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Variations in Accumulation of Terrigenous and Biogenic Materials in the Northwest Pacific Ocean since the Last Interglacial Period

SANGMIN HYUN[1, 2], ASAHIKO TAIRA[1], NAOKAZU AHAGON[1] AND SANG-JOON HAN[2]
[1]*Ocean Research Institute, University of Tokyo, Tokyo 164, Japan*
[2]*Marine Geology and Geophysics Division, Korea Ocean Research and Development Institute, Ansan P.O. Box 29, Seoul 425-600, Korea*

Three cores were taken from the northwest Pacific Ocean (Shikoku Basin) to determine the accumulation rates of both biogenic and terrigeneous fractions since the last penultimate interglacial period. The sediment is characterized by large amounts of terrigenous materials with low biogenic fractions and intermittent volcanic-ash layers, suggesting a hemipelagic origin. Composition of major elements shows no significant differences among sites. Relatively small variation of TiO_2/Al_2O_3 ratios with respect to SiO_2 content is the strong evidence for the common origin of terrigenous materials. The fraction of biogenic carbonates varies from near 0% in ash layers to about 35%, with a gradual increase toward the south (St. 4 through St. 6 to St. 20). However, carbonate contents show step-wise increasing tendency from St. 4 through St. 6 to St. 20, which suggests a southward increase of carbonate production. The color reflectance indicates that the sediment of the southern sites contains relatively higher amounts of biogenic carbonates. The mass accumulation rate of terrigenous fractions during the glacial period was 2—3 times higher than that of interglacial period. This enhanced mass accumulation rate of terrigenous materials was concomitant with the high accumulation rate of biogenic fractions. The total sediment accumulation rate is considered as the most important factor controlling mass accumulation rates of the biogenic and terrigenous materials. The enhanced sediment accumulation during the glacial periods is interpreted as a consequence of climate-induced change in the supply of eolian dust from the Asian continent. Enhanced wind strength during the glacial time may have increased transportation of terrigenous materials to the ocean. Thus, variation of sediment accumulation is highly linked with climatic variations.

INTRODUCTION

Hemipelagic sediment provides a good opportunity to investigate processes of sedimentation and detailed environmental changes due to the relatively high sedimentation rate. Even though the continental shelf and slope only comprise about 10% of the world ocean, global environmental change in the world is highly linked with these areas, because the continental shelf and slope are responsible for about half of the world ocean's productivity (Berger *et al.*, 1989), which in turn influences the composition of the ocean sediments (Pye, 1987; Dersch and Stein, 1994). The wind-transportd materials through westerly winds and connected jet stream activities are highly correlated with a recent distribution of the Pacific sediments (Windom, 1975). The hemipelagic sediments, thus, can provide powerful informations on climatic variation during glacial and interglacial periods.

It has been recognized that there exists a striking correlation between the fluxes of biogenic and terrigenous fractions in the sediment trap (Honjo, 1982; Deuser *et al.*, 1983). Furthermore, the biogenic flux, calcareous skeletal remains, and refractory aluminosilicate contents show a strong biogeochemical fluctuation in sediment trap at the circumpolar water of the Drake Passage (Wefer *et al.*, 1982). This strongly indicates that the biogenic and terrigenous fluxes are highly related with the surface water properties such as nutrient concentration and output of biogenic components as well as climatic conditions. Thus, it is necessary to investigate the relationship between terrigenous materials and biogenic components from hemipelagic sediments because they can provide a detailed information concerning the change of climatic and paleoceanographic environments.

In this study, we present the geochemical data from the hemipelagic sediment in the northwest Pacific and estimate the variations of accumulation rates in the biogenic and terrigeneous materials. The variations of accumulation rates of biogenic and terrigenous materials are speculated in terms of their source and relation with climatic and oceanographic variations since the last interglacial period.

MATERIALS AND METHODS

Three piston cores (St. 4, St. 6 and St. 20) were taken from the eastern edge of the Shikoku Basin (Fig. 1) during the cruises KT93-7 and KT92-17 with R/V *Tansei-maru* of the Ocean Research Institute, University of Tokyo, under the project of International Geosphere and Biosphere Program (IGBP). Each sediment core was dissected at 10 cm interval, and sediment subsamples were dried at 105°C for 24 hrs. Before drying sediment samples, sediment color was measured by the Minolta chroma meter, CR-33, for the rapid determination of sediment character. Untreated powdered samples were analyzed using CHN analyzer (Yanakimoto MT-2 type) to produce the total carbon (TC: carbonate carbon plus organic carbon) content. For total organic carbon (TOC) determination, the sedimet samples were treated with 1*N* hydrochloric acid to eliminate inorganic carbon. The amount of inorganic carbon was calculated by subtracting TOC from TC content. The carbonate content of the sediment was expressed as the weight percentage of the bulk sample: $CaCO_3$ (%) = (TC − TOC) × 8.33.

Biogenic silica contents were determined by a time-series dissolution experiment in 2M Na_2CO_3 solution at 85°C following the procedure of Montlock and Froelich (1989). Major and minor element concentrations were determined using X-ray fluorescence (XRF) (Hyun, 1997). To calculate mass accumulation rate (MAR) of each biogenic fraction and major and minor elements, we used linear sedimentation rate (LSR; cm/ka) and dry bulk

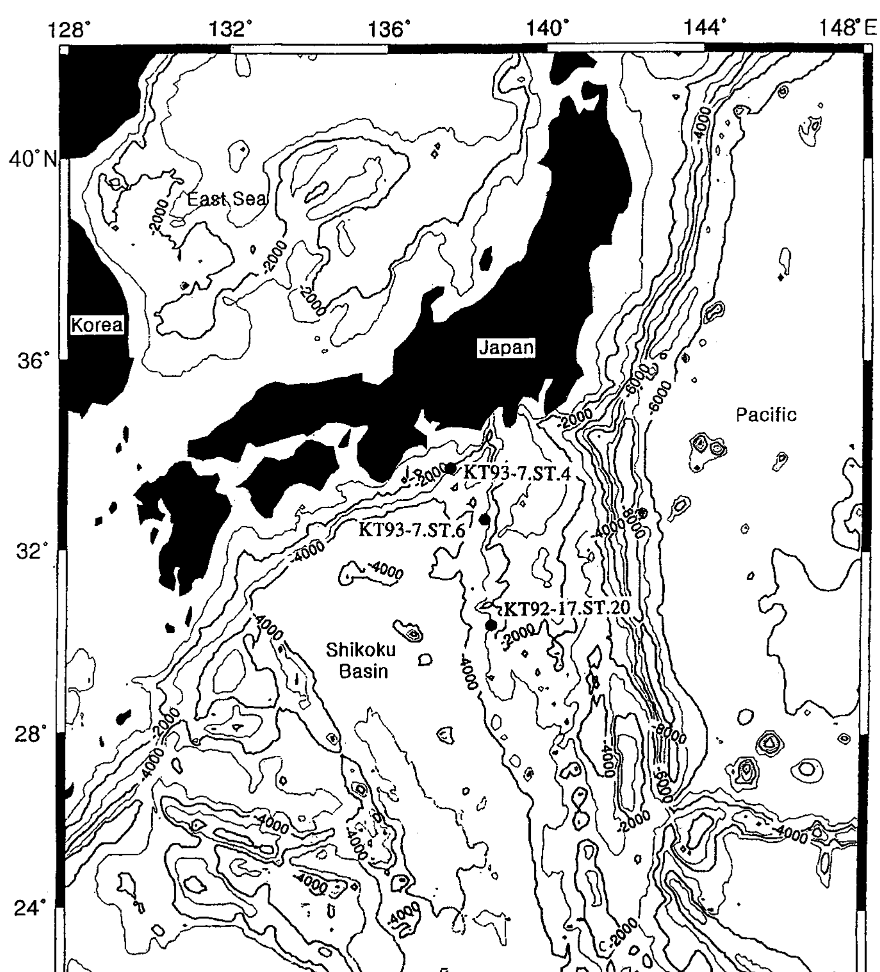

Fig. 1. Bathymetry and three core sites from the northwest Pacific Ocean (Shikoku Basin). The Izu-Bonin Arc is situated to the east of the sampling sites. Contours are in meters.

density (DBD; g/cm^3). The linear sedimentation rate was determined on the basis of stable oxygen isotope and AMS ^{14}C age (Hyun *et al.*, 1996). The mass accumulation rate (MAR) of single sediment component (*e.g.* organic carbon) is calculated as follows:

$$MAR_{organic\ carbon}\ (g/cm^2 \cdot ka) = Organic\ carbon/100 \times MAR_{bulk},$$

where MAR is LSR (cm/ka) × DBD (g/cm^3).

Geological setting and oceanographic conditions

The eastern edge of the Shikoku Basin, where the three piston cores (St. 4, St. 6 and St. 20) were taken, is characterized by relatively high sedimentation rate due to both the steep slope and moderate accumulation of biogenic sediment (Ahagon, 1997). Three cores are located across the Kuroshio pathway along the north-south transect of the Shikoku Basin (Fig. 1).

The Kuroshio current, a major north Pacific western boundary current, is thought to have repeatedly changed its course through glacial and interglacial times, causing significant oceanographic environmental changes in this region (Nitani, 1972; Chinzei *et al.*, 1987). Judging from the sediment character which is thought to be typically hemipelagic, the sediment of this location is suitable for comparing sedimentation rates between the glacial and interglacial periods.

RESULTS AND DISCUSSION

Composition of sediments

From the microscopic smear slide observation, it is revealed that the component of the sediments in three cores are basically alike. The components include terrigenous particles (quartz, feldspar, and clay minerals) of silt and clay size, and biogenic opal. Typical portion of these components is 60—90% of terrigenous particles, 7—35% of carbonate, 2—7% of biogenic opal, and minor component of organic matters, carbonate and intermittently occurred volcanic ash (Hyun, 1997).

The step-wise increase of biogenic carbonate content from the north site (St. 4) to south site (St. 20) is the main character of these sediments (Fig. 2). Decreasing terrigenous materials into the south site is probably attributed to the increased supply of biogenic materials; that is a dilution effect by biogenic materials, especially by biogenic carbonate. The color reflectance of two cores (St. 4 and St. 6) is similar to each other, suggesting a similar compositional characteristics. However, both L* (psychometric lightness) and b* (psychometric yellow-

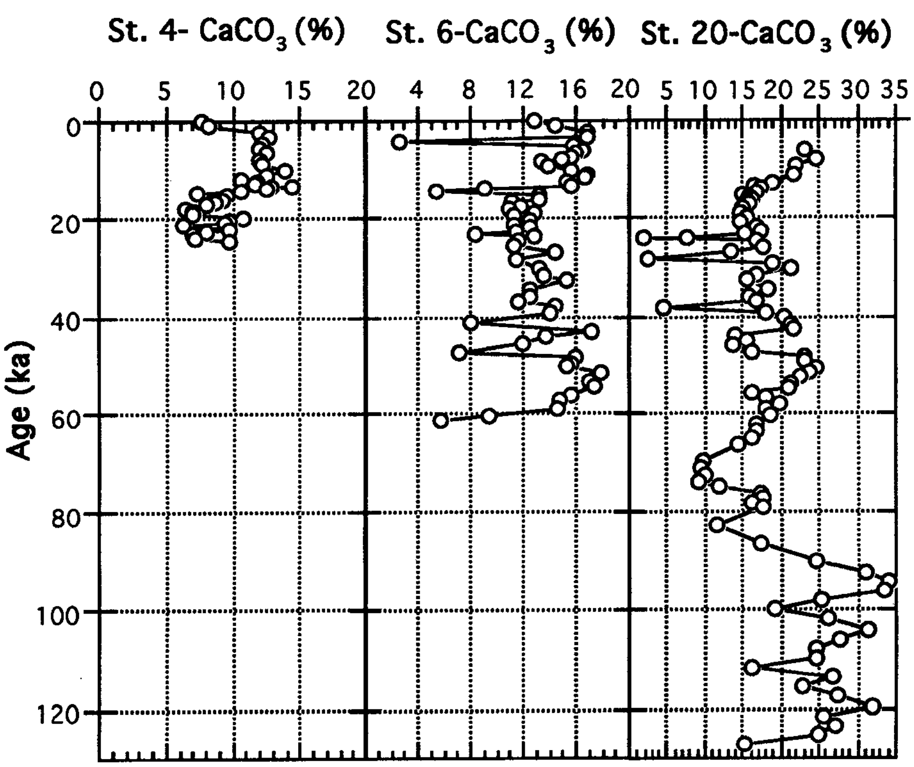

Fig. 2. Biogenic carbonate contents at three cores. The carbonate contents increase stepwise from north to south (St. 4 to St. 20).

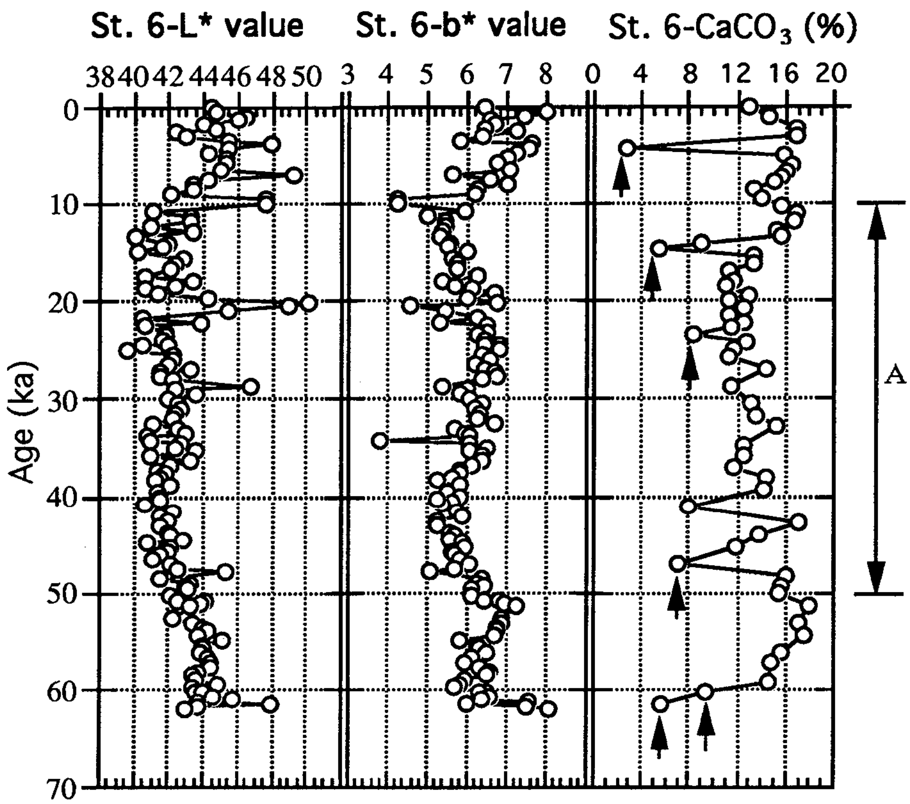

Fig. 3. Profiles of L* and b* values and $CaCO_3$ at St. 6. L* value indicates lightness and b* value yellow-blue chromatiness. Arrows point out the sediment including volcanic ash and scoriaceous materials. The interval A indicates the period of low carbonate content and low L* and b* values.

blue chromaticness) values at St. 6 are slightly higher than those of St. 4 (Hyun, 1997). This strongly supports the increase of carbonate content, which will be discussed in the following section. These indices rely on sediment lightness which is usually determined by the carbonate, organic matter, iron and manganese contents (Nagano and Nakashima, 1992; Mix *et al.*, 1992). As shown in Fig. 3, the high values of reflectance indices of L* and b* are well matched with carbonate content. Especially, the sediment between 10 and 50 ka (Interval A in Fig. 3) has a relatively low carbonate content, and low L* and b* values. This, at least, suggests that both L* and b* values vary in accordance with the carbonate content.

Terrigenous fractions, sedimentation rate and mass accumulation rate

Terrigenous fractions are defined as the difference between the total amount of sediment and biogenic fractions including TOC, carbonate and biogenic opal. The MAR of the total sediment and terrigenous fractions of three cores are illustrated in Fig. 4. The linear sedimentation rates at three cores are illustrated in Fig. 5. The sedimentation rate is 2—3 times higher during glacial period than that of interglacial period. Figures 4 and 5 show a strong correlation between the sedimentation rate and the mass accumulation rate of total sediments. These figures also show a cyclic variation in accumulation rate between glacial and interglacial periods with increasing during glacial period and decreasing during interstadial and interglacial periods.

Glacial period is generally characterized by intense aridification and prevailing westerly winds (Windom, 1975; CLIMAP Project Members, 1976). Thus, the climatic condition of the glacial period is thought to be more favorable for transporting the Asian continental loess to the ocean floors. Therefore, the cyclic variations in biogenic and terrigenous materials during the glacial and interglacial periods are interpreted as the consequence of climatic variation. Large amounts of mass accumulation in terrigenous materials in the cores during glacial period reflect climatic characteristics.

The accumulation rate of biogenic fractions, which will be discussed in the following section, also shows an increasing tendency during the glacial period. Thus, the sediment at this area is characterized by enhanced terrigenous and biogenic materials, and concordant variation during glacial

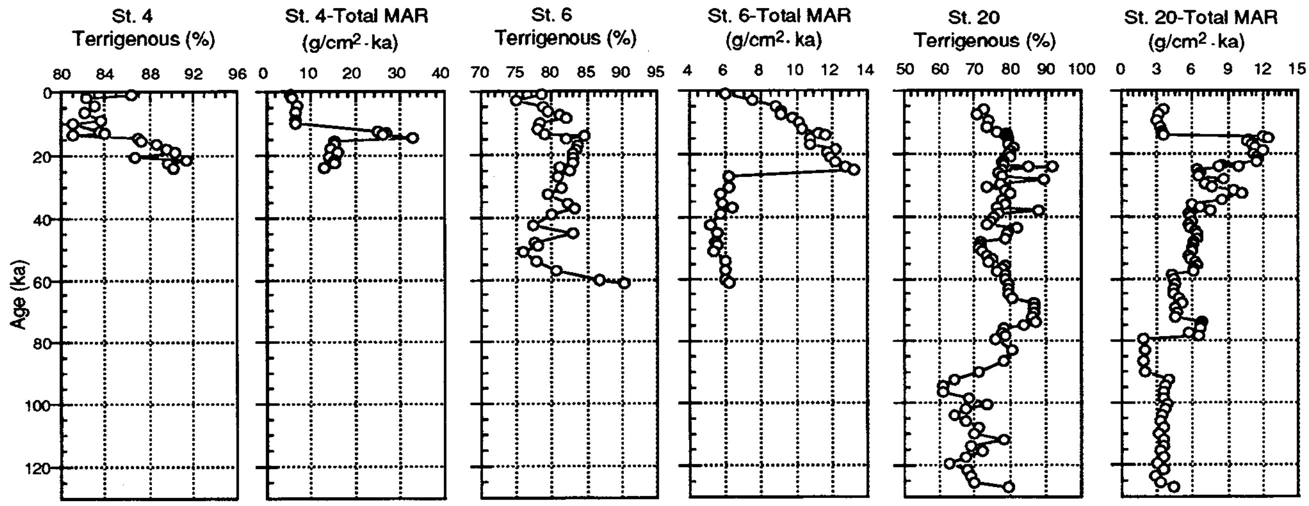

Fig. 4. Total sediment accumulation rates and content variations of terrigenous fractions at three cores.

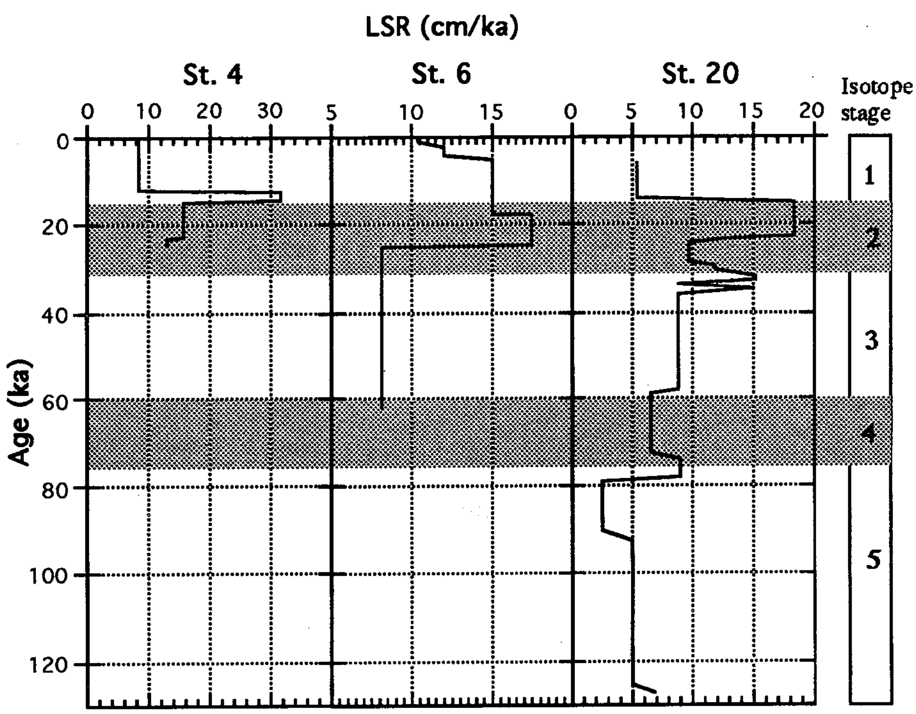

Fig. 5. Average sedimentation rates (cm/ka) of the three cores. Dotted areas indicate glacial periods and LSR is linear sedimentation rate (cm/ka). Ages were determined on the basis of oxygen isotopes and several AMS ^{14}C ages.

period. This is the most important information on the sedimentation processes of the northwest Pacific Ocean (Shikoku Basin). This supports that the accumulation of biogenic fractions in water column is strongly related with the supply of the abiogenic materials due to the increased wind stress, especially during the last glacial period (Ittekkot, 1993).

Biogenic fractions and mass accumulation rate

The downcore variations of biogenic fractions, TOC, biogenic carbonate and opal, and their MAR at three cores are shown in Fig. 6. Figure 6 shows that the total MAR variation acts as a main function of variation in TOC mass accumulation rate.

The nature of organic carbon was examined by C/N ratio and OCSR (organic carbon/sedimentation rate) diagram (Muller, 1977; Stein, 1990). These results indicate that most of organic carbon is considered as marine source since the C/N ratios are below 10 in most sediments. However, between 80 and 100 ka at St. 20, C/N ratio exceeds 10 implying some admixture of terrigenous organic carbon.

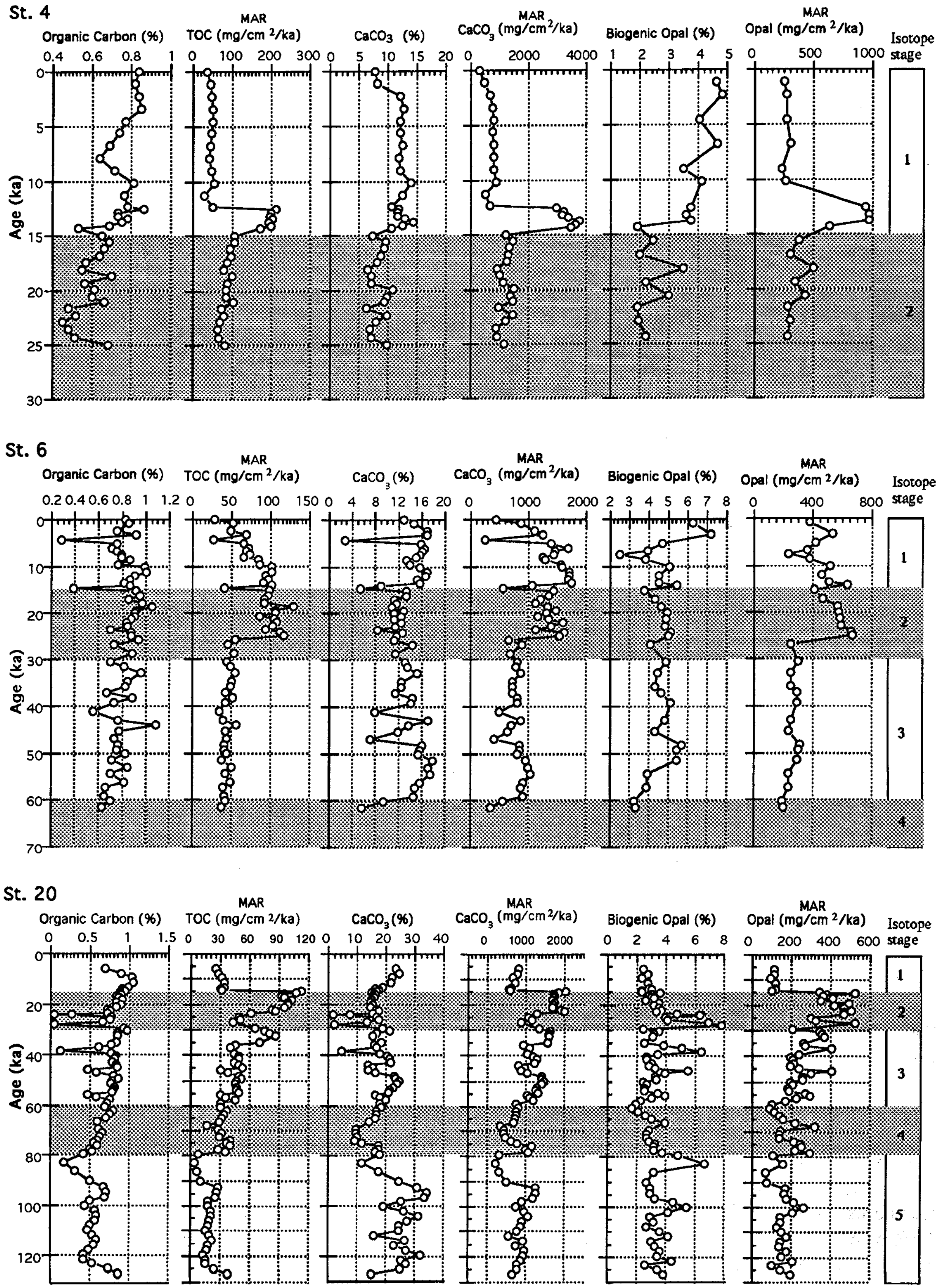

Fig. 6. Downcore variations of biogenic fractions (organic carbon, carbonate and biogenic opal) and their mass accumulation rates at three cores. Dotted areas indicate glacial period.

Detailed information on C/N ratio and OCSR diagram in these three cores can be found elsewhere (Hyun *et al.*, 1996).

A striking increase of carbonate contents is recognized in the lower part (older than 80 ka) of the sedimemt at St. 20. This increase is probably attributed to the increase of surface carbonate productivity because the sampling point is situated above the carbonate compensation depth (CCD) which is usually believed to be approximately 4000 m depth in the Pacific Ocean. High carbonate mass accumulation rate during the last glacial maximum at St. 20 is probably related with both climatic variation and oceanographic condition, such as the Kuroshio Current of this region. A difference in carbonate dissolution between glacial and interglacial periods may be considerable in general, but the entire carbonate dissolution at this site may be not significant (Hyun, 1997).

Carbonate contents at three cores are generally low during glacial time and increase toward the south. Carbonate content in the Holocene is twice higher than that of the last glacial period. There are three possible explanations: (1) an increase of the terrigenous materials and its resultant dilution, (2) lowered carbonate production, and (3) carbonate dissolution effect. However, MAR of carbonate at three cores are characterized by an abrupt change at the boundary of glacial and interglacial periods. The most conspicuous feature of the carbonate variation is a sudden increase around 15 ka at St. 20 (Fig. 5). This sudden fluctuation of carbonate MAR is probably attributed to the change of either calcareous productivity variation, carbonate dissolution rate, or total mass accumulation rate. However, carbonate MAR apparently shows the same patterns as observed in total sediment MAR and MAR of TOC from three cores. Thus, the significant and sharp fluctuation of carbonate MAR is thought to be mainly attributed to change in total sediment accumulation rate.

No significant downcore variation in biogenic opal content is observed from all three cores. The highest value of biogenic opal content occurs around 28 ka, while the lowerest about 60 ka at St. 20. However, as a whole, MAR of the biogenic opal at three cores show a similar trend to those observed in MAR of other biogenic components (Fig. 6). The other two cores (St. 4 and St. 6) also show essentially the same pettern in MAR of each biogenic fraction during both glacial and interglacial periods.

Synthetically, TOC content shows downcore decreases at St. 20 and St. 4, and relatively constant variation at St. 6. However, MAR of TOC, carbonate and opal show conspicuous trend, and are characterized by an increase during the last glacial period. This matches well with the trend of total sediment mass accumulation rate.

Major element concentration and provenance

The important conclusion drawn from the smear slide observation is that the hemipelagic intervals in the three cores are very similar in terms of composition of particles. As shown in Fig. 7, the variation of ratios of the two major elements (TiO_2, Al_2O_3) with respect to SiO_2 clearly indicates that the compositional variations of sediment among three cores are not significant. Since two major elements (TiO_2, Al_2O_3) are considered as typical terrigenous origin (Goldberg and Arrhenius, 1958; Spears and Kanris-Sotirios, 1976; Moorby, 1983), it is believed that the contribution of terrigenous components from three cores is not very significant except for several volcanic ash layers and the period between 80 and 100 ka at St. 20. The extremely narrow range of the ratio (0.040 to 0.045) of the two major elements is a strong evidence for the common origin of terrigenous materials, which is essential information to estimate sediment components and their

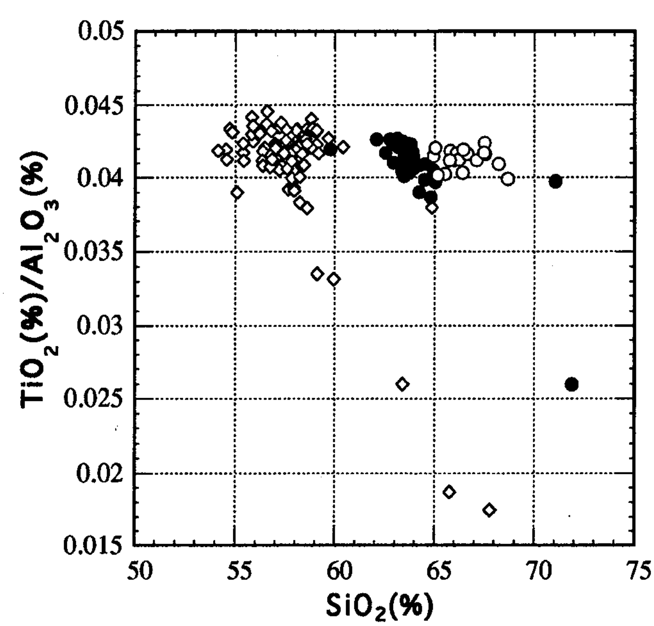

Fig. 7. TiO_2 (%)/Al_2O_3 (%) ratio vs. SiO_2 (%) variation. Open squares, black circles and open circles indicate data from St. 20, St. 6 and St. 4, respectively.

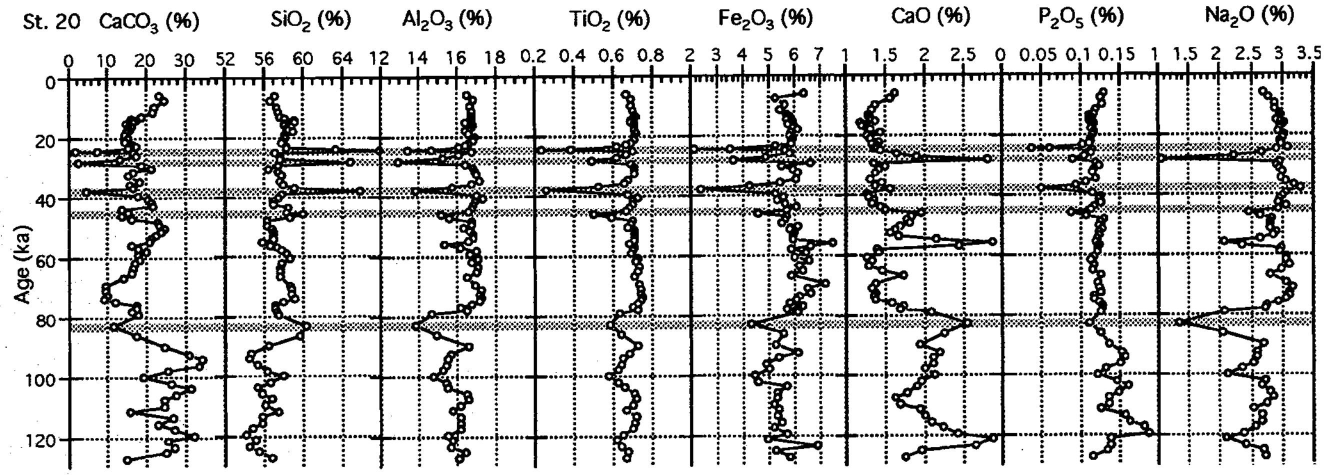

Fig. 8. Profiles of the $CaCO_3$ (%) and seven major elements at St. 20. Dotted areas indicate ash layers.

compositional variation since the last interglacial period.

Increasing carbonate content southward indicates that the increasing amount of aluminosilicate content in north sites is almost terrigenous materials. The SiO_2 content in St. 4 is about 10% higher than that in St. 20, and about 5% higher than in St. 6 (Fig. 7). The contribution of terrigenous materials from Japanese Islands may be significant through the downward transport.

The variation in seven major elements (Si, Al, Ti, Fe, Ca, P, Na) and carbonate content at St. 20 are illustrated in Fig. 8. The vertical profiles of these seven major elements show a slight difference around 80 ka, probably due to the dilution by increasing carbonate content. Increase in carbonate older than 80 ka corresponds to the decrease in SiO_2 and Al_2O_3. Increase of CaO older than 80 ka may also reflect high amount of carbonate contents. However, the source of terrigenous materials would be the same because the TiO_2/Al_2O_3 ratio versus SiO_2 shows a small variation as shown in Fig. 7.

Chemical aspect of eolian dust from Asian continent and resultant climatic changes have been investigated (*e.g.* Rea and Janecek, 1982; Taylor *et al.*, 1983; Rea, 1990). The flux of Chinese loess is directly linked with climatic variation deduced by marine $\delta^{18}O$ records in the north Pacific (Hoven *et al.*, 1989). These previous works clearly demonstrated that the eolian dust is the main source of the sediments in this region and even in the East Sea (Sea of Japan) and the Pacific Ocean. Dersch and Stein (1994) have shown that a dramatic shift to higher eolian sediment supply would be reflected in glacial/interglacial and arid/humid climatic cycles in the East Sea. A comparative study between the component of loess and the selected sediment based on the major and minor elements shows no significant difference in genetic character even though the minor element is not dealt in this paper (Hyun, 1997).

Overall, the concentration of these major elements does not show any distinctive breaking point except for several volcanic-ash layers in turbidite sediment in the eastern part of Shikoku Basin (Nozaki and Ohta, 1993), supporting the idea of the same origin for sediment components. Only the increased carbonate content in the lower part of core at St. 20 probably due to the enhanced carbonate production was likely to affect the sediment lithology.

Relationship among MAR of the total sediment, TOC, $CaCO_3$ and biogenic silica

The MAR of several biogenic fractions is very closely related to each other and the relative proportion of each component is fairly constant, at least, from the last interglacial to glacial period (Fig. 9). The correlation coefficient between MAR of the organic carbon and the carbonate accumulation is 0.76, and 0.64 between MAR of TOC and opal accumulation rate. Figure 9 is plotted for exceptional evidence of volcanic ash layer. Both Figs. 9A and 9B show relatively strong positive correlations among TOC, biogenic carbonate and biogenic opal. This indicates that overall compositional proportion of settling particles stays essentially constant. The variation in MAR of each component through time is thus controlled by total sediment mass accumulation rate, *i.e.*, total flux of particles. This uniform

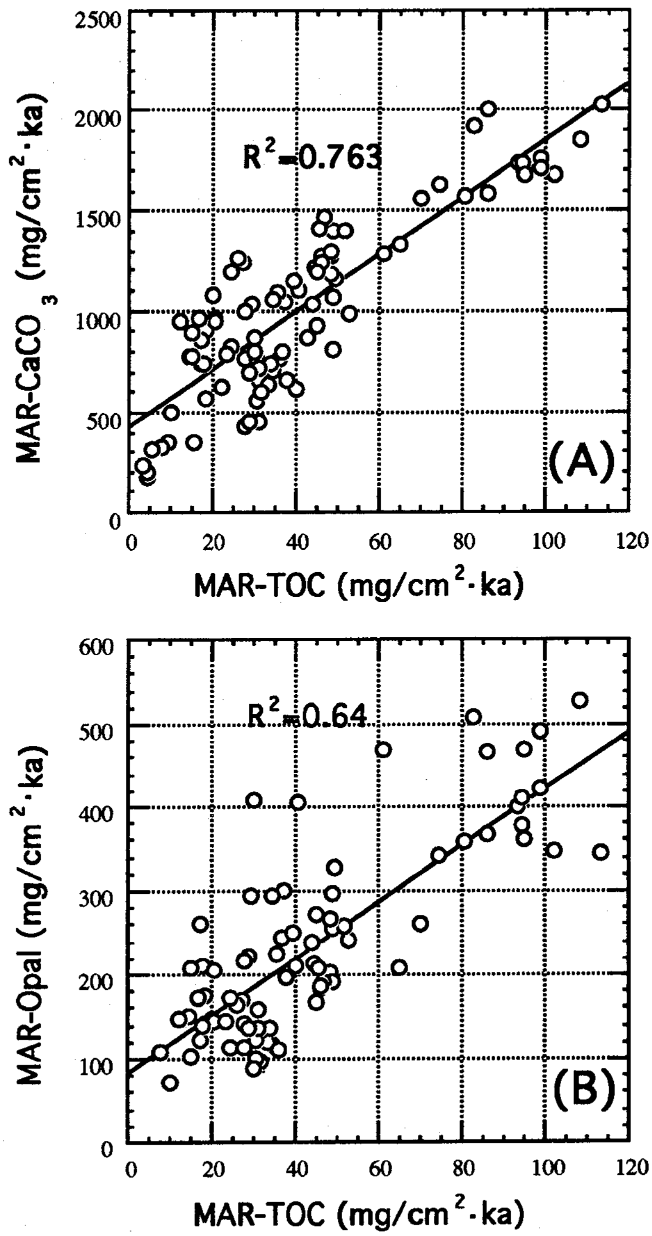

Fig. 9. Correlation of mass accumulation rates (**A**) between carbonate and total organic carbon and (**B**) between opal and total organic carbon.

composition of settling particles provides a unique opportunity to test variation in both biogenic and terrigeneous fractions in terms of overall paleoceanographic environmental change during glacial and interglacial periods.

The variation of biogenic fractions in sediment trap shows a strong relationship with that of terrigenous fraction. As suggested by previous works (Honjo, 1982; Ittekkot, 1993), the flux of biogenic material is highly correlated with the flux of terrigeneous material, and *vise versa*, indicating that the fine particles in water column are removed by aggregation of sinking organic particles. Furthermore, the flux shows a seasonal variation, which implies that sinking particles in water column are primarily controlled by biological activities and climatic variations. The high sedimentation rate is well explained by covariance between the supply of terrigeneous and abiogenic materials since the penultimate interglacial periods.

CONCLUSIONS

The sediments from the northwestern Pacific Ocean (Shikoku Basin) are characterized by high amounts of terrigenous materials and the low amounts of biogenic materials and the volcanic ashes. The vertical distributions of most major elements clearly shows no significant compositional differences among three sites (St. 4, St. 6 and St. 20). Relatively narrow range of TiO_2/Al_2O_3 ratio with respect to SiO_2 is a strong evidence for the origin of the terrigenous materials.

The fraction of biogenic carbonate varies from near 0% in ash layer to about 35% in core bottom at St. 20. However, carbonate content shows step-wise increasing tendency from north to south, which suggests an increase of carbonate production rate in the study area, especially at St. 20.

The mass accumulation rate of biogenic fractions shows distinctive variation between glacial and interglacial periods. The total sediment accumulation rate is considered as the most important factor controlling mass accumulation rate of biogenic fractions. Total mass accumulation rate is two to three times higher during the last glacial maximum than that in the Holocene. This striking difference in mass accumulation rate between glacial and interglacial periods is probably related with glacial-interglacial climatic variation. This pattern of glacial high and interglacial low sedimentation rate is a characteristic of hemipelagic sediment, which is probably related with climate variation.

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