman glass microfibre filters using a Satorius PTFE pressure filter holder and a Masterflex wriggle pump. The filters were kept frozen at  $-20^\circ\text{C}$  until analysis. The mud fraction of the shelf sediments

and the top 4 cm of the slope sediments were freeze-dried and ground using a mortar grinder.

### 2.2. Extraction and separation

Internal standards (1-heptadecanol and *n*-C<sub>24</sub>D<sub>50</sub>) were added to the sediment and suspended matter and the spiked samples were extracted with benzene/methanol (1:1) in a Soxhlet apparatus for 24 h. The extract was concentrated (using N<sub>2</sub> gas) and hydrolyzed with methanolic KOH. The neutral lipids were extracted with *n*-hexane (4 times). The nonsaponifiable lipids were subjected to silica gel (deactivated with 5% H<sub>2</sub>O) column chromatography. Aliphatic hydrocarbons were eluted with *n*-hexane. Less polar lipids were eluted with *n*-hexane/dichloromethane (2:3, v/v), and the fraction containing fatty alcohols and sterols was isolated by using a mixture of dichloromethane/methanol (4/1, v/v). The isolated alcohols and sterols were taken to dryness, redissolved in benzene, and derivatized with NO-*bis*(trimethylsilyl)acetamide.

### 2.3. Analysis

Alcohols/sterols (as TMS ethers) and aliphatic hydrocarbons were analyzed by capillary gas chromatography using an HP 5890A gas chromatograph equipped with a split/splitless injector and an FID. An SGE (Australia) OCI-5 cool on-column injector was also fitted in the gas chromatograph for quantification. Separation was achieved by an SE-30 capillary column (30 m  $\times$  0.25 mm i.d.). Oven temperature programming was: 45–90 $^\circ\text{C}$  at 15 $^\circ/\text{min}$ , 90–270 $^\circ\text{C}$  at 3 $^\circ/\text{min}$ , and 270 $^\circ\text{C}$  for 30 min. The detector was at 300 $^\circ\text{C}$ , and H<sub>2</sub> was used as the carrier gas. Identification was accomplished by coinjection with authentic standards; positive confirmation was obtained using an HP 5890 gas chromatograph coupled to an HP 5970B mass selective detector. Quantification was achieved by capillary GC using an electronic integrator (Chromatopac C-R6A, Shimadzu, Japan). The analytical precision of *n*-alkane, *n*-alkanol and sterol concentrations was estimated to be 2–8%.

### 2.4. Common names of sterols

The following are some common names (in parentheses) of sterols: cholesta-5,22E-dien-3 $\beta$ -ol (22-dehydrocholesterol), cholest-5-en-3 $\beta$ -ol (cholesterol), 5 $\alpha$ -cholestan-3 $\beta$ -ol (cholestanol), 5 $\alpha$ -cholestan-3 $\alpha$ -ol (epi-cholestanol), 5 $\beta$ -cholestan-3 $\beta$ -ol (coprostanol), 5 $\beta$ -cholestan-3 $\alpha$ -ol (epicoprostanol), 22,23-methylene-23,24-dimethyl-cholest-5-en-3 $\beta$ -ol (gorgosterol), 22,23-methylene-23,24-dimethyl-5 $\alpha$ -cholestan-3 $\beta$ -ol (gorgostanol), 24-methylcholesta-5,22E-dien-3 $\beta$ -ol (diatomsterol, brassicasterol), 24-methylcholesta-5,24(28)-dien-3 $\beta$ -ol (24-

Table 1  
Grain size distribution (in percent) of the continental shelf sediments off northeastern Taiwan

Station	< 0.063 mm ( < 230 mesh)	0.063–0.59 mm (230–30 mesh)	> 0.59 mm ( > 30 mesh)
46	0.77	52.9	46.3
47	0.03	40.6	59.3
48	0.17	68.3	31.5
49	0.21	50.1	49.7
50	0.11	50.1	49.8
51	0.06	99.94	–

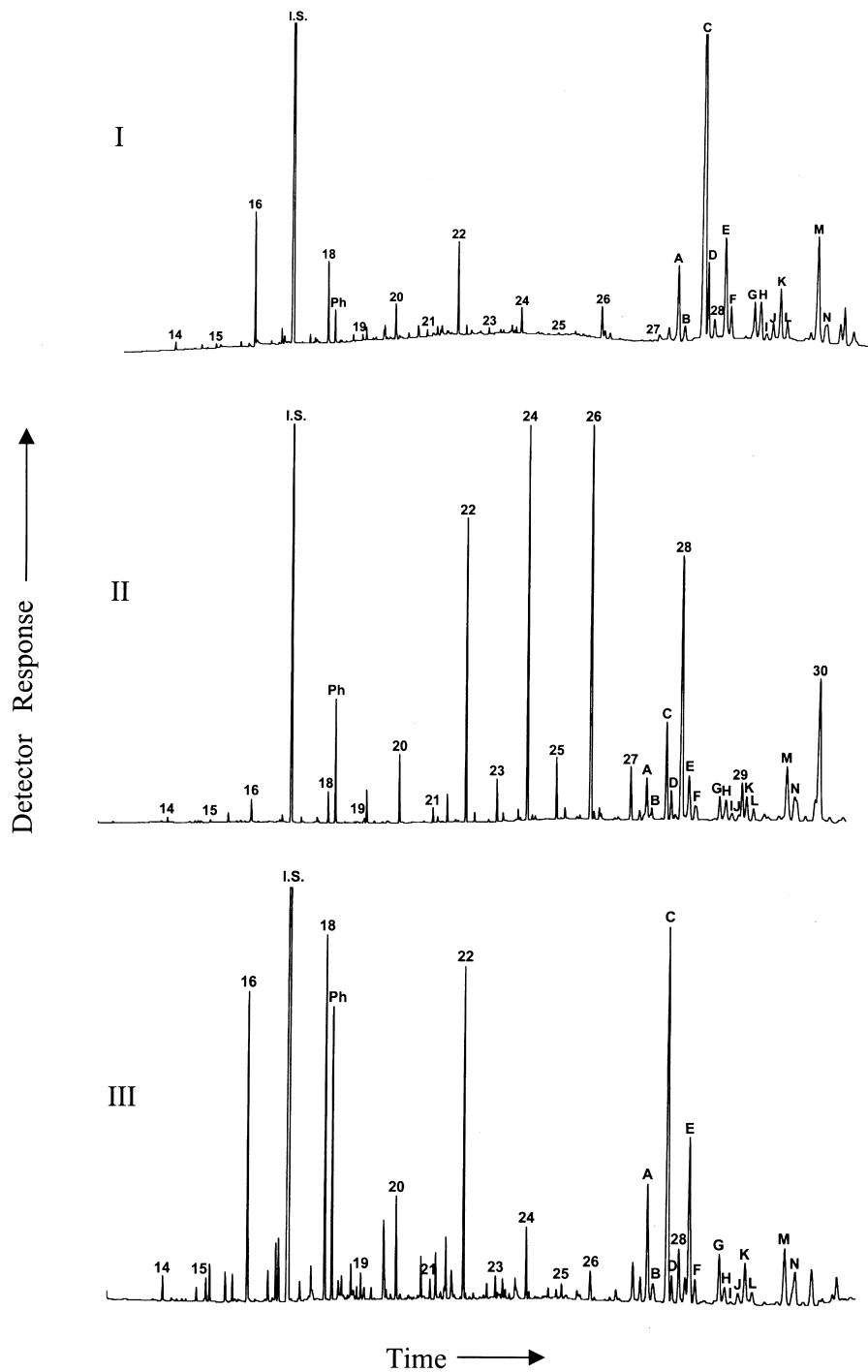


Fig. 2. Representative GC traces of the alkanol/sterol fraction for surface sediments from the shelf and slope and total suspended matter off northeastern Taiwan (I) shelf (Sta. 48), (II) slope (Sta. 4) and (III) total suspended matter (Sta. 22). Numbers above peaks refer to C number of *n*-alkanols, Ph = phytol, and IS = internal standard (*n*-C<sub>17</sub>OH). Letters above peaks refer to sterols: (A) cholesta-5,22E-dien-3 $\beta$ -ol (22-dehydrocholesterol); (B) 5 $\alpha$ -cholest-22E-en-3 $\beta$ -ol; (C) cholest-5-en-3 $\beta$ -ol (cholesterol); (D) 5 $\alpha$ -cholestan-3 $\beta$ -ol (cholestanol); (E) 24-methylcholesta-5,22E-dien-3 $\beta$ -ol (diatomsterol); (F) 24-methyl-5 $\alpha$ -cholest-22E-en-3 $\beta$ -ol; (G) 24-methylcholesta-5,24(28)-dien-3 $\beta$ -ol (24-methylenecholesterol); (H) 24-methylcholest-5-en-3 $\beta$ -ol (campesterol) + 24-methyl-5 $\alpha$ -cholest-24(28)-en-3 $\beta$ -ol(?); (I) 24-methyl-5 $\alpha$ -cholestan-3 $\beta$ -ol; (J) 23,24-dimethylcholesta-5,22E-dien-3 $\beta$ -ol; (K) 24-ethylcholesta-5,22E-dien-3 $\beta$ -ol (stigmasterol); (L) 24-ethyl-5 $\alpha$ -cholest-22E-en-3 $\beta$ -ol; (M) 24-ethylcholest-5-en-3 $\beta$ -ol ( $\beta$ -sitosterol); and (N) 24-ethyl-5 $\alpha$ -cholestan-3 $\beta$ -ol + unknown.

Table 2  
Sterol concentrations in coastal marine sediments on the shelf and slope and in total suspended matter of the water column off northeastern Taiwan

Sta.	1	2	3	4	5	6	7	8	9	10	11	12	13	14	Total	Ratio <sup>b</sup>
Shelf sediments (concentration in ng/g)																
45	386 (0.21)	156 (0.09)	1810 (1.00)	247 (0.14)	583 (0.32)	152 (0.08)	317 (0.18)	240 (0.13)	45 (0.02)	121 (0.07)	328 (0.18)	110 (0.06)	789 (0.44)	282 (0.16)	5570	1.33
46	327 (0.18)	150 (0.08)	1820 (1.00)	308 (0.17)	652 (0.35)	245 (0.13)	228 (0.13)	281 (0.15)	63 (0.03)	150 (0.08)	425 (0.23)	139 (0.08)	805 (0.44)	347 (0.19)	5920	1.42
47	3840 (0.19)	1060 (0.05)	20,500 (1.00)	2000 (0.10)	6400 (0.31)	1400 (0.07)	2200 (0.11)	1910 (0.09)	366 (0.02)	907 (0.04)	2360 (0.12)	955 (0.05)	5600 (0.27)	2270 (0.11)	51,770	1.05
48	850 (0.14)	234 (0.04)	6200 (1.00)	645 (0.10)	1480 (0.24)	365 (0.06)	587 (0.09)	609 (0.10)	92 (0.01)	237 (0.04)	710 (0.11)	231 (0.04)	1750 (0.28)	562 (0.09)	14,550	0.85
49	1080 (0.18)	379 (0.06)	6120 (1.00)	692 (0.11)	1830 (0.30)	438 (0.07)	596 (0.10)	627 (0.10)	102 (0.02)	269 (0.04)	805 (0.13)	315 (0.05)	1900 (0.31)	658 (0.11)	15,810	1.04
50	949 (0.14)	238 (0.03)	6910 (1.00)	721 (0.10)	1750 (0.25)	359 (0.05)	541 (0.08)	655 (0.09)	98 (0.01)	256 (0.04)	757 (0.11)	352 (0.05)	1870 (0.27)	390 (0.06)	15,850	0.81
51	954 (0.12)	93 (0.01)	8030 (1.00)	775 (0.10)	1240 (0.15)	192 (0.02)	409 (0.05)	361 (0.04)	43 (0.01)	157 (0.02)	441 (0.05)	159 (0.02)	1070 (0.13)	169 (0.02)	14,090	0.52
Avg.	4.8 (0.13)	1.7 (0.05)	35.8 (1.00)	8.6 (0.24)	13.0 (0.36)	4.9 (0.14)	2.8 (0.08)	6.5 (0.18)	2.9 (0.06)	3.6 (0.10)	14.3 (0.40)	4.7 (0.13)	21.8 (0.61)	12.1 (0.34)	138	1.63
46 <sup>c</sup>	8.6 (0.02)	2.8 (0.04)	68.1 (1.00)	11.8 (0.17)	22.8 (0.33)	9.7 (0.14)	6.7 (0.10)	9.0 (0.13)	2.7 (0.04)	3.4 (0.05)	13.4 (0.2)	4.7 (0.07)	28.1 (0.41)	14.4 (0.21)	206	1.17
Slope sediments (concentration in ng/g)																
1	187 (0.45)	104 (0.25)	420 (1.00)	146 (0.35)	266 (0.63)	144 (0.34)	123 (0.29)	193 (0.46)	58 (0.14)	74 (0.18)	232 (0.55)	111 (0.26)	488 (1.16)	287 (0.68)	2830	3.57
2	762 (0.41)	265 (0.14)	1880 (1.00)	323 (0.17)	903 (0.48)	316 (0.17)	851 (0.45)	519 (0.28)	132 (0.07)	224 (0.12)	578 (0.31)	248 (0.13)	1980 (1.05)	696 (0.37)	9680	2.51
3	102 (0.45)	44 (0.19)	229 (1.00)	71 (0.31)	135 (0.59)	59 (0.26)	63 (0.28)	75 (0.33)	31 (0.14)	108 (0.47)	74 (0.32)	42 (0.18)	244 (1.07)	138 (0.60)	1420	3.47
4	258 (0.50)	62 (0.12)	514 (1.00)	152 (0.30)	332 (0.65)	143 (0.28)	165 (0.32)	178 (0.35)	72 (0.14)	43 (0.08)	178 (0.35)	85 (0.17)	434 (0.84)	342 (0.67)	2960	3.23
5	122 (0.30)	61 (0.15)	408 (1.00)	103 (0.25)	168 (0.41)	97 (0.24)	99 (0.24)	109 (0.27)	56 (0.14)	28 (0.07)	106 (0.26)	50 (0.12)	268 (0.66)	211 (0.52)	1890	2.44
7	59 (0.47)	24 (0.19)	125 (1.00)	37 (0.30)	64 (0.51)	41 (0.33)	42 (0.34)	59 (0.47)	25 (0.20)	27 (0.22)	48 (0.38)	27 (0.22)	259 (2.07)	126 (1.01)	963	3.79
8	129 (0.51)	80 (0.32)	252 (1.00)	94 (0.37)	202 (0.80)	76 (0.30)	73 (0.29)	84 (0.33)	39 (0.15)	67 (0.27)	123 (0.49)	92 (0.37)	232 (0.92)	167 (0.63)	1710	4.01
Avg.	129 (0.51)	80 (0.32)	252 (1.00)	94 (0.37)	202 (0.80)	76 (0.30)	73 (0.29)	84 (0.33)	39 (0.15)	67 (0.27)	123 (0.49)	92 (0.37)	232 (0.92)	167 (0.63)	1710	3.29 ± 0.61
Suspended matter (concentration in µg/g)																
17	68.4 (0.33)	13.9 (0.07)	209.0 (1.00)	14.0 (0.07)	97.4 (0.47)	18.5 (0.09)	40.8 (0.20)	15.6 (0.07)	6.7 (0.03)	6.1 (0.03)	37.9 (0.18)	11.1 (0.05)	74.1 (0.35)	36.2 (0.17)	650	1.51
22	114.0 (0.28)	21.2 (0.05)	402.0 (1.00)	24.5 (0.06)	195 (0.49)	28.8 (0.07)	58.5 (0.15)	26.0 (0.06)	3.2 (0.01)	15.3 (0.04)	60.2 (0.15)	19.9 (0.05)	89.4 (0.22)	67.8 (0.17)	1130	1.37
27	123.0 (0.35)	24.5 (0.07)	355.0 (1.00)	35.6 (0.10)	203.0 (0.57)	39.9 (0.11)	60.2 (0.17)	31.2 (0.09)	6.4 (0.02)	12.6 (0.04)	50.8 (0.14)	22.3 (0.06)	80.8 (0.23)	92.3 (0.26)	1140	1.75
Avg.	123.0 (0.35)	24.5 (0.07)	355.0 (1.00)	35.6 (0.10)	203.0 (0.57)	39.9 (0.11)	60.2 (0.17)	31.2 (0.09)	6.4 (0.02)	12.6 (0.04)	50.8 (0.14)	22.3 (0.06)	80.8 (0.23)	92.3 (0.26)	1140	1.54 ± 0.19
Oyster ( <i>Isognomon legumen</i> , concentration in µg/g)																
Tai-tung	451 (0.32)	—	1480 (1.00)	24.3 (0.02)	419 (0.29)	9.8 (0.01)	260 (0.18)	303 (0.21)	—	14.1 (0.01)	170 (0.12)	10.1 (0.01)	75.7 (0.05)	20.5 (0.01)	3190	0.85

<sup>a</sup> Sterol identification: (1) cholesta-5,22E-dien-3β-ol (22-dehydrocholesterol); (2) 5α-cholest-22E-en-3β-ol (cholesterol); (3) cholest-5-en-3β-ol (cholestanol); (4) 5α-cholestan-3β-ol (cholestanol); (5) 24-methylcholesta-5,22E-dien-3β-ol (diatomsterol); (6) 24-methyl-5α-cholest-22E-en-3β-ol; (7) 24-methylcholesta-5,24(28)-dien-3β-ol (24-methylencholesterol); (8) 24-methylcholest-5-en-3β-ol (campesterol) + 24-methyl-5α-cholest-24(28)-en-3β-ol(?); (9) 24-methyl-5α-cholestan-3β-ol; (10) 23,24-dimethylcholesta-5,22E-dien-3β-ol; (11) 24-ethylcholesta-5,22E-dien-3β-ol (stigmasterol); (12) 24-ethyl-5α-cholest-22E-en-3β-ol; (13) 24-ethylcholest-5-en-3β-ol (β-sitosterol); and (14) 24-ethyl-5α-cholestan-3β-ol + unknown. Numbers in parentheses indicate each sterol concentration normalized to cholesterol concentration as 1.00.

<sup>b</sup> Ratio = Σ(algal sterols/cholesterol), where algal sterols = total sterols — (campesterol + stigmasterol + β-sitosterol).

<sup>c</sup> Sand fraction.

methylenecholesterol), 24-methylcholest-5-en-3 $\beta$ -ol (campesterol, or 22,23-dihydrobrassicasterol), 24-ethylcholesta-5,22E-dien-3 $\beta$ -ol (stigmasterol, or poriferasterol), and 24-ethylcholest-5-en-3 $\beta$ -ol ( $\beta$ -sitosterol, or clionasterol).

### 2.5. Total organic carbon (TOC)

Sediment samples were oven-dried in air for 48 h. Total organic C was determined by the dichromate-acid oxidation method (Gaudette et al., 1974) modified by addition of Ag<sub>2</sub>SO<sub>4</sub> to H<sub>2</sub>SO<sub>4</sub> at the rate of 15 g/l. Titration was carried out with an automatic titrator (Metrohm 702 SM Titrino, Switzerland). The relative standard deviation of TOC determination was generally < 1%.

### 3. Results

The GC traces of the alkanol/sterol fraction for representative shelf and slope sediment samples and suspended matter are shown in Fig. 2. In the shelf sediments, cholesterol is a dominant component, and

alkanols and sterols have about the same proportions. In comparison, cholesterol is not a predominant component in the slope sediments, and *n*-alkanols of higher molecular weight (> C<sub>20</sub>) outweigh those of lower molecular weight (for stations 3, 4, 5 and 7) and sterols. As for the suspended matter, it is dominated by cholesterol, phytol and normal C<sub>16</sub>, C<sub>18</sub> and C<sub>22</sub> alkanols.

In order to facilitate the following comparison of sterol composition, 3 common sterols in vascular plants (namely, campesterol, stigmasterol and  $\beta$ -sitosterol) were excluded. This is in view of the large variation of terrestrial inputs of organic matter to the study area, as shown by the predominance of  $\beta$ -sitosterol in the slope sediments (stations 1, 2, 3, and 7, Table 2). The concentration of each of the remaining sterols, termed algal sterols, was normalized to that of cholesterol and the ratios were summed up. The resulting  $\Sigma$ (algal sterols/cholesterol) ratios varied from 0.52 to 1.42 (average  $1.00 \pm 0.31$ ;  $n = 7$ ) for the shelf samples, and from 2.44 to 4.01 (average  $3.29 \pm 0.61$ ;  $n = 7$ ) for the slope samples. The ratios for the slope sediments are similar to those in coastal marine sediments on the continental slope off southwestern Taiwan (Jeng et al., 1997) and other areas around the

Table 3  
Summary of lipid data

Station	$\Sigma$ Sterols (ng/g)	LMW <sup>a</sup> <i>n</i> -alkanols (ng/g)	HMW <sup>b</sup> <i>n</i> -alkanols (ng/g)	Phytol (ng/g)	TOC (%)	$\Sigma$ Sterols/TOC ( $\mu$ g/gC)	CPI <sup>c</sup>
Shelf sediments							
45	5570	648	328	71	0.65	857	2.87
46	5920	980	452	85	0.63	940	2.29
47	51,770	3470	2690	600	0.91	5690	1.80
48	14,550	1130	1120	206	0.55	2650	1.88
49	15,810	993	571	191	0.67	2360	1.40
50	15,850	772	394	106	0.88	1800	1.79
51	14,090	870	469	44	–	–	1.32
Average	17,650 $\pm$ 15,690	1266 $\pm$ 984	861 $\pm$ 848	186 $\pm$ 192	0.70 $\pm$ 0.15	2380 $\pm$ 1780	1.91 $\pm$ 0.53
Slope sediments							
1	2830	315	2180	472	0.66	429	2.87
2	9680	358	1830	806	1.05	922	2.36
3	1420	260	2850	162	0.45	316	3.91
4	2960	297	4880	291	0.74	400	5.34
5	1890	221	2260	320	0.54	350	4.71
7	963	155	2320	49	0.62	150	4.47
8	1710	102	783	36	0.29	590	3.64
Average	3065 $\pm$ 3005	244 $\pm$ 91	2443 $\pm$ 1249	305 $\pm$ 270	0.62 $\pm$ 0.24	452 $\pm$ 245	3.90 $\pm$ 1.05
Suspended matter (concentration in $\mu$ g/g)							
17	650	114.8	116.2	58.6	–	–	–
22	1130	373.7	286.5	134.3	–	–	–
27	1140	187.6	90.0	86.0	–	–	–
Average	973 $\pm$ 280	225 $\pm$ 134	164 $\pm$ 107	93.0 $\pm$ 38.3	–	–	–

<sup>a</sup> LMW *n*-alkanols =  $\Sigma$  C<sub>14</sub>, C<sub>16</sub>, C<sub>18</sub>, C<sub>20</sub>.

<sup>b</sup> HMW *n*-alkanols =  $\Sigma$  C<sub>20</sub>, C<sub>22</sub>, C<sub>24</sub>, C<sub>28</sub>.

<sup>c</sup> CPI =  $\frac{1}{2} \left[ \frac{C_{25}+C_{27}+C_{29}+C_{31}}{C_{24}+C_{26}+C_{28}+C_{30}} + \frac{C_{25}+C_{27}+C_{29}+C_{31}}{C_{26}+C_{28}+C_{30}+C_{32}} \right]$ .

world (Harvey, 1994; Mudge and Norris, 1997). However, the relatively low ratios for the shelf sediments are rather unusual as compared to common coastal marine sediments.

The CPI values of the *n*-alkanes (Table 3) varied between 1.32 and 2.87 (average  $1.91 \pm 0.53$ ) for the shelf sediments, and between 2.36 and 5.34 (average  $3.90 \pm 1.05$ ) for the slope sediments. The former is similar to the CPI value of 1.80 in southern Taiwan Strait sediments with fossil hydrocarbon contamination (Jeng, 1986), whereas the latter is among the highest for coastal marine sediments surrounding Taiwan. The shelf and slope sediments have higher plant alkane (HPA) indices (Table 4) varying over a wide range. The corresponding stanol/stenol ratios in the 3 sets of samples follow the order: suspended matter < shelf sediments < slope sediments (Table 5).

## 4. Discussion

### 4.1. Aliphatic hydrocarbons

The difference shown in aliphatic hydrocarbon GC traces (Fig. 3) suggests that lipids in the shelf sediments are weathered (Clayton and Swetland, 1978; Reed, 1977). By contrast, lipids in the slope sediments are associated with more recently deposited material (or unweathered) (Han et al., 1968; Reed, 1977; Venkatesan and Kaplan, 1982). The *n*-alkanes of the shelf sediments (average CPI=1.91) suggest weathering or relatively smaller contributions of higher plant waxes (Eglinton and Hamilton, 1967). An unresolved complex mixture (UCM) can be noted in the shelf sediments. Considering that no major oil spill or ship wreck occurred in this area over the past two decades, the UCM is most likely the result of weathering

(Bjørøy et al., 1979; Clayton and Swetland, 1978). By contrast, the slope sediments show an *n*-alkane profile indicating more input of higher plant waxes (average CPI=3.90). However, comparatively lower CPI values are found at stations 1 and 2 (2.87 and 2.36, respectively) that can be attributed to some eroded material from the nearby Lanyang River. A recent report shows that modern C in the POC discharged from the main channel of the Lanyang River was <30% (Kao and Liu, 1996).

### 4.2. Distribution of sterols

It can be seen that the  $\Sigma$ (algal sterols/cholesterol) ratios of the shelf sediments decrease roughly toward the east (from stations 45 to 51). The CPI values of *n*-alkanes also show the same trend, with higher values at stations 45 and 46 and lower values at stations 49 and 51. This may result from input of re-suspended recent sediments as a source of cholesterol from the East China Sea shelf (Milliman et al., 1985), Taiwan Strait, or small islands just located to the south of stations 45 and 46.

In the slope sediments (samples from stations 1, 2, 3 and 7), concentrations of  $\beta$ -sitosterol are higher than those of cholesterol. Since  $\beta$ -sitosterol is generally considered to be derived from higher plants (Huang and Meinschein, 1976; Meyers, 1997) and possibly algae as well (Volkman, 1986), it may be attributed to riverine input from eastern Taiwan followed by Kuroshio' northward transport. Also worthwhile noting is that stations 2 and 47 have much higher sterols than the adjacent stations (Fig. 4) which can be tied to high phytoplankton chlorophyll *a* in the upper water column (ca. top 50 m) near these two sites (Chen Lee, 1992).

The organic C normalized total sterol concentrations

Table 4  
Higher plant alkane (HPA) indices of shelf and slope sediments

Shelf sediments	Sta. 45	Sta. 46	Sta. 47	Sta. 48	Sta. 49	Sta. 50	Sta 51	Average
HPA <sup>a</sup>	0.209 (-0.44)	0.186 (-0.60)	0.492 (1.60)	0.451 (1.30)	0.193 (-0.55)	0.201 (-0.50)	0.158 (-0.81)	0.270 $\pm$ 0.139
Water depth (m)	126	105	163	108	107	110	99	
Slope sediments	Sta. 1	Sta. 2	Sta. 3	Sta. 4	Sta. 5	Sta. 7	Sta. 8	Average
HPA <sup>a</sup>	0.558 (-0.20)	0.469 (-1.55)	0.559 (-0.18)	0.638 (1.02)	0.642 (1.08)	0.513 (-0.88)	0.621 (0.76)	0.571 $\pm$ 0.066
Water depth (m)	1608	1380	726	451	558	238	830	

<sup>a</sup> HPA is the ratio of concentration of  $\Sigma C_{24}$ ,  $C_{26}$ ,  $C_{28}$  *n*-alkanols to that of  $\Sigma C_{24}$ ,  $C_{26}$ ,  $C_{28}$  *n*-alkanols plus  $\Sigma C_{27}$ ,  $C_{29}$ ,  $C_{31}$  *n*-alkanes. Numbers in parentheses are *z* score.

in the slope area (Table 3) show the lowest value at station 7 and higher values at stations 2 and 8. The distribution suggests that direct deposition from the overlying water column is probably less important than down-slope transport as the source of sterols. Stations 2 and 8 are situated at the base of Mien-hua Canyon (not shown on Fig. 1), which serves as an effective conduit for the transport of re-suspended matter from the shelf (Biscaye and Anderson, 1994; Monaco et al., 1990). On the other hand, the sediment from station 7 is poorly sorted and hence unfavorable for the accumulation of high levels of lipids.

#### 4.3. Sterol sources of the shelf sediments

Cholesterol is an important sterol in animals (Tissot and Welte, 1978). It is also widespread in plants (Volkman, 1986) and usually the most abundant sterol in coastal marine sediments (Harvey, 1994; Mudge and Norris, 1997). Jeng and Chen (1995) pointed out that

grain size effect (i.e., enrichment in fine sediments) is more important for predominant sterols than for less abundant ones. Since the present data for the shelf sediments are based on the mud fraction, one may question if the results are biased. To address this question, the sterols in the sand fraction (30–230 mesh) of samples 46 and 49 were also analyzed, resulting in  $\Sigma(\text{algal sterols/cholesterol})$  ratios of 1.63 and 1.17, respectively (Table 2). These compare favorably with the same ratios in the mud fraction for samples 46 and 49 (1.42 and 1.04, respectively; Table 2), suggesting that the grain size effect plays a minor role in this case.

The  $\Sigma(\text{algal sterols/cholesterol})$  ratios for the shelf sediments and slope sediments are obviously different. The significantly lower  $\Sigma(\text{algal sterols/cholesterol})$  ratios for the shelf sediments ( $1.00 \pm 0.31$ ) are different from common shelf sediments. The cause for this unusual value may lie in the sources of sterols. As mentioned earlier, shelf sediments analyzed were isolated from shell fragments and debris, where cholesterol is

Table 5  
The stanol/stenol ratios of 3 sterol pairs for suspended matter, shelf and slope sediments off northeastern Taiwan

Shelf sediments stanol/stenol <sup>a</sup>	Sta. 45	Sta. 46	Sta. 47	Sta. 48	Sta. 49	Sta. 50	Sta. 51	mean $\pm$ s.d.
2/1	0.40 (0.43)	0.46 (0.86)	0.28 (-0.43)	0.28 (-0.43)	0.35 (0.07)	0.25 (-0.64)	0.10 (-1.71)	0.30 $\pm$ 0.12 (-0.26 $\pm$ 0.83)
4/3	0.14 (-0.36)	0.17 (-0.09)	0.10 (-0.73)	0.10 (-0.73)	0.11 (-0.64)	0.10 (-0.73)	0.10 (-0.73)	0.12 $\pm$ 0.03 (-0.57 $\pm$ 0.25)
6/5	0.26 (-0.47)	0.39 (0.40)	0.22 (-0.73)	0.25 (0.53)	0.24 (-0.60)	0.21 (-0.80)	0.15 (-1.20)	0.25 $\pm$ 0.07 (-0.56 $\pm$ 0.49)
Slope sediments stanol/stenol <sup>a</sup>	Sta. 1	Sta. 2	Sta. 3	Sta. 4	Sta. 5	Sta. 7	Sta. 8	mean $\pm$ s.d.
2/1	0.56 (1.57)	0.35 (0.07)	0.43 (0.64)	0.24 (-0.71)	0.50 (1.14)	0.41 (0.50)	0.62 (2.00)	0.44 $\pm$ 0.13 (0.74 $\pm$ 0.92)
4/3	0.35 (1.55)	0.17 (-0.09)	0.31 (1.18)	0.30 (1.09)	0.25 (0.64)	0.30 (1.09)	0.37 (1.73)	0.29 $\pm$ 0.07 (1.03 $\pm$ 0.61)
6/5	0.54 (1.40)	0.35 (0.13)	0.44 (0.73)	0.43 (0.67)	0.58 (1.67)	0.64 (2.07)	0.38 (0.33)	0.48 $\pm$ 0.11 (1.00 $\pm$ 0.72)
Suspended matter stanol/stenol <sup>a</sup>	Sta. 17	Sta. 22	Sta. 27	mean $\pm$ s.d.				
2/1	0.20 (-1.00)	0.19 (-1.07)	0.20 (-1.00)	0.20 $\pm$ 0.01 (-1.02 $\pm$ 0.04)				
4/3	0.07 (-1.00)	0.06 (-1.09)	0.10 (-0.73)	0.08 $\pm$ 0.02 (-0.94 $\pm$ 0.19)				
6/5	0.19 (-0.93)	0.15 (-1.20)	0.20 (-0.87)	0.18 $\pm$ 0.03 (-1.00 $\pm$ 0.18)				

<sup>a</sup> Sterol identification: (1) cholesta-5,22E-dien-3 $\beta$ -ol (22-dehydrocholesterol); (2) 5 $\alpha$ -cholest-22E-en-3 $\beta$ -ol; (3) cholest-5-en-3 $\beta$ -ol (cholesterol); (4) 5 $\alpha$ -cholestan-3 $\beta$ -ol (cholestanol); (5) 24-methylcholesta-5,22E-dien-3 $\beta$ -ol (diatomsterol); (6) 24-methyl-5 $\alpha$ -cholest-22E-en-3 $\beta$ -ol. Numbers in parentheses are *z* score. The mean of the sample of cores and sample standard deviation ( $n = 17$ ) for sterol 2/sterol 1 are 0.34 and 0.14, those for sterol 4/sterol 3 are 0.18 and 0.11, and those for sterol 6/sterol 5 are 0.33 and 0.15, respectively.



usually the predominant sterol of most mollusks and other shell-containing animals (Ballantine et al., 1983; and references therein). For instance, a wild oyster sample, taken from the unpolluted rocky coast of southeast Taiwan was also analyzed, which gave a  $\Sigma(\text{algal sterols/cholesterol})$  ratio of 0.85 (Table 2). The cholesterol and other sterols in the shelf sediments are most likely the remaining cholesterol and other sterols of the mollusks (rather than sediments) from the last glaciation (11.8 ka ago).

Sterols in the shelf sediments can also be derived from near-bottom transport or from the water column. As mentioned earlier, the shelf sediments have significantly lower  $\Sigma(\text{algal sterols/cholesterol})$  ratios (average

1.00); therefore, sediment transport from the surrounding area like the slope with significantly higher  $\Sigma(\text{algal sterols/cholesterol})$  ratios (average 3.29) is unlikely. Similarly, the average CPI of *n*-alkanes for the shelf sediments (1.91) is about one half of that for the slope sediments (3.90). So, sediments that move from the slope up to the shelf by re-suspension followed by the intrusion of the Kuroshio onto the shelf in the winter and spring time (Chern and Wang, 1992; Hsueh et al., 1992) cannot be the major source and pathway of sterols in the shelf area. Additional input of sterols from the water column to the shelf is required.

Cholesterol is known to have the largest decay constant (e.g., Sun and Wakeham, 1994) among 4 sterols,

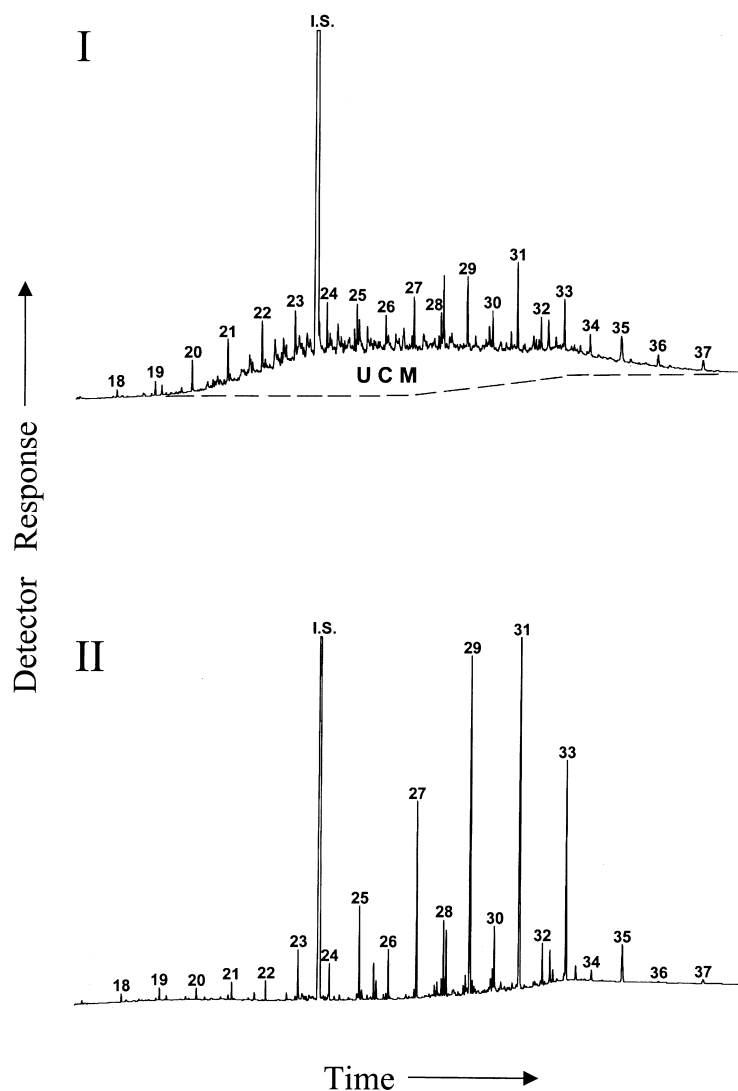


Fig. 3. Representative GC traces of the aliphatic fraction for surface sediments from the shelf and slope off northeastern Taiwan. (I) Shelf (Sta. 49) and (II) Slope (Sta. 4). Numbers above peaks refer to carbon number of *n*-alkanes, IS=internal standard (*n*-C<sub>24</sub>D<sub>50</sub>).

so the proportion of cholesterol relative to other sterols is expected to decrease with time after burial. However, we have observed higher proportions of cholesterol in the older shelf sediments than in the younger slope sediments. This seems to be at odds with conventional wisdom and suggests that residual sterols derived from mollusks should not be the only source of sterols in the shelf sediments. The sterol composition of suspended matter in seawater needs to be examined. Samples of total suspended matter from 1 m below the sea surface at 3 shelf stations were collected and analyzed. Their  $\Sigma$ (algal sterols/cholesterol) ratios (1.51, 1.37, and 1.75; Table 2) are substantially closer to the average ratio of shelf sediments (1.00) than that of slope sediments (3.29). So, settling of suspended particles from the water column is a more

probable source for sterols in the shelf sediments. Sterols may have been added to the shelf sediments, mainly in the mud fraction, from the water column even though there is no long-term, apparent sedimentation for the relict sediments. Sterol input (accompanied by sedimented suspended matter) to the shelf sediments in very trace amounts appears most probable and reasonable. In this case, sterol addition and degradation (or possibly output), although the rate of addition may be extremely slow, are in a dynamic state. One may ask what the contribution of suspended matter brought to the shelf area during river (e.g. Langyang River) floods is. The contribution is probably minimal. The reason is that the main stream and branch of the Kuroshio Current may confine terrigenous particles to the inshore area and may

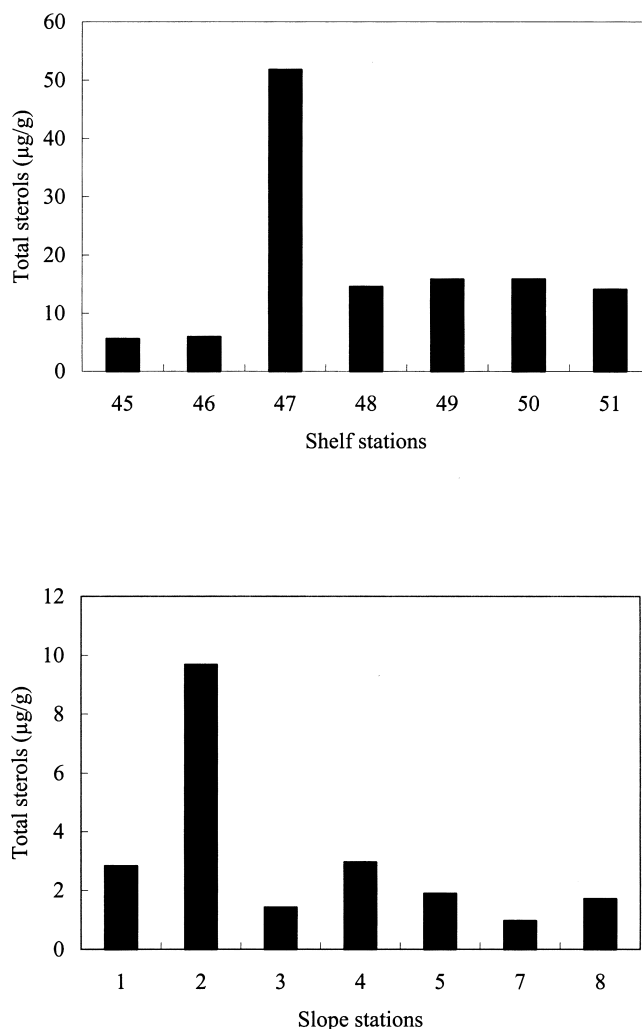


Fig. 4. Histograms of total sterol concentrations in the shelf and slope sediments.

act as a barrier to the offshore transport of terrigenous particles to regions farther away from the coast, toward the north and east (Hsu et al., 1998).

#### 4.4. Evidence of recent lipid inputs to the shelf sediments

The presence of extractable phytol in the shelf sediments points to recent inputs of lipids. Compared to *n*-alkanols, phytol is relatively unstable. For instance, in a sediment core (sedimentation rate 0.33 cm/a) from off southwestern Taiwan, extractable phytol decreases abruptly from 1049 ng/g (0–4 cm) to 375 ng/g (4–8 cm), but bound phytol increases sharply from 43 to 58%. This might suggest that part of the extractable phytol was transformed into the bound form (Jeng et al., 1997). In a field study, Sun et al. (1998) demonstrated that solvent-extractable phytol was rapidly converted into highly-bound phytol (in sediment matrix) after deposition. Because of its rapid transformation, the presence of extractable phytol in the shelf sediments (range from 44 to 600 ng/g) is an indication of recent inputs of phytol (and other lipids — sterols) to the sediments. In other words, if there were no phytol input from the surroundings to the shelf sediments (i.e. closed system) in the recent past, there would not be any extractable phytol left at present.

Higher plant waxes tend to contain mainly longer chain (>C<sub>22</sub>) saturated alcohols (Cranwell and Volkman, 1981; Eglinton and Hamilton, 1967). By contrast, the dominance of even-chain *n*-alkanols in the range C<sub>14</sub>–C<sub>20</sub> is indicative of marine sources, probably derived from phytoplankton and bacteria (Simoneit, 1978; Smith et al., 1983). In what follows, the *n*-alkanols are divided into two groups — C<sub>14</sub>, C<sub>16</sub>, C<sub>18</sub>, and C<sub>20</sub> as the lower-molecular-weight (LMW) fraction and C<sub>22</sub>, C<sub>24</sub>, C<sub>26</sub>, and C<sub>28</sub> as the higher-molecular-weight (HMW) fraction. In the shelf sediments, concentrations of LMW *n*-alkanols are somewhat higher than those of HMW *n*-alkanols (1266 vs 861 ng/g on average; see Table 3). The distribution is clearly reversed for the slope sediments (244 vs 2443 ng/g). As for suspended matter, concentrations of the LMW and HMW fractions are equally high (225 vs 164 μg/g) and the proportion is very close to that in the shelf sediments. Thus, it appears that the water column as a source of *n*-alkanols (and other lipids — sterols) in bottom sediments is more important in the shelf region than in the slope region.

#### 4.5. An estimate of the relict material in the shelf sediments

Based on the C preference index (CPI) of *n*-alkanes, it is possible to estimate the relative contribution of relict material and higher plants in the mud fraction of the shelf sediments. The average CPI value for the

shelf sediments is 1.91, and those for relict material and higher plant waxes are assumed to be 1 and 7 (Hedges and Prahl, 1993), respectively. Using a two-end member mixing model, it can be written that

$$1X + 7(1 - X) = 1.91$$

where  $X$  is the fraction of relict material and  $(1-X)$  is the fraction of higher plant waxes in the shelf samples. It then follows that relict material contributes approximately 85% to the mud fraction analyzed, with the remaining 15% derived from higher plant waxes through the water column. From these percentage values, the  $\Sigma(\text{algal sterols/cholesterol})$  ratio of the relict material can be estimated. As given earlier, the  $\Sigma(\text{algal sterols/cholesterol})$  ratios of shelf sediments and the suspended matter are 1.00 and 1.54, respectively. The  $\Sigma(\text{algal sterols/cholesterol})$  ratio of the relict material,  $Y$ , could be estimated from the equation

$$0.85Y + 0.15 \times 1.54 = 1.00.$$

The value of  $Y$  is 0.90.

#### 4.6. The higher plant alkane (HPA) index

Sedimentary *n*-alkanes are more resistant to degradation than *n*-alkanols. The higher plant alkane (HPA) index is defined as

$$\text{HPA} = [n\text{-alkanol}] / ([n\text{-alkanol}] + [n\text{-alkane}])$$

and has been used as a degradation signal to estimate the extent of degradation of geolipids (Poynter and Eglinton, 1990, 1991; Westerhausen et al., 1993). The assumption of using the HPA index is that the ratio of primary fluxes of long-chain *n*-alkanes and *n*-alkanols is approximately constant over the total study area. In the present study, the HPA index is used to indicate if the input ratio of *n*-alkanols to *n*-alkanes is approximately constant for each area. In the shelf area, water depths for the 7 stations vary within a small range (99–163 m). So, the difference of lipid degradation during sedimentation through the water column is minimal. It is seen from Table 4 that the two extremes of HPA indices (0.492 vs 0.158) for the shelf sediments differ by a factor of about 3, indicating that the input ratio of *n*-alkanols to *n*-alkanes changes over the shelf. For sediments of varying water depths in an area, the HPA index for the surface sediment has been shown to fall as water depth increases (Poynter and Eglinton, 1991; Westerhausen et al., 1993). For the slope area, there is no correlation between HPA index and water depth, implying that the input ratio of *n*-alkanols to *n*-alkanes is not constant over the slope, such as an extra terrestrial input from the Langyang River.

#### 4.7. Comparison of stanol/stenol ratios of the shelf and slope sediments

The stanol/stenol ratios of 3 sterol pairs for the 3 sets of samples in the study area are listed in Table 5. The data show that suspended matter, which probably represents the ratios of stanol-containing marine plankton (Volkman, 1986), has the lowest stanol/stenol ratios because the conversion of stenol to stanol by bacteria is expected to be slow in the oxic surface waters. Similarly, lower stanol/stenol ratios of surface water compared to those of surface sediments from the Black Sea were reported by Gagosian et al. (1979). The high stanol/stenol ratios of the shelf sediments may be caused by selective degradation of stenols relative to stanols at the sediment/water interface (Nishimura and Koyama, 1977). The slope area is mostly anoxic, as indicated by the presence of plenty of mottled and filamented authigenic pyrite (Chen et al., 1995). Therefore, relatively higher stanol/stenol ratios for the slope sediments (Table 5) are probably due to greater input of terrestrial material and microbiological hydrogenation of stenols (Edmunds et al., 1980; Taylor et al., 1981). The present result is similar to that reported by Gagosian et al. (1980), who showed that the lowest stanol/stenol ratios were found in the oxic sediments of the western North Atlantic, and the highest ratios were found in the anoxic sediments of the Black Sea and Walvis Bay. As mentioned earlier, the shelf sediments are older than the slope sediments. The stanol/stenol ratios are lower for the older shelf sediments than for the younger slope sediments. This seems to be inconsistent with the trend in sediment cores in which downcore increase of the stanol/stenol ratio is usually observed (Nishimura and Koyama, 1976). The reason for this is that their respective physicochemical conditions are totally different. The shelf sediments are in an oxic environment and an open system, as discussed earlier. However, sediment cores generally are in relatively less oxic environments (except the top few cm), and material exchange with the sediment is, to some degree, limited once it is buried in the sediment column. The lower stanol/stenol ratios in the shelf sediments can be attributed to (1) the oxic environment at shallow water depths — which is unfavorable for bacteria-mediated transformation of stenol into stanol and (2) sterol input from suspended matter with the lowest stanol/stenol ratios.

In the present study, the term “degradation” is used for discussion. Degradation is defined as the decrease of compound concentration by transformation into other molecules (such as conversion of stenols to stanones to sterenes), decomposition into smaller molecules, or incorporation into high molecular weight components, all of which would remove extractable sterols from the sediment.

## 5. Conclusion

In summary, the significantly lower  $\Sigma$ (algal sterols/cholesterol) ratios for the shelf sediments are unusual as compared to common coastal marine sediments. The major source for sterols in the shelf sediments is most likely mollusks from the last glaciation. Suspended matter in the water column probably is another sterol source for the shelf sediments although there is no apparent sedimentation to verify it. However, evidence of recent lipid inputs to the shelf sediments can be provided by the presence of extractable phytol and the comparison of *n*-alkanol distributions among the shelf and slope sediments and suspended matter. Further, sterol addition in trace amounts from the water column to the shelf is likely to occur, and sterol input and degradation on the shelf are probably in a dynamic state. Lipids in the shelf sediments are relatively more degraded than lipids in the slope sediments; this may indicate that lipid input rates are slower in the shelf area than in the slope area. The input ratios of long-chain *n*-alkanols to *n*-alkanes (HPA index) to the shelf and slope vary in respective areas. The shelf sediments have lower stanol/stenol ratios than the slope sediments because of the oxic shelf environment and the input of the lowest ratios of suspended matter to the shelf.

## Acknowledgements

We are grateful to the captain, crew and technicians of *R/V Ocean Researcher I* for their assistance with sample collection. Our thanks go to Prof B.R.T. Simoneit and two anonymous reviewers for their helpful comments. This study was financially supported by National Science Council grant NSC87-2611-M-002A-004-K2.

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