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A paleoenvironmental record during 7～21 Ka BP in the sediments off northeastern Taiwan

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Abstract: A sediment core collected from the western slope of the South Okinawa Trough off northeastern Taiwan recorded the last glacial/postglacial changes of paleoenvironments. The sediments that had higher content of biogenic material ($CaCO_3$ and opal), lower terrigenous /biogenic ratio and higher salinity in the last glacial period suggested lower river water input to the East China Sea shelf. On the contrary, the sediments in the postglacial period contained higher terrigenous detritus and organic matter, and might reflect the warmer and more humid climate or higher river discharge.

1. Introduction

The abundance of chemical elements in recent pelagic clays has been measured by many investigators in order to study their geochemical cycles and processes in the marine environment (GOLDBERG and ARRHENIUS, 1958; KRISHNASWAMI, 1976). The temporal changes of chemical compositions in the sediment are partly due to the diagenesis and partly due to the change in the source functions (e. g., terrigenous detritus, calcium carbonate, hydrogenous ferromanganese oxide, biogenic silica) and are considered to be an useful indicator of the paleo-depositional environment (GOLDBERG, 1961).

The western slope of the South Okinawa Trough off northeastern Taiwan is the transition zone between the shallow East China Sea shelf and the deep Okinawa Trough (Fig. 1). After turning northeastward here, the main current of the Kuroshio flows along the Okinawa Trough. UJIIE *et al.* (1991) proposed that the main course of the present (postglacial) Kuroshio was similar to that of the interglacial period, but the Kuroshio moved to the east of the Ryukyu Island Arc during the last glacial period. Thus, the mineral and chemical compositions of the sediments on the slope may change with flow patterns or the input of the source material.

The purpose of the present study is to obtain the paleoenvironmental information concerning the glacial/postglacial changes based on the variations of chemical elements in the sediments.

2. Material and methods

A piston core was collected from the lower part (water depth: 1480 m; Lat. 25°00′N; Long. 122°42′E; Fig. 1) of the trough slope by R/V Ocean Researcher I on cruise 120 in 1987. The upper 370 cm segment of the core is the target of this study. Sampling was made at approximately 10 cm intervals.

The grain size distribution of the sediment was measured with a Coulter LS-100 Counter. The sediments were prepared for chemical analysis by washing three times with distilled water and then drying for several days at 50 ℃. The desalted sediments were ground into powder in an agate mortar and the total carbon and total nitrogen (TN) contents were measured using an element analyser (LECO-CHN 932) calibrated with the NIST standard material (SRM-2704, C: 3.348 wt% and S: 0.397 wt%) and the LECO standard (EDTA, C: 41.1 wt% and N: 9.59 wt%). Dried samples were combusted at 450℃ for 3 hours to remove the organic carbon and the inorganic carbon contents measured by the element analyser. The inorganic carbon was then subtracted

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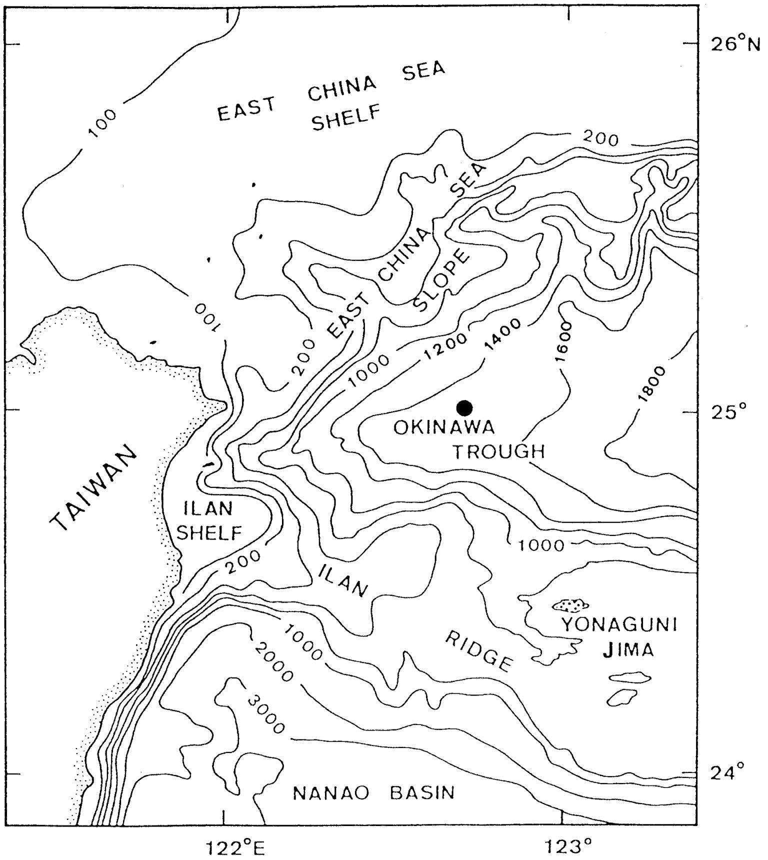

Fig. 1. Location map of the sampling site (●). (modified from Yu and Hong, 1992)

from the total carbon to obtain the total organic carbon (TOC).

For X-ray fluorescence (XRF) analysis, the sediment powder was palletized at 20 tons/30mm dia. to form a cake with cellulose as backing. The contents of 27 elements were measured using the Rigaku RIX- 2000 XRF machine. Eight standard samples, including MAG-1, NIES (no.2), NIST (1646), NIST (2704), BCSS (BD001), MESS (MD001), PACS (PD004) and GBW (07314) were used for calibration. The precisions were within ± 5% for Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Rb, Sr, Zr and Pb; ± 10% for C, N, Ga, Cd and Ba; ± 30% for Ce and As. The carbon- 14 dating of bulk organic matter was done by both the AMS (accelerator mass spectrometry) method at the Institute of Geological & Nuclear Sciences in New Zealand and by the conventional method at the National Taiwan University. The samples were sieved and treated consecutively with hot solution of acid, alkali, and acid before measurement. A conventional radiocarbon age implies correction for isotope fractionation through the normalization on δ^{13}C (PDB) value of-25‰. We subtracted the reservoir correction of 400 years (Bard, 1988) from the conventional radiocarbon age, and then the age was transformed to the calibrated age with the formula of Stuiver and Reimer (1993). The quartz contents were measured using the SIEMENS D5000 X-ray diffraction with a Cu target tube at 40kV, 30mA.

3. Results and discussion

3.1 Chronology and organic matter stratigraphy

The core was dated as between 7 and 21 Ka BP (Fig. 2a), thus the last glacial/postglacial changes could be identified. Piston coring often loses the uppermost loose sediments because of

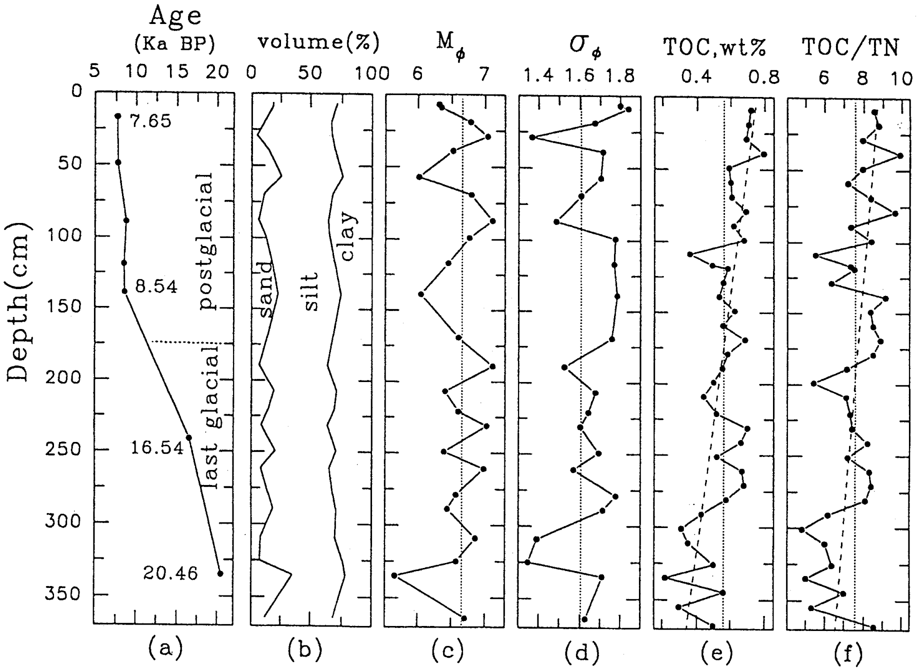

Fig. 2. The profiles of (a) C-14 dates, "●" denotes those measured by the AMS method and "▲" denotes the one measured by the conventional method; (b) sand, silt and clay contents; (c) mean grain diameters (M_ϕ); (d) sorting coefficients (σ_ϕ); (e) TOC (total organic carbon) contents, and (f) atomic TOC/TN (total organic carbon/total nitrogen) ratios. The dashes lines show the mean values and the broken lines show the trends.

wrong setting of corer. But CHUNG and CHANG (1995) studied some box cores and suggested that the surface sediments on the upper and middle slopes were too old to detect the excess ^{210}Pb activities. SHIEH (1993) also found that the age of the coretop sediments was about 5 Ka BP and the sedimentation rates varied in the ranges of 10~46 cm/Ka since 10 Ka BP from another core on the slope. It is possible that there is no recent sediments that cover the study area because of the erosion of bottom currents. If the sedimentation rate was constant in each segment, the rate of the upper 150 cm segment (140cm/Ka) would be about 8 times higher than that below (16.8 cm/Ka). The sedimentation rate of the upper segment is very high and the possibility of deposit by slumping could not be ruled out.

There is little fluctuation in the grain size distribution of the sediments throughout the core. The sediments contain about 55% silt, 30% clay and 15% sand (Fig. 2b). The mean diameter (M_Φ) is in the range of 5.6~7.1 Φ (phi) and belongs to the fine silt class (Fig. 2c). The sorting coefficients (σ_Φ) calculated with the formula of Folk and Ward (1957) are in the ranges of 1.3~1.8, and suggest that all sediments are poorly sorted (Fig. 2d). The coarser sediments show poorer sorting than the finer ones.

The TOC contents and the atomic TOC/TN ratios seem to have slightly higher values in the upper segment than the lower one (Figs. 2e and 2f). Usually, older sediments lose more organic matter because of consumption by oxidation or decomposition by the microbials. Our TOC results (Fig. 2e) are consistent with this trend. The gradual diagenetic decomposition of sedimentary organic matter with preferential loss of nitrogen (RAISWELL and TAN, 1985) also should have resulted in a gradual increase in the C/N ratios with depth in the Recent sediments but Fig. 2f shows a reverse trend. The reason is as follows:

The C/N ratios of the marine phytoplankton are about 6. The TOC / TN ratios of the

Table. 1 The elemental composition of the sediments

element	range	average	(1)	(2)	element	range	average	(1)	(2)
TC(%)	1.28–1.67	1.41	1.82	3.94	Mn(ppm)	377–471	434	520	2020
TOC(%)	0.22–0.79	0.55	0.61	0.97	Fe(%)	2.88–4.51	3.72	3.15	3
TN(ppm)	510–1100	870	615	1130	Ni(ppm)	38–50	42	25	36
Na(%)	1.19–1.76	1.43	1.33	1.93	Cu(ppm)	30–52	41	14	26
Mg(%)	1.37–1.52	1.47	1.07	1.15	Zn(ppm)	77–111	99	66	82
Al(%)	6.83–8.21	7.91	5.31	5.93	Ga(ppm)	15–21	18	13	14
Si(%)	26.7–28.5	27.3	29.2	19.2	As(ppm)	nd*–14	7	8	9
P(ppm)	500–563	525	500	630	Br(ppm)	12–44	26	13	39
S(ppm)	748–1394	1051	472	730	Rb(ppm)	101–143	127	93	87
Cl(ppm)	600–7400	2800	3000	5700	Sr(ppm)	160–208	180	269	415
K(%)	2.11–2.59	2.47	1.82	1.83	Zr(ppm)	178–397	220	180	120
Ca(%)	2.42–3.80	2.87	4.15	11.4	Cd(ppm)	nd*–4	2	0.07	0.16
Ti(ppm)	3800–4200	4000	3500	3100	Ce(ppm)	56–101	79	63	46
V(ppm)	90–121	107	71	78	Ba(ppm)	391–557	472	396	440
Cr(ppm)	70–95	81	61	58	Pb(ppm)	46–87	58	21	27

(1) Taken from Zhao (1983) for the East China Sea
(2) Taken from Zhao and Yan (1994) for the Okinawa Trough
*nd ≦ 1 ppm

sediments in the core are mostly between 5～9, similar to those of the marine phytoplankton (KNAUER *et al*., 1979; CHEN *et al*., 1996). However, the trend of the TOC/TN ratios, decreasing with depth rather than increasing (Fig. 2f), is opposite to the effects of early diagenesis and seems to indicate the change of organic matter sources. The higher TOC/TN ratios in the upper segment of the core may reflect the slight increase of the terrigenous organic matter input in the postglacial period. MULLER (1977) found that in the case of most of continental margin sediments with 1～4% organic carbon, clay-bound nitrogen substances including inorganic ammonium does not significantly affect the C/N ratios. But in some deep sea sediments containing little organic matter as in our case, the effect of inorganic nitrogen on the total nitrogen is drastic and cannot be neglected as inorganic ammonium may represent 20～45% of the total nitrogen. Because the end product of the TOC decomposition may be lost to the overlaying water, yet the end product of decomposing organic nitrogen is bound to the clay particles, the TOC/TN ratios may decrease to below 6 (Fig. 2f).

3.2 Distribution of elements

The sediments deposited on the south East China Sea shelf are characterized as coarse-grained, mud-free, with high carbonate contents and shell fragments. Quartz and feldspars are abundant in the shelf sand which are primarily discharged from mainland China (CHEN *et al*., 1992). The terrigenous material of the muddy sediments in the South Okinawa Trough is also mainly derived from mainland China through the shelf but partly from Taiwan and the Ryukyu Island Arc (LIN and CHEN, 1983). These terrigenous and local biogenic material may control the elemental composition in the sediments.

Table 1 shows the abundance of the chemical elements in the core, and in the surface sediments of the East China Sea shelf (ZHAO, 1983) and the Okinawa Trough (ZHAO and YAN, 1994). The contents of most elements in the core are similar to those on the East China Sea shelf and in the Okinawa Trough. The major chemical components in the sediments are SiO_2 (59%), Al_2O_3 (15%), $CaCO_3$ (7%), Fe_2O_3 (5%), K_2O (3%), MgO (2.5%), Na_2O (2%), total organic matter (1.5%) and TiO_2 (0.7%).

The correlation coefficients of elements are shown in Table 2. Aluminum is a typical element from the aluminosilicate minerals and is considered as of terrigenous origin. Mg, K, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Ga, Br, Rb and Ba all show strong positive correlation with Al, suggesting that they are essentially of terrigenous origin. Calcium and Sr are mainly from biogenic origin in the East China Sea (ZHAO and YAN, 1994) thus Sr has a high correlation coefficient (0.91) with Ca, and both have strong negative correlation with the terrigenous elements. Although Si is largely from the aluminosilicate minerals, it has strong positive correlation with the biogenic elements (Ca and Sr) and has negative correlation with the terrigenous elements. The likely explanation is that the variation in the biogenic silica content is larger than the differences in the terrigenous silicate. As a result, the fluctuation of Si behaves as a biogenic element.

The TOC, TOC/TN ratio as well as the terrigenous elements have strong negative correlation with the biogenic elements such as Ca and Sr (Tab. 2). It may indicate that higher TOC amount was supplied by the land plants. The Al content decreases with depth, but the ratios of Sr, Ca and Si to Al increase with depth (Figs. 3a and 3b). It seems to reveal a higher detritus input and lower biogenic content in the postglacial period compared with the last glacial period.

Total silica is composed of a minerogenic/aluminosilicate component and a biogenic component. The large variation in the Si/Al ratios of the sediments seems to indicate the changes of these components or their sources. ZHAO (1983) proposed that the high Si/Al ratios were induced by the high quartz contents in the coarse sediments of the East China Sea. The input of volcanic glasses or eolian dust with high quartz content may also increase the Si/Al ratio. However, there are less than 5 wt% changes in quartz, equaling abut 2.6 wt% changes in Si in the sediments. In addition, the higher Si/Al ratio in the lower segment of the core is not associated with the coarser material or higher quartz content (Figs. 2c and 3c), thus the higher ratio may be a result of an increase in the biogenic opal input during the last glacial period. Diatoms are the major source of biogenic opal in the marine sediments thus biogenic opal can be a proxy of local surface water productivity because its content in the sediments is an indicator of surface nutrients. Therefore, local paleoproductivity can be reconstructed with biogenic opal content in the sediments (BROECKER and PENG, 1982; CHARLES *et al.*, 1991). The portion of biogenic silica (opal) was determined on the basis of the assumption that the Si/Al ratio in the aluminosilicate fraction of the sediments is about 3 (LEINEN, 1977). Thus we subtracted three times of Al content from the total Si and obtained the biogenic Si content. The contents of biogenic silica were calculated as between 5 and 17 wt%, higher during the last glacial period (Fig. 3d).

Assuming that Ca is mainly from the calcium carbonate, the content of $CaCO_3$, ranging between 7 and 10 wt% (Fig. 3d), is very close to the result from the measurement of inorganic carbon. The content of the total inorganic biogenic material (opal and calcium carbonate combined) ranges between 11 and 26 wt% and increases with depth (Fig. 3d). The T/B (terri genous/biogenic) ratio rises gradually from around 4 at the bottom to ca. 8 at the surface (Fig. 3e).

3.3 Paleoceanographic significance

During the last glacial period, a growth of the continental ice sheets caused a global drop in sea level by 100～120m (FAIRBANKS, 1989). As a result, large parts of the modern East China Sea shelf were exposed, when the main course of the Kuroshio moved to the east of the Ryukyu Island Arc (UJIIE *et al.*, 1991; AHAGON *et al.*, 1993). But SHIEH and CHEN (1995) suggested that the Kuroshio did not change its path flowing into the Okinawa Trough until about 7.5 Ka BP during the Holocene period. Nevertheless, these authors all agreed that the Kuroshio moved away from the current path during the last glacial period. The change of current patterns would affect the characteristics of the sediments.

The C/N ratios can assist in defining the nature of the organic matter. Although the TOC/TN ratios of the sediments are similar to those of the marine phytoplankton, the close relationships between the contents of TOC, terrigenous elements and the TOC/TN ratio in this core reflect the importance of terrigenous organic material input. If the sedimentation rate was indeed higher in the postglacial period, the higher values of these parameters presented seem to indicate higher terrigenous detritus/organic material inputs because of higher river discharge. We can not decide reliable flux of biogenic materials without reasonable sedimentation rates. The organic carbon and the biogenic opal contents are directly related to the surface productivity (HEBBELN and BERNER, 1993). But the organic carbon in this core is partly mixed with the terrestrial organic material thus is not suitable for calculating the productivity. Higher productivity in the glacial time was found in the northwestern margin of the Pacific (YAN *et al.*, 1991; AHAGON *et al.*, 1993), in the Northeast Pacific (KARLIN *et al.*, 1992), and in the Equatorial Pacific (REA *et al.*, 1991), and was discussed in detail by HONDA (1994). AHAGON *et al.* (1993) considered that there was greater amount of particles and nutrients supplied from the more exposed land-areas in addition to increased production in the Okinawa Trough at that time. During the postglacial period, the sampling site was near the main course of the Kuroshio which was characterized by its deficiency in nutrient and by the low primary productivity, thus contributing little biogenic material content. Meanwhile, the warmer/wetter climate triggers off higher river runoff, therefore more terrigenous organic/inorganic detritus is supplied to the ocean. On the other hand, the Kuroshio moved away during the last glacial period and the more eutrophic near-shore water enhanced the primary productivity which resulted in higher opal and $CaCO_3$ contents, and also lowered the T/B ratio in the sediments (Fig. 3e).

The concentration of Mg in the sediments may reflect the salinity of water in the sedimentary environment. The contents of Mg ion and Mg-containing substances increase with salinity in water (ZHANG, 1992). The Mg/Al ratio averaged 0.183 in the upper segment and

Table. 2 The correlation coefficients among the elements.

	TOC	TN	TOC/TN	Na	Mg	Al	Si	P	S	Cl	K	Ca	Ti	V	Cr
TN	0.74	1.00													
TOC/TN	0.83	0.26	1.00												
Na	0.16	−0.08	0.28	1.00											
Mg	0.61	0.73	0.33	−0.01	1.00										
Al	0.76	0.76	0.49	0.11	0.80	1.00									
Si	−0.75	−0.74	−0.48	−0.21	−0.82	−0.89	1.00								
P	−0.04	−0.17	0.03	0.23	−0.30	−0.26	0.09	1.00							
S	0.02	0.21	−0.09	−0.17	0.16	0.12	−0.03	−0.31	1.00						
Cl	0.38	0.15	0.40	0.90	0.20	0.35	−0.48	0.23	−0.29	1.00					
K	0.73	0.77	0.45	0.07	0.85	0.97	−0.91	−0.34	0.08	0.33	1.00				
Ca	−0.75	−0.72	−0.52	−0.09	−0.81	−0.89	0.85	0.27	0.01	−0.35	−0.93	1.00			
Ti	0.47	0.41	0.34	0.16	0.55	0.74	−0.63	−0.14	−0.06	0.28	0.70	−0.68	1.00		
V	0.58	0.58	0.35	−0.04	0.65	0.78	−0.71	−0.53	−0.09	0.17	0.84	−0.80	0.66	1.00	
Cr	0.64	0.58	0.45	0.14	0.60	0.75	−0.72	−0.02	−0.17	0.42	0.73	−0.78	0.52	0.60	1.00
Mn	0.47	0.42	0.33	0.14	0.45	0.55	−0.61	0.29	−0.10	0.36	0.51	−0.45	0.37	0.26	0.54
Fe	0.64	0.71	0.37	−0.10	0.83	0.75	−0.81	−0.30	0.00	0.17	0.85	−0.91	0.51	0.73	0.62
Ni	0.65	0.54	0.46	0.37	0.44	0.65	−0.70	0.19	−0.16	0.58	0.62	−0.59	0.37	0.45	0.74
Cu	0.56	0.58	0.30	−0.15	0.53	0.65	−0.57	−0.31	0.00	0.15	0.66	−0.53	0.28	0.61	0.54
Zn	0.78	0.75	0.52	0.17	0.75	0.92	−0.87	−0.20	0.03	0.40	0.92	−0.88	0.66	0.79	0.77
Ga	0.53	0.48	0.38	0.18	0.70	0.79	−0.71	−0.24	0.07	0.31	0.80	−0.74	0.58	0.72	0.60
As	0.10	0.23	−0.05	−0.13	−0.01	0.09	−0.04	−0.36	0.38	−0.09	0.10	0.08	−0.17	0.01	−0.13
Br	0.69	0.55	0.53	0.63	0.53	0.68	−0.81	0.13	−0.12	0.81	0.68	−0.70	0.54	0.48	0.65
Rb	0.67	0.77	0.37	0.02	0.67	0.81	−0.65	−0.50	0.35	0.16	0.81	−0.72	0.51	0.69	0.48
Sr	−0.59	−0.56	−0.41	−0.04	−0.75	−0.74	0.77	0.14	0.15	−0.31	−0.80	0.91	−0.59	−0.71	−0.75
Zr	−0.59	−0.57	−0.41	0.01	−0.57	−0.56	0.57	0.24	0.01	−0.21	−0.62	0.61	−0.02	−0.49	−0.56
Cd	−0.01	−0.11	0.08	−0.07	−0.14	−0.24	0.18	0.32	−0.09	−0.02	−0.24	0.15	−0.25	−0.36	0.02
Ce	0.22	0.08	0.23	0.22	0.32	0.15	−0.26	−0.06	0.07	0.20	0.17	−0.11	0.04	0.23	−0.03
Ba	0.58	0.64	0.33	−0.06	0.60	0.63	−0.56	−0.20	0.30	0.05	0.61	−0.54	0.36	0.50	0.40
Pb	0.32	0.37	0.14	−0.25	0.24	0.37	−0.34	−0.06	0.13	0.02	0.36	−0.26	0.00	0.23	0.40

0.188 in the lower segment (Fig. 3f), suggesting higher salinity during the glacial than during the postglacial period. The average Mg/Al raito was 2.7% with a maximum of 10% higher during the glacial period (Fig. 3f). It means that the salinity of the water column might be on the average 1 unit and with a maximum of 3.4 units higher than the postglacial period. SHIEH (1993) suggested that the salinity of surface seawater was as high as 37.8 during the last glacial maximum period because of the low precipitation on land and the low river runoff. The surface salinity of the study site is presently at about 34.5. An increase of 1 to 3 would result in a good agreement with Shieh's data for the last glacial period.

Further, the salinity of the global seawater also increased during the last glacial period when fresh water was extracted from the ocean and was locked up in the form of ice sheets. The present ocean has a mean depth of 3800 meters and a mean salinity of 34.7. The removal of a 120 meter thick layer of water would increase the salinity by about 1.1, in good agreement with our Mg data. The fluctuation of the river discharge might also influence the salinity of surface water. Although the sampling site was closer to the shore during the last glacial period, there was relatively lower fresh water input. As mentioned above, there was higher river discharge during the postglacial period, but the Kuroshio controlled the salinity. According to the distribution of the Mg/Al ratio, we conclude that the variations in the salinity in the South Okinawa Trough was mainly controlled by the global sea-level fluctuations during the glacial/postglacial period.

Table. 2 (continued)

	Mn	Fe	Ni	Cu	Zn	Ga	As	Br	Rb	Sr	Zr	Cd	Ce	Ba
TN														
TOC/TN														
Na														
Mg														
Al														
Si														
P														
S														
Cl														
K														
Ca														
Ti														
V														
Cr														
Mn	1.00													
Fe	0.42	1.00												
Ni	0.56	0.45	1.00											
Cu	0.41	0.48	0.54	1.00										
Zn	0.51	0.75	0.74	0.66	1.00									
Ga	0.45	0.61	0.57	0.48	0.75	1.00								
As	−0.08	−0.03	−0.15	0.41	0.00	−0.15	1.00							
Br	0.58	0.58	0.77	0.32	0.73	0.59	−0.12	1.00						
Rb	0.27	0.61	0.37	0.61	0.75	0.65	0.38	0.46	1.00					
Sr	−0.42	−0.88	−0.60	−0.44	−0.79	−0.66	0.33	−0.63	−0.43	1.00				
Zr	−0.21	−0.60	−0.50	−0.58	−0.59	−0.56	−0.15	−0.39	−0.56	0.53	1.00			
Cd	0.02	−0.17	0.02	−0.05	−0.19	−0.23	−0.06	−0.10	−0.28	0.08	0.05	1.00		
Ce	0.06	0.14	0.14	0.18	0.19	0.31	−0.07	0.20	0.18	−.11	−0.21	−0.32	1.00	
Ba	0.18	0.49	0.38	0.47	0.63	0.55	0.12	0.33	0.68	−0.39	−0.51	−0.23	0.20	1.00
Pb	0.35	0.24	0.35	0.76	0.37	0.07	0.52	0.08	0.33	−0.19	−0.36	0.14	−0.03	0.19

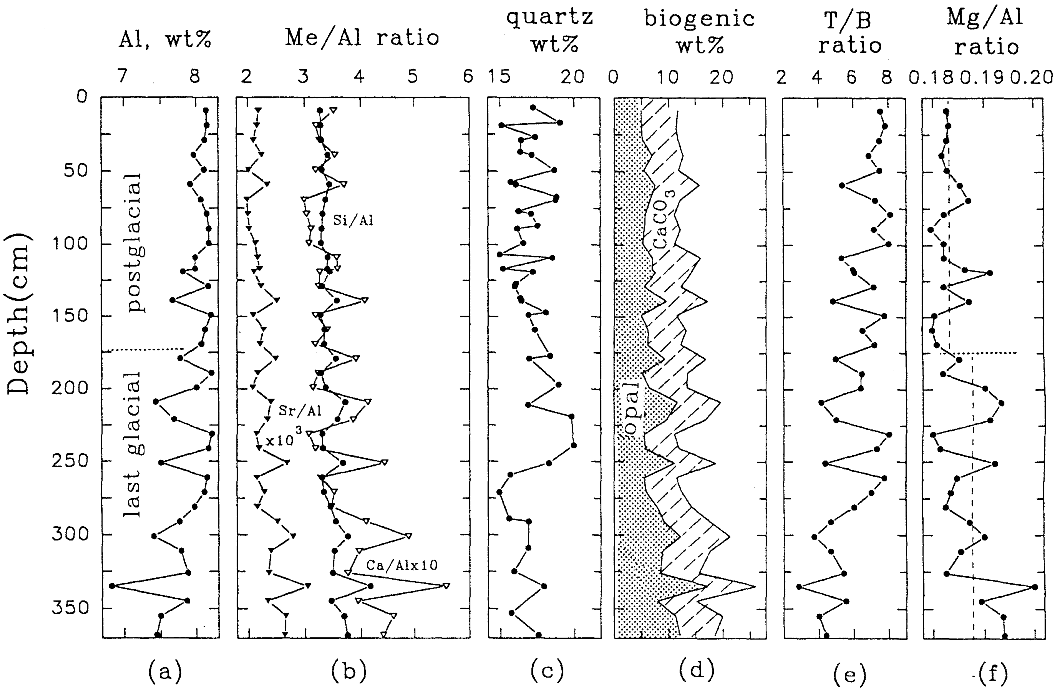

Fig. 3. The profiles of (a) Al ; (b) ratios of Sr/Al, Si/Al and Ca/Al ; (c) quartz ; (d) biogenic material (opal and $CaCO_3$) ; (e) T/B (terrigenous/biogenic) ratio ; and (e) Mg/Al ratio. The broken lines are average values for each segment.

4. Conclusions

The sediment core collected from the South Okinawa Trough revealed the last glacial/postglacial changes. The change in the relative abundance of the terrigenous and biogenic inputs was the major factor that controlled the distribution of these elements. There were about 74~89 wt% terrigenous and 11~26 wt% biogenic materials (including $CaCO_3$ and biogenic silica) in the sediments. The content of biogenic material in the glacial period was about 10 wt% higher than that in the postglacial period.

These data suggest higher terrigenous detritus/organic material input or higher river discharge in the postglacial period. The Mg/Al ratio indicated that the salinity of the seawater was related to the global sea-level fluctuation, and was on the average 1 unit higher in the last glacial period.

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