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Past sea surface temperature of the East Sea inferred from alkenone

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We measured the alkenone concentration of bulk sediments from a piston core collected from the Ulleung Basin in the East Sea in order to reconstruct past sea surface temperatures (SST). Sediment ages are well constrained by AMS ^{14}C dates of the planktonic foraminifera *Globigerina bulloides*. Coretop alkenone SST calibration with modern surface temperatures and sediment trap data (Hong *et al.*, 1996) indicate that the SST estimated from alkenones most likely represent the temperatures of late fall. Downcore variations in the alkenone saturation index indicate that between 19 and 15 kyr BP the surface waters were about 3°C warmer than today. Between 15 and 11 kyr BP, the temperatures were about 3°C lower than today. A rapid SST increase of about 3°C occurred at approximately 10 kyr BP. After considering the factors which might influence the SST reconstruction from the $U^{k'}_{37}$ values, we conclude that the alkenone temperature estimates are reliable. The reason for glacial warming in the East Sea is not clear, although there is a possibility that it could be caused by shift in the season of maximum alkenone production from summer during the last glaciation to late fall during the Holocene. Cooling between 15 and 11 kyr BP may be due to inflow of cold water into the East Sea such as via the Oyashio Current or ice-melt water. Warming at the early Holocene could be due to inflow of the Tsushima Current into the East Sea through the Korea Strait.

Key words: paleotemperature, alkenone, East Sea

INTRODUCTION

The East Sea is a semi-enclosed marginal sea connected to the North Pacific and adjacent seas through four shallow and narrow Straits (Fig. 1). The warm waters of the Tsushima Current flow into the East Sea through the Korea Strait and flow out through the Tsugaru and Soya Straits, with a subpolar front bounding the warm waters in the central part of the East Sea (Moriyasu, 1972). The East Sea waters below several hundred meters are considered to be formed offshore Vladivostok by severe cooling in wintertime (Kim *et al.*, 2002). The waters are cold (<1°C) and highly oxygenated (~250 μM) and have uniform salinity (34.1‰).

According to previous studies (Oba *et al.*, 1991, Keigwin and Gorbarenko, 1992), the East Sea experienced large environmental changes from the glacial to the interglacial periods. A global sea level fall of about 120 m during the last glaciation would have restricted the inflow of the Tsushima Current into the East Sea, which might have enhanced cooling of surface seawater. Micropaleontological studies (e.g. Oba *et al.*, 1991) suggest that the East Sea surface waters were colder during the last glaciation than today. The warm-water species of planktonic foraminifera, diatom, and radiolaria were far more abundant during the Holocene period than during the last glaciation, indicating a relatively warm Holocene and possibly a cold glacial period.

Recently, however, Ishiwatari *et al.* (2001) have reconstructed the past sea surface temperatures (SST) in the East Sea using alkenones. According to them, glacial SST in the East Sea was abnormally warmer than today. Planktonic foraminiferal $\delta^{18}O$ records from the East Sea present isotopically lighter values during the last glaciation, indicating glacial warming and/or lowered salinity (Gorbarenko and Southon, 2000). In addition, a close examination of microfossil data shows that the glacial climate condition in the East Sea was not as cold as expected from previous studies. The cold-water species of calcareous nan-

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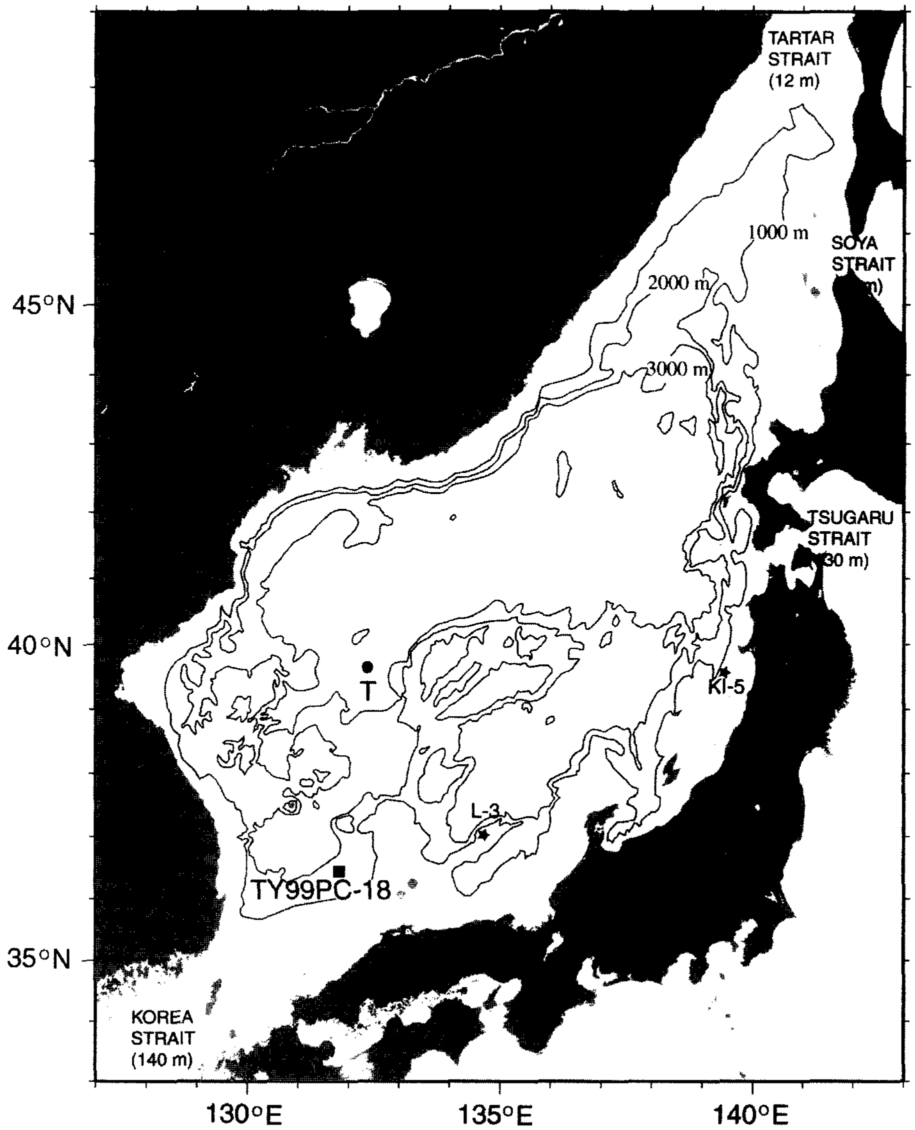

Fig. 1. Bathymetric map of the East Sea with the locations of cores. Rectangle indicates core used for this study. Circle indicates the location of sediment trap from Hong *et al.* (1996). Stars indicate cores from Ishiwatari *et al.* (2001).

noplankton, *Coccolithus pelagicus* and diatom, *Thalassiosira nordenskioldii* disappeared during the glacial period, and they occurred after the glacial period (Oba *et al.*, 1991).

Paleotemperature reconstruction by alkenone is based on the widely accepted hypothesis that some haptophyte microalgae produce long chain (C_{37}) unsaturated ketones whose degree of unsaturation changes with environmental temperature. Since Brassell *et al.* (1986) showed that the relative abundance of C_{37} alkenones is related to variations in sea surface temperature, numerous studies were conducted to verify the applicability of this new technique to paleotemperature estimates. The relationship between alkenone unsaturation index ($U^{k'}_{37} = [C_{37:2}]/([C_{37:2}]+[C_{37:3}])$) and temperature of seawaters in which algae grow has been calibrated using results from laboratory batch culture experiments of *Emiliania huxleyi* (e.g. Prahl and Wakeham, 1987; Prahl *et al.*, 1988), and results of analysis of surface water and sediments trap samples (e.g. Prahl *et al.*, 1993) and coretop sediments (e.g. Müller *et al.*, 1998). These studies suggest that the relationship between $U^{k'}_{37}$ measured in sediments and seawater temperature is generally linear.

In this study we measured the alkenone concentration of bulk sediments in the piston core recovered from the East Sea in order to reconstruct past SST. For age control, radiocarbon dating of single foraminifera species *Globigerina bulloides* was performed. Accurate reconstruction of SST in the East Sea since the last glaciation will help to understand the history of ocean environmental changes in this region.

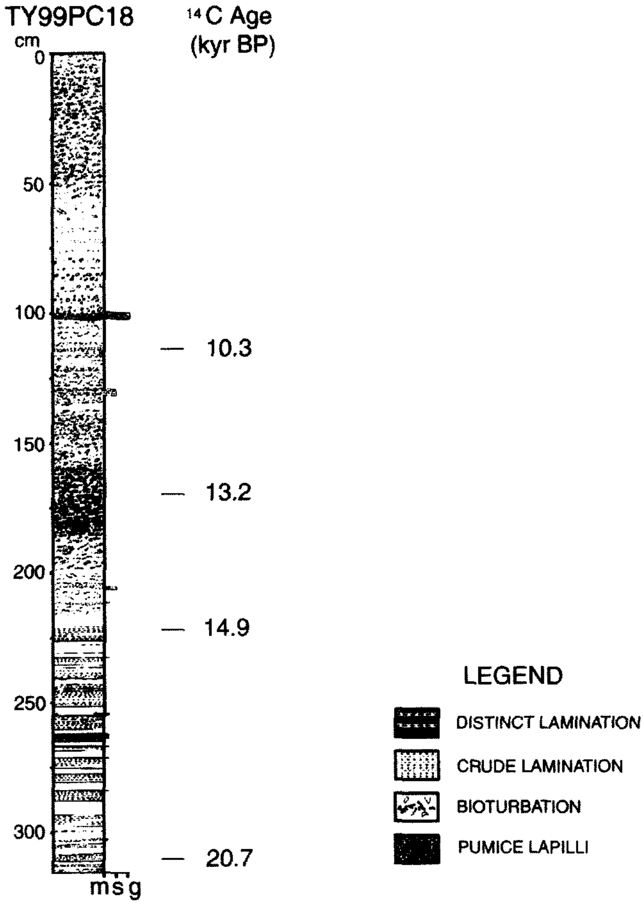

Fig. 2. The columnar section of core TY99PC18 with ^{14}C ages. (m, s, g denote mud, sand, gravel, respectively.)

MATERIALS AND METHODS

Sediment samples

We used the marine sediments of a piston core TY99PC18 (131°39.63'E, 36°29.51'N) recovered at a water depth of 1867 m in the southeastern part of the Ulleung Basin, East Sea (Fig. 1). According to a previous study (Lee *et al.*, 1996), slope failures occurred frequently on the basin slope in the late Pleistocene due to degradation of the regional slope stability by glacio-eustatic sea-level lowering. This caused a wide variety of mass-flow movements on the slope, and triggered sedimentation of laminated turbidites in the deep basin. In this study we investigated the sedimentary characteristics of core sediments in detail in order to avoid mass-flow influences.

Core TY99PC18 consists of a thick sequence of fine-grained sediments: the topmost 2 m thick bioturbated mud is underlain by alternating layers of several centimeter thick crudely laminated mud and homogenous mud (Fig. 2). Bioturbated mud is either distinctly or indistinctly burrowed mud, and includes abundant siliceous microfossils such as diatom, silicoflagellates and sponge spicules (Bahk, 2001). This was most likely deposited by hemipelagic/pelagic settling. In crudely laminated mud, the poorly sorted diatomaceous mud matrix and the randomly scattered foraminiferal sand as well as the absence of systematic vertical variation in texture and laminae thickness indicate that these are mainly formed by hemipelagic sedimentation (Bahk, 2001). Homogenous mud usually lacks visible primary structures and bioturbation, but occasionally shows grading at the basal zones indicative of fine-grained turbidites (Bahk, 2001).

Guided by X-radiograph imagery, we focused our attention on core sediments that appeared unaffected by down-slope processes and we eventually collected samples from the bioturbated mud and crudely laminated mud that well preserved the history of oceanic conditions in the East Sea.

The ages of core sediments were determined by radiocarbon dating. Accelerator Mass Spectrometry (AMS) ^{14}C dates of monospecific planktonic foraminifera (*G. bulloides,* size >212 μm, 7~22 mg) from 4 depths in core TY99PC18 were measured at the Leibniz Laboratory (Table 1).

Analytical Methods

Bulk sediment samples (1–2 g) were taken from the cores for the analysis of alkenones. They were analyzed at Brown University. Organic extracts were obtained by using a Dionex automated fluid extractor (ASE 2000) with a small volume (~26 ml) of solvent (methanol:hexane, 3:1 v/v) at high temperature (150°C) and pressure (2000 psi). After evaporation at 50°C under N_2 stream and dilution with a small amount (0.2–1.0 ml)

Table 1. ^{14}C ages for Core TY99PC18.

Sample Depth (cm)	Materials*	Laboratory Number	AMS ^{14}C Ages (yr B.P.)
115–116.5	*G. bulloides*	KIA11587	10335±50
171–173	*G. bulloides*	KIA11588	13210±80
222–224	*G. bulloides*	KIA11589	14940±70
309–311	*G. bulloides*	KIA11590	20660±130

Globigerina bulloides (size >212 μm, 7~22 mg) were analyzed for radiocarbon dating at the Leibniz Laboratory, Germany.

of toluene, these extracts were analyzed by a gas chromatograph (HP 5890 series II) with a flame-ionized detector and DB-1 column (60 m×0.32 mm×0.10 μm film thickness, J&W Scientific). During this process, samples are injected on-column and the temperature was increased by 25°C/min from 90–250°C and 1°C/min from 250–290°C, then 1°C/min from 290–310°C with an isothermal holding step at 310°C for 15 minutes. $U^{k'}_{37}$ values are calculated from peak areas on the GC chromatograph. We achieved a reproducibility of 0.008 $U^{k'}_{37}$ units (±1σ), which corresponds to a temperature uncertainty of ±0.2°C based on multiple extractions of sediment standards from the Santa Barbara Basin and the central California margins. In this study, $U^{k'}_{37}$ temperatures were estimated using the calibration of Prahl *et al.* (1988) ($U^{k'}_{37}$=0.034T + 0.039).

RESULTS

Chronology

^{14}C ages of pelagic sediments were estimated by linear interpolation assuming constant sedimentation rates between ^{14}C dates. AMS ^{14}C dating of planktonic foraminifera gives an age of 20,660 kyr at a depth of 309–311 cm, and 14,940 kyr at a depth of 222–224 cm of core TY99PC18 (Table 1 and Fig. 2). These results indicate that the laminated sequences (alternation of pelagic sediments and turbidites) were deposited during the last glacial period between about 15 and 21 kyr BP. Also, the core contains a distinctive volcanic tephra layer known as Ulleung-Oki at a depth of 100 cm. The interpolated radiocarbon age of this layer is about 9 kyr BP, which is consistent with the estimated eruption age of 9,300 yr BP (Machida and Arai, 1983).

Coretop SST calibration

The coretop SST estimated from the $U^{k'}_{37}$ value is 18°C. We compared the coretop alkenone temperature with the modern observed SST. The monthly mean SST derived from the COADS climatological database (Woodruff *et al.*, 1987) at a location (36°30'N, 131°30'E) close to the core site is presented in Fig. 3. A comparison of the coretop alkenone SST with COADS data collected from 1960 to 1997 shows that the coretop SST correspond to the observed SST for June and November. In the meantime, according to sediment trap data from the southwestern Japan Basin

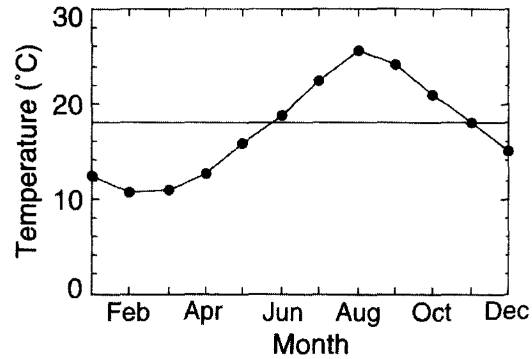

Fig. 3. Annual variation of modern sea surface temperature at a location close to TY99PC18. Alkenone SST for coretop sediment (18°C) is indicated as a horizontal line. Data from COADS (Woodruff *et al.*, 1987).

(Fig. 1), the highest flux of total organic carbon and inorganic carbon occurs during October and November, indicating that the maximum primary production occurs during this period (Hong *et al.*, 1996). Since it is highly likely that the season of maximal production for haptophyte microalgae, the alkenone producer, coincides with the season for maximum primary production, we assume that maximum alkenone production occurs during the late fall. If our assumption is correct, the coretop SST from alkenone most likely represent the late fall SST.

Downcore variation in alkenone SST

GC chromatography indicates that sediments in our core contain a series of long chain (C_{37} and C_{38}) alkenones enough to be analyzed. Total C_{37} alkenone concentrations are low during the glacial period (15–20 kyr) (Fig. 4). During the deglacial period (10–15 kyr) the alkenone content increased slightly. Abnormally high concentrations of alkenones occur during the early Holocene (5–10 kyr). Alkenone concentrations are relatively high during the Holocene compared to the glacial period.

In TY99PC18, the temperature estimated from the $U^{k'}_{37}$ was 15°C approximately 21 kyr BP (Fig. 4). Then, the temperatures increased rapidly up to 21°C 19 kyr BP. The average temperature for the interval between 15–18 kyr BP was 21°C. Around 15 kyr BP, the temperatures rapidly decreased to 15°C, and they fluctuated between 16°C and 14°C between 11–14 kyr BP. About 11 kyr BP the temperatures rapidly increased again, then remained between 18°C and 19°C during the period between 8 to 10 kyr BP, and slowly decreased to 16°C 5 kyr BP. For the coretop, the tem-

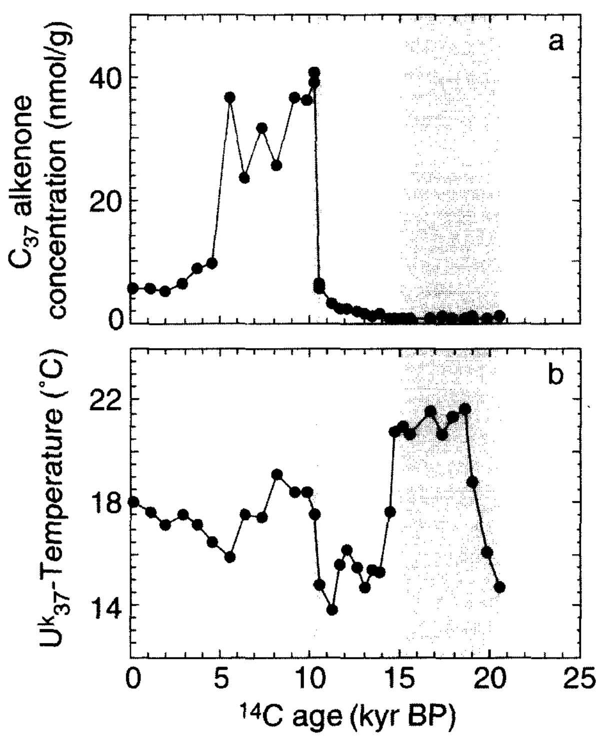

Fig. 4. (a) Total C_{37} alkenone concentration versus ^{14}C ages in core TY99PC18. Shading indicates the interval of alternating layers of crudely laminated mud and homogenous mud. (b) $U^{k'}_{37}$-based temperature estimates versus ^{14}C ages in core TY99PC18.

perature estimated from the $U^{k'}_{37}$ was 18°C.

DISCUSSION

Our results suggest that the alkenone-based temperatures in the East Sea were about 3°C warmer during the last glaciation than today. This precisely matches the results of the alkenone study in the East Sea by Ishiwatari *et al.* (2001). They used piston cores from the western margin of Honshu, near Akita (Fig. 1), and found that the glacial SST in the region were about 2–3°C warmer than today. Comparison of our results with their estimates suggests that this warming during the glacial period could be a basin-scale phenomenon if the estimates were correct. However, in order to use the $U^{k'}_{37}$ index as a proxy for paleotemperature, the influence of several factors on SST reconstruction from $U^{k'}_{37}$ values must be considered. These factors include compositional changes in alkenone producer, variations in salinity, temporal changes of nutrient concentration and growth rate, and shift in the season of maximum alkenone synthesis.

Factors influencing alkenone SST estimates

Compositional changes in alkenone producer: The various calibration studies of cultural experiments (Prahl and Wakeham, 1987; Prahl *et al.*, 1988; Brassell, 1993; Volkman *et al.*, 1995) show different correlations between $U^{k'}_{37}$ and ocean water temperature for different species. *E. huxleyi* is widely regarded as the most likely source of long chain alkenones in the present day but other sources as such *Gephyrocapsa oceanica* also produce this compound (Marlowe *et al.*, 1990; Volkman *et al.*, 1995). A culture experiment with *G. oceanica* showed that the relationships between $U^{k'}_{37}$ and temperature are different from those of *E. huxleyi* (Volkman *et al.*, 1995). Recently, however, Conte *et al.* (1998) indicated that the relationships between the $U^{k'}_{37}$ and temperature for the *G. oceanica* strain are within the range of those observed for the different *E. huxley* strains, suggesting that the calibration equation for *G. oceanica* is significantly different from that of *E. huxley*. Therefore, it is uncertain that a different correlation between $U^{k'}_{37}$ and ocean water temperature exists for different species.

If a different relationship between $U^{k'}_{37}$ and temperature exists for different species, the compositional changes in the alkenone producer in the East Sea over the last 20 kyr could make the temperature estimates biased depending on the calibration equation used. Recently, Ishiwatari *et al.* (2001) investigated the relative abundance of *E. huxleyi* and *G. oceanica* from the sediments of piston cores of the East Sea over the last 36 kyr. Their results show that the relative abundance of *E. huxleyi* was close to or above 50% throughout this time except for the intervals between 9–10 and 19–20 kyr BP. Since coretop temperature estimates with the calibration equation for *E. huxleyi* are consistent with the observed temperature, application of the calibration equation for *E. huxleyi* to the entire core appears to be reasonable given the compositional changes in alkenone producer.

Salinity changes: An examination of live coccolithphore indicates that *Emiliania huxleyi*, the source organism of alkenone, is one of the most euryhaline coccolithophore species (Winter *et al.*, 1994). It can easily withstand the high salinity of 41‰ in the Red Sea (Winter *et al.*, 1979) and the low salinity of 11‰ in the Sea of Azov and 17–18‰ in the Black Sea (Bukry, 1974). In order to check the influence of salinity on the $U_{37}^{k'}$ index, Sonzogoni *et al.* (1997) investigated the coretop sediment samples from the Bay of Bengal and the Arabian Sea where they are characterized by a large salinity gradient (31–36‰) and almost no temperature changes. They found that salinity changes do not appear to affect $U_{37}^{k'}$ values. Also Sikes *et al.* (1991) determined the SST recorded by alkenones of the Black Sea sediments, and found that it was very close to modern SST. Considering the surface salinity of 18–22‰ in the Black Sea, consistency between alkenone-based temperatures and modern temperatures indicates that salinity does not affect $U_{37}^{k'}$. Recently, however, Rosell-Melé (1998) suggested that $U_{37}^{k'}$ is not a reliable paleothermometer due to the effect of salinity especially at the region where the relative abundance of $C_{37:4}$ to the total abundance of C_{37} alkenones (37:4%) is greater than 5. If his hypothesis is correct, 37:4% can be used as an indicator of the effect of salinity on $U_{37}^{k'}$. The concentrations of 37:4 in core TY99PC18 are negligible during the last glaciation, suggesting that the effects of salinity changes on $U_{37}^{k'}$ values seem to be insignificant.

Changes in nutrient concentration and growth rate: In order to use $U_{37}^{k'}$ index as a proxy for paleotemperature, the influence of other factors such as nutrient concentration and growth rate on $U_{37}^{k'}$ values must be considered. Based on chemostat cultures of *E. huxleyi* at constant temperature with variable growth rates, Popp *et al.* (1998) concluded that nutrient-limited growth rate effects do not produce significant error in $U_{37}^{k'}$-based paleotemperature estimates. However, isothermal culturing experiments with single clone of *E. huxleyi* (Epstein *et al.*, 1998) show that $U_{37}^{k'}$ values vary with nutrient availability and cell division rate, suggesting that alkenone-based paleotemperature estimates could be influenced by variations in nutrient concentration as well as seawater temperature. According to Epstein *et al.* (1998), a decrease of nutrient content from 40 μM NO_x to less than 1 μM results in an increase of 0.10–0.19 in $U_{37}^{k'}$ unit which corresponds to a temperature increase of 1.8–4.4°C. It is unknown if the nutrient concentration in the East Sea surface waters has changed from the last glaciation to today. At present, the nutrient concentrations of the East Sea surface waters are significantly low. During the last glacial period, stratification of the water column would have limited the transport of nutrients from the deep to the surface, and nutrient concentrations would have still been low at the surface at that time. Therefore, it is highly unlikely that the magnitude of any conceivable glacial-interglacial change in nutrient concentration would be sufficient to significantly bias $U_{37}^{k'}$-based temperature estimates.

Changes in season of maximum alkenone production: The alkenone SST estimate for the coretop sediment is consistent with the observed SST for late fall (see the section on coretop SST calibration). Also sediment trap data confirm this conclusion by indicating that maximum primary production occurs during this period (Hong *et al.*, 1996). The reason why maximum production occurs at the late fall is not clear. One possibility is that nutrient supply from the deep to the surface by deep convection during that season could lead to increased productivity.

Then, what could have happened in the East Sea during the glacial period? Was the flux of alkenone from the surface to the deep highest in the late fall during the glacial period, or did the season of peak flux alter? Paleoceanographic conditions of the East Sea were quite different during the last glaciation, so it is highly unlikely that the same physiographic condition would exist during the glaciation. The East Sea waters during the glaciation are characterized by severe stratification of water column and anoxic conditions at the bottom waters (e.g. Crusius *et al.*, 1999). Even severe winter cooling was not effective enough to break the stratification of the water column and to transfer nutrients from the deep to the surface. Therefore, it appears that the season for maximum primary production was not confined to the late fall and that there is a possibility that the estimates could be biased toward summer temperatures.

Paleoceanographic Implications

According to the results of alkenone analysis, the East Sea surface waters were warm during the last glaciation (15–20 kyr BP). The SST rapidly increased from 15°C to 21°C at 19 kyr BP. The warming during the glacial period appears to be consistent with microfossil records. A cold-water species of calcareous nannoplankton, *Coccolithus pelagicus,* as well as a cold-water diatom species, *Thalassiosira nordenskioldii* disappeared during the glacial period, and they reappeared after the glacial period (Oba *et al.*, 1991). These facts indicate that the glacial climate in the East Sea was not as cold as expected from previous studies. Further studies with other proxy records are required to reconstruct accurate paleotemperatures. The reason for warming at this time is not clear. However, we can not rule out the possibility that the season for maximum production of haptophyte microalgae shifted from summer during the glaciation to late fall during the Holocene. The variations of alkenone concentration indicate that the productivity in the region decreased during the glaciation probably due to intensified water-column stratification.

During the deglaciation (11–15 kyr), the surface temperatures rapidly decreased from 21°C to 15°C. This cooling was also supported by microfossil data. The occurrence of cold-water species *C. pelagicus* and *T. nordenskioldii* (Oba *et al.*, 1991) confirms cooling during this time. It could be possible that during this period the East Sea was under the strong influence of inflow of cold seawater such as that brought by the Oyashio Current. Also, it might be possible that inflow of cold ice-melt water from the north was significant during this interval. During this period productivity slightly increased.

At about 10 kyr BP, rapid environmental changes occurred in the East Sea. The surface temperatures rapidly increased from 15°C to above 18°C. Other geological records suggest that the warm surface waters were probably caused by the inflow of waters from the south through the Korea Strait (Oba *et al.*, 1991). Abnormally high concentrations of alkenone are indicative of high productivity during this period. Approximately 5 kyr BP, there was a slight cooling at the surface seawater. Subsequently, conditions have been similar to what is observed today.

SUMMARY

In order to reconstruct the variations in SST over the last 20 kyr, C_{37} alkenones were measured using the marine sediments from the Ulleung Basin in the East Sea. Sediment ages were determined by AMS ^{14}C dating of the planktonic foraminifera *Globigerina bulloides*. The results of alkenone analysis show that total C_{37} alkenone concentrations are low during the last glaciation compared to the Holocene. Anomalously high concentrations of C_{37} alkenones occurred in the early Holocene. Results from sediment trap study (Hong *et al.*, 1996) and comparisons of coretop alkenone temperature with modern surface temperatures indicate that the alkenone SST estimates most likely represent those of late fall.

Downcore variations in the $U^{k'}_{37}$ values indicate that the surface waters were about 3°C warmer during the glaciation (15–20 kyr BP) than today. During the deglacial period (11–15 kyr BP), the temperatures were about 3°C lower than today. A rapid SST increase of about 3°C occurred at the early Holocene (about 10 kyr BP). We examined several factors which might affect the estimation of alkenone temperatures. Such factors include compositional changes in alkenone producer, variations in salinity, temporal changes of nutrient concentration and growth rate, and shift in the season of maximum alkenone synthesis. The results indicate that alkenone SST estimates are reliable. The glacial warming in the East Sea could be caused by a shift of the season of maximum alkenone production from summer during the last glaciation to late fall during the Holocene. Cooling during the deglacial period may be due to inflow of cold water such as that brought by the Oyashio Current or ice-melt water. Warming in the early Holocene could be due to inflow of warm water from the south.

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