

## Alkenone sea surface temperature in the Southern Ocean for the last two deglaciations

Minoru Ikehara <sup>1</sup>, Kimitaka Kawamura <sup>2</sup>, Naohiko Ohkouchi <sup>2</sup>, Katsunori Kimoto <sup>1</sup>, Masafumi Murayama <sup>3</sup>, Toshio Nakamura <sup>4</sup>, Tadamichi Oba <sup>3</sup>, and Asahiko Taira <sup>1</sup>

**Abstract.** A piston core taken from the Tasman Plateau in the Southern Ocean has been studied for organic compounds to reconstruct the late Pleistocene marine environments. Here we report paleo sea surface temperature (SST) for the last two deglaciations using the unsaturation degree of alkenones ( $U^{k_{37}}$ ) preserved in the marine sediments. The  $U^{k_{37}}$  record indicates that the SST was at least 4°C lower than the present SST at the last glacial maximum (LGM), and the amplitude of paleo-SST is at most 5.2°C from the penultimate glacial (MIS-6) to the last interglacial warm period (the Eemian). Our results also demonstrate that the Eemian warm period in the Southern Ocean lasted only 3,000 years followed by a sharp cooling at around 120 kyr BP. The sharp cooling in the Southern Ocean seemed to occur a few millennia (2-3 kyrs) before the beginning of continental ice-sheet growth.

### Introduction

The Southern Ocean occupies a significant position of world ocean and is characterized by several unique features: high-nutrient and low-chlorophyll (HNLC) conditions [e.g. Coale *et al.*, 1996], development of polar zonation surrounding Antarctica, deep convection to intermediate and deep waters. Reconstruction of the late Pleistocene sea surface temperature (SST) of the Southern Ocean is important for understanding paleoceanographic conditions such as the shift of the polar frontal zonation and its link to the Earth's climate system. In the early 1980s, the CLIMAP Project estimated SST for the last glacial maximum (LGM) and the last interglacial period (the Eemian) based on the planktonic microfossil assemblages in a large number of deep sea sediments [CLIMAP Project Members, 1981, 1984]. One of the significant findings was that the estimated SST in the low- to mid-latitudes at the LGM was not much different (<2°C) from the present SST. Further, the difference in the SST for the high-latitudes was not large (2-3°C) except in the North Atlantic and northwest Pacific. CLIMAP Project Members [1984] also concluded that SST of the last interglacial ocean was not significantly different from that of the modern ocean. However, the paleo-SST reconstructed at high-latitudes based

on zooplanktonic microfossils contains uncertainty, because the diversity of foraminifera is much lower and their living depth ranges widely.

A geochemical thermometer, referred as  $U^{k_{37}}$  [Brassell *et al.*, 1986], is based on the analysis of long-chain alkenones ( $C_{37}$ ) synthesized by certain haptophyte microalgae [Marlow *et al.*, 1990], and is characterized by a linear relationship to its growth temperature [Prahl and Wakeham, 1987]. This ratio is also unaltered through the food web in the water column and early diagenesis in the sediments [Prahl *et al.*, 1989, 1993]. The  $U^{k_{37}}$  ratios have so far successfully been used to reveal the past-SST variations in the equatorial Pacific [Prahl *et al.*, 1989; Ohkouchi *et al.*, 1994], North Pacific [Prahl *et al.*, 1995; Herbert *et al.*, 1995], equatorial Atlantic [Brassell *et al.*, 1986; Jasper and Gagosian, 1989; Sikes and Keigwin, 1994; Schneider *et al.*, 1995], and North Atlantic [Eglinton *et al.*, 1992; Zhao *et al.*, 1993, 1995; Sikes and Keigwin, 1996]. Here we report for the first time alkenone SST records for the last two deglaciation periods in the Southern Ocean.

### Samples and methods

A piston core (KH94-4 TSP-2PC; 48°08.18'S, 146°52.45'E, 2321m water depth) and a multiple corer sample (TSP-2MC; 48°07.60'S, 146°53.99'E, 2283m water depth) were collected from the Tasman Plateau in the Southern Ocean during the R/V *Hakuho-Maru* cruise [Ikehara *et al.*, 1996; Fig. 1]. Sediments are mainly composed of calcareous foraminifera-nanno ooze (CaCO<sub>3</sub> content: 87-94%). It is verified that the multiple core TSP-2MC corresponds to the very surface sediments of piston core TSP-2PC based on comparison with magnetic susceptibility, color data, and oxygen isotopic values of these two core sediments.

A  $\delta^{18}O$  stratigraphy was established for the subsurface dwelling planktonic foraminifera *Globigerina bulloides* (250 to 300  $\mu$ m in test size; 40 specimens). These tests were reacted with 100% H<sub>3</sub>PO<sub>4</sub> at 60°C and analyzed in a Finnigan MAT 251 mass spectrometer. The  $\delta^{18}O$  results are expressed as per mil deviations from PDB (Pee Dee Belemnite) standard. The core chronology was determined based on graphic correlation to the SPECMAP  $\delta^{18}O$  record [Imbrie *et al.*, 1984] and AMS-<sup>14</sup>C dating of planktonic foraminifera *G. bulloides* (see Fig. 3a). The AMS-<sup>14</sup>C dates were obtained using the graphitization method [Nakamura *et al.*, 1987; Kitagawa *et al.*, 1993] and were corrected for a constant surface water age of 400 years [Bard, 1988]. The averaged sedimentation rate was estimated to be 1.5 cm/kyr.

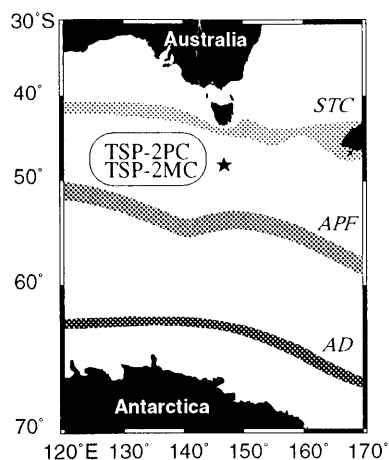
The  $U^{k_{37}}$  values were obtained by the following procedure. The frozen sediment samples were cut at 2 cm intervals. Approximately 20 g of wet sediments were extracted for lipids with methanol/dichloromethane (3:1) and then

<sup>1</sup> Ocean Research Institute, University of Tokyo, Nakano-ku, Tokyo, Japan

<sup>2</sup> Institute of Low Temperature Science, Hokkaido University, Sapporo, Hokkaido, Japan

<sup>3</sup> Graduate School of Environmental Earth Science, Hokkaido University, Sapporo, Hokkaido, Japan

<sup>4</sup> Dating and Material Research Institute, Nagoya University, Chikusa-ku, Nagoya, Japan



**Figure 1.** Location map of the deep sea cores TSP-2PC and TSP-2MC collected from the Tasman Plateau in the Southern Ocean and hydrographic conditions. STC : Subtropical Convergence zone, APF : Antarctic Polar Front zone, AD : Antarctic Divergence zone. Subantarctic Water properties between STC and APF are characterized by relatively warm and saline waters. In contrast, Antarctic Surface Water between APF and AD are characterized by cool and less saline waters.

dichloromethane/methanol (10:1) using an ultrasonic homogenizer. The extracts were isolated by a centrifuge and washed with 50 ml of 0.15M HCl to remove salts contained in the sediments, and then saponified with 0.5M KOH/methanol under a reflux. Neutral components were separated by extraction with dichloromethane/*n*-hexane (10:1). They were further divided into four sub fractions on a Pasteur pipette column packed with silica gel (BIO-SIL A, 200-400 mesh) which was deactivated with 1 % water. The fraction containing long-chain alkenones was analyzed by a Carlo Erba 5160 gas chromatograph (GC) equipped with a cold on-column injector, a HP-5 fused silica capillary column (30 m x 0.32 mm i.d.; 0.25  $\mu$ m film thickness), and a flame ionization detector (FID). Hydrogen was used as a carrier gas, and the column oven temperature was programmed from 70°C to 120°C at 30°C/min. and then to 320°C (30 min.) at 6°C/min. The GC peaks were processed using a Shimadzu Chromatopac C-R7A integrator. Identification of alkenones was conducted by comparing gas chromatographic retention times with previous work [Ohkouchi *et al.*, 1994].  $U^{k_{37}}$  values were obtained with the following definition [Prah *et al.*, 1988]:

$$U^{k_{37}} = [C_{37:2}] / ([C_{37:2}] + [C_{37:3}])$$

where  $[C_{x:y}]$  denotes the concentration of the alkenone with *x* carbon number and *y* double bonds. Triplicate analyses of composite sediments showed that the analytical errors in the procedures were  $\pm 0.001$  for  $U^{k_{37}}$  values (i.e.  $\pm 0.03^\circ\text{C}$ ).  $C_{37:4}$  alkenone has been detected in some sediment samples, however, its concentrations were generally low.

## Results and Discussion

### Core top alkenone SST

Long-chain  $C_{37}$ - $C_{39}$  alkenones detected in the surface sediments (TSP-2MC, 0-1 cm) had a  $U^{k_{37}}$  value of 0.448. Comparison of the annual SST cycle from Levitus [1982] with

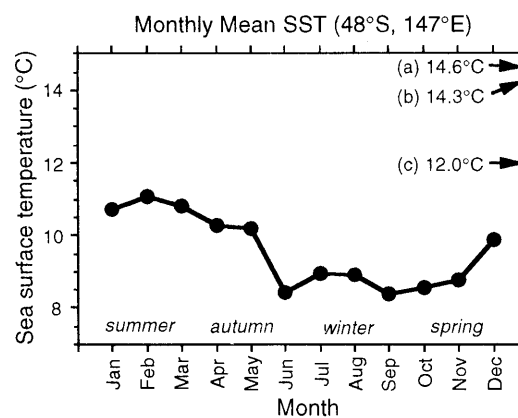
that derived from three calibrations of the  $U^{k_{37}}$  method (Fig. 2) shows that the SST calculated using the calibration of Prah *et al.* [1988] is similar to the summer SST near the sampling site. Therefore, we decided to use the following equation for the calculation of the past SST, whose precision is about 0.5  $^\circ\text{C}$  [Prah *et al.*, 1988].

$$T (^\circ\text{C}) = (U^{k_{37}} - 0.039) / 0.034$$

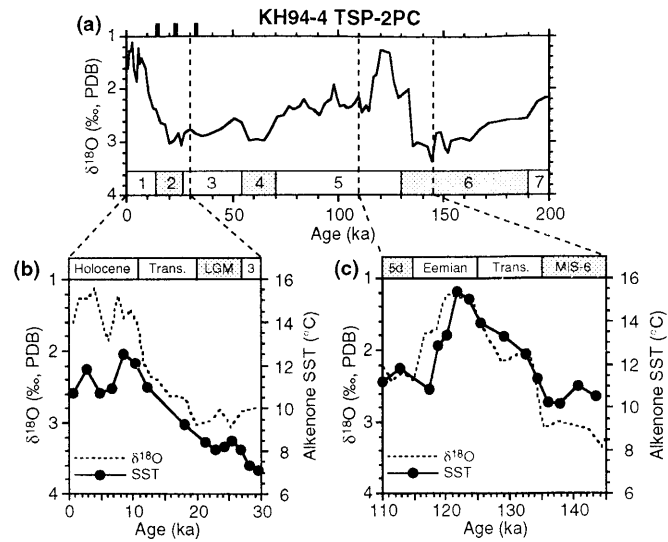
A slight difference between the alkenone SST (12.0°C) and the summer Levitus SST (11.1°C; see Fig. 2) may be caused by a difference of the data source: the former represents an average SST for a few millennia whereas the latter represents at best few decades.

### Glacial-interglacial variation of alkenone SST

Our  $U^{k_{37}}$  values ranged from 0.281 to 0.465 during the last deglaciation (last 30 ka; MIS-3 to Holocene) and from 0.385 to 0.563 during the penultimate deglaciation (110-145 ka; MIS-6 to MIS-5d). Figure 3 demonstrates changes in the alkenone SST and planktonic  $\delta^{18}\text{O}$  [Ikehara, 1997]. The alkenone SST between LGM and Holocene varied from 8.1 to 12.5°C (Fig. 3b), whereas those between MIS-6 and MIS-5e ranged from 10.2 to 15.4°C (Fig. 3c). The SST difference between LGM and Holocene is 4.4°C, and that for the penultimate deglaciation is 5.2°C. Thus, we conclude that the alkenone SST shifted by ca. 5°C during the last two glacial/interglacial climate variations in the Tasman Plateau region. These results suggest that SST variations in this region were greater by ca. 2°C than the CLIMAP results [CLIMAP Project Members, 1981]. This larger amplitude of glacial/interglacial SST change agrees with the SST changes reconstructed from diatom assemblages in the sediments from the Indian sector Southern Ocean, which showed 5-6°C difference for the glacial/interglacial periods [Pichon *et al.*, 1992]. The SST decrease in the glacial Southern Ocean may be



**Figure 2.** Annual surface temperature cycle of a location (48°S, 147°E) close to TSP-2PC and TSP-2MC [Levitus, 1982]. Alkenone SST for the surface sediments (TSP-2MC, 0-1cm) calculated by three calibrations are shown, as follows; (a) the linear field data calibration of Sikes and Volkman [1993] in the Southern Ocean ( $U^{k_{37}} = 0.0414T - 0.156$ ), (b) the calibration of Brassell [1993] based on global combined field data ( $U^{k_{37}} = 0.037T - 0.083$ ), and (c) the calibration of Prah *et al.* [1988] based on cultures of *Emiliania huxleyi* ( $U^{k_{37}} = 0.034T + 0.039$ ).



**Figure 3.** (a) Changes of the planktonic foraminiferal  $\delta^{18}\text{O}$  in the sediment core (TSP-2PC) collected from the Tasman Plateau in the Southern Ocean. Odd numbers mean interglacial periods and shaded even numbers indicate glacial periods. AMS- $^{14}\text{C}$  age levels are shown by bold marks on the top of the figure. (b) Changes in the planktonic foraminiferal  $\delta^{18}\text{O}$  and sea surface temperature (SST) reconstructed from the  $\text{U}^{k_{37}}$  index during the last deglaciation (MIS-3 to Holocene; 0–30 ka). (c) Same as (b) but for penultimate deglaciation (MIS-6 to MIS-5d; 110–145 ka). For the definition of  $\text{U}^{k_{37}}$ , see the text. Estimated SST was calculated by using the following equation:  $T (^{\circ}\text{C}) = (\text{U}^{k_{37}} - 0.039) / 0.034$  [Prah et al., 1988], where T denotes a haptophyte growth temperature.

caused by northward shift of the Antarctic Polar Front (APF) at that time.

Although the alkenone SST curve from LGM to Holocene is similar to that from MIS-6 to MIS-5e, the reconstructed SST values of the last deglaciation are systematically lower by ca. 3 °C compared to those for the penultimate deglaciation. In contrast, the foraminiferal results did not show a systematic shift of  $\delta^{18}\text{O}$  values, which has been fluctuated mainly by ice volume. Thus, the systematic shift of alkenone SST between the last and penultimate deglaciations suggests that main contributor (coccolithophorid) to the  $\text{C}_{37}$  alkenones was different in the Southern Ocean for the last two deglaciations. In fact, the abundance of *E. huxleyi* decreased during the penultimate deglaciation in comparison with those of the last deglaciation [M. Horikoshi and H. Okada, pers. commun., 1997]. Alternatively, an excursion of the polar frontal zone and salinity change probably caused by evaporation to precipitation balance may contribute to the systematic shift of the alkenone SST between two deglaciations.

### Eemian SST changes

The highest SST (15.4°C) in the Eemian was recorded at around 122 kyrs BP when the  $\delta^{18}\text{O}$  values became minimum, and the alkenone SST dropped abruptly by 4.5°C (similar to glacial SST) in the middle of the Eemian (Fig. 3c). Based on the analyses of grain size and composition of sediments, no significant change was suggested in sedimentary processes at this period. Assuming a constant sedimentation rate during the Eemian, we estimated that the warmest period higher than 15°C lasted only ca. 3,000 years and ended at around 120 kyrs BP. The climate of the Eemian was thought to be stable and similar to that of the Holocene [e.g. CLIMAP Project Members, 1984]. However, our results suggest that the Eemian SST in the Southern Ocean is not stable throughout this period, and that rapid cooling occurred at around 120 kyrs BP. It is likely that

this abrupt cooling took place entirely in the Southern Ocean, because similar cooling of mid-Eemian have been found in sediment cores recovered from other regions in the Southern Ocean [e.g. Martinson et al., 1987; Pichon et al., 1992].

However, the cooling event is unlikely explained by any significant decrease in the continental ice volume, because planktonic  $\delta^{18}\text{O}$  values did not show a positive peak during the mid-Eemian (Fig. 3c). These results suggest that the cooling of the Southern Ocean preceded the significant increase of the continental ice sheets by 2–3 kyrs and that the global cooling from the last interglacial to glacial, which is recorded in the oxygen isotope signal of foraminifera, was triggered by an abrupt cooling of the high latitudes. We believe that this documented offsets in time between SST and  $\delta^{18}\text{O}$  shifts are real records and cannot be explained by a bioturbation process, which could cause a selective redistribution of foraminiferal fossils and coccolithophorid grains [Hutson, 1980], because the abundance of both *G. bulloides* and alkenones were observed to stay almost constant throughout the sediment sequence from 110 to 130 kyr BP.

### Conclusions

Southern Ocean sediment cores were studied for long-chain alkenones to reconstruct the SST for the last two deglaciation periods in relation to the global glacial/interglacial cycles. Our results demonstrate that:

(1) The alkenone SST difference between glacial and interglacial in the Southern Ocean was found to be approximately 5°C, indicating that the amplitude of Southern Ocean SST fluctuation was twice greater than that reported by the CLIMAP projects.

(2) The SST for the Eemian appear to be higher than that of the Holocene by ca. 3°C.

(3) The Eemian warmest period lasted only 3,000 years followed by a sharp cooling at around 120 kyrs BP. This cooling event occurred a few millennia before the beginning of a large continental ice-sheet growth.

**Acknowledgments:** We thank the on-board scientists and crew of the R/V *Hakuho-Maru* cruise KH94-4 for their cooperation in collecting the samples. We also acknowledge F. PrahI and one anonymous reviewer for their critical and useful comments, T. Nakatsuka, P. Hesse, and N. Ahagon for fruitful discussions, Y. Imai and Y. Ishimura for analytical help. This study was partly supported by a grant from the JSPS (Japan Society for the Promotion of Science) for Junior Scientists to M. I.

## References

- Bard, E., Correction of accelerator mass spectrometry  $^{14}\text{C}$  ages measured in planktonic foraminifera: paleoceanographic implications, *Paleoceanogr.*, **3**, 635-645, 1988.
- Brassell, S. C., G. Eglinton, I. T. Marlowe, U. Pflaumann, and M. Sarnthein, Molecular stratigraphy: a new tool for climatic assessment, *Nature*, **320**, 129-133, 1986.
- Brassell, S. C., Applications of biomarkers for delineating marine paleoclimatic fluctuations during the Pleistocene, in *Organic Geochemistry*, edited by M. H. Engel and S. A. Macko, Plenum Press, New York, pp. 699-738, 1993.
- CLIMAP Project Members, Seasonal reconstructions of the Earth's surface at the Last Glacial Maximum, *Geol. Soc. Am. Map Chert, MC-36*, 1-18, 1981.
- CLIMAP Project Members, The last interglacial ocean, *Quat. Res.*, **21**, 123-224, 1984.
- Coale, K. H., K. S. Johnson, S. E. Fitzwater, R. M. Gordon, S. Tanner, F. P. Chavez, L. Ferioli, C. Sakamoto, P. Rogers, F. Millero, P. Steinberg, P. Nightingale, D. Cooper, W. P. Cochlan, M. R. Landry, J. Constantinou, G. Rollwagen, A. Trsvina, and R. Kudela, A massive phytoplankton bloom induced by an ecosystem-scale iron fertilization experiment in the equatorial Pacific Ocean, *Nature*, **383**, 495-501, 1996.
- Eglinton, G., S. A. Bradshaw, A. Rosell, M. Sarnthein, U. Pflaumann, and R. Tiedemann, Molecular record of secular sea surface temperature changes on 100-year timescales for glacial terminations I, II and IV, *Nature*, **356**, 423-426, 1992.
- Jasper, J. P., and R. B. Gagosian, Alkenone molecular stratigraphy in an oceanic environment affected by glacial freshwater events, *Paleoceanogr.*, **4**, 603-614, 1989.
- Herbert, T. D., M. Yasuda, and C. Burnett, Glacial-interglacial sea-surface temperature record inferred from alkenone unsaturation indices, Site 893, Santa Barbara Basin, *Proc. ODP, Sci. Results*, **146**, 257-264, 1995.
- Hutson, W. H., Bioturbation of deep-sea sediments: Oxygen isotopes and stratigraphic uncertainty, *Geology*, **8**, 127-130, 1980.
- Ikehara, M., N. Ahagon, M. Murayama, H. Matsuoka, T. Niwai, Y. Shirayama, M. Shimanaga, C. Tamura and C. Igarashi, Description of piston core samples in the Southern Ocean, *Preliminary Report of the R/V Hakuho Maru Cruise KH94-4*, pp.69-71, Ocean Research Institute, University of Tokyo, 1996.
- Ikehara, M., Late Pleistocene Southern Ocean paleoceanography based on organic and isotope geochemistry, Ph.D. Thesis, Univ. of Tokyo, 1997.
- Imbrie, J., J. D. Hays, D. G. Martinson, A. McIntyre, A. C. Mix, J. J. Morley, N. G. Pisias, W. L. Prell, and N. J. Shackleton, The orbital theory of Pleistocene climate: Support from a revised chronology of the marine  $\delta^{18}\text{O}$  record, in *Milankovitch and Climate, Part 1*, edited by A. L. Berger et al., pp. 269-305, 1984.
- Kitagawa, H., T. Masuzawa, T. Nakamura, and E. Matsumoto, A batch preparation method for graphite targets with low background for AMS  $^{14}\text{C}$  measurements, *Radiocarbon*, **35**, 295-300, 1993.
- Levitus, S., Climatological atlas of the world ocean, *NOAA Prof. Pap.* **13**, 173pp., U.S. Govt. Print. Office, Washington D. C., 1982.
- Marlowe, I. T., S. C. Brassell, G. Eglinton, and J. C. Green, Long-chain alkenone and alkyl alkenoates and the fossil coccolith record of marine sediments, *Chem. Geol.*, **88**, 349-375, 1990.
- Martinson, D. G., N. G. Pisias, J. D. Hays, J. Imbrie, T. C. Moore, and N. J. Shackleton, Age dating and the orbital theory of the ice ages: Development of a high-resolution 0 to 300,000-year chronostratigraphy, *Quat. Res.*, **27**, 1-29, 1987.
- Nakamura, T., N. Nakai, and S. Ohishi, Techniques of tandem accelerator mass spectrometry and their applications to  $^{14}\text{C}$  measurements, in *Proceedings of the 4th International Symposium on Accelerator Mass Spectrometry, Nuclear Instruments and Methods in Physics Research B29*, edited by H. E. Cove, A. E. Litherland, and D. Elmore, pp. 335-360, 1987.
- Ohkouchi, N., K. Kawamura, T. Nakamura, and A. Taira, Small changes in the sea surface temperature during the last 20,000 years: Molecular evidence from the western tropical Pacific, *Geophys. Res. Lett.*, **21**, 2207-2210, 1994.
- Pichon, J. -J., L. D. Labeyrie, G. Bareille, M. Lagracherie, J. Duprat, and J. Jouzel, Surface water temperature changes in the high latitudes of the southern hemisphere over the last glacial-interglacial cycle, *Paleoceanogr.*, **7**, 289-318, 1992.
- PrahI, F. G., and S. G. Wakeham, Calibration of unsaturation patterns in long-chain ketone compositions for paleotemperature assessment, *Nature*, **330**, 367-369, 1987.
- PrahI, F. G., L. A. Muehlhausen, and D. L. Zahnle, Further evaluation of long-chain alkenones as indicators of paleoceanographic conditions, *Geochim. Cosmochim. Acta*, **52**, 2303-2310, 1988.
- PrahI, F. G., G. J. De Lange, M. Lyle, and M. A. Sparrow, Post-depositional stability of long-chain alkenones under contrasting redox conditions, *Nature*, **341**, 434-437, 1989.
- PrahI, F. G., R. B. Collier, J. Dymond, M. Lyle, and M. A. Sparrow, A biomarker perspective on prymnesiophyte productivity in the northeast Pacific Ocean, *Deep-Sea Res.*, **40**, 2061-2076, 1993.
- PrahI, F. G., N. Pisias, M. A. Sparrow, and A. Sabin, Assessment of sea-surface temperature at  $42^{\circ}\text{N}$  in the California Current over the last 30,000 years, *Paleoceanogr.*, **10**, 763-773, 1995.
- Schneider, R. R., P. J. Müller, and G. Ruhland, Late Quaternary surface circulation in the east equatorial South Atlantic: Evidence from alkenone sea surface temperatures, *Paleoceanogr.*, **10**, 197-219, 1995.
- Sikes, E. L. and J. K. Volkman, Calibration of alkenone unsaturation ratios ( $\text{U}_{k37}$ ) for paleotemperature estimation in cold polar waters, *Geochim. Cosmochim. Acta*, **57**, 1883-1889, 1993.
- Sikes, E. L., and L. D. Keigwin, Equatorial Atlantic sea surface temperature for the last 30 kyr: A comparison of  $\text{U}_{k37}$ ,  $\delta^{18}\text{O}$  and foraminiferal assemblage temperature estimates, *Paleoceanogr.*, **9**, 31-45, 1994.
- Sikes, E. L., and L. D. Keigwin, A reexamination of northeast Atlantic sea surface temperature and salinity over the last 16 kyr, *Paleoceanogr.*, **11**, 327-342, 1996.
- Zhao, M., A. Rosell, and G. Eglinton, Comparison of two  $\text{U}_{k37}$ -sea surface temperature records for the last climatic cycle at ODP Site 658 from the sub-tropical Northeast Atlantic, *Palaeogeogr., Palaeoclimatol., Palaeoecol.*, **103**, 57-65, 1993.
- Zhao, M., N. A. S. Beveridge, N. J. Shackleton, M. Sarnthein, and G. Eglinton, Molecular stratigraphy of cores off northwest Africa: Sea surface temperature history over the last 80 ka, *Paleoceanogr.*, **10**, 661-675, 1995.

M. Ikehara, Ocean Research Institute, University of Tokyo, Nakano-ku, Tokyo 164, Japan (e-mail: ikehara@ori.u-tokyo.ac.jp)

K. Kawamura, Institute of Low Temperature Science, Hokkaido University, Sapporo, Hokkaido 060, Japan (e-mail: kawamura@soya.lowtem.hokudai.ac.jp)

K. Kimoto, Ocean Research Institute, University of Tokyo, Nakano-ku, Tokyo 164, Japan (e-mail: kimopy@ori.u-tokyo.ac.jp)

M. Murayama, Graduate School of Environmental Earth Science, Hokkaido University, Sapporo, Hokkaido 060, Japan (e-mail: murayama@geoeco.hokudai.ac.jp)

T. Nakamura, Dating and Materials Research Center, Nagoya University, Chikusa-ku, Nagoya 464-01, Japan (e-mail: g44466a@nucc.cc.nagoya-u.ac.jp)

T. Oba, Graduate School of Environmental Earth Science, Hokkaido University, Sapporo, Hokkaido 060, Japan (e-mail: oba-tad@geoeco.hokudai.ac.jp)

N. Ohkouchi, Institute of Low Temperature Science, Hokkaido University, Sapporo, Hokkaido 060, Japan (e-mail: ohkouchi@soya.lowtem.hokudai.ac.jp)

A. Taira, Ocean Research Institute, University of Tokyo, Nakano-ku, Tokyo 164, Japan (e-mail: ataira@ori.u-tokyo.ac.jp)

(Received December 5, 1996; revised January 27, 1997; accepted January 27, 1997)