

PALEO-ENVIRONMENT STUDY AT YIHJU, SOUTHWESTERN TAIWAN: A CASE STUDY ON GEOCHEMICAL ANALYSIS OF SULFUR AND CARBON

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ABSTRACT

A systemic determination of abundance and isotopic composition of sulfur and carbon allowed us to reconstruct the paleo-environment of the southwestern coastal plain of Taiwan over the past one hundred thousand years. High sulfur content, S/C ratio and $\delta^{34}\text{S}$ values at two intervals of the studied core show that the study area has undergone two transgressional events. Low concentrations of total organic carbon were generally observed except for a short period of a marsh environment. Heavier $\delta^{13}\text{C}$ values of organic carbon indicate that C4 plant was a relatively important source of organic matters as a result of arid depositional environment during the regression.

Key words: paleo-environment, organic matter, $\delta^{13}\text{C}$, $\delta^{34}\text{S}$, transgressional event

INTRODUCTION

A coastal area is a transitional zone between two totally different environments: the land and sea. Due to their physio-chemical differences, biogeochemical processes and associated products are significantly discernible. A coastal area is further quite sensitive to climate, sea level, and sedimentary source change. Thus, sedimentary strata may record the history of environmental change. Abundance of sulfur and carbon as well as their isotopes has been used as proxies to demonstrate such changes (e.g., Peterson & Howarth, 1987; Berner, 1982). Sedimentary sulfur normally appears in the form of pyrite but also of monosulfide, elemental sulfur, sulfate compound and organic sulfur. Diagenesis and mineral transformation are mainly controlled by the prevailing biogeochemical process, sulfate reduction, in marine sediments (Chambers & Trudinger, 1979; Goldhaber & Kaplan, 1980; Fry *et al.*, 1986). Coastal

environments usually have sufficient sulfate. Active sulfate reduction is found in anoxic sediments where there is abundant sulfate and organic matters. Sulfate reduction bacteria use sulfate as an electron donor to oxidize organic carbon and produce hydrogen sulfide, which subsequently react with iron to become pyrite (Berner, 1970, 1984). This biogeochemical process prefers ^{32}S ; thus the product (mainly pyrite) has a significant light isotope value (Harrison, 1957, 1958; Lyn *et al.*, 1975). On the other hand, organic sulfur may be transformed from the dissolved hydrogen sulfide or directly disintegrated from plant debris. Therefore, both inorganic and organic sulfurs could provide valuable information regarding the degree of sulfate reduction and its influence on the sediment record (e.g., Summons, 1993).

Organic carbon retains the syn-depositional signals for source identification (Arthur *et al.*, 1988; Muller & Voss, 1999). Organic carbon preserved in coastal sediments may be derived from two major sources: fragments of terrestrial plants and marine organisms with different isotopic values (Deines, 1980). The terrestrial C3 and C4 plants, through different photosynthesis processes, show different $\delta^{13}\text{C}$ values: C3: $-26\sim-28\%$; C4: $-12\sim-14\%$ (Deines, 1980; O'Leary, 1981). In general, C3 plant grows under a humid condition, while C4 plant grows at a relatively arid climatic condition. The $\delta^{13}\text{C}$ values of organic matters in modern marine sediments have reportedly a wide range (Deines, 1980); however, Hsieh and Chen (1999) pointed out that the modern values are relatively constant in a local area. Therefore, the $\delta^{13}\text{C}$ signature could be used not only as an indicator of plant populations but also a good proxy of the history of sea-level change in the coastal environment.

Since carbon and sulfur cycles are coupled to each other, the ratio of sedimentary organic carbon to pyrite (C/S ratio) was used as a proxy to distinguish deposits formed in submarine or subaerial environments (Berner & Raiswell, 1983; 1984). Typical modern marine sediments have an average C/S ratio of 2.8 ± 0.8 (Berner, 1982), whereas the freshwater sediments have relatively higher C/S ratio due to its low sulfur content. However, its use as an indicator of deposition environment was limited when iron becomes limited (Raiswell & Berner, 1985; Calvert & Karlin, 1991; Lyons & Berner, 1992). A complete sulfidation of Fe may limit the sulfide content in Fe-poor rocks. In addition, unreasonable C/S values have been reported in freshwater sediments with almost no sulfur content. In this paper we use S/C ratio instead of C/S ratio to avoid such a problem. The boundary S/C ratio adopted in this study to identify the depositional environments is 0.36 ± 0.10 (Fig. 5; Berner, 1982).

The studied core is drilled at Yihju, a small town in the coastal plain of southwestern Taiwan, geologically speaking, a foreland basin (Fig. 1; Hsu, 1984). The core length is about 200m recording at least 50kyr sedimentary history. With an attempt to unravel detailed sedimentary history, measurement of concentrations and isotopic values of both sulfur and carbon were carried out. Based on published radiocarbon ages, the Holocene transgression here started at about 10ka and ended at 6.5ka. This transgressive tract consists of intercalated silt, mud, sand, and also contains fossils of foraminifers and mollusks, indicating that it was formed in a coastal environment (Lin *et al.*, 1998; Liu and Hsia, 1998). In an attempt to diagnose the geochemical records of the transgression, organic and inorganic sulfur concentration and their $\delta^{34}\text{S}$ values were analyzed. The $\delta^{13}\text{C}$ of the organic material in the studied strata was detected in order to understand the sources of these organic matters, and to reconstruct the paleoclimatic condition.

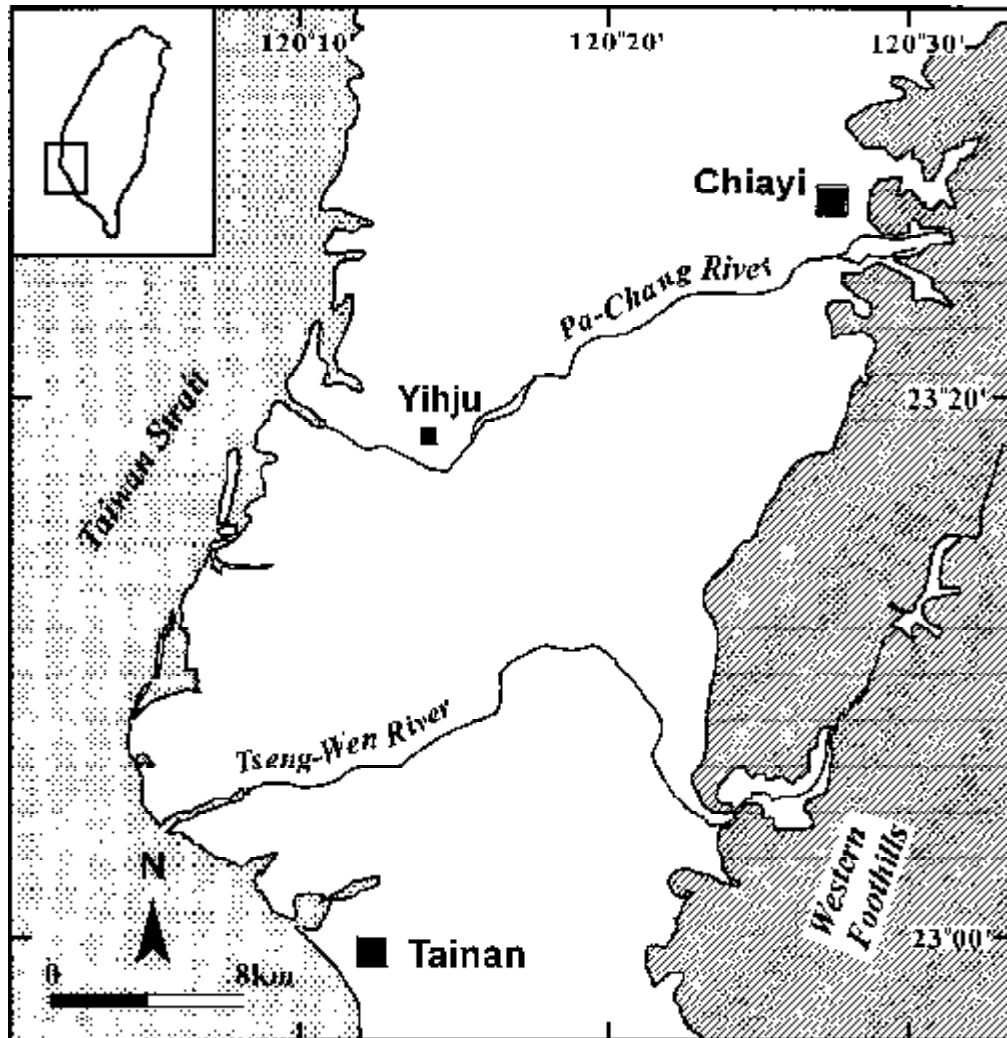


Figure 1. Map showing the study area and location of Yihju core in southwestern coastal plain, Taiwan. The area is bounded by western Foothills (right shaded area) and Taiwan Strait.

The sampling interval is every 10m. Sediments were separated into two aliquots with adequate amounts for both sulfur and carbon analyses. For sulfur analysis, the diffusion method (Hsieh & Yang, 1989; Hsieh & Shieh, 1997) was applied to extract the inorganic sulfur of Pyrite-S, AVS and elemental S concentrations in sediments. The concentrations of these inorganic sulfur species were all derived by titration method. For the sulfur isotopic analyses, these inorganic sulfur were sequentially transformed into Ag_2S , later reoxidized into SO_2 in a vacuum system (Fritz *et al.*, 1974; Yanagisawa & Sakai, 1983; Ueda & Krouse, 1986). The residual sediments were mixed with Eschka compound and fused in high temperature about 570°C . Organic sulfur, then, will be extracted by rinsing the mixture with hot water, and finally it will be precipitated as BaSO_4 and transformed into SO_2 in a vacuum system. The errors for

concentrations of inorganic and organic sulfur are $\pm 5\%$ and $\pm 10\%$ respectively. On the other hand, TOC was first measured on EA1110 elemental analyzer with the aliquot for carbon analysis. The error of TOC measurement was $\pm 2\%$. For $\delta^{13}\text{C}$ analysis, samples were first treated with 2N HCl to remove the inorganic carbon. The organic carbon is transformed into CO_2 gas via combustion in a vacuum system. The C and S isotopes were analyzed on Delta Plus and Nuclides mass spectrometers. Results are expressed in per mil (‰) as $\delta^{13}\text{C}$ relative to Pee Dee Belemnite (PDB), and $\delta^{34}\text{S}$ relative to troilite sulfur from the Canyon Diablo meteorite (CDT). The errors for carbon and sulfur isotope values are $\pm 0.1\text{‰}$ and $\pm 0.3\text{‰}$.

RESULTS

Inorganic Sulfur

As shown in Table 1 and Figure 2a, the concentrations of acid-volatile sulfur (AVS) and elemental sulfur (ES) are both much lower than chromium (II) reducible sulfur (i.e., Cr-S, mostly pyritic sulfur) throughout the entire core. The concentrations of Cr-S vary with depth as well as their $\delta^{34}\text{S}$ values. At depths of less than 100m and between 180m and 200m, high concentrations of Cr-S, associated with lighter values of $\delta^{34}\text{S}$ were observed, indicating that these sediments might be deposited under a marine environment during the last transgression (Fig. 2b).

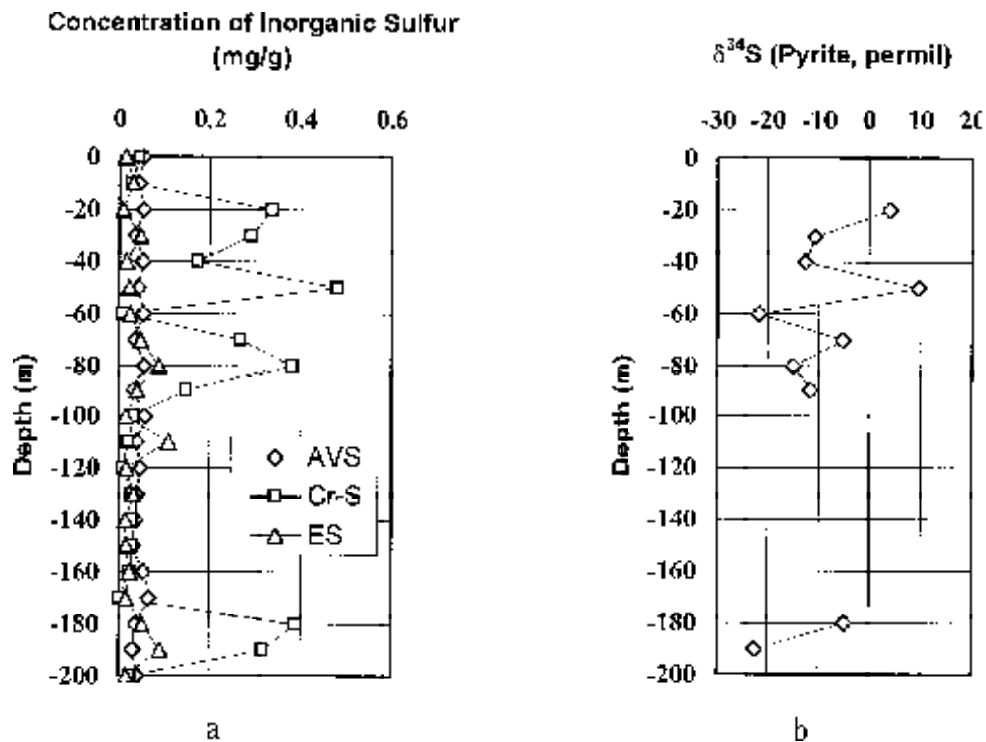


Figure 2. Vertical profiles of (a) AVS (acid-volatile sulfur), Cr-S (pyritic sulfur), and ES (elemental sulfur) and (b) $\delta^{34}\text{S}$.

Table 1. Analytical data of inorganic, organic sulfur concentration and their stable carbon and sulfur isotopes.

Depth (m)	AVS (mg/g)	Cr-S (mg/g)	ES (mg/g)	$\delta^{34}\text{S}_{\text{inorg-S}}$ (‰)	Org-S (mg/g)	$\delta^{34}\text{S}_{\text{org-S}}$ (‰)	TOC (%)	$\delta^{13}\text{C}$ (‰)	S/C
0	0.05	0.04	0.01	--	0.67	7.71	0.49	-22.48	0.09
-5	--	--	--	--	--	--	--	-22.73	--
-10	0.04	0.03	0.03	--	0.46	8.24	0.15	-22.74	0.09
-15	--	--	--	--	--	--	--	-23.54	--
-20	0.06	0.34	0.01	4.05	0.52	8.34	0.15	-24.32	1.76
-30	0.04	0.29	0.05	-10.70	0.48	8.22	0.16	-24.27	1.12
-35	--	--	--	--	--	--	--	-23.76	--
-40	0.05	0.17	0.02	-12.59	0.40	7.46	0.55	-24.05	1.04
-45	--	--	--	--	--	--	--	-23.50	--
-50	0.04	0.48	0.02	9.70	0.74	7.82	0.42	-23.78	1.25
-55	--	--	--	--	--	--	--	-23.75	--
-60	0.05	0.01	0.02	-21.58	0.69	7.74	0.46	-14.34	0.03
-70	0.03	0.27	0.05	-5.27	0.57	3.09	0.25	-23.46	0.54
-75	--	--	--	--	--	--	--	-23.66	--
-80	0.06	0.38	0.09	-15.00	0.69	5.29	6.36	-20.29	0.29
-85	--	--	--	--	--	--	--	-23.15	--
-90	0.03	0.15	0.04	-11.55	0.59	8.22	0.36	-22.13	0.22
-95	--	--	--	--	--	--	--	-19.45	--
-100	0.06	0.03	0.01	--	0.75	8.84	0.45	-21.44	0.06
-105	--	--	--	--	--	--	--	-18.14	--
-110	0.04	0.02	0.11	--	0.50	8.70	0.72	-20.96	0.06
-115	--	--	--	--	--	--	--	-15.34	--
-120	0.05	0.01	0.02	--	0.44	8.74	0.84	-21.65	0.01
-125	--	--	--	--	--	--	--	-14.38	--
-130	0.04	0.03	0.03	--	0.48	7.71	0.44	-18.66	0.06
-135	--	--	--	--	--	--	--	-15.83	--
-140	0.04	0.03	0.01	--	0.65	8.72	0.28	-25.34	0.09
-145	--	--	--	--	--	--	--	-26.00	--
-150	0.03	0.03	0.01	--	0.58	8.55	0.46	-24.21	0.18
-155	--	--	--	--	--	--	--	-25.48	--
-160	0.05	0.02	0.02	--	0.45	8.44	0.18	-24.92	0.08
-165	--	--	--	--	--	--	--	-24.78	--
-170	0.07	0.00	0.01	--	0.68	8.58	0.36	-24.33	0.00
-175	--	--	--	--	--	--	--	-21.16	--
-180	0.04	0.39	0.05	-4.85	0.61	8.75	0.22	-25.49	1.43
-185	--	--	--	--	--	--	--	-24.56	--
-190	0.03	0.32	0.09	-22.43	0.46	8.12	0.29	-26.01	0.87
-195	--	--	--	--	--	--	--	-26.59	--
-200	0.04	0.02	0.02	--	0.43	3.63	0.20	-26.14	0.19

Organic Sulfur

The average organic sulfur concentration is 0.56 ± 0.11 (Tab. 1 and Fig. 3a). Higher concentration of organic sulfur is observed at depth intervals of 50~110m and 140~170m. Its $\delta^{34}\text{S}$ remained at a relatively constant value of 8‰ with a few exceptionally low values at depths of 70m and 200m (Fig. 3b), indicating the influence of ^{34}S -depleted sulfate during the time of transgressions.

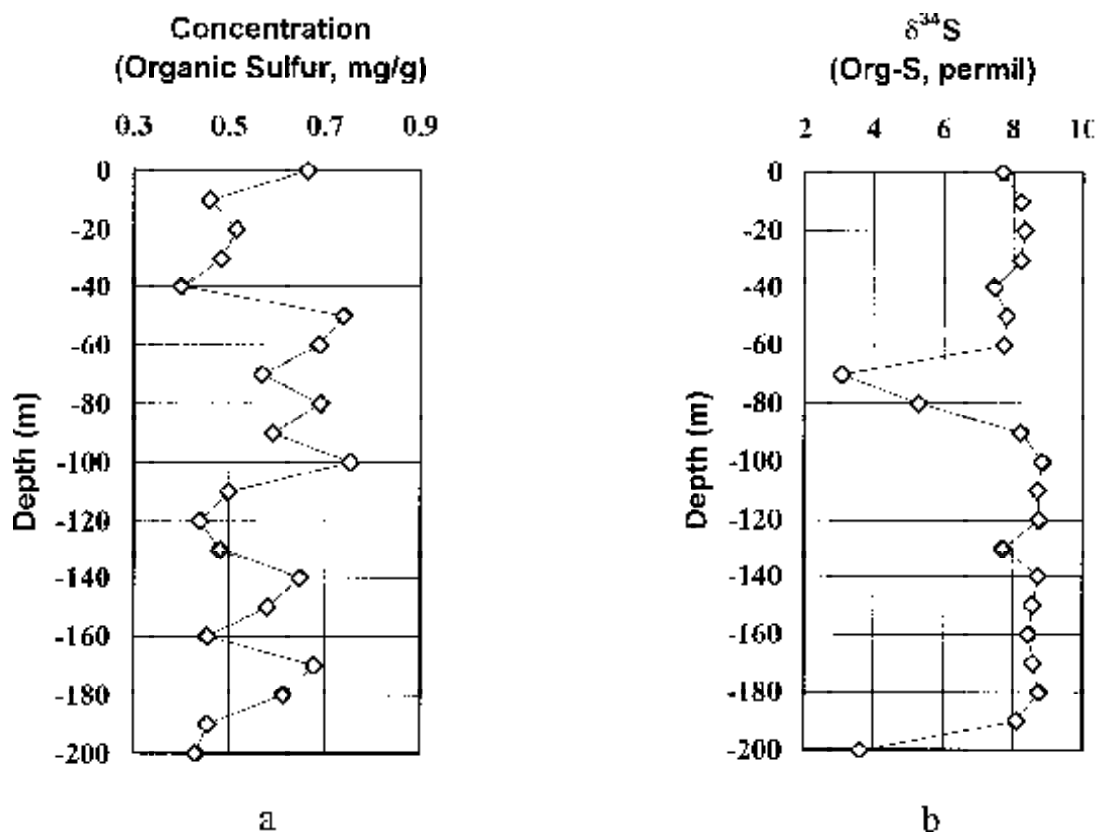


Figure 3. Vertical profiles of organic sulfur concentrations (a) and $\delta^{34}\text{S}$ (b).

Organic Carbon

The total organic carbon (TOC) contents remain relatively stable between 0.1~0.5% except for an extremely high value of 1.3% at 80m (Table 1 and Fig. 4a). At the interval of -140 to -200m, the $\delta^{13}\text{C}$ values are about 3‰ lighter than modern marine value of -22‰ probably a

result of mixing with marine phytoplankton and terrestrial organic materials (Fig. 4b; Hsieh & Chen, 1999). At the interval of -60 to -130m, the relatively heavier $\delta^{13}\text{C}$ values imply that C4 plant becomes an important source of organic matter. Above the depth of 60m, the $\delta^{13}\text{C}$ values turn into the range of modern shallow marine and lagoon sediments (Fig. 4b).

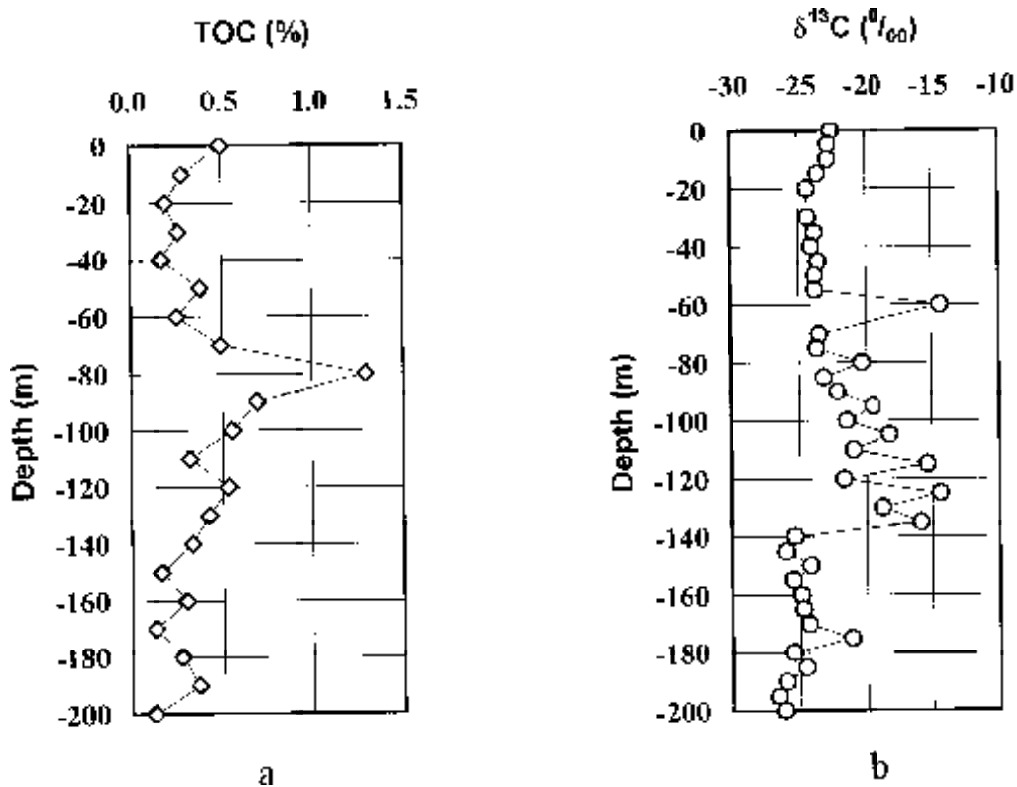


Figure 4. Vertical profiles of organic carbon concentration (a) and $\delta^{13}\text{C}$ (b).

S/C Ratio

As shown in Table 1 and Figure 5, the S/C ratios show a similar trend as Cr-S concentration. On the basis of available age determinations, the upper 100m associated with higher S/C ratio can be correlated to Holocene. A second higher S/C ratio is observed at depth of 170-180m as an old transgression, which might indicate the transgressional event of the high sea-stand during OIS 5 (oxygen isotope stage 5, constructed by deep-sea foraminiferal analysis (Morley & Hays, 1981).

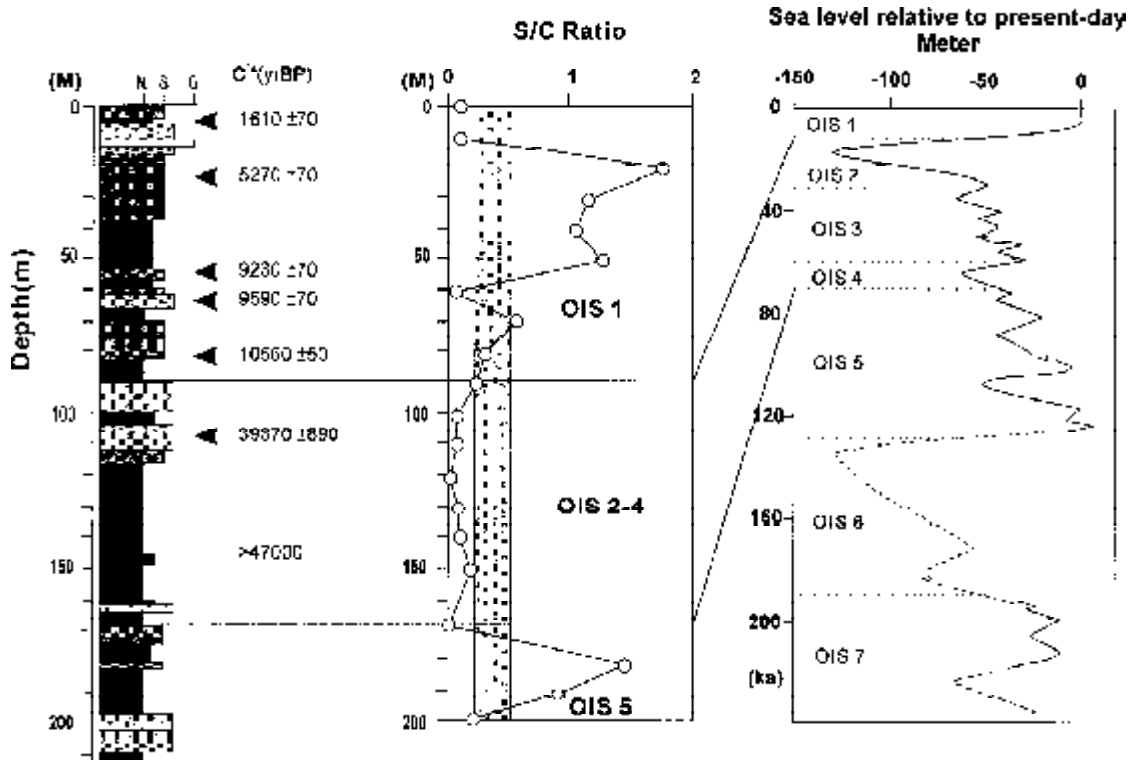


Figure 5. The S/C ratio profile of Yihju core correlated with lithostratigraphic columnar section, ^{14}C ages and the global sea level over the past 200 kyrs. The shaded area represents the S/C ratio (0.26-0.46) boundary dividing subaerial and normal marine sediment. Sea level curve used in this paper is adopted from Chappell & Shackleton (1986).

DISCUSSIONS

Holocene Transgression

Based on our inorganic sulfur result, the uppermost 100m shows relatively high concentration associated with light $\delta^{34}\text{S}$ (Figs. 4a & 4b). High inorganic sulfur concentration usually represents a marine environment where the sulfate reduction prevails to transform seawater sulfate into hydrogen sulfide and that ^{32}S is significantly enriched. As a result, the uppermost 100 meters are most likely to represent deposits during Holocene transgression. In addition, the high S/C ratios, generally greater than 0.36 except for the depth of 60 m, reveal that this interval is developed under a marine environment (Fig. 5). On the other hand, the heavier $\delta^{13}\text{C}$ of lower depths (i.e., 100~140m) extends into very early Holocene (Fig. 4b). This indicates that this area has not been dominated by C3 plant, indicator of humid period such as present-day, until very early Holocene.

Late Pleistocene Terrestrial Environment

The inorganic sulfur concentrations between the depths of 100 to 170m exhibit values much less than its overlying sediments. Less inorganic sulfur is accumulated in a terrestrial environment as a result of no sulfate reduction due to rather low sulfate concentration in the freshwater environment. Since this interval shows low inorganic sulfur concentration and low S/C ratios, sediments in this interval were most likely to deposit in the subaerial condition. It is reasonably correlated to the time period of OIS 2-4, where the sea stand was generally lower than those of Holocene and OIS 5. The $\delta^{13}\text{C}$ gradually shifting to a heavier value from depth of 140m upward to very early Holocene indicates that the concurrent climate was relatively dryer than its upper and lower parts of the core. Since this arid event has been found in the Taipei basin of northern Taiwan (Wei, 1997), the dry event is probably a coeval phenomenon in the entire western Taiwan.

OIS 5 Transgression

Our data also show another high inorganic sulfur interval at depths of 180-200m, where the $\delta^{34}\text{S}$ value is low and the S/C ratios are higher than 0.36 (Figs. 1 and 5). Therefore, we suggest that an older transgressional period has occurred in this area. Because of lack of age control under 120m, we assume that these sediments were deposited during OIS 5. Based on their lighter $\delta^{13}\text{C}$ values, the dominant vegetation of Yihju area was C3 plant during this period (Fig. 4b). This conclusion is consistent with that OIS 5 is a well-known humid and warm period.

Special Environments at -60m and -80m

An unusually low value of inorganic sulfur concentration appears at -60m. In a marine or coastal environment, three possible conditions can result in such a value: (1) lack of iron; (2) decrease in supply of organic carbon, and (3) reoxidation of already formed pyrite. Iron is usually abundant in the coastal plain of southwestern Taiwan (Pi, 1995; Hsia, 1998). Based on our TOC result (Fig. 4a), the organic carbon supplement at this depth remains similar during its sedimentary history. Thus, the organic carbon and iron concentrations are unlikely to be responsible for this unusually low inorganic sulfur concentration. Lin *et al.* (1998) showed that this area was a sand bar environment based on grain-size analysis. This is also confirmed by the C4 signal of a very heavier carbon isotopic value, indicating that this area might be emerged and vegetated by C4 plants.

An extraordinarily high TOC at -80m (Fig. 4a), as well as the lowest $\delta^{34}\text{S}$ of the organic matter, shows that an abrupt change of organic deposition during this short period. The unusually low $\delta^{13}\text{C}$ of the organic carbon shows that C4 plant, an indicator of an arid environment, was predominating during deposition. A marsh environment may account for such characteristics.

The Sources of Organic Sulfur

There are three major types of organic sulfurs incorporated in the sediments: nonchromium-reducible organic sulfur (Non-CROS), chromium-reducible organic sulfur (CROS) and hydrolyzable organic sulfur (HYOS) (Canfield *et al.*, 1998). The organic sulfur concentration includes mostly Non-CROS using our extracting procedure. The light $\delta^{34}\text{S}$ organic sulfur observed at depths between 70-80m and 200m (Fig. 3b) can easily be transformed from the diagenetic addition of sulfide or polysulfide into unsaturated organic compounds (Sinninghe Damste *et al.*, 1989a,b; Kohnen *et al.*, 1990, 1991; Wakeham *et al.*, 1995). The sulfide or polysulfide is usually depleted in ^{34}S in an open system, shifting CROS to a relatively light $\delta^{34}\text{S}$ value of Non-CROS.

The marine organic material, which is lipid-dominant, could easily have this light $\delta^{34}\text{S}$ signature. At the periods during their deposition, the marine organic material was predominant with very little organic matter during the commencing stage of a transgression event.

CONCLUSIONS

1. Two intervals of high inorganic sulfur concentration at the upper 100m and 180 to 200m are probably resulted from the transgression events (Holocene and OIS 5).

2. Low inorganic sulfur content and light $\delta^{34}\text{S}$ value at 60m was a result of limited sulfate in a sand bar environment. The previously formed pyrite can be easily oxidized in this subaerial environment.

3. The extraordinarily high organic carbon at -80m implies that probably a marsh environment was formed, where high biological productivity enhanced the organic carbon deposition. Excepting this, however, the deposition of organic carbon at Yihju area has been generally stable and limited in a range of 0.1~0.5% since late Pleistocene.

4. From depths of 60 to 130m, the $\delta^{13}\text{C}$ values shift to a heavier C4 plant, implying that C4 plant was an important source of organic matters. The flourishing of C4 plant indicates that the climate was relatively arid. Based on age data, this stratum was developed from early OIS 2 to early OIS 1.

5. The light $\delta^{34}\text{S}$ values of organic sulfur around -70~-80m and -200m are noticeably influenced by ^{34}S depleted sulfide of marine source. As a result, the marine organic source was dominant and the terrestrial source decreased at these periods during the beginning stage of transgression.

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