Research Article

Effect of Sea-Level Change on Deep-Sea Sedimentary Records in the Northeastern South China Sea over the past 42 kyr

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We integrated multiple geochemical analysis of a 13.75 m-long core 973-4 recovered from the northeastern South China Sea (SCS) to detect the response of deep-sea sediment archives to sea-level change spanning the last 42 kyr. The age-depth model based on AMS ¹⁴C dating, together with the sediment grain size, shows an occurrence of turbidity current at around 14 kyr, which was associated with submarine landslides caused by gas hydrate dissociation. A dominantly terrigenous sediment input was supplied from southwestern Taiwan rivers. By synthesizing environment-sensitive indexes, four distinct stages of paleoenvironmental evolutions were recognized throughout the studied interval. Well-oxygenated condition occurred during the stage I (42.4–31.8 kyr) with low sea-level stand below -80 m, accompanied by flat terrigenous input. The largest amounts of terrigenous sediment input occurred during the late phase of stage II (31.8–20.4 kyr) with the lowest sea-level stand below -120 m because of a short distance from paleo-Taiwan river estuaries to the core location. An occurrence of Ca-enriched turbidity current disturbed the original sediments during the stage III (20.4–13.9 kyr). The stepwise elevated sea-level stand resulted in an enclosed (semi-enclosed) system and contributed to a relatively low-oxygen environment in deep ocean during the stage IV (13.9 kyr—present). Temporal variations of TOC and CaCO₃ display contrary pattern synchronously, indicating a decoupled relationship between organic carbon burial and carbonate productivity. Our results highlight that these sedimentary records as reflected in the paleoenvironmental changes in the northeastern SCS were mainly driven by sea-level fluctuations and later, since the mid-Holocene, the strengthening East Asian summer monsoon (EASM) overwhelmed the stable sea level in dominating the environmental changes.

1. Introduction

As the largest marginal sea of the western Pacific, the South China Sea (SCS) receives large amounts of sediment annually, with possible terrigenous supplies from the Pearl, Red, and Yangtze Rivers, as well as the island of Taiwan and Luzon [1–4]. It is considered that more than 700 Mt/year of fluvial sediments are transported to the SCS from surrounding rivers [3], amounting to 3.7% of estimated global fluvial sediment discharge to the world ocean [5]. Among various fluvial drainage systems, the mountainous rivers from southwestern (SW) Taiwan discharge the largest number of

suspended sediments directly to the SCS, with a total load of 176 Mt/year, South China contributes about 102 Mt/year, and the supply from Luzon Island is more than 13 Mt/year (Figure 1) [3]. Recently, the area in the east of the Pearl River (EPR) has been considered as another potential source for sediment supply to the northern SCS [6]. These fluvial sediments are further transported by various coastal, surface, and deep currents after entering the SCS, such as the East Asian monsoon (EAM), which is the major control on the surface circulation. Meanwhile, the deep-water current from the Western Pacific through the Luzon Strait and the intrusion of shallower Kuroshio Current are considered to be the

FIGURE 1: Overview map of the northern part of the SCS, including core location 973-4, monsoon and current systems, fluvial drainage systems, and their annual suspended sediment discharge to the South China Sea. Core 973-4 is labeled by red star. Monsoon and current systems are labeled by colored arrows. Monsoon winds after Webster [11], Guangdong coastal currents after Fang et al. [8], Kuroshio Current after Caruso et al. [7], deep water currents after Zhao et al. [12]. Fluvial drainage systems and their annual discharge to SCS are represented by grey arrows with numbers (in million metric tons, Mt/yr), and the size represents the flux (summarized by [3]). The base map was generated from GeoMapApp.

main pathways that can transport sediments derived from Taiwan to the northern SCS. Moreover, the Guangdong Current impacted by EAM flows northeastward in summer and southwestward in winter, which is important for sediment transport from the Pearl River to the northern SCS (Figure 1) [3, 7–12]. Consequently, the SCS is commonly regarded as the largest receptacle of fluvial sediments among semi-enclosed marginal seas around the world [5], and this area provides a natural laboratory for deciphering paleoenvironmental changes from high-resolution sedimentary records.

In comparison with continental basin affected by significant modification of diagenetic processes [13–15], marine sediments in ocean basin have more clearer fingerprints for the deposition history; hence, they are robust archives for reconstructing the past variations of both climate and environment. Over the last decade, many studies attempted to elucidate the paleoenvironmental changes by using multiple paleoproxies, such as elemental geochemistry [16, 17], foraminiferal characteristics ([18, 19]), magnetism [20], clay minerals [3, 9, 21], and granularity [22, 23]. The key conclusion from these results is that sea-level fluctuations placed a

significant impact on climate change and dominated the environmental development. For example, during the Last Glacial Maximum (LGM) when the global sea level dropped below -120 m, the coastline retreated, and larger areas of continental shelf were exposed; fluvial sediments could be discharged into outer shelf and deep ocean more easily [24].

The continental margin off SW Taiwan is located in the northeastern SCS, which is a greatly suitable area for studying the sedimentary responses to the sea-level change because this area has received large amounts of fluvial sediments. A recent research demonstrated that Taiwan-sourced sediments could be delivered into the SCS for a long distance [25]. In such a case, frequent turbidity currents induced by sea-level changes have been observed [26], and lowstand is more likely to trigger seafloor instability [27, 28]. As a result, the exogenous materials carried by paroxysmal currents are able to disrupt the original sequence during the process of sedimentation. Because of active methane seepage, published reports have paid more attention to the dissociation of deep methane-hydrated reservoirs [29–31], few studies concentrated on the paleoenvironmental changes in the northeastern SCS over the studied time scales, and the sedimentary

FIGURE 2: Downcore variations in lithology (a), calibrated age-depth model (b) and mean grain size (c). Lithological characteristics were summarized from [51]. The calibrated AMS ^{14}C dating was collected from [3, 19, 31, 52]. The yellow circle represents the abnormal intervals resulted from turbidity current, and the calibrated age presents great linear feature and high deposition rate (32.167 cm/kyr) except for several turbidite-influenced data. Grain size data were taken from [31], and units 1-4 are defined correspondingly.

records related to sea-level variations remained ambiguous. In this work, detailed sedimentary records from core 973-4 are used to better constrain the paleoenvironmental changes related to the sea-level variations since the last glaciation.

2. Materials and Methods

2.1. Materials and Age-Depth Model. Due to special location and tectonic characteristics, the SCS was greatly affected by surrounding units such as Eurasian, Pacific, and Indo-Australian plates, which resulted in complicated submarine topography. The study area is located in the SW Taiwan Basin of the northeastern South China Sea, where submarine faults, bottom channels, and mud diapirs were developed [32]. Many studies have analyzed the hydraulic properties of rock fractures [33–40] and suggested that these fracture systems in strata are favorable pathway for fluid migration [41–45], but the fracture systems develop heterogeneously in natural environments, and the fracture prediction is quite difficult and needs more experimental investigations [46–49]. Additionally, large-scale seep carbonates associated with gas hydrate and CH_4 -enriched fluid migration, named as “Jiulong methane reef”, were discovered nearby the study

area in 2004 [50]. The studied core 973-4 (Figure 1) ($118^\circ 49.0818'\text{E}$, $21^\circ 54.3247'\text{N}$; 1666 m water depth; 13.75 m long) was retrieved from the lower continental slope in the northeastern SCS conducted by the “Haiyang-6” vessel in 2011. After core retrieval, the collected sediments were divided into segments and then freeze-dried for the further analysis. Simultaneously, the core was described immediately. The lithology of core 973-4 is characterized by dark-green silty clay and grey silty clay, and the abnormal intervals between 450 and 605 cm consist of a silty layer, accompanied by enhanced abundances of foraminifera as described by Qu [51] in details (Figure 2(a)).

On the basis of the geochronological framework (Figure 2(b)), core 973-4 had a high linear deposition rate with an average of 32.167 cm/kyr, and the oldest sediments were from the phase of the Marine Isotope Stage (MIS) 3 recorded at the age of 42.4 kyr. An obvious discontinuity in the age-depth model was observed at the intervals between 450 and 605 cm aged from around 14 to 20 kyr, in accordance with the core observation, and the younger strata was disturbed by older sediments. The inconsecutive sequence was identified as the potential occurrence of turbidity currents, which have been simply mentioned before [29, 31].

2.2. Analytical Methods. Samples were taken at 30 cm intervals, and 40 samples were obtained from top to bottom of core 973-4. The samples were freeze-dried and then crashed into powders with an agate pestle and mortar. To remove the organic matter and carbonate, 10% H_2O_2 at 60°C for 1 h and 0.5 N HCl at 60°C for 2 h were treated to react with sediments, respectively. Afterwards, sediments were washed three times with deionized water and then were freeze-dried for further treatment. For each sample, about 30 mg freeze-dried sediments were placed into the digestion vessel and microwaved (Top Wave) along with an acid mixture of 3 ml HF and 6 ml HNO_3 . After cooling, the remaining solutions were transferred into the Teflon Crucible on a hotplate for heating overnight until the samples were evaporated to dryness. Subsequently, the residues were dissolved in 2 ml HNO_3 (2%) and diluted to 20 ml with Milli-Q water. The obtained solutions were collected for the determination of major metal concentrations (Al, Ba, Ca, Fe, K, Mg, Mn, Na, P, Ti) by using an Inductively Coupled Plasma-Optical Emission Spectrometer (ICP-OES) at the Institute of Soil Science, Chinese Academy of Sciences (Nanjing). The precisions for chemical analysis of major elements were better than 3%, and the results were reliable. For the analysis of trace element concentrations (Sr, Li, Be, V, Cr, Co, Ni, Cu, Zn, Rb), the digested solution was determined at the Institute of Earth Environment, Chinese Academy of Sciences (Xi'an) by using an Inductively Coupled Plasma Mass Spectrometry (ICP-MS). Within the elemental detection ranges, precision and accuracy were both better than 5% for trace elements.

Several sedimentary segments were collected for the analysis of total nitrogen (TN) and nitrogen isotope ($\delta^{15}\text{N}$). For the TN measurement, dry samples were dealt with 1 N HCl to react with carbonate and then rinsed with deionized water thoroughly, followed by drying at 40°C for 24 h. Subsequently, about 3 mg processed subsamples were analyzed for TN using a PE 2400 SERIES-II Elemental Analyzer. About 20 mg each subsample was taken for the measurements of nitrogen isotopes ($\delta^{15}\text{N}$) by a Thermo Scientific Delta V Advantage mass spectrometer. All the measurements were conducted at the State Key Laboratory of Marine Environment Science, Xiamen University. The analytical precision was better than 0.2‰ for TN and 0.3‰ for $\delta^{15}\text{N}$.

In addition, published data from core 973-4 were also synthesized in this study to detail the dominated controls of sea level on sedimentary records.

3. Results

According to the downcore variations in grain size, the overall core of 973-4 has four distinctive horizons (Figure 2(c)). Unit 4 (40–450 cm) exhibits the grain size ranging from 5.67 to 8.32 μm with a mean grain size of 7.07 μm and mainly consists of celadon silty clay. There is a rapid increase in grain size in unit 3 (450–610 cm) to an average of 7.95 μm due to the influence of turbidity current, and the sediments are foraminifera-enriched silt with plant fragments. Unit 2 (610–980 cm) shows the lowest grain size value with a mean of 6.45 μm and consists of grey silty clay. A gradual increase in grain size is presented in the lower unit 1 (980–1370 cm),

ranging between 6.24 and 10.06 μm with an average of 7.77 μm , and this interval constitutes grey silt and grey clay (Table 1).

The measured major and trace elements for core 973-4 in this study are mainly composed of Al (5.57–9.59%), Fe (3.03%–4.61%), Ca (2.00%–10.25%), Mg (1.07%–2.07%), Na (1.38%–1.92%), K (1.69%–3.21%), Ti (0.32%–0.55%), Mn (273.89–827.10 ppm), P (432.86–879.41 ppm), V (92.65–157.29 ppm), Cr (52.72–94.58 ppm), Rb (55.28–156.27 ppm), and Sr (125.05–411.20 ppm). As similar with variability of grain size, the elemental profiles have also been divided into the same four intervals from top to bottom (Figure 3). Vertical variations of Al, K, Fe, Mg, Ti, V, and Cr show a similar temporal pattern, which exhibits more depleted in unit 3 while a dramatic increase in concentrations is observed in unit 2. Al concentrations decreased with depth from 7.22% at the top to 6.19% at the depth of 600 cm, following by an increase to 9.59% at the depth of 830 cm, and then the Al values vary little until the bottom. Ti concentrations show stable trend in unit 1 and unit 4, but fluctuations are observed in unit 2 and unit 3 ranging between 0.32% and 0.55%. Similarly, V and Cr show steady variations in unit 1 and unit 4, while nonsteady trends are displayed in unit 2 and unit 3, increasing from 92.65 $\mu\text{g/g}$ to 157.29 $\mu\text{g/g}$ for V and 52.57 $\mu\text{g/g}$ to 94.58 $\mu\text{g/g}$ for Cr with depth, respectively. Mn concentrations generally increase with depth from 0.04% to 0.08% except for the intervals of unit 3 featuring a minimum of 0.03%. The profiles of Ca and Sr display a similar temporal pattern, which is characterized by extraordinarily high values in unit 3 with a peak of 10.25% for Ca and 411.20 $\mu\text{g/g}$ for Sr, while the concentrations in other depths are low and vary slightly, with an average of 2.57% for Ca and 152.77 $\mu\text{g/g}$ for Sr, respectively. The more detailed variations of elemental contents in core 973-4 are presented in Table 1 and Figure 3.

Accordingly, it is noticeable that these vertical profiles exhibit special variations in unit 3 relative to other intervals, which may be related to the modification by the turbidity current. In order to better constrain this anomaly, the major element concentrations from core 973-4 sediments, together with those from the Pearl River [54], the Southwestern Taiwan Rivers [55], and the Luzon River [56] were all used for comparison after normalizing them to the upper continental crust (UCC) (Figure 4) [57].

Our results show that most of the normalized elements including Al, Ca, Fe, Mg, Mn, Na, P, and Ti in core 973-4 are depleted with the exception of K. In addition, the values in unit 3 affected by the turbidity current are significantly different from those deposited in normal strata (unit 1, unit 2 and unit 4), especially for the Ca content. The normalized concentrations of Ca in core 973-4 within unit 3 are greatly high, such an abnormal variation significantly deviates from normal deposition sequence and other river sediments. In addition, there are no obvious similarities in UCC-normalized concentrations between those from core 973-4 and other river samples (Figure 4).

Biogenic proxies in sediments including TN, $\delta^{15}\text{N}$, TOC, and CaCO_3 are mainly driven by marine productivity and oceanic processes [58]. In this work, the TN and $\delta^{15}\text{N}$ values within selected depths ($n = 20$) were mainly used to

TABLE 1: Average (avg) and standard deviation (std) of geochemical parameters in different intervals of the core 973-4. Major elements are given in weight percent (%) while trace elements are in $\mu\text{g/g}$.

Parameters		Unit 4 (70-450 cm)		Unit 3 (450-610 cm)		Unit 2 (610-980 cm)		Unit 1 (980-1280 cm)	
		Avg	Std	Avg	Std	Avg	Std	Avg	Std
Major elements (%)	Al	7.13	0.38	6.10	0.26	7.59	0.62	7.37	0.21
	Ca	2.49	0.43	7.64	1.38	2.64	0.16	2.59	0.08
	Fe	3.82	0.14	3.13	0.07	4.06	0.20	4.06	0.09
	K	2.26	0.05	1.90	0.13	2.54	0.21	2.43	0.04
	Mg	1.23	0.09	1.21	0.06	1.64	0.14	1.52	0.06
	Mn	0.04	0.00	0.03	0.00	0.06	0.01	0.07	0.00
	Na	1.78	0.11	1.53	0.07	1.57	0.10	1.54	0.06
	P	0.06	0.00	0.05	0.00	0.06	0.01	0.07	0.01
Trace elements ($\mu\text{g/g}$)	Ti	0.46	0.01	0.36	0.02	0.46	0.03	0.45	0.01
	V	123.57	6.24	102.46	6.15	130.03	8.30	129.59	4.91
	Cr	71.04	4.58	56.95	2.70	79.02	4.68	79.58	2.51
	Rb	79.32	16.14	89.03	4.87	99.42	22.08	87.68	4.62
	Sr	162.70	25.70	320.33	50.14	154.37	13.87	141.25	7.23
Grain size(μm)		7.07	0.75	7.95	0.38	6.45	0.31	7.77	0.76
$\delta^{13}\text{C}_{\text{TOC}}(\text{‰})$		-25.08	0.88	-24.11	0.76	-25.18	0.49	-25.36	1.04
TOC(%)		0.65	0.25	0.74	0.13	0.88	0.16	0.64	0.19
$\text{CaCO}_3(\text{‰})$		6.82	3.00	21.32	4.64	4.47	1.33	7.50	1.72
$\delta^{15}\text{N}(\text{‰})$		4.29	0.79	4.38	1.21	4.75	0.96	4.77	0.91
TN(%)		0.09	0.13	0.07	0.08	0.08	0.10	0.06	0.07

Data sources: grain size data and CaCO_3 content [31], $\delta^{13}\text{C}_{\text{TOC}}$ and TOC data [53], and other data (this study).

FIGURE 3: Variations in major and trace elemental concentrations, together with TOC (yellow line), CaCO_3 (red line) content, and their normalized to Al values (black line) at core 973-4 in the northeastern SCS over the past 42 kyr.

determine the source of organic matter, the TN values vary in a narrow range from 0.06% to 0.10% (average 0.08%), and TN contents in unit 1 are apparently lower than upper layers. The $\delta^{15}\text{N}$ values exhibited in four units are between 3.97‰ and 5.24‰ (average 4.59‰), and a gradually increasing trend of the mean $\delta^{15}\text{N}$ value is observed with respect to depth.

The content of TOC and CaCO_3 has been reported before [31, 53]. Over the past 42 kyr, the content of TOC varies in the range between 0.39% and 1.26%, which shows relatively higher values in unit 2 and unit 3 but lower values in unit 1 and unit 4. Temporal variations of CaCO_3 present extremely high values in unit 3 and reach a maximum of 30.19%, while lower values are displayed in unit 2, and relatively higher

FIGURE 4: UCC-normalized major element concentrations at core 973-4 since the past 42 kyr, showing comparisons with those from the Pearl River [54], Luzon River [56], and offshore of SW Taiwan [55].

values are showed in unit 1 and unit 4. The Al normalized ratio of biogenic records (TOC/Al and CaCO_3/Al) displays a consistency in vertical changes with their content data (Figures 3(n) and 3(o)).

4. Discussion

4.1. The Occurrence of Turbidity Current. Based on sedimentological and geochemical evidences, the unit 3 at core 973-4 was characterized by the largest mean grain size and the highest CaCO_3 content; additionally, the sedimentary strata were disturbed by ex situ sediments, resulting in a sedimentary discontinuity along the normal deposition sequence. Previous studies have confirmed this phenomenon and mentioned the occurrence of turbidity current event briefly [19, 31], but no synthetical description was focused on the overall processes. In this study, we attempt to explain the inducing factors, development process, and the impact of the turbidite event on sedimentation, which could provide further detailed information for investigating environmental evolution in the northeastern SCS.

Core 973-4 is located in the northeastern SCS, where methane hydrate has been considered to accumulate into deep-sea sediments [59]. Generally, gas hydrate is stable under low-temperature and high-pressure environment. When hydrated reservoir forms, an enhanced strength of gas hydrate-bearing sediments has been indicated from laboratory simulation [60], yet the hydrate-free sediment is unconsolidated and differs from the tight reservoirs, such as coal and oil shale [61, 62, 63]. Zhang et al. [31] reported the methane hydrate dissociation history by using stable carbon and oxygen isotope composition of carbonate in bulk sediment, the results revealed that during past 14 kyr, methane hydrate decomposition events were significant and led to the largest methane flux from deep sediments to the overlying layer, and the experimental study indicated that tectonic zones may serve as a favorable pathway for fluid migration [64, 65].

At around 14 kyr, a significant increase in water temperature was concluded [66], which was likely to promote the gas hydrate decomposition, and the high values of oxygen isotopes of benthic foraminifera during this interval [31] were closely related to the warm seawater temperature. Besides, the relatively low sea-level condition (-90 m) during this period might be another crucial factor to accelerate the dissociation process. As a consequence, intense release of gas from deep reservoirs can directly destabilize the seafloor, resulting in the loss of reservoir strength and the occurrence geological hazards, such as earthquake, submarine landslides, and turbidity current [67, 68]. Moreover, both steep slope and developed submarine channel can contribute to the hyperpycnal flow and sediment transportation to the deep ocean.

The intervals of turbidite at core 973-4 range from 450 cm to 605 cm, where mean grain size and stratigraphic chronology vary significantly due to instantaneously enhanced hydrodynamic condition. Furthermore, the concentration of all major elements deposited in turbidite-influenced strata, with the exception of calcium, is less than that of normal deposition strata (Figure 4), and the extreme enrichment in Ca content is attributed to the external input of biogenic content and coincides with an increasing abundance and diversity of foraminiferal assemblages [19]. In return, those materials could result in the enhanced dilution effect to other elements. Chen [69] reported that the nearby core 973-3 (1026 water depth) (Figure 5) occurred turbidity current within the intervals between 220 cm and 770 cm. Interestingly, the strata from these two adjacent sites was overturned at nearly the same time according to the age-depth model. Yet, the shallow-water core 973-3 received more turbidites than that at core 973-4, resulting in much thicker turbidite-influenced layer. Therefore, we can infer that the location of the submarine landslides was situated in the area closed to the river mouth.

At around 14 kyr, both low sea-level condition and rising temperature of seawater were responsible for dissociating the

FIGURE 5: A conceptual picture showing the possible processes, including the occurrence of turbidite, sediment delivery, and sedimentation due to the slope failure induced by gas hydrate dissociation. The three drawings in the bottom display the evolution of the hydrated reservoir from a microscopic aspect during gas hydrate dissociation owing to unstable environmental condition. (a). When formed within sediment, gas hydrate filled into pores and cemented particles with high saturation, resulting in an increase in bulk sediment strength (b). When the perturbations to the thermodynamic equilibrium of the gas hydrate system occurred due to the change of sea level and seawater temperature, hydrate began to dissociate from the stability zone (hydrated zone). This process released water and free gas, which diffused upward to the subsurface layer or even to the seafloor shaped as gas plumes, accordingly, the strength in sediments decreased, and sediment particles showed a tendency to be compressed (the dotted arrows) (c). As the hydrate dissociates and gas releases, the sediment particles were redistributed, and the reservoirs became instability. Consequently, those factors were sufficient to trigger an occurrence of submarine landslide and then induced the turbidity current. The turbidity current transported large block of sediments into the deep ocean along the steep slope; finally, the fine materials in suspension deposited on the top of coarse particles. The exogenous components mixed with in situ marine sediments and redeposited on the seafloor, which caused the destruction of the original deposition sequence with certain depth.

hydrated reservoirs, which subsequently led to the release of gas and slope failure, and the contemporaneous turbidity current began to form. During the early stage of the turbidity current formation, plenty of materials were transported from the parent location to deep sea along the steep slope. After delivering for a long distance, turbidity current evolved to a relatively low-density flow. In the form of sediment plume, fine components in suspension were finally deposited on the top of coarse-particle layer due to weakened hydrodynamic condition, which was reflected in the profile of sediment granularity that a sharp drop in mean grain size overlay the turbidite-influenced strata (Figure 2). Below the turbidite-influenced layer, age-depth discontinuity caused by episodically active turbidity currents within some intervals was also speculated during the low sea-level period, but the scale was so small that it had little effect on the environmental reconstruction throughout the studied timescale.

Turbidity current could disturb the original sedimentation rate, and the concomitant materials would mix with local layer; as a consequence, the newly assembled sediments redeposited on the top of normal deposition strata, which led to a discontinuity in the chronological framework. The overall process including the occurrence of submarine landslide, formation of turbidity current, sediment delivery, and deposition process is conceptually illustrated in Figure 5. Large-scale submarine landslides associated with dissociation of gas hydrate have been reported world-

wide, such as the West African margin [70]. Gas hydrate dissociation controlled by climate change has been recognized as a geohazard, which is related to the fate of human activity, and it has been drawn increasing international concern for a long time.

4.2. The Sources of Organic Matter and Sediment Provenance.

The high deposition rate was identified from the age-depth model, and some plant fragments and rotten wood were also observed during core recovery [31], which indicated that core 973-4 was influenced by the input of terrigenous matters. More specifically, the TOC/TN ratio and organic carbon isotope values ($\delta^{13}\text{C}_{\text{TOC}}$) are widely used to distinguish the source of organic matter [71]. Generally, MOM (marine organic matter) has more positive $\delta^{13}\text{C}_{\text{TOC}}$ values and lower TOC/TN ratios than TOM (terrestrial organic matter) [72–74]. By contrast, the TOC/TN ratios in core 973-4 (average 9.39) (Figure 6) are distinctly higher than those from adjacent region of the Dongsha area (average 4.86) [75], presenting a significant contribution from the terrestrial origin. Moreover, the $\delta^{15}\text{N}_{\text{TN}}$ values in core 973-4 fluctuate in the range from 3.97‰ to 5.24‰, which are in accordance with nitrogen isotope compositions from surface sediments in SCS (3.4‰–6.6‰) [76]. $\delta^{15}\text{N}_{\text{TN}}$ values in sediment are mainly controlled by the availability of nitrogen and the original nitrogen isotope compositions [77]. Due to the instability during cyclic process, nitrogen isotope ratios ($\delta^{15}\text{N}$) are prone to generate disagreement. In comparison, $\delta^{13}\text{C}_{\text{TOC}}$ is

FIGURE 6: $\delta^{13}\text{C}_{\text{org}}$ values vs. $\text{C}_{\text{org}}/\text{N}_{\text{total}}$ ratios of sediments from core 973-4. Data fields after Lamb et al. [71].

a more dependable proxy for indicating the organic-matter sources. Previous research reported that the $\delta^{13}\text{C}_{\text{TOC}}$ of MOM typically varies between -19‰ and -22‰ [78]. The studied core confirmed a mainly terrestrial-sourced carbon source as indicated by depleted $\delta^{13}\text{C}_{\text{TOC}}$ values ranged from -22.69‰ to -26.94‰ with an average of -25.04‰ . Additionally, a simple balance model of carbon isotope was used to calculate the relative proportion of the organic matter contribution between TOM and MOM to core 973-4 [79]:

$$\delta^{13}\text{C}_{\text{sedi}} = W_T \times \delta^{13}\text{C}_{\text{terr}} + W_M \times \delta^{13}\text{C}_{\text{mari}}, \quad (1)$$

$$100\% = W_T + W_M,$$

where $\delta^{13}\text{C}_{\text{terr}}$ and $\delta^{13}\text{C}_{\text{mari}}$ are $\delta^{13}\text{C}$ end-member for terrestrial (-27‰) and marine (-19.5‰) organic origin, respectively; $\delta^{13}\text{C}_{\text{sedi}}$ is the carbon isotope ratios from core 973-4 samples; W_T and W_M represent the TOC contribution ratios from terrestrial and marine source, respectively.

As a consequence, about 73.9% of the calculated TOM in sediment from core 973-4 was contributed to the total organic materials, suggesting a dominantly terrestrial sedimentary environment spanning the long-term time scale. Besides, some marine-derived organic matter was also contributed to the core 973-4 during the certain depositional cycles (Figure 6).

It is widely accepted that REE compositions are considered as a reliable indicator for provenance because of their conservative behavior during the sediment transportation and formation [57, 80]. Wu [81] measured the REE compositions from the same core to decipher the influence of cold seeps on the geochemical characteristics, but not to discuss the sediment provenance. Thus, we normalize the REE concentrations at core 973-4 to the upper continental crust (UCC-normalized) [57]. Accordingly, the UCC-normalized REE compositions of three potential provenances including sediments from the Pearl River, SW Taiwan, and Luzon Island, which present a distinct difference to discriminate the sediment sources in the northern SCS, are displayed for comparison. As a result, it is evident that most of the sediment samples in core 973-4 have similar REE patterns to

the SW Taiwan River sediments characterized by relatively flat variation with a slightly positive anomaly in middle REEs (Sm, Eu, Gd, Tb, Dy) and a negative anomaly in other REEs (La, Ce, Pr, Nd, Ho, Er, Tm, Yb, Lu) (Supplementary Figure S1), coinciding with the clay mineral results [3]. The generally high REE values in the Pearl River relative to the SW Taiwan Rivers and Luzon Island may be related to the strong chemical weathering and stable morphology in the Pearl River basin [82]. In addition, two discrimination plots of δEu vs. δCe and $(\text{La}/\text{Sm})_{\text{UCC}}$ vs. $(\text{Gd}/\text{Lu})_{\text{UCC}}$ are effectively used to reflect sediment sources [80], and the δEu and δCe are defined as $\text{Eu}_N/(\text{Sm}_N \times \text{Gd}_N)^{1/2}$ and $\text{Ce}_N/(\text{La}_N \times \text{Pr}_N)^{1/2}$, respectively, where N indicates chondrite normalization (data from [57]). Notably, the REE pattern from core 973-4 samples over the last 42 kyr is also similar with the SW Taiwan Rivers with higher values of δCe and $(\text{La}/\text{Sm})_{\text{UCC}}$ (Supplementary Figure S2).

To summarize, the accumulated sediments in core 973-4 were predominantly originated from Taiwanese rivers over the past 42 kyr, and no REE pattern is similar to the Pearl River or Luzon Island sediments, indicating little sediment contribution from these two sources to the investigated area. Due to the formation of the Taiwan orogen and strengthening deep-water current, the fluvial-derived sediments are considered to be the significant sedimentary inputs to the northern SCS since 3 Ma [10]. Various oceanic currents, such as surface Asian monsoon, intrusion of the subsurface Kuroshio Current, and deep-water current via Luzon Strait (Figure 1), are believed to further promote the transport process of Taiwan-sourced sediments into northern SCS thousands of kilometers away from the source region [52].

4.3. Reconstruction of Paleoenvironments. According to our data above, sea-level fluctuations have a dominated impact on the sedimentary archives responding to the environmental change, such as the occurrence of turbidite. Here, reliable paleorecords, such as grain size, major, and trace elements, together with biogenic proxies (Figure 7), are used to detect the detailed evolution process about how the sea-level variations have an influence on the paleoenvironmental change at core 973-4 in the northeastern SCS since the last glaciation, including the terrigenous input, redox condition, organic matter burial, and ocean productivity.

The fact that Al and Ti are mainly from terrigenous input and behave conservatively during the transport process, which serve as valid signs for estimating the amount of terrigenous input during the geological past. Generally, elevated terrigenous input indicated by higher Al and Ti content occurred during the glacial episodes with low sea-level stand because of the short distance from paleoestuary to the deep ocean. However, in warming stage with relative high sea level, most terrestrial sediment carried by various currents deposited in the inner shelf close to the river mouth. The similar pattern was also confirmed at core 973-4 throughout the past 42 kyr in this study (Figures 7(h) and 7(i)).

Ratios of Rb/Sr and K/Na are applied to reflect the chemical weathering. The increasing chemical weathering intensity rapidly leaches Sr compared to Rb [89]. Similarly, the K/Na ratio is also indicative of sediment recycling, which

FIGURE 7: Temporal variations of paleorecords at core 973-4 sediments over the past 42 kyr. (a). Carbon isotope of total organic carbon [53]. (b) The TOC content [53]. (c) The CaCO_3 content [31]. (d)–(i) Elements and elemental ratios (this study). (j) mean grain size [31]. (k) sedimentation rate (summarized by [19, 29, 31, 83]), and the dotted line during stage III represents the average sedimentation rate. (l) Oxygen isotope of *Uvigerina* spp. [31]. (m) Sea-level curves for global change, represented by the blue line [84], Mekong River Estuary, represented by the pink line [85], and Sunda Shelf represented by the orange line [86]. Last Glacial Maximum (LGM) and Holocene are marked in green and blue shades, respectively [87, 88]. The dashed lines are the boundary between each evolutionary stage. LGM: Last Glacial Maximum. “+” and “-” in two sides represent increase and decrease, respectively.

FIGURE 8: Correlation plots for TOC vs. CaCO_3 of samples from core 973-4. Colored patterns represent the data in different stages over the past 42 kyr.

increases with weathering due to more liable nature of plagioclase relative to K-feldspar [89]. Therefore, higher ratios of Rb/Sr and K/Na are indicative of intense chemical weathering. As discussed above, relatively higher Rb/Sr and K/Na ratios in core 973-4 are displayed during the glaciation, especially LGM; but lower ratios are observed in the early Holocene, afterward, the ratios gradually increase since middle Holocene (Figures 7(f) and 7(g)). The variability in Rb/Sr and K/Na ratios confirms that the sediments at core 973-4 had experienced a distinct change in chemical weathering over the past 42 kyr, and we attribute the controlling factors on weathering to the climatic alternation and sea-level variations. The rates of chemical weathering and physical erosion are often coupled, and higher chemical weathering rates are always together with rapid erosion [90]. Enhanced erosion is marked by global cooling due to the increased atmospheric CO_2 [91]. Moreover, Kump [92] presented a clear correlation between chemical erosion rates and river runoff, and as a result, higher chemical weathering intensity in low sea-level period was potentially associated with the strong hydraulic condition. To adequately explain the link between weathering rates and climatic conditions is really a hard work; additionally, the interference from Taiwan makes it more tough to define the substantially erosion rates, monsoon strength, and intensity of chemical weathering in the investigated region [93].

CaCO_3 is an indicative proxy for ocean productivity [94], whereas its content is generally low in deep ocean owing to the dissolution process [95]. Core 973-4 lies above the current carbonate compensation depth (CCD) of about 3400 m [96], so weak dissolution of CaCO_3 in study area is likely. To eliminate the detrital dilution effect, the marine biogenic proxies (TOC and CaCO_3) are normalized to Al concentration. For core 973-4, consistency in vertical variations between Al normalized ratios (TOC/Al and CaCO_3 /Al) and

their biogenic contents is illustrated spanning the last 42 kyr, which suggests that the dilution effect can be neglected (Figures 3(n) and 3(o)). Recently, Cartapanis et al. [97] reported the global organic carbon burial in marine sediments over the last 150 kyr, and the result showed that higher accumulation rate of organic carbon was observed during the glacial period relative to the interglaciation. Core 973-4 showed the similar variability over the past 42 kyr (Figure 7(b)), with higher values during the LGM and deglacial period and lower values during the interstadial and Holocene. Interestingly, the content of organic carbon deviated from the productivity records, e.g., CaCO_3 . The temporal variations of TOC and CaCO_3 displayed polar trend over the studied time scales, indicating a decoupled relationship between organic carbon burial with ocean productivity.

As illustrated in Figure 8, there is a quite excellent negative correlation between TOC and CaCO_3 content on orbital time scales with some exceptions for abnormal values within the turbidite-influenced layer. During the cooling period with low sea level, higher TOC content was ascribed to the increasing terrigenous input, accompanied by low content of CaCO_3 , which was related to the low temperature cooled by intensified winter monsoon. In contrast, the synchronous concurrence of higher CaCO_3 content and lower TOC content was exhibited during the warm episode with sea-level highstand. This phenomenon may be environment-specific and has been proved in much of the marginal seas [58, 97, 98].

Trace metal records in sediment are good indicators for redox condition in oceanic systems, and these metal concentrations are often depleted under well-oxygenated conditions and accumulated in sediment under low-oxygen conditions [99]. In oxic conditions, V (V) and Cr (IV) are soluble phase in aquatic systems, while in the euxinic conditions, reduced V (III) is enriched in the sediments [99]. Jones [100] proposed that the V/Cr ratio is sensitive to the redox change; generally,

higher V/Cr ratios suggest a more reduced condition in the sediment-water interface. Moreover, Mn is also a redox-sensitive and labile element. Under oxygen-enriched conditions, oxidized Mn (III) and Mn (IV) as the insoluble phase are accumulated in the sediment; however, under oxygen-limited conditions, it will be reduced to the Mn (II) existed in the aquatic systems [101]. Hence, the higher Mn/Al ratios suggest an oxygenated condition, and lower Mn/Al ratios suggest a low-oxygen environment. During the glacial stage with low sea-level stand, oxygenated conditions in the water-sediment interface documented by higher Mn/Al and lower V/Cr ratios were displayed, such an environment was not favoring for organic carbon preservation. After entering the Holocene with sea-level highstand, a gradual shift to reducing environment occurred in the seafloor as indicated by lower Mn/Al and higher V/Cr ratios. Bottom water redox condition was controlled by the deep-water ventilation induced by the sea-level change [58]. Low sea-level stand was more likely to promote the vertical and advective water circulation, resulting in higher bottom water O₂ concentrations, but in sea-level highstand, weakening of vertical water mixing led to low-oxygen conditions and generated a relatively enclosed system in deep ocean.

Based on these records, we recognized four conceptual stages of evolution of the paleoenvironmental change at core 973-4 in the northeastern SCS throughout the last 42 kyr, and each proxy is clearly correlated to the sea-level variations (Figure 7).

4.3.1. Stage I: Weak Warm Period in Last Glaciation with Slightly Low Sea-Level Stand (42.4–31.8 kyr). During 42.4–31.8 kyr, which was an interstadial and belonged to a special period characterized by a weak warm episode during the last glaciation, sea level was low below -80 m. The core 973-4 was typical of stable Al and Ti content, accompanied by decreasing $\delta^{13}\text{C}_{\text{TOC}}$ values (Figure 7(a)), indicating an accelerated contribution of organic matter from terrestrial origin. Meanwhile, the oxygenated condition in bottom water was exhibited by high Mn/Al ratios and low V/Cr ratios compared to other stages (Figures 7(d) and 7(e)), which was not favoring for TOC preservation, resulting in low TOC content (Figure 7(b)). Due to sensibility to the redox conditions in the process of diagenesis, the burial efficiency of organic carbon is still questioned [58]. The biogenic CaCO₃ recorded in sediments showed moderate to high values, which demonstrated a relatively high paleo-productivity during this stage (Figure 9(a)). High CaCO₃ content indicated that the weak warm climate during the interstadial increased the seawater temperature and further promoted the calcareous biological activity. The decoupled relationship between organic carbon and ocean paleoproductivity has been confirmed above.

4.3.2. Stage II: Cold Period with Significantly Sea-Level Drop (31.8–20.4 kyr). The episode of 31.8–20.4 kyr belonged to the glacial period. When sea level dropped below -120 m during the Last Glacial Maximum (LGM) in the late phase of this stage, the abrupt environmental change was showed from the sedimentary records. The clearly higher Al and Ti content, especially during the LGM, indicated a greatly high terrige-

nous sediment input to the core 973-4 off the Taiwan Shelf, which was also demonstrated by the markedly high sedimentation rate (Figure 7(k)). Extremely low sea-level stand shortened the distance between the core site location and paleo-Taiwan river mouth during the last glacial period, resulting in an increasing flux of fluvial sediments to the lower continental slope (Figure 9(b)). The fluctuated $\delta^{13}\text{C}_{\text{TOC}}$ values and low C/N ratios supported an additional contribution from marine organic matter (Figure 6). A shift to weak oxygenated condition relative to the stage I in a water-sediment interface documented by redox-sensitive elemental ratios was still not favorable for organic matter preservation. However, the enhanced terrigenous inputs brought large amount of terrigenous debris matter (TDM) to the core location; consequently, the TOC content during this stage was high (Figure 7(b)). The oxygenated condition was a possibility that the low sea-level stand induced a strong deep-water ventilation and then led to higher O₂ concentration in the bottom water. Additionally, the higher CaCO₃ content was apparently interrupted, and its low values were synchronous with the high TOC values. Owing to the rapid sea-level drop and the polar front entrance from the North Pacific to the SCS, the southwest SCS passage was closed, which caused a significant decrease of the seawater temperature [24, 102]. As a result, low temperature produced low productivity in the ocean.

4.3.3. Stage III: Alternation of Cold to Warm Period with Sea-Level Rise (20.4–13.9 kyr). During the 20.4–13.9 kyr, this stage belonged to the late glacial period and the overall deglaciation with stepwise sea-level rise. Nevertheless, a significant turbidity current occurred during the late period of this stage and then disturbed the in situ sedimentary layer. As a result, sedimentary records exhibited abrupt changes, and the original information within sediments was covered up by the extraneous materials. Therefore, it is impossible to reconstruct the paleoenvironment during this interval. However, we can infer some potential information from the variations of several geochemical parameters. It is noteworthy that Al displayed an obviously decreased trend after the occurrence of turbidity current (13.9 kyr) compared to that in the late phase of stage II (20.4 kyr), implying a potential decline in terrigenous sediment input to the lower continental slope in the northeastern SCS, which may be associated with the rapid sea-level rise (Figure 7(m)). Continuous sea-level rise resulted in most terrigenous sediments to be deposited in the proximal area on the inner continental shelf rather than the outer continental slope [16]. The elevated $\delta^{13}\text{C}_{\text{TOC}}$ values indicated an increasing marine-sourced sediment input during this period; moreover, the content of CaCO₃ reached a peak due to the increasing abundance of foraminifer. As reported by previous research [103], calcareous foraminiferal fauna could contribute to about 55% of total CaCO₃ in the marine environment with the water depth shallower than CCD. Accordingly, further evidence was concluded to demonstrated that turbidity current brought Ca-enriched oceanic materials to accumulate in the studied area. Therefore, the sea-level fluctuations had a dominant influence on terrigenous sediment input and sources of organic matter in core 973-4.

FIGURE 9: Schematic cartoons showing the terrigenous input, organic carbon burial, redox condition in the water-sediment interface, and ocean productivity at core 973-4 located in the northeastern SCS at the (a) interstadial phase during the last glaciation with low sea level (stage I), (b) period of glaciation with the lowest sea level (stage II), and (c) Holocene in sea-level highstand with warm and humid climate (stage IV), with the exception of the stage III influenced by the turbidity current. The size of shallow green arrows indicates the flux of terrigenous input. Core 973-4 is labeled as a red pillar. The ocean productivity in the surface water is expressed by bottle green circles, and higher productivity as indicated by more circles represents the enhanced biogenic CaCO₃ input to deep sea and the seafloor. Dashed area with color from blue to white represents dissolved O₂ content from low to high in the ocean documented by the phase of manganese.

4.3.4. Stage IV: Cold to Warm and Humid Holocene with Further Sea-Level Rise until the Modern Coastline (13.9 kyr—Present). The climate began to be warm and humid since 13.9 kyr, except for several transient cold events, such as the Younger Dryas events (YD) in the early phase of this stage. The sea level gradually rose and reached to the present-day highstand at around 8 kyr [104]. As discussed above, the terrigenous sediment input responding to the sea-level change has been well documented by the variability of Al and Ti. Since the middle Holocene, coastline gradually reached to the modern level, and large amount of sediments

discharged from Taiwan-sourced rivers were restricted to the estuarine areas and inner shelf. Lower flux of terrestrial input resulted in low TOC content, although a gradual change to oxygen-depletion environment supported by higher V/Cr and lower Mn/Al ratios, such a condition was a favorable condition for TOC preservation. The reducing condition was attributed to the relatively enclosed system in the bottom water resulted from the high sea-level stand (Figure 9(c)). Enhanced TOM was contributed to the core location in the early Holocene, but elevated MOM was supplied since the middle Holocene as suggested by the gradually

increasing $\delta^{13}\text{C}_{\text{TOC}}$ values (Figures 6 and 7(a)). During the early Holocene at around 10 kyr, a significant drop of CaCO_3 content (Figure 7(c)) might indicate a carbonate minimum event (CM), featuring a pattern with a rapid decrease following by a slow increase in the CaCO_3 content, while the TOC content varied inversely. The decoupled relationship between TOC and CaCO_3 reminded us that organic matter preservation is not a simple function of ocean productivity [58].

Since the middle Holocene, stable sea-level stand exerted little effect on sediment flux into the sea, but the Al and Ti content increased slightly. Meanwhile, the chemical weathering also increased as indicated by K/Na and Rb/Sr ratios (Figures 7(f) and 7(g)). Therefore, we proposed that the effect of rapidly strengthening East Asian summer monsoon (ESAM) overwhelmed that of the stable sea level and dominated the environmental changes in the northeastern SCS during the Holocene with sea-level highstand.

5. Conclusion

In summary, a detailed sedimentary analysis for core 973-4 collected from northeastern SCS spanning the past 42 kyr suggested that sea-level changes dominated the deposition history. Due to warm seawater temperature and relatively low sea level, the dissociation of gas hydrated reservoir caused a significant turbidity current at around 14 kyr. The organic matter was primarily from terrestrial supply as indicated by negative $\delta^{13}\text{C}_{\text{TOC}}$ values and higher TOC/TN ratios, and Taiwanese rivers were considered to be the dominant sediment provenance. We recognized four distinct stages responding to sea-level change on the basis of geochemical variations recorded in sediments: stage I (42.4–31.8 kyr) was characterized by low sea-level stand with weak warm climate, lowstand promoted the water ventilation and then induced the oxygenated condition in bottom water, which was not favoring for the organic matter burial, accompanied by consistent terrigenous input; during the period of stage II (31.8–20.4 kyr), the prominently high terrigenous input to the study area was presented by high Al and Ti content due to a short distance from the paleo-Taiwan river mouth to core location when the sea level stood below -120 m, the large TDM input resulted in the increasing TOC content though the redox condition in water-sediment interface was still oxygenated, while the CaCO_3 content decreased during this stage, and the contrary pattern showed that ocean productivity was decoupled from organic carbon burial; the turbidity current occurred during the late period of stage III (20.4–13.9 kyr), which brought large amount of Ca-enriched oceanic materials and subsequently disturbed the original sedimentary sequence, and we inferred that the terrigenous input gradually declined due to stepwise rise in sea-level stand as showed by potential decrease of Al content; during the episode of stage IV (13.9 kyr–present), high sea-level stand promoted a reducing environment in the bottom water because of weak ventilation with well-oxygenated surface water. Warm climate induced the enhanced productivity while the TOC content was low. Since the middle Holocene, the variations of terrigenous input, redox condition, and

chemical weathering were mainly controlled by East Asian summer monsoon when sea level reached to the modern coastline.

Data Availability

The data used to support the findings of this study are included within the manuscript and the supplementary materials.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

Authors' Contributions

Bin Wang and Huaiyan Lei contributed equally to this work.

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Supplementary Materials

Supplementary material contains Figure S1 and Figure S2, which show the sediment provenance derived from REE compositions. (*Supplementary Materials*)

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