

# Biomarker reconstruction of phytoplankton productivity and community structure changes in the middle Okinawa Trough during the last 15 ka

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**Biomarkers have been widely used to reconstruct phytoplankton productivity and community structure changes, and this method has been applied for the first time in the middle Okinawa Trough during the transition from the last deglaciation to the Holocene. The total content of all marine phytoplankton biomarkers, used as a total productivity indicator, reveals higher productivity during the deglaciation. The ratios of the biomarkers are used as community structure indicators which show that, compared with the Holocene, the contribution from haptophytes decreased while the contributions from diatoms and dinoflagellates increased during the deglaciation. The increased productivity during the deglaciation was likely caused by the stronger winter monsoon. Also increased nutrient supply from terrestrial sources contributes to the higher productivity due to lower sea-level, which is consistent with higher terrestrial biomarker (long-chain *n*-alkanols) content. These changes in the nutrient supply also contributed to the community structure changes in the Okinawa Trough.**

biomarkers, Okinawa Trough, community structure, deglaciation, Holocene

Atmospheric CO<sub>2</sub> level variations can both cause and respond to climate changes<sup>[1]</sup>. The ocean is an important carbon reservoir containing 55 times more carbon than the atmosphere, and hence, ocean carbon cycle can influence both atmospheric CO<sub>2</sub> concentration and climate. The “biological pump” is the most important process of the ocean carbon cycle, and as primary producers, the phytoplankton largely control the “biological pump” efficiency. Phytoplankton total productivity and community variations can both change the biological pump and its control on carbon cycles<sup>[2]</sup>. Therefore, understanding the geological evolution of phytoplankton productivity and community structure could help us to decipher carbon cycle variability and mechanisms.

Previous reconstructions have revealed spatial variability regarding productivity changes during glacial-interglacial transitions. For example, while productivity

of the upwelling region of the Arabian Sea was higher during the glacials than during the interglacials<sup>[3]</sup>, productivity for the mid- and low-latitude regions showed an opposite trend<sup>[4]</sup>. Studies on glacial-interglacial community structures also revealed two modes: relatively stable versus major changes. Schubert et al.<sup>[5]</sup> reconstructed community structure changes for the monsoon-influenced upwelling regions of the Arabian Sea and found that while total productivity changed dramatically during the last 200 ka, phytoplankton community structure remained relatively stable on a millennial timescale. Schulte and Bard’s<sup>[6]</sup> study of a site on the Maldives platform of the Indian Ocean also suggested

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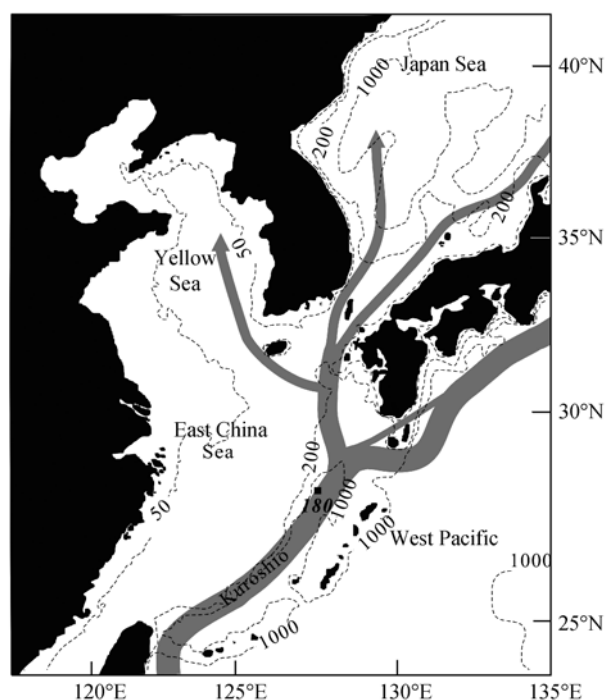
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that the community did not change significantly during the last 320 ka. In contrast, other studies revealed large glacial-interglacial changes in community structure<sup>[7–12]</sup>, for example, Werne et al.<sup>[9]</sup> observed a shift from a diatom-dominated phytoplankton community in the Cariaco Basin during the Younger Dryas (YD) to a coccolithophorid-dominated community during the Holocene. Such community changes were likely caused by changes in both concentration and composition of nutrients. Even with these reconstructions, there is still a lack of a clear understanding of glacial-interglacial changes in productivity due to the use of different proxy methods<sup>[13]</sup> and the limited number of records from different oceans<sup>[14]</sup>. This is especially true for the mid- to low-latitude western Pacific, thus productivity reconstruction employing the multi-biomarker approach has important implications in understanding the regional patterns of glacial-interglacial productivity changes and their role in the global carbon cycle.

This paper focuses on productivity reconstruction using a sediment core from the middle Okinawa Trough off the continental shelf of the East China Sea (Figure 1). The location is influenced by air-sea interaction, and high sedimentation rates offer high resolution records to register both marine and terrestrial climate changes. Nutrients and productivity for this region have been influenced by the Kuroshio Current variability and by river discharge changes associated with sea-level fluctuations<sup>[15]</sup>. Productivity reconstruction of the Okinawa Trough has been carried out by Chinese scientists using microfossils, and it was found that productivity was higher during the last full glacial compared with that during the late glacial and Holocene<sup>[16–18]</sup>. There are few records of Okinawa paleoenvironmental variability using biomarkers. Most are sea surface temperature records (SST) using the  $U_{37}^{K'}$  index, but some are concerned with sedimentary origins using *n*-alkanes and redox conditions using isoprenoids<sup>[19,20]</sup>. High resolution records of paleoceanographic conditions and community structure are sparse. Thus, multi-biomarker analyses from the same core can provide new insight into the variability of both total and individual phytoplankton productivity, as well as the corresponding changes in community structure. This paper reports the first reconstruction of deglaciation to Holocene productivity and community structure changes for the middle Okinawa Trough. The following biomarkers are used as productivity proxies:



**Figure 1** Location of core 180 (■) modified from Li et al.<sup>[26]</sup>. The shaded arrows indicate the Kuroshio Current and its branches.

brassicasterol for diatoms, alkenones for coccolithophorids, dinosterol for dinoflagellates, cholesterol for zooplankton, and long-chain alkyl diols and ketols for eustigmatophytes<sup>[5–12,21]</sup>.

## 1 Material and methods

Core 180 (305 cm) was recovered by the First Institute of Oceanography, State Oceanic Administration in the deep basin of the middle Okinawa Trough (Figure 1). The core consists mostly of hemipelagic muds, and the lithology ranges from clay to silt with foraminiferal calcareous oozes<sup>[22]</sup>. Based on mineralogical and elemental analyses, it was concluded that intervals 70–130 cm and 210–250 cm contained volcanic ash deposits. According to the two  $^{14}\text{C}$  dates of foraminifers<sup>[23]</sup>, the 70–130 cm layer corresponds to the K-Ah volcanic eruption<sup>[24]</sup>. However, it is not possible to correlate the 210–250 cm layer to any volcanic eruption event, due to the lack of any age control points covering this interval. The age model for core 180 is constrained by the foraminiferal  $\delta^{18}\text{O}$  curve and the two  $^{14}\text{C}$  dates, converted to calendar ages using calendar 5.1 (<http://radiocarbon.pa.qub.ac.uk/calib/>)(Table 1). According to this age model, the bottom of core 180 is approximately 15

**Table 1** AMS<sup>14</sup>C dates for Core 180 from the middle Okinawa Trough

Depth (cm)	Foraminifers	Measured <sup>14</sup> C age (a)	Corrected <sup>14</sup> C age (a)	Calendar age (a)
117–122	<i>G.sac</i> <sup>a)</sup> and <i>N.dut</i> <sup>b)</sup>	10520 ± 240	10120	11681 ± 409
285–290	<i>G.sac</i> and <i>N.dut</i>	13120 ± 110	12720	15006 ± 193

a) *Globigerinoides sacculifer*; b) *Neogloboquadrina dutertrei*.

ka. Due to the lack of detailed age control, we have divided the records into the deglacial and the Holocene (separated at 107 cm), and we emphasize the deglacial and Holocene differences in our discussion of the biomarker records.

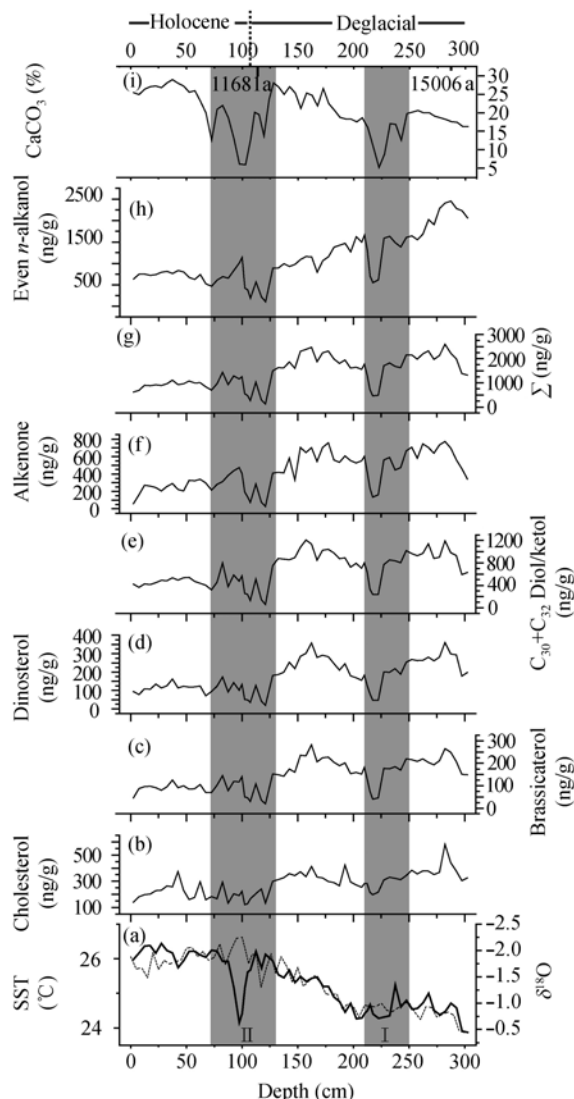
Samples of 1 cm each were taken at 5 cm intervals, resulting in 65 samples. The analyses of planktonic foraminifer *Globigerinoides sacculifer*  $\delta^{18}\text{O}$  and  $\text{CaCO}_3$  content were reported<sup>[23]</sup>. Biomarker determination involved sample extraction and instrumental analysis. Freeze-dried samples of about 2 g were extracted with a mixture of dichloromethane and methanol (3:1) by ultrasonication four times, after adding  $\text{C}_{24}$  deuterium-substituted *n*-alkane and  $\text{C}_{19}$  alcohol as internal standards (IS).

The extracts were first hydrolyzed in KOH-MeOH solution and preliminarily separated using silica gel chromatography. The neutral lipid fraction, which contains the alkanols, alkenones and the sterols, was dried and derivatized for instrumental analyses. Biomarker identification and structure verification were performed on a Thermo Gas Chromatograph-Mass spectrometer (GCMS). Quantification of the biomarkers was done on an Agilent 6890N GC, using a HP-1 column (50 m),  $\text{H}_2$  as carrier gas at 1.2 mL/min. The biomarker content was calculated by ratioing its GC peak integration to that of the IS. SST was calculated using the Müller equation,  $\text{SST} = (U_{37}^{K'} - 0.044)/0.033$ <sup>[25]</sup>.

## 2 Results and discussion

### 2.1 Sea surface temperature

The SST (Figure 2(a)) oscillated between 23.9 and 25°C during the first part of the deglaciation (302.5–200 cm) with an average value of 24.5°C; SST began to increase at 200 cm and reached 26.2°C by 112.5 cm, followed by an abrupt cooling and reached 24.1°C at 97.5 cm. SST fluctuated around 26°C during the late Holocene (87.5–2.5 cm). The average Holocene SST for this core is slightly higher than the modern annual SST for this region, a feature that has been noted in several locations of the Okinawa Trough<sup>[24,27,28]</sup>. Nevertheless, the  $U_{37}^{K'}$



**Figure 2** Contents of biomarkers and other proxy records for core 180 plotted against depth. (a)  $U_{37}^{K'}$  SST (solid line) and planktonic foraminiferal  $\delta^{18}\text{O}$  (dashed line) records; (b) content of cholesterol (ng/g); (c) content of brassicasterol (ng/g); (d) content of dinosterol (ng/g); (e) content of  $\text{C}_{30}+\text{C}_{32}$  diols and ketols (ng/g); (f) content of  $\text{C}_{37}$  alkenones (ng/g); (g) total contents of brassicasterol, dinosterol,  $\text{C}_{30}+\text{C}_{32}$  diols and ketols, and  $\text{C}_{37}$  alkenones (ng/g); (h) content of the *n*-alkanols ( $\text{C}_{26}+\text{C}_{28}+\text{C}_{30}+\text{C}_{32}$ , ng/g); (i) content of  $\text{CaCO}_3$  (%). The shaded vertical bars indicate the two ash-containing layers I and II. The numbers of 11681a and 15006a indicate the two <sup>14</sup>C dates.

SST is best regarded as reflecting annual SST. Because core 180 did not reach the Last Glacial Maximum (LGM), its record could not be used to estimate the full glacial-interglacial SST difference. The deglacial-

Holocene SST difference is approximately 2°C, consistent with previous  $U_{37}^{K'}$  results from the Okinawa Trough<sup>[24,27,28]</sup>. With the exception of the cold event around 100 cm, the variations of SST and  $\delta^{18}\text{O}$  records parallel each other, i.e., increasing SST corresponds to more negative  $\delta^{18}\text{O}$  values. To date, most paleoceanographic researchers seek the phase relationship between SST and deglaciation changes by comparing SST and  $\delta^{18}\text{O}$  records<sup>[27,29]</sup>. When these two records are in phase, it is often interpreted that SST increase and glacier melting were synchronous. To some extent, our records indicate the Okinawa Trough SST increase was also synchronous with sea-level changes during the last deglaciation. However, recent results indicate that alkenones on fine-grained particles and the coarser-grained foraminifers from the same sedimentary layer could have different ages, due to their different sedimentary geochemical processes<sup>[30]</sup>. But the age differences are often within one thousand years<sup>[31]</sup>, and because our sampling resolution is relatively low, this age offset does not significantly affect our discussion of events longer than one thousand years. Our preliminary conclusion of synchronous SST and deglaciation changes can only be verified by future  $^{14}\text{C}$  dating of both alkenones and foraminifers from the same samples.

In addition to major influences by sea-level changes, the foraminiferal  $\delta^{18}\text{O}$  is also controlled by both SST and local salinity, two factors which are not easily separated. Normally, lower SST corresponds to more positive  $\delta^{18}\text{O}$ , but the cold event around 100 cm correlates to a positive  $\delta^{18}\text{O}$  excursion. Thus, this  $\delta^{18}\text{O}$  event could not have been caused by the SST change, and it was most likely caused by a lower salinity event, probably due to increased river discharge. Therefore, data from core 180 indicate that this could be a cold and wet event. However, this event is recorded within the volcanic ash-containing layer, and it is not possible to eliminate the possible influence of the volcanic eruption on the  $\delta^{18}\text{O}$  record. Since the long-chain even-carbon alcohols (*n*-alkanols) are produced by higher land plants, their sedimentary contents can be used as proxies for terrigenous inputs<sup>[12]</sup>. Although the *n*-alkanol content was lower within the broad ash-containing layer of 130–70 cm, there is a peak corresponding to the 100 cm cold event. Since volcanic ashes do not contain *n*-alkanols, this peak reflects increased terrigenous input (Figure 2(h)), most likely a result of increased river dis-

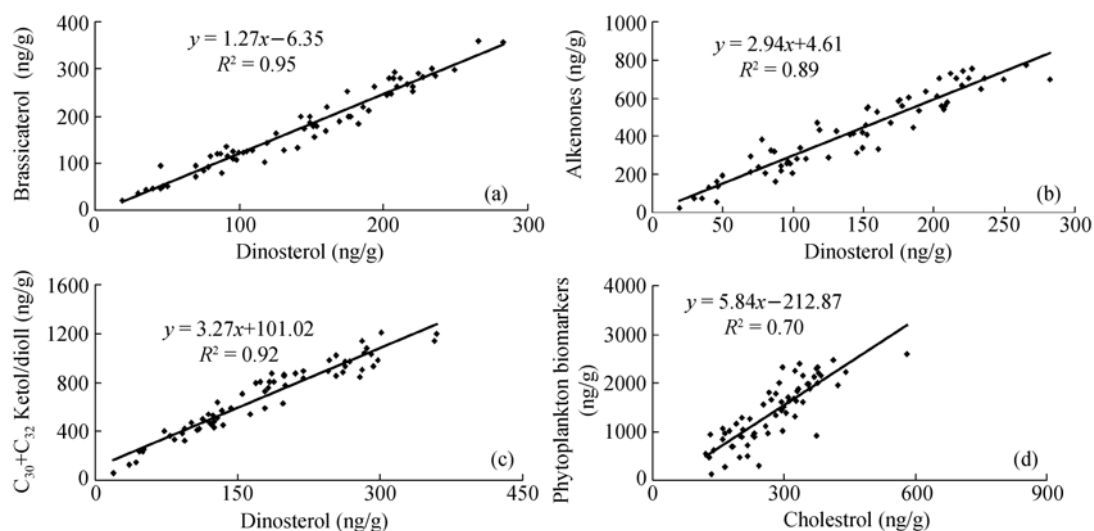
charge. The *n*-alkanol data provide indirect evidence that the negative  $\delta^{18}\text{O}$  event around 100 cm was caused by increased freshwater input.

## 2.2 Biomarker content

The biomarker content records are plotted along with the previously published  $\text{CaCO}_3$  content record<sup>[23]</sup> in Figure 2(a)–(i). From the deglaciation to the Holocene, the contents for all biomarkers reveal a decreasing trend, brassicasterol changed from 282.6 ng/g to 19.1 ng/g, dinosterol from 359.5 to 19.6 ng/g,  $\text{C}_{30}+\text{C}_{32}$  diols-ketols from 1207.6 to 57.5 ng/g, alkenones from 771 to 27.5 ng/g, and cholesterol from 581.1 to 121.2 ng/g. However,  $\text{CaCO}_3$  content shows an opposite (increasing) trend. In the first ash-containing layer (I in Figure 2) biomarker contents show minimum values, but in the second ash-containing layer (II in Figure 2) biomarker contents reveal a trend of decreasing values followed by increases. Within the second layer, biomarker and  $\text{CaCO}_3$  contents also have differences. This is most evident for the 100 cm cold event where biomarkers reveal an increasing trend while  $\text{CaCO}_3$  has minimum values. One explanation for some of the lower content values of these proxies in the two ash-containing layers is the “dilution effect” by the volcanic ashes. However, this does not exclude the deleterious effect on marine productivity by volcanic eruption-induced climate changes. Since the higher biomarker content values within the second ash-containing layer could not be a result of the “dilution effect”, a more plausible explanation for the higher biomarker values is increased productivity due to the increases of river transported nutrients. On the other hand, increased river input of terrigenous materials diluted sedimentary  $\text{CaCO}_3$  content. Sedimentary  $\text{CaCO}_3$  content dilution by terrigenous input has been recognized in the South China Sea, where during glacial times, increased productivity resulted in higher contents of TOC and biomarkers, but the “dilution effect” caused a decrease in  $\text{CaCO}_3$  content<sup>[32]</sup>.

The similarity of the biomarker content variations is shown in *X*-*Y* linear plots in Figure 3(a)–(d). It can be seen that brassicasterol and dinosterol have the best correlation ( $R^2=0.95$ ) (Figure 3(a)), but the correlations among the others are also very good. For example, the  $R^2$  between alkenones and dinosterol is 0.89 and 0.92 between diols-ketols and dinosterol. These high correlations indicate that the productivity of these phytoplanktons for the site generally co-varied, implying relatively





**Figure 3** X-Y plots of biomarker contents.

minor changes in phytoplankton community structure. Both content and mass accumulation rate (MAR) have been used for productivity reconstruction at other sites<sup>[7,9,12]</sup>. MAR is a better proxy, since it includes the effect of bulk sedimentation rate change on biomarker content. However, accurate calculation of MAR needs a well-constrained age model. The lack of such an age model for our core would result in large uncertainties for MAR calculation, so we prefer to use biomarker content as productivity proxies. To some extent, the summation of the contents of the four biomarkers can be used as a total productivity proxy, and it shows a similar trend as those for the individual biomarkers, i.e., higher values during deglaciation compared with the Holocene (Figure 2(g)). Cholesterol is produced by zooplankton, and to some extent can be used as a proxy for zooplankton biomass. The variation of this biomarker has a similar trend compared with the four phytoplankton biomarkers, and with the total biomarker content. An X-Y plot shows that cholesterol and summation of the phytoplankton biomarkers have a good correlation ( $R^2=0.70$ ) (Figure 3(d)). It can be concluded that both primary production and zooplankton biomass reveal higher values during deglaciation and lower values during the Holocene. It has been reported that sedimentary  $\text{CaCO}_3$  for the region is mostly biogenic<sup>[23]</sup>, thus it could also be used as a productivity proxy. However, for core 180, it has almost an inverse relationship with biomarkers, indicating the terrigenous “dilution effect” had more influence on sedimentary  $\text{CaCO}_3$  content. On the other hand, terrigenous materials also contain some  $\text{CaCO}_3$ , which

makes the sedimentary  $\text{CaCO}_3$  variations more difficult to interpret. Just as in the case for the South China Sea,  $\text{CaCO}_3$  content is not a good productivity proxy for core 180.

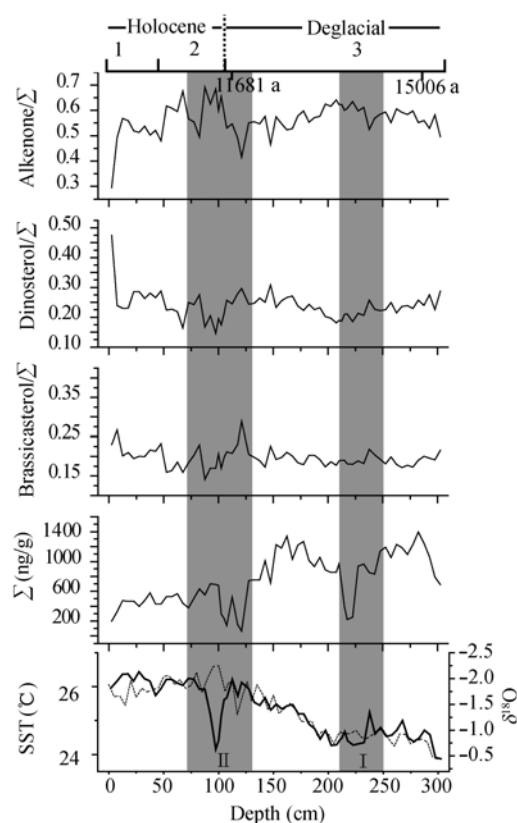
The above discussion shows that core 180 productivity was higher during deglaciation compared with the Holocene. This trend is in agreement with previous results from the Western Equatorial Pacific, the East China Sea and the Northwestern Pacific, but is opposite to the trend from the Eastern Pacific. Comparison of these results can shed some light on the forcing mechanisms of Pacific productivity changes. Previous reports found that productivity was higher during the glacial than the interglacials for the Western Pacific, and these changes were attributed to stronger East Asia winter monsoons during the glacial<sup>[33]</sup>. For example, a study by Chang et al.<sup>[18]</sup> showed that productivity for the Southern Okinawa Trough decreased since the LGM, revealing a trend of higher values during the glacial-deglaciation and lower values during the Holocene. A study of two cores from the Shatsky Rise of the mid-latitude North Pacific also showed higher productivity during the glacial<sup>[34]</sup>. In contrast, all records off California in the East Pacific reveal that productivity during the cold periods was lower compared with the warm periods, and dissolved oxygen in bottom water was higher during the cold periods. Higher productivity during the warm periods is linked with increased upwelling, caused by the strengthened subtropical high pressure cell<sup>[35]</sup>. In addition, for the East Equatorial Pacific, the east-west productivity gradient north of the Equator decreased during

the last glacial, indicating lower productivity for the eastern part<sup>[36]</sup>. Therefore, productivity change during the last glacial-interglacial transition for the eastern and western Pacific had opposite trends. These changes are linked with the north-south migration of the inter-tropical convergence zone (ITCZ) and the location of the subtropical high pressure cell during the glacial-interglacial transition. During the glacial, the ITCZ migrated southward, trade winds weakened, the East Asia winter monsoon strengthened, and the increased dust input could have increased productivity for the western Pacific region. In addition, a stronger winter monsoon would deepen the mixed layer, bringing nutrients to the surface layer to increase productivity. For site 180, the influence of sea-level changes on productivity is probably more significant. Lowering of sea-level during the glacial meant that the site was closer to the coast, thus terrigenous input of nutrients would increase and productivity would be higher compared with the Holocene. Results from our biomarker contents and ratio provide additional evidence supporting this model for the glacial-interglacial changes in nutrient input and productivity for the Okinawa Trough.

### 2.3 Phytoplankton community structure change

Biomarker content can not only be used to estimate individual phytoplankton productivity, but the ratio of the three main phytoplankton biomarkers (brassicasterol, dinosterol and alkenones) can also be used to indicate community structure changes for diatoms, dinoflagellates and coccolithophorids, respectively. The application of the biomarker ratios is based on the following assumptions. First, the three phytoplanktons with similar productivity would produce similar amounts of the corresponding biomarkers, and the amounts do not vary with environmental factors significantly. Although a few measurements show that the cellular contents of these biomarkers are similar<sup>[37,38]</sup>, this assumption is far from being verified due to the lack of cultured samples. In a more strict sense, the biomarker ratios should not be used to represent the absolute ratios of the phytoplankton, but the changes in the biomarker ratios could reflect the relative changes in the phytoplankton percentage<sup>[5,7,9]</sup>. Secondly, the diagenetic effect on these biomarkers is similar, so that their ratios in the photic zone could be preserved in sediments. Since these biomarkers have similar chemical properties, this assumption generally applies<sup>[39]</sup>. Plotted in Figure 4 are the diatom,

dinoflagellate and coccolithophorid biomarkers ratios to the total biomarker amounts. According to the biomarker ratios, coccolithophorids have been the most abundant phytoplankton for this core. The distribution of phytoplankton in modern oceans indicates that coccolithophorids are favored in high temperature, lower nutrient environments, while diatoms thrive in high nutrient environments<sup>[40,41]</sup>. Since the core location lies beneath the subtropical low productivity region, coccolithophorids are normally most abundant. The paleo-community structure derived from biomarker ratios is consistent with modern surveys. Figure 4 also reveals obvious community structure changes during the last 15 ka, which can be divided into three intervals based on the changing patterns. During the deglaciation, the community structure was relatively stable, but there were some minor changes in the ash-containing layer (210–250 cm) when total productivity was decreasing. Within this layer, diatom and dinoflagellate contributions first increased,



**Figure 4** Biomarker content percentage (normalized to the total content) and the  $U_{57}^{K'}$  and  $\delta^{18}\text{O}$  records for core 180.  $\Sigma$  = the total contents of brassicasterol, dinosterol and  $\text{C}_{37}$  alkenones. The records are divided into three intervals labeled on the top panel. 1. Late Holocene; 2. early Holocene; 3. deglaciation. The shaded vertical bars indicate the two ash-containing layers I and II. The numbers of 11681 a and 15006 a indicate the two  $^{14}\text{C}$  dates.

followed by decreases. During the early Holocene (107.5 – 47.5 cm), community structures underwent clear changes. When total productivity generally decreased, the most obvious individual changes were the increase in coccolithophorids and decreases in both diatoms and dinoflagellates. For the ash-containing layer (II in Figure 4), there were also some changes in community structure. Diatoms and dinoflagellates increased near the bottom of layer II, but their contributions decreased near the cold event (ca. 100 cm) when coccolithophorids increased. During the late Holocene, coccolithophorid contribution decreased while total productivity remained low. There was a major change in community structure near the coretop, when coccolithophorid contribution decreased rapidly, dinoflagellate contribution increased rapidly, and there were small increases in diatoms.

These results indicate that community change is not only related to total productivity, but also related to other environment factors, especially nutrient changes. Previous results reveal that dinoflagellates and diatoms can outcompete coccolithophorids under high nutrient environmental conditions, and diatoms are especially favored when dissolved silicon concentration is high<sup>[41]</sup>. Thus, when total productivity was higher during the deglaciation, the coccolithophorid relative contribution was lower. When total productivity was lower under the lower-nutrient-supply environment during the Holocene, coccolithophorid contribution was higher. Recent changes in coccolithophorids (rapid decrease) and dinoflagellates (rapid increase) indicate major changes in nutrient supply. These changes could be related to anthropogenic activity, but this inference can only be verified with accurate dating in the future. Community structures in the two ash-containing layers could be related to silicon supply by the eruptions, but the eruption-induced environmental changes could also play some role.

Previous studies found that there were major differences in the calcareous and siliceous productivities during the Holocene in the northern Okinawa Trough<sup>[18]</sup>. The calcareous plankton productivity peak occurred around 7 ka, followed by a significant decrease ~7.0–6.4 ka, but remained steady thereafter. Siliceous plankton productivity showed increasing trend prior to 3.0 ka, peaked at 3.0 ka, and then decreased afterwards. This process was proposed to be influenced by the Kuroshio

current variability and the associated eddy and upwelling strength. Detailed comparison of our biomarker records with these reports is not possible, due to the lack of a well-constrained age model for the Holocene for core 180. However, our biomarker records do reveal that the coccolithophorid (calcareous) contribution was higher during the early Holocene and lower during the late Holocene, in agreement with the published results. The rapid increases in dinoflagellate and diatom contributions near the core top in 180 are also consistent with previous reports. Our results provide biomarker evidence that increased total productivity and the increased diatom and dinoflagellate contribution during the deglacial could help to explain the lower CO<sub>2</sub> levels during the deglacial and glacial periods.

### 3 Conclusions

Biomarkers show that for the middle Okinawa Trough, total productivity and individual diatom, dinoflagellate and coccolithophorid productivity were all higher during the deglaciation compared with the Holocene.

Community structure reconstruction using biomarkers reveal that coccolithophorids have been the dominant phytoplankton during the last 15 ka, however, community structure did change. From the deglaciation to the early Holocene transition, the relative contribution from coccolithophorids increased, but the diatom and dinoflagellate contributions decreased. For the core top (late Holocene), nutrient input changes resulted in a rapid decrease in coccolithophorid contribution, rapid increases in dinoflagellates, and some increase in diatom contribution.

Productivity and community structure changes were most likely controlled by nutrient supplies. The reason for the higher productivity during the deglaciation could be the strengthened winter monsoon compared with the Holocene. Changes in terrigenous nutrient supply were also important, as the terrestrial biomarkers (*n*-alkanols) reveal a decreasing trend from the deglaciation to the Holocene. This decrease in terrigenous supply was mostly related to sea-level changes, but it was also influenced by the strengthening of the Kuroshio current and the weakening of the East Asia winter monsoon.

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