

Variations of terrestrial input and marine productivity in the Southern Ocean (48°S) during the last two deglaciations

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Abstract. Various biomarkers (*n*-alkanes, *n*-alcohols, and sterols) have been studied in a piston core TSP-2PC taken from the Southern Ocean to reconstruct the paleoenvironmental changes in the subantarctic region for the last two deglaciations. Mass accumulation rates of terrestrial (higher molecular weight *n*-alkanes and *n*-alcohols) and marine (dinosterol and brassicasterol) biomarkers increased significantly at the last two glacials and stayed low during interglacial peaks (early Holocene and the Eemian). These records indicate that the enhanced atmospheric transport of continental materials and the increased marine biological productivity were synchronously linked in the Southern Ocean at the last two glacials. This suggests that increased glacial dust inputs have relieved iron limitation in the subantarctic Southern Ocean. These two processes, however, were not linked at the cooling phase from the Eemian to marine isotope stage (MIS) 5d. During this period, paleoproductivity may have been influenced by the latitudinal migration of the high-production zone associated with the Antarctic Polar Front.

1. Introduction

Martin [1990] proposed the "iron hypothesis," indicating that iron is a limiting factor to control the phytoplankton productivity and consequent CO₂ drawdown from the atmosphere. Since then, the hypothesis has been tested by in situ field experiments in the subarctic Pacific [e.g., *Martin and Fitzwater*, 1988; *Coale*, 1991], the equatorial Pacific [e.g., *Coale et al.*, 1996], and the South Pacific gyre [*Behrenfeld and Kolber*, 1999]. To evaluate whether the past marine productivity was enhanced in the Southern Ocean during the glacial periods, several approaches have been performed using inorganic chemical tracers and microfossils [e.g., *Mortlock et al.*, 1991; *Charles et al.*, 1991; *Shemesh et al.*, 1993; *Kumar et al.*, 1995; *Nürnberg et al.*, 1997]. However, there are no reported studies on marine biomarkers, which have been recognized as useful in reconstructing past marine biological productivity [e.g., *Prahl*, 1992; *Ohkouchi et al.*, 1997a]. The Southern Ocean is characterized by high-nutrient and low-chlorophyll (HNLC) surface waters [e.g., *Martin*, 1990; *Coale et al.*, 1996], development of the polar zonation surrounding Antarctica, and deep convection to form intermediate and deep waters.

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On the basis of the study of alkenones in a Southern Ocean sediment core, *Ikehara et al.* [1997] reported, for the first time, variations of sea surface temperatures (SSTs) and showed that the amplitude of paleo-SSTs are 4°-5°C from the glacials (Last Glacial Maximum and marine isotope stage 6) to the interglacials (early Holocene and the Eemian). To better understand the relationship between local SST and global climate changes which may be associated with the marine productivity during the glacial-interglacial cycles, it is necessary to study various biomarkers, including hydrocarbons, alcohols, and sterols in the Southern Ocean sediments. In this study, we present the analytical results of lipid class compounds in a carbonate-rich deep-sea sediment core recovered from the Tasman Plateau in the Southern Ocean.

2. Oceanographic Setting

The Southern Ocean is delimited northward by the mean latitude of the Subtropical Convergence Zone (~45°S). Figure 1 illustrates the core location and hydrographic systems in the Australian sector of the Southern Ocean. Three major water mass fronts exist between 40°S and Antarctic coast, i.e., from north to south, Subtropical Convergence (STC), Antarctic Polar Front (APF), and the Antarctic Divergence (AD). The APF and the STC are key components of the Southern Ocean hydrographic system. The STC, which extends along 40°-45°S, is located at the boundary between the Subantarctic and the East Australian Currents, with surface expressions of increased gradients of SST and salinity. The present STC Zone is generally characterized by an abrupt latitudinal gradient of SST around the 15°C summer surface isotherm and the 10°C winter isotherm [e.g., *Gamer*, 1959]. The position of this zone is recognized by the surface pattern of isotherms and an increased concentration of nutrients derived from subtropical to subantarctic waters [*Buttler et al.*, 1992].

The APF, extending along 50°-55°S in the southern Pacific, exists at the boundary between the Subantarctic and Antarctic

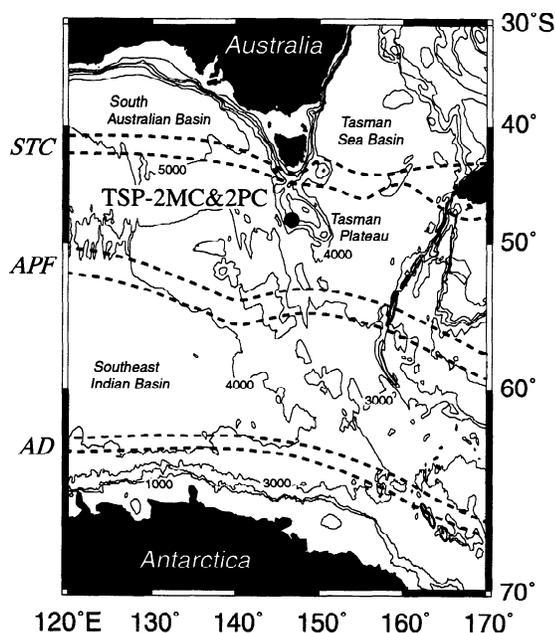


Figure 1. Location map of a multiple core (TSP-2MC) and a piston core (TSP-2PC) recovered from the southwest slope on the Tasman Plateau, the Southern Ocean. Three frontal systems are also shown; STC is the Subtropical Convergence zone, APF is the Antarctic Polar Front zone, and AD is the Antarctic Divergence zone.

Circumpolar Currents (ACC). These surface currents are driven by the west wind drift. At present, the APF is found around 53°S in the latitudinal section between Tasmania and Antarctica [Rintoul et al., 1997].

Sea ice is another important component of the Antarctic oceanographic system. In the Australian sector of the Southern Ocean the sea ice extends up to ~61°S in winter and retreats nearly down to the coast (66°S) in summer. The seasonal variation of sea ice coverage is linked to the location of the STC and the APF, whose migration is in response to the circumpolar westerly winds [Oglesby, 1990].

3. Samples and Methods

3.1. Sediment Samples

Piston core TSP-2PC and multiple core TSP-2MC were collected from the southwest slope of the Tasman Plateau

(South Tasman Rise) during the KH94-4 cruise of the *R/V Hakuho-Maru* [Ikehara et al., 1996] (Table 1 and Figure 1). Sediments are mainly composed of calcareous foraminifera-nannofossil ooze. The multiple core TSP-2MC was confirmed to correspond to the very surface sediments of the piston core TSP-2PC by comparison with magnetic susceptibility, color data, and oxygen isotopic values of these two core sediments [Ikehara, 1997]. The sediment samples were stored at -15°C until analysis.

3.2. Oxygen Isotopic Stratigraphy and Radiocarbon Ages

A $\delta^{18}\text{O}$ stratigraphy was established for the subsurface-dwelling planktonic foraminifera *Globigerina bulloides* (d'Orbigny) (M. Ikehara et al., manuscript in preparation, 1999). The core chronology was determined based on graphic correlation to the Spectral Mapping Project (SPECMAP) normalized $\delta^{18}\text{O}$ curve [Imbrie et al., 1984; Martinson et al., 1987] and accelerator mass spectrometer (AMS) ^{14}C ages of planktonic foraminifera *G. bulloides* shell whose diameters are >300 μm . The SPECMAP turning is also supported by the calcareous nannofossil biostratigraphy [Horikoshi, 1998]. For the last deglaciation period the AMS ^{14}C ages were obtained for the top of the cores TSP-2PC and TSP-2MC (Table 2) at Nagoya University using the graphitization method [Nakamura et al., 1987; Kitagawa et al., 1993]. The AMS ^{14}C ages, which were calculated using a half-life of 5568 years, were corrected for a reservoir age of 480 years in this latitudinal region [Bard, 1988]. A calibration of the ^{14}C ages was also done on the program CALIB 4.0 [Stuiver et al., 1998] and the basis of equation proposed by Bard et al. [1998] in order to define a calendar chronology (Table 2).

3.3. Lipid and CNS Analyses

Lipids were extracted from the wet sediments (~20 g) three times with methanol/ CH_2Cl_2 (3:1), CH_2Cl_2 /methanol (10:1), and CH_2Cl_2 /methanol (10:1) using an ultrasonic homogenizer. The extracts were combined, washed with 0.15 M HCl, and saponified with 0.5 M KOH/methanol for 2 hours under reflux. Neutral lipids were extracted with CH_2Cl_2 /*n*-hexane (10:1). The neutral components were further separated into four subfractions (N-1 to N-4) by silica gel column chromatography [Kawamura, 1995]. The fatty alcohol fraction was treated with bis-trimethyl-silyl-trifluoroacetamide (BSTFA) to derive TMS ethers. Each fraction was analyzed using a Carlo Erba 5160 gas chromatograph (GC) installed with a cool on-column injector,

Table 1. List of Core Samples

Core Name ^a	Latitude	Longitude	Water Depth, m	Core Length, cm	Location	Sources
TSP-2MC	48°07.6'S	146°54.0'E	2283	10	Southern Ocean	this study
TSP-3MC	48°33.5'S	146°24.7'E	2897	14	Southern Ocean	this study
TSP-2PC	48°08.2'S	146°52.5'E	2321	713	Southern Ocean	this study
KH92-1-5cBX	3°32.0'N	141°51.6'E	2282	31.5	western tropical Pacific	Ohkouchi et al. [1997a]
MANOPC	0°57.2'N	138°57.3'W	4287	330	central tropical Pacific	Prahl et al. [1989]

^a TSP, Tasman Plateau; MC, multiple core; PC, piston core; BX, box core.

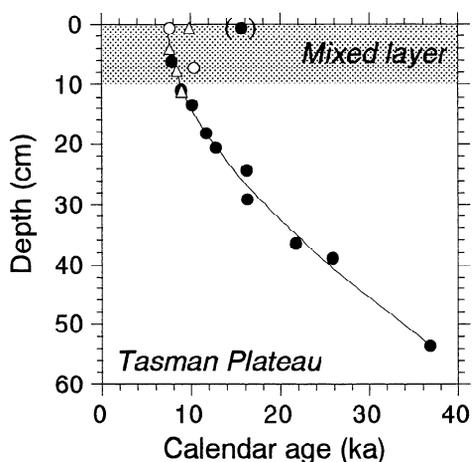


Figure 2. Vertical profile of calendar ages in multiple cores TSP-2MC (open circles), TSP-3MC (open triangles), and a piston core TSP-2PC (solid circles). Shaded interval shows a potential mixing layer due to bioturbation.

a HP-5 fused silica capillary column (30 m x 0.32 mm inner diameter (ID); 0.25 μm film thickness), and a flame ionization detector (FID). Hydrogen was used as a carrier gas. GC-mass spectrometric analyses were performed with a Finnigan MAT ITS-40 system with similar GC conditions.

During the experimental procedures we checked the recovery of lipids by using both internal and external standards. Internal standard recovery averaged $78.4 \pm 13.7\%$ (1σ , $n=31$) for $n\text{-C}_{19}$ alcohol. The reported concentrations of alcohols were corrected for the recoveries. However, the concentrations of aliphatic hydrocarbons and alkenones were not corrected for the recovery because recovery experiments showed that the recoveries of $\text{C}_{18}\text{-C}_{36}$ n -alkanes were generally higher than 90%. Triplicate analyses of composite sediments showed that the analytical errors in the experiments were 6.2% for C_{31} n -alkane and 5.6% for C_{24} fatty alcohol. Blank experiments that were performed in parallel with sample analyses showed no serious contamination peaks. The blank to sample ratios are usually $<3\%$ for C_{25} n -alkanes. Data presented here are also corrected for the procedural blanks.

The dried samples were ground to a fine powder and were split into two fractions. One fraction was analyzed for the measurement of total carbon content using a Fisons CNS Elemental Analyzer (NA1500). The other was treated with 3 M HCl for a few hours to remove carbonate carbon and analyzed for total organic carbon, nitrogen, and sulfur using the CNS analyzer. Total inorganic carbon contents were calculated from the difference between total carbon and total organic carbon contents. CaCO_3 contents were calculated from the total inorganic carbon content, assuming that all carbonate occurs as calcium carbonate.

4. Results

4.1. Surface Sediment Ages of Tasman Plateau and Core Chronology

A calendar age (15.8 ka) for the core top section (0-2.3 cm) of piston core TSP-2PC is apparently older than a subsurface

sample (7.82 ka and 4.8-7.3 cm; see Table 2 and Figure 2). This discrepancy is probably caused by the artificial disturbance of surface soft sediment layers by subsurface older sediments during a recovery process on the ship deck. On the other hand, a calibrated age of the surface sediments (0-1 cm) taken by a multiple corer (TSP-2MC) which prevents the coring disturbance is calculated to be 7.56 ka (Table 2 and Figure 2). A similar age is also obtained from another surface sample (TSP-3MC; see Table 2 and Figure 2) recovered from the southwest slope of the Tasman Plateau, suggesting that the combination of low sedimentation rate and bioturbation in this region causes the older ages (7-8 ka) at the surface sediments, except for the core top section of the piston core. Age-depth plots in cores TSP-2MC, TSP-3MC, and TSP-2PC on the Tasman Plateau indicate that the thickness of the bioturbated layer is ~ 10 cm (Figure 2), which correspond to ~ 9 ka in TSP-2PC. The vertical profile of these calendar ages in core TSP-2PC shows a smooth trend with a third-order polynomial fitting from subsurface sediments (6.1 cm) to the marine isotope stage (MIS) 3 section (53.4 cm; see Figure 2). Therefore the upper ~ 10 cm (~ 9 ka) section was not used in the discussion below.

4.2. Variations of Bulk Parameters

As shown in Figure 3a, total organic carbon (TOC) concentrations decrease from the Last Glacial Maximum (LGM) (average is 0.12%) to the last deglaciation (average is 0.10%) and stay relatively constant during the deglaciation. Total nitrogen (TN) contents vary from 0.015 to 0.023% with a pattern similar to the TOC contents. Molar C/N ratios are relatively low in the last glacial (average is 7.1) and increase

Table 2. Accelerator Mass Spectrometer (AMS) Radiocarbon Results on Planktonic Foraminifera from the Cores TSP-2MC, TSP-3MC, and TSP-2PC

Core	Sample Interval, cm	Middepth, cm	^{14}C Age, ^a years BP.	Calendar Age, ^b years BP.	Error, years
2MC	0.0-1.1	0.6	7,190	7,560	180
	6.7-7.8	7.2	9,810	10,320	130
3MC	0.0-1.2	0.6	9,300	9,820	130
	3.5-4.7	4.1	7,200	7,560	110
	7.0-8.2	7.6	8,140	8,450	110
	10.5-11.7	11.1	8,630	8,970	115
2PC	0.0-2.4	1.2	13,760	15,800	110
	4.8-7.3	6.1	7,490	7,820	140
	9.7-12.1	10.9	8,480	8,890	110
	12.1-14.5	13.3	9,510	10,120	160
	16.9-19.4	18.2	10,700	11,670	130
	19.4-21.8	20.6	11,220	12,730	160
	23.0-25.4	24.2	14,050	16,140	110
	27.9-30.3	29.1	14,180	16,290	120
	35.1-37.6	36.4	18,910	21,730	190
	37.6-40.0	38.8	22,400	25,820	190
52.1-54.6	53.4	32,130	36,790	530	

^a The ^{14}C ages were calculated with a 5568 year half-life.

^b AMS ^{14}C ages were converted to calendar years by CALIB 4.0 calibration program [Stuiver *et al.*, 1998] for the calendar years younger than 24,000 years BP. For the interval 24,000 to 40,000 calendar years BP; calendar years BP. are $3.0126 \times 10^{-6} (^{14}\text{C} \text{ years BP.})^2 + 1.2896 (^{14}\text{C} \text{ years BP.}) - 1005$ [Bard *et al.*, 1998].

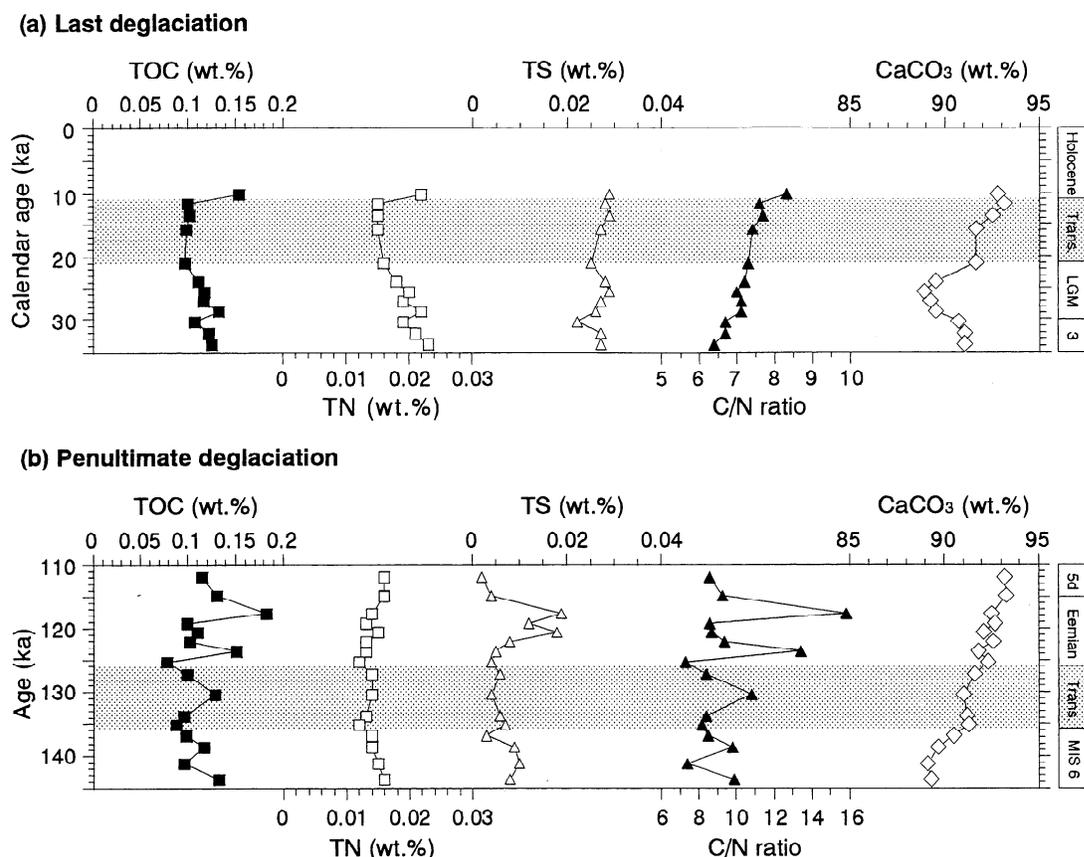


Figure 3. Down-core profiles of total organic carbon (TOC), total nitrogen (TN), total sulfur (TS) contents, C/N molar ratio, and CaCO₃ contents for (a) the last deglaciation and (b) the penultimate deglaciation in the core TSP-2PC. Shaded areas indicate each deglaciation period from glacial to interglacials.

toward the early Holocene (average is 8.5). They were relatively constant during the last transition (average is 7.5). The C/N ratios suggest that the marine-derived organic carbon dominates the source of sedimentary organic carbon during the last deglaciation, although C/N ratios of low percent TOC sediments are often anomalously low owing to the adsorption of ammonia on clay minerals [e.g., Müller, 1977]. Total sulfur (TS) (0.022–0.029%) also showed a profile similar to TOC and TN. Calcium carbonate contents range from 88.9 to 93.1% for the last deglaciation with a slight negative excursion at around 25 ka.

For the penultimate deglaciation, TOC shows a concentration range of 0.08 to 0.18% with positive peaks during the Eemian (Figure 3b). TN contents are rather constant in a range of 0.012–0.016%. Molar C/N ratios indicate short-term positive excursion during the Eemian. TS shows a distinct increase during the Eemian. Calcium carbonate content ranges from 89.1 to 93.3% with a gradual increase from the penultimate glacial to interglacial periods.

4.3. Molecular Distributions of Lipid Class Compounds

Homologous series of C₁₄–C₃₅ *n*-alkanes and C₁₂–C₂₈ fatty alcohols have been detected in the sediments. The molecular distributions of C₂₅–C₃₅ *n*-alkanes showed an odd-carbon

number predominance with a maximum at C₃₁ (Figures 4a and 4b). These distributions suggest that the alkanes are of continental higher-plant origin. It should also be noted that no anthropogenic signs such as unresolved complex mixture (UCM) were observed. The carbon preference indices (CPIs) ratio of the amounts of odd-carbon *n*-alkanes to those of even-carbon *n*-alkanes [Peltzer and Gagosian, 1989] for C₂₅–C₃₄ *n*-alkanes stayed ~8 throughout the two deglaciation periods, whereas small negative excursions were observed in the early Holocene and the Eemian. The CPI values of *n*-alkanes are generally known to be 5–10 for terrestrial higher-plant waxes and 1 for petroleum hydrocarbons and combustion residues of fossil fuels [e.g., Simoneit and Mazurek, 1982]. The higher CPIs have been observed in the marine aerosols collected at New Zealand, which is thought to be derived from terrestrial plants in Australia [Gagosian et al., 1987]. Molecular distributions of *n*-alkanes in our Tasman Plateau sediments are similar to that of Australia-derived aerosols, suggesting that the terrestrial organic compounds in the sediments mainly originated from the Australian continent.

Molecular distributions of C₂₄–C₂₈ fatty alcohols generally showed an even-carbon number predominance with a maximum at C₂₆ (Figures 4c and 4d). The CPIs for C₂₃–C₂₈ fatty alcohols varies between 5.3 and 7.2 during the two deglaciation periods. The CPI values of fatty alcohols are generally known to be 4–

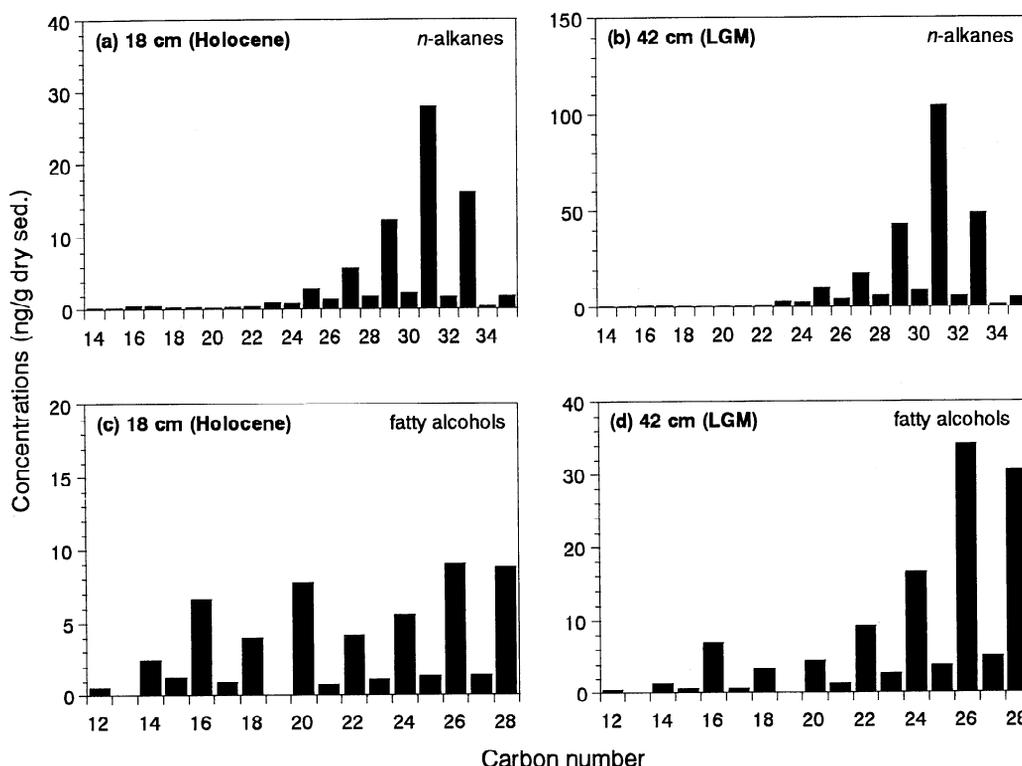


Figure 4. Chain length distributions of (a) *n*-alkanes (Holocene), (b) *n*-alkanes (Last Glacial Maximum (LGM)), (c) fatty alcohols (Holocene), and (d) fatty alcohols (LGM) in the sediment core (TSP-2PC). Notice the differences in the vertical scales in each plot.

20 for terrestrial higher-plant waxes [Simoneit and Mazurek, 1982]. Thus the results of *n*-alcohols also indicate that the organic materials in the core TSP-2PC are contributed greatly from the terrestrial higher plants [e.g., Eglinton and Hamilton, 1967; Simoneit, 1977].

Molecular compositions of *n*-alkanes and *n*-alcohols did not show significant change during the last two deglaciation periods. Average carbon chain lengths (ACLs) of higher molecular weight (HMW) *n*-alkanes is defined as follows [see Ohkouchi et al., 1997a]:

$$ACL_{alk} = (25[C_{25}] + 27[C_{27}] + 29[C_{29}] + 31[C_{31}] + 33[C_{33}] + 35[C_{35}]) / ([C_{25}] + [C_{27}] + [C_{29}] + [C_{31}] + [C_{33}] + [C_{35}]),$$

where $[C_n]$ is concentration of C_n *n*-alkanes. ACLs of *n*-alkanes (C_{25} – C_{35}) and fatty alcohols (C_{24} – C_{28}) stayed nearly constant around 30.5 and 26.3 for the last two deglaciations, respectively. These results indicate that the sources of terrestrial biomarkers did not change during each deglaciation period.

4.4. Terrestrial Biomarker Flux During the Last Two Deglaciations

Mass accumulation rates (MARs) calculated for terrestrial biomarkers (total C_{25} – C_{35} *n*-alkanes and total C_{24} – C_{28} fatty alcohols) in the last deglaciation (9–35 ka) are shown in Figure 5a. These compounds are derived from leaf waxes of terrestrial

higher-plants [e.g., Eglinton and Hamilton, 1967; Tulloch, 1976; Simoneit, 1977; Ohkouchi et al., 1997b]. Their MARs exhibit a distinct positive peak at around 25–28 ka in the middle of the LGM, and a gradual decrease toward the latest deglaciation period (Figure 5a). In contrast, MARs of these compounds show lower values at the boundary between the deglaciation and postglacial periods (around 11–12 ka). Averaged MARs of C_{25} – C_{35} *n*-alkanes and C_{24} – C_{28} fatty alcohols in the LGM (22–29 ka) were 1.7 and 1.3 times higher than those of the latest deglaciation period (12–16 ka), respectively. Although MAR of terrestrial biomarkers increased in the early Holocene, this may be an overestimation due to bioturbational smoothing and/or diagenesis effects in the surface sediments.

For the penultimate deglaciation, MARs of HMW *n*-alkanes are distinctly high in MIS 6 and show a rapid decrease at around 136 ka, where the boundary between MIS 6 and the penultimate deglaciation period exists (Figure 5b). Such a rapid decrease of terrestrial biomarkers at the latest MIS 6 is consistent with the decrease found at the end of the LGM (around 22 ka). On the other hand, the MARs of *n*-alkanes stayed constant with low values around 110 ng/cm²/kyr from the Eemian to MIS 5d. MARs of fatty alcohols also show a trend similar to HMW *n*-alkanes (Figure 5b), although the amplitude of alcohols is less than that of *n*-alkanes. Averaged MARs of *n*-alkanes and fatty alcohols in the MIS 6 were 2.5 and 1.6 times higher than those of the Eemian warmest period between 120 and 126 ka, respectively.

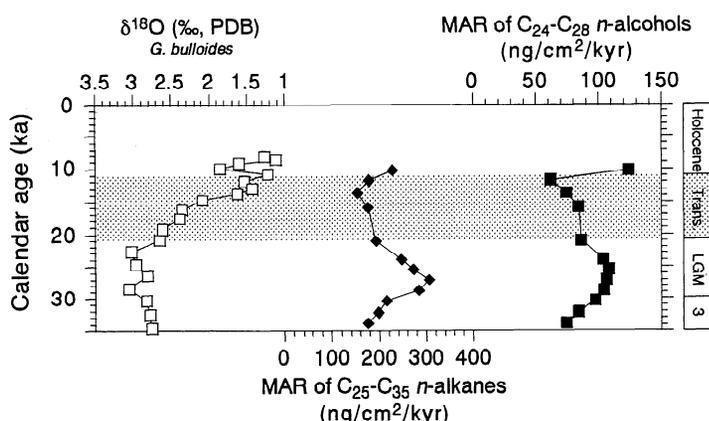
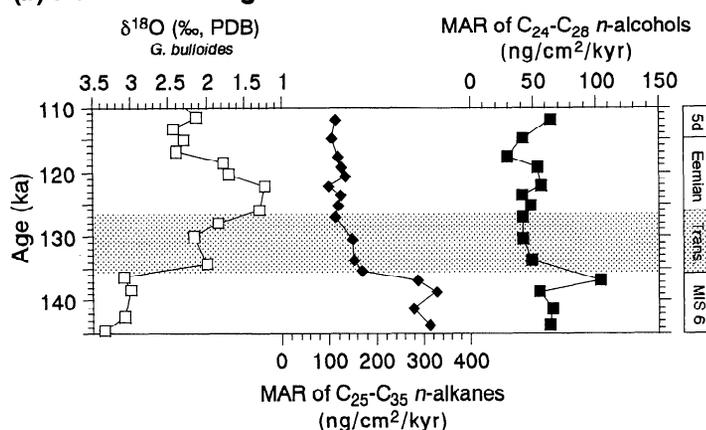
(a) Last deglaciation**(b) Penultimate deglaciation**

Figure 5. Down-core profiles of the $\delta^{18}\text{O}$ record of *Globigerina bulloides*, mass accumulation rates (MARs) of $\text{C}_{25}\text{-C}_{35}$ *n*-alkanes, and MARs of $\text{C}_{24}\text{-C}_{28}$ fatty alcohols for (a) the last deglaciation and (b) the penultimate deglaciation in core TSP-2PC. Shaded areas indicate each deglaciation period from glacial to interglacials.

4.5. Marine Biomarker Flux During the Last Two Deglaciations

MARs of dinosterol, brassicasterol (24-methylcholesta-5,22-dien-3 β -ol), and total $\text{C}_{37}\text{-C}_{39}$ long-chain alkenones are plotted during the last two deglaciation periods in Figures 6a and 6b, respectively. Dinosterol is specific to dinoflagellates [Boon *et al.*, 1979; Robinson *et al.*, 1984], whereas brassicasterol is a marker for diatom inputs to marine sediments [Volkman, 1988]. Long-chain alkenones are biosynthesized specifically by haptophyte algae [Volkman *et al.*, 1980; Marlowe, 1984; Marlowe *et al.*, 1990]. The MARs of dinosterol and brassicasterol exhibit distinct positive peaks at the LGM and a gradual decrease toward the latest transition period (Figure 6a). MARs of these compounds also show lower values at the boundary between the deglaciation and postglacial periods (around 11-12 ka).

For the penultimate deglaciation, MAR profiles of the sterols are characterized by relatively higher values in the glacial stage (MIS 6), gradual decrease toward the penultimate deglaciation period, and significant increase in the MIS 5d

(Figure 6b). It is of interest to note that these marine biomarkers exhibit a minimum MAR during the early Eemian between 122 and 126 ka. Averaged MARs of dinosterol and brassicasterol in the MIS 6 were 1.9 and 2.3 times higher than those of the early Eemian, respectively.

In contrast, the MARs of $\text{C}_{37}\text{-C}_{39}$ alkenones show a depth profile somewhat different from other marine biomarkers (Figure 6). Their MAR values in the early last deglaciation (15-21 ka) are as high as those of the MIS 5d and early MIS 6. The late last deglaciation (11-14 ka) and early Eemian (120-126 ka) exhibit a markedly low MARs. These results indicate that past changes in haptophyte algal production is not linked to the glacial-interglacial climate cycle in the Southern Ocean. The sedimentary C_{37} alkenone fluxes in the North Atlantic are controlled by the precessional cycle (23 kyr) during the late Quaternary [Villanueva *et al.*, 1998]. The alkenone production may also be controlled by their physiological condition [Brassell, 1993]. For better understanding the production mechanism of haptophyte algae in the southern high-latitude ocean, a long-term study of alkenone production and coccolithophorid assemblages in the Southern Ocean is needed.

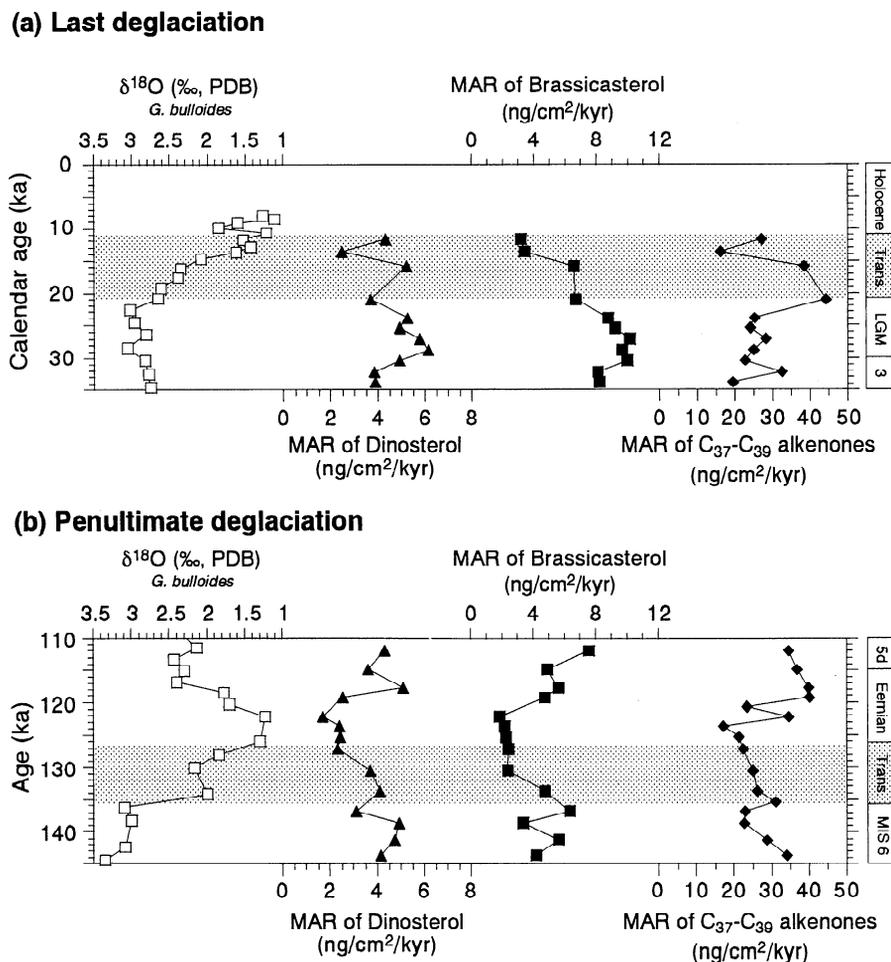


Figure 6. Down-core profiles of the $\delta^{18}\text{O}$ record of *Globigerina bulloides*, and mass accumulation rates (MARs) of dinosterol, brassicasterol, and C₃₇-C₃₉ alkenones for (a) the last deglaciation and (b) the penultimate deglaciation in core TSP-2PC. Shaded areas indicate each deglaciation period from glacial to interglacials.

5. Discussion

5.1. Atmospheric Transport of Terrestrial Biomarkers

Long-range atmospheric transport of terrestrially derived organic compounds (*n*-alkanes, *n*-alcohols, and fatty acids) to the open ocean has been studied in the tropical North and South Pacific Ocean [Gagosian *et al.*, 1982; Gagosian and Peltzer, 1986; Peltzer and Gagosian, 1989; K. Kawamura and K. Tanaka, manuscript in preparation, 1999]. Wax components derived from terrestrial higher-plants were also detected in the marine atmosphere over the North Pacific [Kawamura, 1995] and over the Southern Ocean including the Tasman Plateau (K. Kawamura and T. Niwai, unpublished results, 1996). We consider that atmospheric transport is the major pathway for the terrestrial organic compounds detected in the Tasman Plateau sediments. The core site, located on the southwest slope of the Tasman Plateau, is far away (600 km south) from Tasmania. Furthermore, there is no significant river inflow from the Australian continent over the Tasman Plateau.

Our biomarker MAR studies suggest that the atmospheric transport of terrestrial compounds increased significantly at the last two glacials (LGM and MIS 6) and decreased at the onset of deglaciation (around 22 ka and 136 ka; see Figure 5). The enhanced atmospheric transport is most likely caused by increased wind velocity and/or extended arid areas on the continents during the glacials. Our observations and interpretations agree well with the previous studies in the equatorial Pacific [Prah *et al.*, 1989; Prah, 1992; Ohkouchi *et al.*, 1997a], where the abundance of terrestrial biomarkers decreased at the transition from the last glacial to the Holocene. This is consistent with a sedimentological study, which reported that aeolian dust flux in the Tasman Sea sediments increased during glacial periods by a factor of 1.5-3 compared to the flux during interglacial period [Hesse, 1994]. Recently, Hesse and McTainsh [1999] reported that midlatitude westerlies in the Australian region of the Southern Hemisphere in the LGM were not stronger than those in the Holocene, based on particle size distribution of aeolian dust over the Tasman Sea. Thus increased terrestrial inputs on the glacial Southern Ocean may be associated with expanded source areas rather than enhanced wind speed.

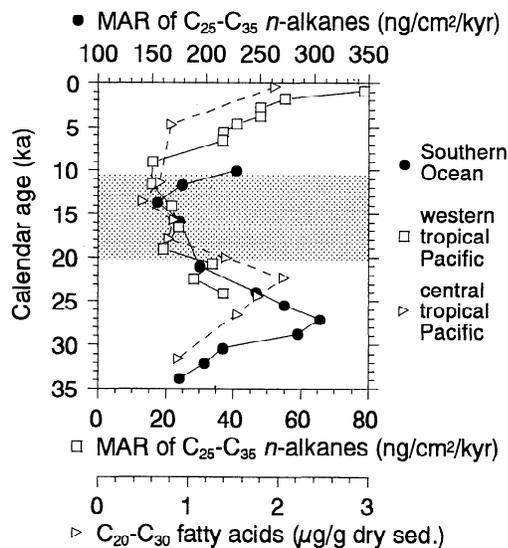


Figure 7. Comparisons of mass accumulation rates (MARs) of terrestrial biomarker (C_{25} - C_{35} n -alkanes) between the Southern Ocean core TSP-2PC (solid circles) and the western tropical Pacific core KH92-1-5cBX (open squares) [Ohkouchi *et al.*, 1997a] during the last deglaciation period. Concentrations of terrestrially-derived fatty acids at the MANOP C (central tropical Pacific) are also plotted by open triangles [Prahel *et al.*, 1989]. Sedimentary ages of the MANOP C records are recalculated for calendar age. Note that the lateral MAR scales are adjusted.

5.2. Comparison of Terrestrial Biomarker Fluxes Between Southern Ocean and Western Tropical Pacific

Ohkouchi *et al.* [1997a] reported that MARs of terrestrial biomarkers such as C_{25} - C_{35} n -alkanes and C_{24} - C_{28} fatty alcohols in the western tropical Pacific were significantly low during the last deglaciation period, which was interpreted to be the result of a depressed atmospheric circulation. Figure 7 shows the comparison of MARs of total C_{25} - C_{35} n -alkanes from the present Southern Ocean sediments with those of the western tropical Pacific [Ohkouchi *et al.*, 1997a] (Table 1). Concentrations of terrestrially derived fatty acids from the central tropical Pacific [Prahel *et al.*, 1989] (Table 1) are also plotted in Figure 7. These terrestrial biomarkers present a very similar signal with a gradual decrease from the late LGM to the boundary between the last deglaciation and post glacial period, although the western tropical Pacific records are limited to the last 24 ka. These comparisons suggest that the variation of atmospheric circulation in the glacial-interglacial cycle was similar between high- and low-latitude Pacific. However, the MARs of C_{25} - C_{35} n -alkanes in the Southern Ocean sediments (TSP-2PC) are 5-7 times greater than those of western tropical Pacific (Figure 7).

A coincidence in time series variation of terrestrial inputs between high- and low-latitude oceans is an important finding for paleoclimatology of the Earth, in spite of the fact that the wind flow patterns are different between the Southern Ocean and tropical Pacific. The core site of TSP-2PC (Southern Ocean) is located under the influence of the zonal westerly winds, while KH92-1-5cBX (western tropical Pacific) is located under

the trade wind belt. Therefore the source regions of terrestrial materials in the two cores should be different and their molecular composition may be unique to the regions. In fact, distributions of n -alkanes were found to be different in these two sediment cores; that is, n -alkane CPI values are 6.4-8.7 in TSP-2PC and 2.7-3.8 in KH92-1-5cBX [Ohkouchi *et al.*, 1997a]. Nevertheless, the similarity in the variation of atmospheric transport in the glacial-interglacial cycle in high- and low-latitude oceans suggests that enhancement and depression of atmospheric circulation occurred on a global scale (rather than regional) in the past.

5.3. Possible Causes for High Productivity in the Glacial Southern Ocean

The increased MARs of dinosterol and brassicasterol during the LGM, stadial MIS 5d, and full glacial MIS 6 are likely due to an enhanced primary productivity in the surface water and subsequent export flux of these compounds to the underlying bottom sediments (Figure 6). However, the productivity was probably reduced significantly at the boundary between the deglaciation and postglacial periods (around 11-12 ka) and during the last interglacial climate optimum (the early Eemian) which occurred between 122 and 126 ka. These results indicate that past productivity of phytoplankton including diatom and dinoflagellate increased significantly in the subantarctic surface water north of the present position of the APF during the glaci-als. Fluxes of marine biomarkers to glacial sediments were greater than in interglacials by more than a factor of 2 (Figure 8), raising the question, Why did the paleoproductivity increase during the glacial periods? Two reasonable explanations could be offered. One explanation is that an enhanced atmospheric input of dusts caused an increased productivity in the HNLC surface water in the Southern Ocean. The other is a northward migration of the high-production zone associated with the polar front.

The second iron enrichment experiments (IronEx II) were performed in the equatorial Pacific Ocean, which is one of the HNLC regions in the world ocean [Coale *et al.*, 1996; Cooper *et al.*, 1996; Behrenfeld *et al.*, 1996; Turner *et al.*, 1996]. On the basis of the results of IronEx II, Coale *et al.* [1996] concluded that the phytoplankton growth in this region is limited by iron bioavailability. The iron fertilization experiment also induced phytoplankton growth and subsequent uptake of surface water CO_2 [Cooper *et al.*, 1996]. Turner *et al.* [1996] pointed out that there is an urgent need to test these iron-DMS-climate relationships in the Southern Ocean.

Parts of the Southern Ocean are another important HNLC region. Sedwick *et al.* [1999] reported that phytoplankton growth rates are limited by iron deficiency within the iron-poor subantarctic Southern Ocean, based on the iron and nutrient enrichment experiments in the southwest of Tasmania. If iron bioavailability has changed in the past Southern Ocean, the primary productivity and subsequent export of organic carbon to the sediments could have a significant effect on atmospheric CO_2 partial pressure in the glacial-interglacial cycles [e.g., Sarmiento and Orr, 1991; Siegenthaler and Sarmiento, 1993]. On the basis of the terrestrial and marine biomarker fluxes, the increase in the input of terrestrial materials and the primary productivity were inferred to be

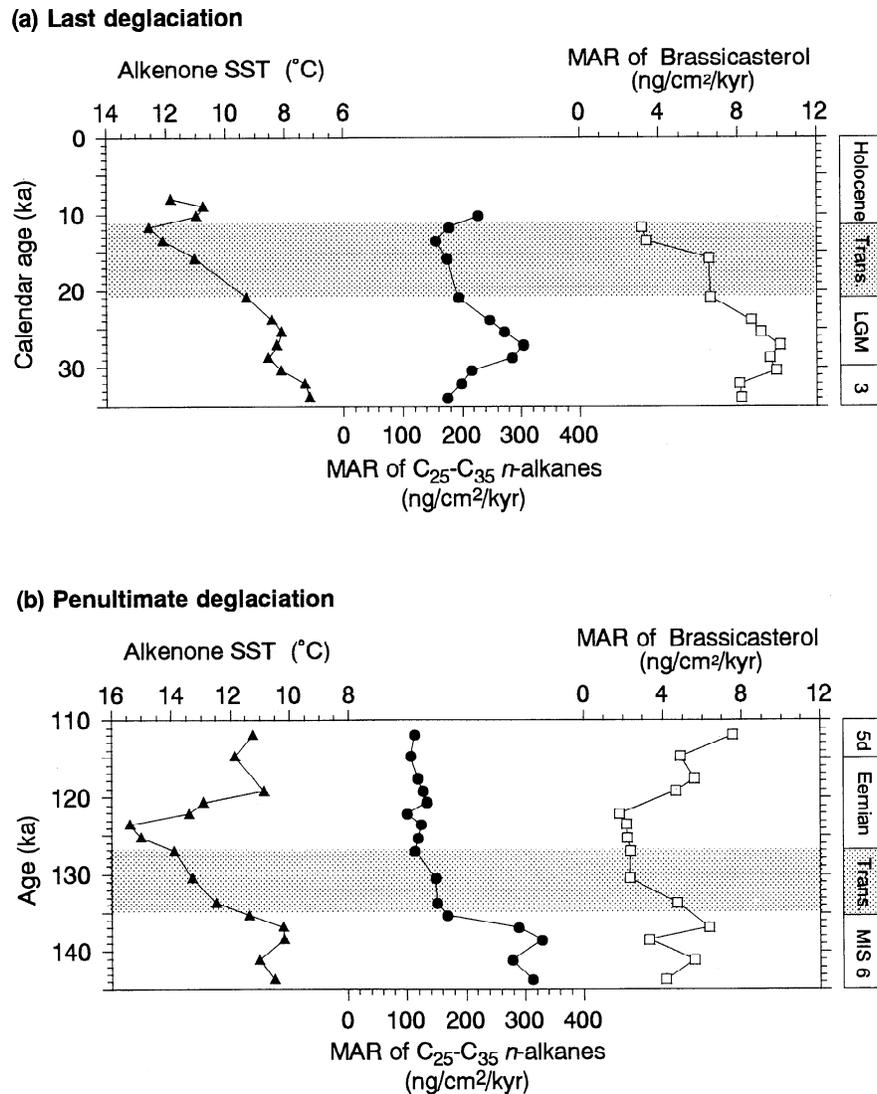


Figure 8. Comparison of alkenone SST records [Ikehara *et al.*, 1997] with the terrestrial inputs (C_{25} - C_{35} n -alkanes) and marine productivity (brassicasterol) in core TSP-2PC for (a) the last deglaciation and (b) the penultimate deglaciation. Shaded areas indicate each deglaciation period.

coupled in the last two glacials in the Southern Ocean (Figure 8), suggesting that fertilization by atmospheric dust input caused an enhanced phytoplankton productivity in the Southern Ocean surface water during the glacials. There is evidence, based on the Vostok ice core study, that the atmosphere of the Southern Hemisphere had elevated concentrations of dust [De Angelis *et al.*, 1987] and MSA [Saigne and Legrand, 1987]. Thus we consider that an increased atmospheric input of continent-derived materials containing iron had led to the propagation of primary production in the subantarctic surface water of the Southern Ocean, although there are many unclear mechanisms on the phytoplankton utilization of iron in seawater [e.g., Hutchins *et al.*, 1999].

A similar trend has been reported, based on opal accumulation records in the South Atlantic, where the marine

productivity was reduced in the south of the present position of the APF and increased in the north of the APF during the last glacial [Mortlock *et al.*, 1991; Charles *et al.*, 1991; Kumar *et al.*, 1993]. Kumar *et al.* [1995] suggested an enhancement of Fe-based glacial productivity north of the present APF in the Atlantic sector of the Southern Ocean. In contrast, Nürnberg *et al.* [1997] postulated higher barium accumulation in the glacial subantarctic zone north of the APF. These studies suggest that marine productivity in the entire subantarctic ocean in the Pacific, Indian, and Atlantic sectors was enhanced under glacial conditions. The increased production in the subantarctic Southern Ocean, where one of the strongest oceanic sinks for atmospheric CO_2 , due in part to biological uptake [Metzl *et al.*, 1999], may have contributed to the lower concentrations of carbon dioxide in the glacial atmosphere.

5.4. Enhanced Productivity During the Stadial MIS 5d

During the MIS 5d, although the terrestrial biomarker fluxes are rather constant, marine biomarker fluxes significantly increased (Figure 8b). There is no report on a significant enhancement of dust flux during the MIS 5d in the Tasman Sea [Hesse, 1994] and the Vostok ice core [Petit et al., 1990]. This indicates that the dust inputs are not linked to the primary productivity at the cooling phase from the Eemian to MIS 5d. Changes in the marine biomarker fluxes in the Southern Ocean are probably involved with the latitudinal migration of the high-production zone ("silica belt") associated with the latitudinal migration of the APF. Ikehara et al. [1997] showed that the alkenone SSTs dropped by $\sim 4^\circ\text{C}$ at the transition from the mid-Eemian to subglacial MIS 5d (Figure 8) and interpreted this as being due to the northward migration of the APF. The higher marine productivity recorded during the late Eemian to the MIS 5d may be initiated with the northward migration of the APF. This interpretation is supported by the higher flux of marine biomarkers in surface sediments underlying the present APF than those of the Tasman Plateau (M. Ikehara et al., unpublished results, 1998).

6. Conclusions

1. Mass accumulation rates (MARs) of terrestrial biomarkers ($\text{C}_{25}\text{-C}_{35}$ *n*-alkanes and $\text{C}_{24}\text{-C}_{28}$ fatty alcohols) showed a significant increase at the last two glacial periods (LGM and MIS 6), suggesting that atmospheric transport was enhanced probably due to an intensified atmospheric circulation and/or an expansion of arid area on the continents at that time.

2. Terrestrial biomarker MARs in the Southern Ocean gave similar time series variations with those of western equatorial

Pacific, suggesting that the variation pattern in the atmospheric circulation for the glacial-interglacial cycle is synchronized in high- and low-latitude oceans. However, MARs of terrestrial biomarkers from the Southern Ocean were 5-7 times greater than those in the western tropical Pacific.

3. Marine biomarker (dinosterol and brassicasterol) MARs also increased at the last two glacials (LGM and MIS 6) and stadial (MIS 5d), suggesting that marine biological productivity was enhanced in these periods. In contrast, their MARs showed a decrease during both warm interglacials (early Holocene and early Eemian).

4. Both terrestrial material inputs and surface biological productivity were found to have synchronously increased in the last two glacials in the Southern Ocean. The increased productivity, which may have contributed to lower the concentrations of glacial atmospheric CO_2 , was accompanied by an enhanced dust input.

5. At the global cooling from the Eemian to stadial MIS 5d, when ice sheets expanded, marine productivity also increased in the Southern Ocean. However, atmospheric circulation does not appear to have changed significantly at that transition. These productivity changes can be interpreted due to a northward migration of the Antarctic Polar Front (APF).

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