

## A record of Miocene carbon excursions in the South China Sea

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**Abstract** High-resolution  $\delta^{13}\text{C}$  records are presented for the Miocene benthic foraminifers *Cibicides wuellerstorfi* and *C. kullenbergi* (24–5 Ma) and the planktonic foraminifer *Globigerinoides sacculifer* (16–5 Ma) from ODP Site 1148A (18° 50.17'N, 116° 33.93'E, water depth 3308.3 m), northern South China Sea. The general pattern of parallel benthic and planktonic  $\delta^{13}\text{C}$  shows a decrease trend of  $\delta^{13}\text{C}$  values from the early-middle Miocene to the middle-late Miocene. Two distinct  $\delta^{13}\text{C}$  positive excursions at 23.1–22.2 and 17.3–13.6 Ma, and two negative excursions at 10.2–9.4 and 6.9–6.2 Ma have been recognized. All these events are cosmopolitan, providing the good data for the stratigraphic correlation of the South China Sea with the global oceans as well as for studying the changes of the global carbon reservoir and its corresponding climate.

**Keywords:** carbon isotope, carbon excursion, Foraminifera, paleoceanography, Miocene, South China Sea.

The fractionation of carbon isotope is the result of a rather complicated biogeochemical process (organic synthesis and decomposition), which is directly related to the changes of the carbon reservoir<sup>[1]</sup>, whereas the changes in oxygen isotope mainly reflect physical processes in a simpler manner (changes in temperature or water-vapor evaporation and precipitation). It is most likely the reason that the carbon isotope is far less applied than the oxygen isotope to the paleoceanographic study. However, the carbon isotope is of special value in tracing the earth system history just as it reflects the changes of the carbon reservoir, particularly the ratio of global organic and inorganic carbon. For examples, the dramatic depletion of sea-water  $\delta^{13}\text{C}$  in the late Cenozoic indicated the decreases of the global organic reservoir and  $\text{CO}_2$  content in the atmosphere<sup>[2]</sup>, and the decrease in sea-water  $\delta^{13}\text{C}$  during the glacial periods recorded the withdrawal of the terrestrial vegetation and the drop of the global biomass<sup>[3]</sup>. The dramatic increases and decreases in sea-water  $\delta^{13}\text{C}$  in some relatively short periods are just the evidence indicating the important changes in the earth surface system. These kinds of carbon excursion events occurred frequently in the Miocene and are the major interest of paleoenvironmental study.

The resolution of previous deep-sea records of the Miocene carbon isotope is generally low, particularly in the West Pacific. The paleoceanographic studies in China seas were mainly made

for the Quaternary and occasionally for the Miocene<sup>[4-6]</sup>. The ODP Leg 184 operated in spring 1999, for the first time, has provided continuous Miocene sequences since 24 Ma. We have done detailed isotopic analysis on benthic and planktonic foraminifers from Site 1148, and this paper is part of the results showing the Miocene  $\delta^{13}\text{C}$  curves of benthic and planktonic foraminifers since 24 Ma with emphasis on the carbon excursion events.

## 1 Material and methods

Hole 1148A is located at the northern slope of the South China Sea ( $18^{\circ}50.17' \text{N}$ ,  $116^{\circ}33.93' \text{E}$ , water depth 3308.3 m). Foraminifers for the isotopic analysis were taken from the section between core depth 188 and 475 mcd. This section has almost 100% recovery and sediments are mainly greenish gray calcareous nannofossil clay and ooze which, upon foraminiferal and calcareous nannofossil biostratigraphic data, were deposited during 23.8—5 Ma<sup>[7]</sup>. A total of 699 benthic and 440 planktonic foraminiferal samples have been analyzed for carbon and oxygen stable isotopic compositions. The methods for sample treatment and isotopic analysis are the same as those mentioned in our another paper<sup>[8]</sup>.

The benthic isotopic analyses were performed on *Cibicidoides wuellerstorfi* within the interval of 15—5 Ma and on *C. kullenbergi* within the interval of 24—15 Ma. *Cibicidoides* spp. or *Oridosalis* spp. and *Uvigerina* spp. were used as alternatives when the above species are absent. The time resolution averages 27.1 ka for the whole benthic samples of 23.8—5 Ma, but it is higher in the intervals of 5—10 Ma and 15—18 Ma with an average time resolution 21 ka and 19 ka respectively. The planktonic analyses were performed on *Globigerinoides sacculifer* for the period of 17.5—5 Ma. The time resolution is higher in the earlier and later intervals with an average resolution 13 ka for 17.5—15 Ma and 22 ka for 10—5 Ma respectively, and as low as 120 ka for the middle interval (15—10 Ma) due to the poor preservation of foraminifer tests.

## 2 Results

Fig. 1 shows the depth distribution of carbon isotopic data of the Miocene benthic *Cibicidoides wuellerstorfi* or *C. kullenbergi* and planktonic *Globigerinoides sacculifer*. The age distribution of isotopic data is shown in fig.2(b) and (c). Ages were calculated by interpolation between nine first and last occurrence time levels of planktonic foraminifera (marked with asterisk in fig.1). In general, both the benthic and planktonic  $\delta^{13}\text{C}$  curves display a parallel decreasing trend from the early-middle Miocene with most values higher than the average to the middle-late Miocene with most values lower than the average. The most dramatic  $\delta^{13}\text{C}$  increases (or  $\delta^{13}\text{C}$  positive excursions) occurred at 23.1—22.2 Ma and 17.3—13.6 Ma, and  $\delta^{13}\text{C}$  decreases (or  $\delta^{13}\text{C}$  negative excursions) at 10.2—9.4 Ma and 6.9—6.1 Ma, respectively. These positive and negative  $\delta^{13}\text{C}$  excursions clearly recorded the changes in the Miocene carbon reservoir and the corresponding fluctuations of the global climate.

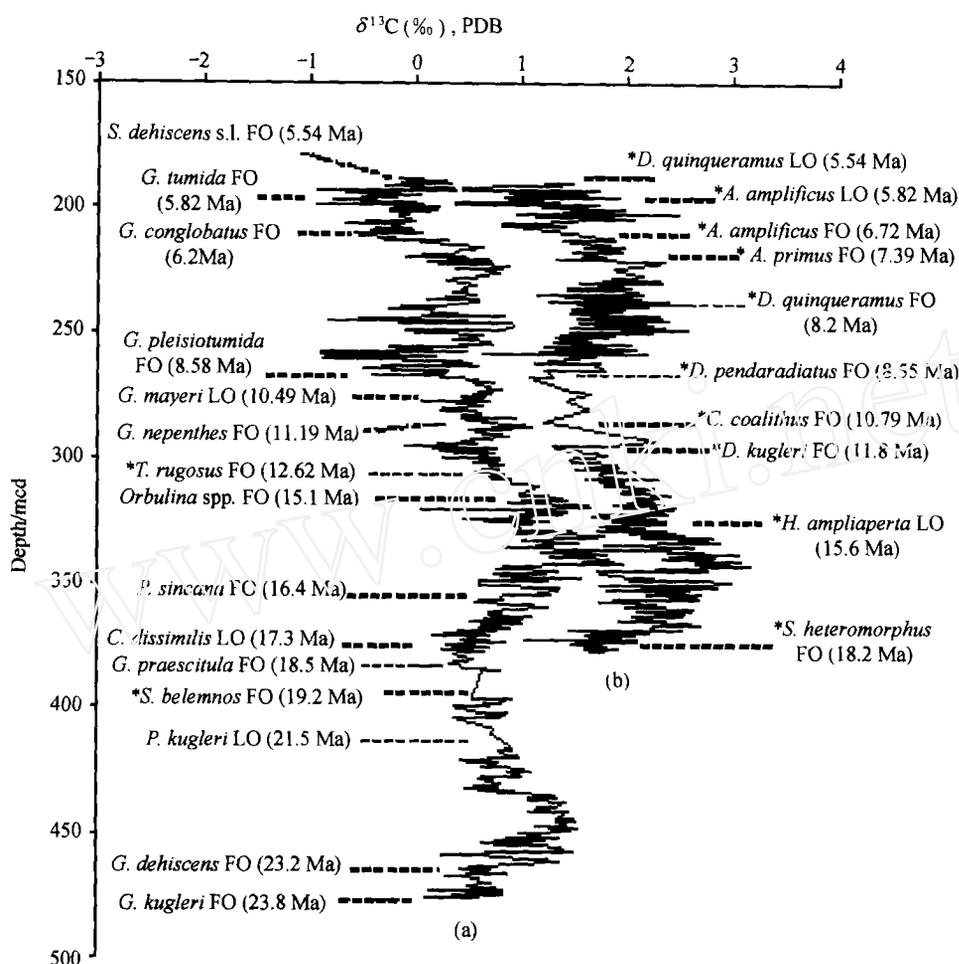


Fig.1. Miocene  $\delta^{13}\text{C}$  data of benthic foraminifers (*Cibicoides wuellerstorfi*, *C. kullenbergi*) and planktonic foraminifer (*Globigerinoides sacculifer*) plotted against composite depth (mcd), and foraminiferal and calcareous nannofossil (with asterisk) biostratigraphic events<sup>[7]</sup> at Hole 1148A, northern South China Sea. Nine planktonic foraminiferal time levels are employed for the age calculation of isotopic samples in the present study.

### 3 Carbon excursion events

#### 3.1 Early Early Miocene positive carbon isotopic excursion

This event is seen in benthic  $\delta^{13}\text{C}$  data (fig.2(b)). The  $\delta^{13}\text{C}$  values rapidly increase from 0.3‰ to 1.5‰ at 23.1 Ma, then remain as high as 1.3‰–1.5‰ during the period of 23.1–22.3 Ma, and then rapidly decline to 0.5‰ at 22.2 Ma. This event has been discovered in the other parts of the world oceans<sup>[9–14]</sup>. The recent studies indicate that this event happened across the boundary of Oligocene and Miocene during the period between 24.4 and 22.6 Ma<sup>[12–14]</sup>. Hodell and Woodruff (1994) called it “a carbon isotopic maximum at the Oligocene/Miocene boundary”<sup>[12]</sup>. A high-resolution isotopic study on the  $\delta^{13}\text{C}$  event at Site 929, western equatorial Atlantic, has revealed that  $\delta^{13}\text{C}$  covaries with  $\delta^{18}\text{O}$ . The coincidence of  $\delta^{13}\text{C}$  maxima with the Mi1  $\delta^{18}\text{O}$

maximum event and a prominent 400 ka cycle indicate a link between the increase in  $\delta^{13}\text{C}$  of the sea water and the global climatic cooling<sup>[13,14]</sup>. It has been suggested that the oceanic productivity and organic burial rate increased during this event with the expansion of East Antarctic ice sheet (EAIS) and the enhancement of the oceanic and atmospheric circulation, and thus resulted in the increase in  $\delta^{13}\text{C}$  of seawater<sup>[13-15]</sup>. The  $\delta^{13}\text{C}$  data from Hole 1148A has further confirmed that the  $\delta^{13}\text{C}$  excursion is a global event. Compared with previous results, however, the event (23.1–22.2 Ma) in the South China Sea obviously happened later, and the most remarkable difference is that  $\delta^{18}\text{O}$  becomes lighter as  $\delta^{13}\text{C}$  increases. The reason is not clear and further study is needed.

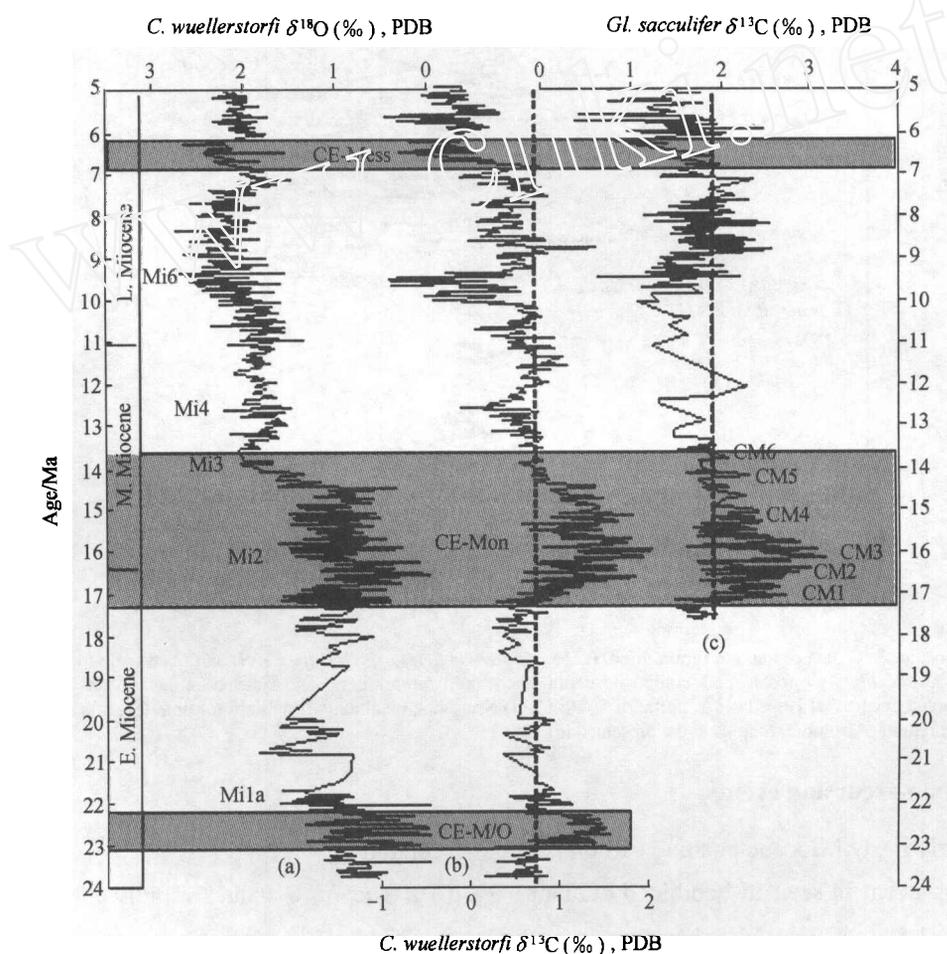


Fig. 2. Age distribution of Miocene  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  records at Hole 1148A, northern South China Sea. (a),  $\delta^{18}\text{O}$  curve of benthic *Cibicidoides wuellerstorfi*, Mi1a, 2, 3, 4 and 6 represent  $\delta^{18}\text{O}$  maxima<sup>[8]</sup>; (b) and (c) are the benthic and planktonic  $\delta^{13}\text{C}$  curves respectively, two dashed lines indicate their average values. CE-O/M = Carbon isotopic excursion crossing the boundary between Oligocene and Miocene, CE-Mon = Monterey carbon isotopic excursion, CE-Mess = Messinian carbon isotopic excursion, and CM1 to CM6 represent 6 carbon maxima<sup>[17]</sup>.

### 3.2 Early-middle Miocene positive carbon isotopic excursion

Both the benthic and planktonic  $\delta^{13}\text{C}$  data at Hole 1148A exhibit a marked increase during

the latest early Miocene and early middle Miocene (fig. 2(b) and (c)), which began at 17.3 Ma and reached peak values at 16.2—16.0 Ma with an increase of 1.6‰, and then gradually decreased to the lowest at 13.6 Ma.

This  $\delta^{13}\text{C}$  excursion occurring in the late-middle Miocene is one of the most important paleoceanographic events in the late Cenozoic. Vincent and Berger (1985) documented a remarked enrichment of foraminifer  $\delta^{13}\text{C}$  at Site 216, tropical Indian Ocean, during the period of 17.5 to 13.5 Ma, and called this heavy  $\delta^{13}\text{C}$  excursion the “Monterey carbon excursion” as it was synchronous with the deposition of diatom-rich sediments around the margin of the North Pacific in the typical Monterey Formation and its equivalent<sup>[16]</sup>. The Monterey carbon excursion has been extensively recorded from the deep-sea sites of the world oceans<sup>[11–13,17,18]</sup>. The definite record of  $\delta^{13}\text{C}$  excursion at Hole 1148A provides further evidence for its world-wide significance.

The timing of the onset, maximum peak and termination of this event had been designated differently in the previous work. However, it is considered by most authors that the Monterey carbon excursion lasted for ca 4 Ma from ca.17.5 to 13.5 Ma and reached the maximum peak at ca.16 Ma<sup>[12,17–19]</sup>. Based on data of more than 10 sites from the world oceans, Woodruff and Savin (1991) designated 7  $\delta^{13}\text{C}$  maxima of the early-middle Miocene as events CM1 to CM7, of which 6 events (CM1—6) occurred in the Monterey carbon excursion<sup>[17]</sup>. The Monterey carbon excursion and its secondary  $\delta^{13}\text{C}$  maxima are of importance in the correlation between the early and middle Miocene stratigraphy. As shown in fig.2, at Hole 1148A of the South China Sea,  $\delta^{13}\text{C}$  reaches its maximum values at 16.2—16.0 Ma and the  $\delta^{13}\text{C}$  maxima centered at 17.0, 16.4, 16.2, 15.0, 14.2 and 13.8 Ma are probably corresponding to CM1 to CM6, respectively. CM5 and CM6 are less developed, particularly in the benthic  $\delta^{13}\text{C}$ .

Generally,  $\delta^{13}\text{C}$  varies with  $\delta^{18}\text{O}$  in a reverse way, but their secondary fluctuations often covary<sup>[17]</sup>. This relationship is confirmed by the data of Hole 1148A. As shown in fig.2, the  $\delta^{18}\text{O}$  depletion of 17.2—14.5 Ma exhibits a minimum at 16.5—16.2 Ma (on the contrary, maximum  $\delta^{13}\text{C}$  at 16.2—16.0 Ma), indicating the Miocene optimum climate and high sea-level, then  $\delta^{18}\text{O}$  shows a rapid enrichment from 14.5 to 13.6 Ma, which is associated with a major EAIS growth and the global cooling. However, the most secondary fluctuations of both  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  vary in the same direction, which is most distinct for the maximum  $\delta^{13}\text{C}$  at 16.2—16.0 Ma corresponding to the maximum  $\delta^{18}\text{O}$  (M11 event) at the same age. The relationship between  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  indicates a global warm and higher sea-level condition during the most time of the Monterey carbon excursion (ca.3 Ma) and a rapid cooling in the last 1 Ma (14.5—13.6 Ma) which occurred in ca.3 Ma after the onset of  $\delta^{13}\text{C}$  enrichment at 17.3 Ma.

Based on the synchronicity of the positive excursion of the seawater  $\delta^{13}\text{C}$  and the diatom-rich deposits around the North Pacific represented by the Monterey Formation, many studies have attributed the former to the latter. However, several hypotheses have been proposed for the large

accumulation of organic material during the Monterey event. Vincent and Berger (1985) suggested a feedback loop to explain the Monterey event, in that the growth of Antarctic ice sheet associated with the increase of meridional temperature contrast, atmospheric circulation and upwelling resulted in the deposition of the  $\delta^{12}\text{C}$ -rich siliceous and organic sediments, leading to the distinct  $\delta^{13}\text{C}$  enrichment in seawater<sup>[16]</sup>. Barron and Baldauf (1990) concluded that the subsidence of the Greenland-Scotland Ridge coupled with rising sea level at 17–15 Ma caused the formation of the North Atlantic Deep Water (NADW), which flowed from the Norwegian Sea, across the ridge, into the North Atlantic. The formation of NADW changed the pattern of the thermal and saline circulation of the ocean, resulting in the basin-basin fractionation of deep waters between the Atlantic and Pacific and the large deposition of biosiliceous and organic sediments in the Pacific<sup>[20]</sup>. Based on a rapid rising in  $^{87}\text{Sr}/^{86}\text{Sr}$  rate during the period of 20–16 Ma, Raymo (1994) inferred that the uplift of the Himalayan-Tibetan Plateau may have enhanced weathering and resulted in a pronounced increase in riverine nutrient influx into the sea, which led to the increasing in marine productivity and enhancement of the burial and preservation of organic materials<sup>[21]</sup>. It is also inferred that the Monterey carbon excursion was caused by the global marine transgression<sup>[22,23]</sup>. During the transgression organic carbon accumulated on the continental shelves, resulting in enriched  $\delta^{13}\text{C}$  in the ocean water. The high temperature and high sea level during the transgression may also cause the expansion of the oxygen-minimum intermediate water which acted as the trap of organic carbon, leading to the increase in  $\delta^{13}\text{C}$  of the ocean water. In summary, all inferences stated above seem to take a same view that the increased productivity and organic burial in the Monterey event have caused the  $\delta^{13}\text{C}$  enrichment in seawater and the decrease in  $\text{CO}_2$  content in atmosphere, resulting in the global climatic cooling. As indicated by isotopic data of Hole 1148 and other sites, however, a Neogene climate optimum state prevailed during the most Monterey events. A rapid cooling just happened in the late period of the event (14.5–13.6 Ma), i.e. the global cooling is 3 Ma later than the onset of  $\delta^{13}\text{C}$  increase or the Monterey organic carbon deposition<sup>[12,16]</sup>. Hodell and Woodruff (1994) suggested that the optimum climate and high weathering were caused by an increased volcanic  $\text{CO}_2$  into the atmosphere from the eruption of the Columbia River Flood Basalts between 17 and 15 Ma. The decrease in the eruptions after 15 Ma and the removal of the atmospheric  $\text{CO}_2$  by continual organic carbon burial in the Monterey deposition led to the global cooling at 14.5 Ma<sup>[12]</sup>.

It seems that the Monterey event has been satisfactorily interpreted by the hypotheses proposed. The benthic and planktonic isotopic data of Hole 1148 further confirm the global significance of the Monterey event and provide the best data for understanding this excursion in the West Pacific.

### 3.3 Early late Miocene negative carbon isotopic excursions

This excursion only exhibits in the benthic  $\delta^{13}\text{C}$  between 10.2 and 9.4 Ma with the minimum

value of  $-0.8\text{‰}$  at 9.7 Ma that is  $1.4\text{‰}$  lighter than the Miocene average value. As shown in fig.2, this  $\delta^{13}\text{C}$  excursion is corresponding to the  $\delta^{18}\text{O}$  maximum event Mi6. This event has been reported in a few literatures. Pagani et al. (1999) provided foraminiferal isotopic profiles of Sites 488 (Southwest Pacific) and 730 (northern Indian Ocean)<sup>[24]</sup>, from which a distinct depletion in benthic  $\delta^{13}\text{C}$  occurring at circa 9.5 Ma can be observed. The negative excursion between 10.2 and 9.4 Ma at Hole 1148 may correspond to it. However, the  $\delta^{13}\text{C}$  depletion ( $<0.8\text{‰}$ ) in Sites 588 and 730 is evidently less than that in the South China Sea.

The planktonic  $\delta^{13}\text{C}$  record does not show a clear covariation with the benthic, which could suggest that the  $\delta^{13}\text{C}$  excursion particularly marked in the benthic record is not due to global changes in carbon reservoirs but also related to other processes.

#### 3.4 Latest Miocene negative carbon isotopic excursion

This event is distinctly indicated by both benthic and planktonic  $\delta^{13}\text{C}$  depletions between 6.9 and 6.2 Ma. The benthic  $\delta^{13}\text{C}$  decreases from  $0.8\text{‰}$  to the minimum  $-0.7\text{‰}$  at 6.6 Ma with a total  $1.5\text{‰}$  depletion. The planktonic  $\delta^{13}\text{C}$  decreases from  $1.9\text{‰}$  at 6.6 Ma (about 0.3 Ma later than the benthic  $\delta^{13}\text{C}$ ) to  $0.8\text{‰}$  at 6.2 Ma with a  $1.1\text{‰}$  drop. The latest Miocene carbon excursion has been extensively reported since 1979 and is presented as a global event<sup>[19,22,25-28]</sup>. It has been called "Messinian carbon shift"<sup>[22]</sup> or "Chron 6 carbon shift"<sup>[28]</sup> with regard to its occurrence in the early Messinian or magnetostratigraphic Chron 6.

Most previous work suggested that this event was assigned to happen between 6.6 and 5.9 Ma<sup>[27]</sup>. However, the earlier age (between 7.1 and 6.8 Ma) of this event has been reported from  $\delta^{13}\text{C}$  data of Morocco<sup>[28]</sup>. Recently, Krijgsman et al. (1999) have applied the stratigraphic technique of astrochronology and dated the Messinian salinity crisis between 5.96 and 5.33 Ma in the Mediterranean<sup>[29]</sup>. At Hole 1148, its occurrence between 6.9 and 6.2 Ma is also different from the previous results.

The negative  $\delta^{13}\text{C}$  excursion is coincident with the  $\delta^{18}\text{O}$  increase in accordance with most of the previous data, and, therefore, many authors have proposed that this carbon event corresponded to the expansion of Antarctic ice sheet, the drop of the sea level, the enhancement of circulations and the increased erosion of organic carbon on the continent<sup>[22,28]</sup>. This negative relationship has been also confirmed in the South China Sea. As shown in fig.2, benthic  $\delta^{18}\text{O}$  at Hole 1148 exhibits a distinct increase during the  $\delta^{13}\text{C}$  excursion (6.9—6.2 Ma), showing the contemporaneous decrease in the benthic and planktonic  $\delta^{13}\text{C}$ . However, this relationship has not been found in all sites, and  $\delta^{18}\text{O}$  in some cores does not show a clear increase in the latest Miocene  $\delta^{13}\text{C}$  excursion<sup>[27]</sup>. In Site 704 of the South Ocean, the  $\delta^{18}\text{O}$  exhibits an increase in the latest Miocene  $\delta^{13}\text{C}$  negative excursion<sup>[26]</sup>. The contradictory phenomenon indicates that further study is needed for this carbon excursion event.

Apart from the above 4 major excursions in  $\delta^{13}\text{C}$ , there are several smaller ones, such as the

positive excursions centered at 11.6, 8.8 and 7.6 Ma, and the negative excursions at 12.6, 10.8 and 5.8 Ma, respectively. The positive excursion at 11.6 Ma is clearly seen in the benthic  $\delta^{13}\text{C}$ , but indefinable in the planktonic  $\delta^{13}\text{C}$  mainly due to the low sample resolution. This event is inferred to CM7 maximum of Woodruff and Savin (1991), an increase in  $\delta^{13}\text{C}$  after the Monterey event<sup>[17]</sup>, which has been reported from the Atlantic<sup>[17,29,30]</sup>, the South Ocean<sup>[12,17]</sup>, the Indian Ocean<sup>[17]</sup> and South-west Pacific<sup>[12,23,24]</sup>. The rest of excursions are lack of data for comparison and need to be confirmed.

#### 4 Conclusions

Hole 1148A has provided the highest resolution and most continuous  $\delta^{13}\text{C}$  data for the whole Miocene, showing general decreasing trend through fluctuations since 24 Ma. Most  $\delta^{13}\text{C}$  values of the early-middle Miocene are heavier than the Miocene average value with two major positive excursions occurring in the intervals of 23.1--22.2 and 17.3--13.6 Ma, respectively. The later excursion (17.3--13.6 Ma) shows the maximum  $\delta^{13}\text{C}$  values in the Neogene, which reaches 2.0‰ at 16.2--16.0 Ma. Most  $\delta^{13}\text{C}$  values of the late Miocene are lighter than the Miocene average value and marked by two negative excursions occurring in intervals of 10.2--9.4 and 6.9--6.2 Ma, respectively. The latest Miocene carbon excursion (6.9--6.2 Ma) is more distinct. These carbon excursions have been proved globally, and applied as an important mark to the stratigraphic correlation. It acts as a significant witness to the changes of global carbon reservoir and its corresponding climate.

The Miocene benthic and planktonic  $\delta^{13}\text{C}$  values show a general covariation, which, in general, parallel to the step-like benthic  $\delta^{18}\text{O}$  curve, reflecting the correlation between continuous decrease of the average content of  $\delta^{13}\text{C}$  in seawater  $\text{CO}_2$  and the global climatic cooling. However, because the foraminiferal  $\delta^{13}\text{C}$  is a complicated process for the carbon migration either within the ocean or between the ocean and the atmosphere or land, its mechanism is not clear so far, and it is still a major topic for further study. The  $\delta^{13}\text{C}$  record of Hole 1148A has provided the most continuous and the highest-resolution  $\delta^{13}\text{C}$  data in the Pacific for the studies of carbon isotopic stratigraphy and global climatic changes.

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