A critical review of marine sedimentary $\delta^{13}C_{org}$-pCO$_2$ estimates: New palaeorecords from the South China Sea and a revisit of other low-latitude $\delta^{13}C_{org}$-pCO$_2$ records

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Abstract. In an attempt to understand better the local biogeochemistry of the South China Sea (SCS) and to unravel the contribution of this marginal low-latitude basin to changes in atmospheric CO$_2$ concentrations, we analyzed the carbon isotopic composition of organic matter ($\delta^{13}C_{org}$) in four sediment cores from throughout the SCS covering the last 220 kyr. Higher values (around -19.5 to -20.5‰) mark glacial stages, while lower values (around -21 to -22.5‰) are characteristic of interglacials. Following well established procedures, the $\delta^{13}C_{org}$ records are converted to local pCO$_2$ estimates. On the basis of these and other low-latitude $\delta^{13}C_{org}$-pCO$_2$ estimates from the literature, we present a critical evaluation of the use of $\delta^{13}C$ of bulk sedimentary organic matter to hindcast past changes in local CO$_2$(aq). Three crucial pitfalls are identified. (1) Given the present inability to quantify precisely the time-varying amount of terrigenous C$_{org}$ input to marine sediments, absolute values of pCO$_2$ estimates based on bulk sedimentary C$_{org}$ are questionable. (2) None of the low-latitude sedimentary $\delta^{13}C_{org}$-pCO$_2$ records shows the expected correlation between temporal changes in upwelling intensity and CO$_2$ estimates, most likely due to the antagonistic influences of CO$_2$(aq) and phytoplankton growth rate on $\delta^{13}C_{org}$. (3) A detailed comparison of marine $\delta^{13}C_{org}$-pCO$_2$ records with the Vostok CO$_2$ record reveals significant differences in phasing, specifically at the end of the last deglaciation and during the oxygen isotope stage 5/4 transition. However, in areas where equilibrium between oceanic and atmospheric CO$_2$ occurs, for example the SCS and the Mediterranean, the timing of changes in $\delta^{13}C_{org}$ should agree with the CO$_2$ record from ice cores if $\delta^{13}C_{org}$ is a reliable proxy for changes in CO$_2$(aq). Taken together, the compilation of records presented here cautions the use of $\delta^{13}C_{org}$ as an unambiguous tracer of dissolved molecular CO$_2$ in the surface ocean and calls for a re-evaluation of the role of the low-latitude ocean on temporal changes in atmospheric CO$_2$.

1. Introduction

Simple mass balance considerations of the global carbon cycle point to the ocean as major player in glacial-interglacial changes in atmospheric CO$_2$ concentrations as recorded in ice cores. In principle, two competing schools of thought that try to characterize and quantify the ocean's impact on atmospheric CO$_2$ have evolved during the last two decades. One invokes changes in marine production to account for the observed variability [e.g., *Broecker*, 1982; *McElroy*, 1983; *Sarnthein et al.*, 1988; *Berger et al.*, 1989; *Martin*, 1990; *Broecker and Henderson*, 1998], while the other attributes the ocean's role mainly to physical processes, such as changes in stratification and/or redistribution of CO$_2$ within the different oceanic reservoirs [e.g., *Boyle*, 1988; *Keir*, 1993; *François et*

al., 1997; *Toggweiler*, 1999; *Stephens and Keeling*, 2000]. Changes in upwelling intensity with time have been proposed as a mediatory mechanism [*Pedersen et al.*, 1991; *Jasper and Hayes*, 1994]. Here, increases in marine production are directly coupled to physical processes that impact on the exchange of CO$_2$ between the atmospheric and the oceanic reservoirs, the net effect, however, being oceanic degassing of CO$_2$ to the atmosphere. In order to differentiate between these different scenarios and to gain a better understanding of the ocean's contribution to changes in atmospheric CO$_2$, a clearer delimitation and quantification of oceanic palaeosources and sinks of CO$_2$ is desirable.

The carbon isotopic composition of marine organic matter ($\delta^{13}C_{org}$) is one method that has shown promise in monitoring the ocean's contribution to changes in atmospheric CO$_2$. Although the exact biological processes determining $\delta^{13}C_{org}$ are still subject to considerable debate, a variety of laboratory and field studies have shown that the $\delta^{13}C$ of the bulk organic fraction of marine plankton generally decreases as the

dissolved molecular CO_2 concentration $[CO_2(aq)]$ increases [Degens et al., 1968; Degens, 1969; Popp et al., 1989; Rau et al., 1989,1991b, 1992, 1997; Freeman and Hayes, 1992; François et al., 1993; Goericke and Fry, 1994; Rau, 1994; Fischer et al., 1998]. In the inferred absence of a significant contribution of terrigenous organic matter to the sediment, which would obscure the bulk sedimentary isotopic signature, this correlation has been widely used to convert bulk sedimentary $\delta^{13}C_{org}$ records to palaeo-pCO_2 estimates [Pedersen et al., 1991; Rau et al., 1991a; Fontugne and Calvert, 1992; Thunell et al., 1992; Müller et al., 1994; Westerhausen et al., 1994; Bentaleb et al., 1996]. Some of these studies have specifically tried to unravel the antagonistic influences of changes in upwelling intensity and in marine production on the direction of CO_2 exchange between the ocean and the atmosphere, a key parameter in understanding the ocean's contribution to changes in atmospheric CO_2 concentrations.

In this paper, we first present new records of the carbon isotopic composition of organic matter in a suite of sediment cores from the South China Sea (SCS) spanning the last climatic cycle. These $\delta^{13}C_{org}$ records are converted to local pCO_2 estimates, following established procedures, and after considering potential biases by terrigenous input, temperature and other effects on the C isotope fractionation as well as possible diagenetic modification of the signal. In the second part of the paper, we discuss the strengths and limitations of low-latitude marine sedimentary pCO_2 estimates based on $\delta^{13}C_{org}$. The discussion is focused on three key criteria for evaluating $\delta^{13}C_{org}$-pCO_2 estimates, (1) absolute values of the estimates, (2) correlation between pCO_2 estimates and indicators of upwelling strength, and (3) a comparison of the temporal evolution of pCO_2 estimates with the ice core (Vostok) record of atmospheric CO_2.

2. Materials and Methods

The sediment cores of this study (Table 1, Figure 1) were recovered during a R/V Sonne cruise to the South China Sea in 1994 and, according to echo-sounding profiles, contain undisturbed hemipelagic sequences [Sarnthein et al., 1994]. The cores were subsampled every 5-20 cm using 10 cm^3 syringes. The surface samples in this study were taken from box cores recovered during the R/V Sonne cruise in 1994 and another R/V Sonne cruise in 1996/1997, and represent the undisturbed uppermost centimeter of the sediment [Sarnthein

et al., 1994; Stattegger et al., 1997]; the overlying fluff layer was carefully sampled using large-volume syringes. For chemical analyses, the samples were freeze dried and manually homogenized in an agate mortar.

The carbon isotopic composition of organic matter was measured on decalcified (10% HCl followed by oven drying without washing) samples using a Fisons NA1500 elemental analyzer coupled to a VG prism mass spectrometer with an analytical precision of ±0.1‰.

Long chain n-alkane and C_{37}-alkenone determinations were carried out following procedures described by Villanueva et al. [1997a]. Briefly, freeze-dried subsamples were extracted with dichloromethane, hydrolized with 6% KOH in methanol, and derivatized with bis(trimethylsilyl)trifluoro-acetaminde. The n-alkanes and the C_{37}-alkenones were quantified gas chromatographically in relation to the peak areas of an internal standard. The n-alkane distribution in SCS sediments is rather uniform, with homologues ranging from tricosane (C_{23}) to pentatriacontane (C_{35}) and maxima at untriacontane (C_{31}). A strong odd-to-even carbon number preference was observed, indicating a higher-plant origin of these compounds [Eglinton and Hamilton, 1967]. No samples with lower odd-even carbon number preference were found, excluding possible microbial reworking [e.g., Grimalt et al. 1985]. This uniform n-alkane distribution justifies the use of one single compound, n-nonacosane (C_{29}), as a proxy for terrigenous input to the sediments.

The chronology of cores 17940-2, 17954-2, and 17961-2, based on oxygen isotope stratigraphy as well as AMS-^{14}C dating, is adopted from Wang et al. [1999]; the age model of core 17924-3 is based on oxygen isotope ratios of G. ruber (M. Sarnthein, written communication, 1999) and will be published elsewhere. In addition, the occurrence of the Toba Ash in core 17961-2 [M. Kienast, unpublished data, 1999; Bühring et al., 2000] is used as an age fix point (74 ka at 781-783.5 cm core depth).

3. Results and Interpretation

The $\delta^{13}C_{org}$ in the four sediment cores located in the southern (core 17961-2), western (core 17954-2) and northern (cores 17924-3 and 17940-2) reaches of the SCS varies between -22.5 and -19.5 ‰ (Figures 2-5). Higher values mark glacial stages, while lower values are characteristic of interglacials. In the following paragraphs, evidence is adduced that this variability is not due to variable terrigenous

Table 1. Core Locations, Water Depth, Core Recovery, and Interglacial/Glacial Sedimentation Rates of the Four Sediment Cores in the South China Sea

Core	Latitude, °N	Longitude, °E	Water Depth, m	Recovery, m	Sedimentation Rates, cm/kyr Interglacial	Glacial
17924-3	19°24.7	118°50.8	3438	19.89	~10[a]	~20[a]
17940-2	20°07.0	117°23.0	1728	13.30	45-85[b]	19[b]
17954-2	14°45.5	111°31.6	1517	11.52	6-7[b]	5-9[b]
17961-2	8°30.4	112°19.9	1968	10.30	3-8[b]	10[b]

[a] M. Sarnthein, unpublished data, 1999
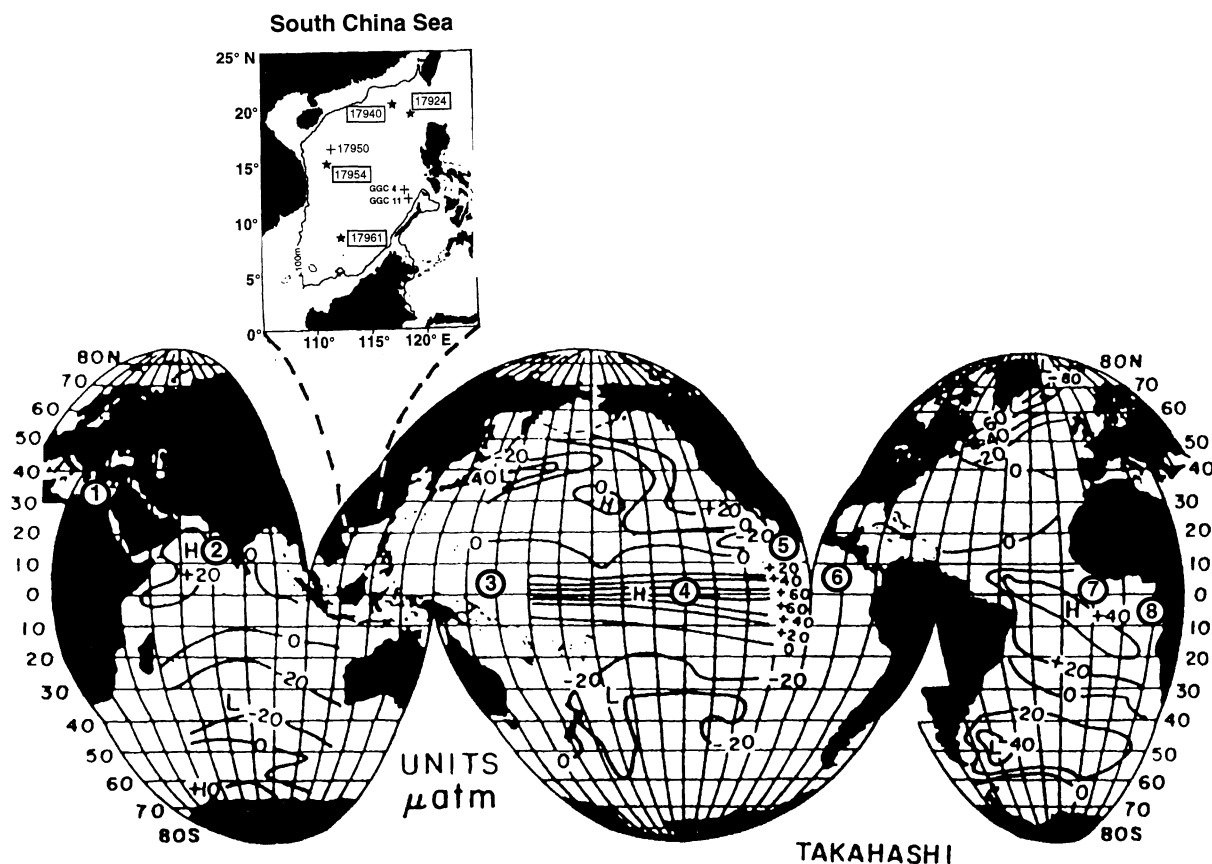[b] From Wang et al. [1999]

South China Sea

Figure 1. Map of global sea-air pCO$_2$ difference (modified from *Broecker and Peng* [1998]) and location of cores discussed in this study; from west to east: (1) eastern Mediterranean Sea [*Fontugne and Calvert*, 1992], (2) Indian Margin [*Ganeshram et al.*, 2000], (3) western equatorial Pacific [*Raymo et al.*, 1996], (4) central equatorial Pacific [*Jasper and Hayes*, 1994], (5) Mexican Margin [*Ganeshram et al.*, 1999], (6) Panama Basin [*Pedersen et al.*, 1991], (7) equatorial Atlantic [*Westerhausen et al.*, 1994], (8) Angola Basin [*Müller et al.*, 1994; *Andersen et al.*, 1999]. Insert map shows location of cores from the SCS presented in this study, including the two stations of *Thunell et al.* [1992], GGC 4 and 11, respectively, and station 17950-2 of *Lin et al.* [1999]. Note that the 100 m isobath shown in the SCS map approximates the location of the coast line at the last glacial maximum.

input to the sediments, direct effects of temperature on planktonic isotope fractionation, changes in marine production, and other biological factors nor to diagenetic modification of the signal.

3.1. Marine Versus Terrigenous Organic Matter

As expected from the palaeogeographic setting of the SCS, concentrations of the terrigenous marker *n*-nonacosane are significantly higher during glacial stages (Figures 2-4) when vast shelf areas in the northern and especially in the southern SCS were emergent (see Figure 1). Given the fact that C$_3$ plant organic matter is characterized by light $\delta^{13}C_{org}$ values (-25 to -27‰ [*Deines*, 1980]), the bulk sedimentary $\delta^{13}C_{org}$ signal of cores 17940-2, 17954-2, and 17961-2 is inversely correlated with this molecular indicator of terrigenous input, ruling out an overwhelming contribution of terrigenous C$_3$ organic matter to the bulk $\delta^{13}C_{org}$ signal at these three sites. Furthermore, since the absolute values of $\delta^{13}C_{org}$ in all four

sediment cores (Figures 2-5) are quite similar, irrespective of their proximity to continental sources and since there is no north-south gradient that could be attributed to changing vegetation patterns, C$_4$ plant organic matter ($\delta^{13}C_{org}$: -12 to -15 ‰; ibid.) most likely constitutes a relatively minor fraction of the organic matter in the cores. According to *Hinrichs et al.* [1999], up to 30% of the bulk sedimentary C$_{org}$ would need to be exclusively of C$_4$ plant origin in order to explain the observed $\delta^{13}C_{org}$ variability. This amount is much larger than any possible C$_4$ contribution as indicated by pollen records from SCS sediments [*Sun and Li*, 1999; *Sun et al.*, 1999]. Moreover, the interglacial concentrations of *n*-alkanes in the SCS cores 17961-2 and 17954-2 (~ 125 and 175 ng/g) are comparable to Holocene values from the central N Atlantic (30-150 ng/g [*Villanueva et al.*, 1997b]) and the central low-latitude Pacific (30-300 ng/g [*Ohkouchi et al.*, 1997]), suggesting comparatively low terrigenous input at least during interglacial periods. In addition, the higher concentrations of *n*-alkanes/nonacosanes in core 17940-2

(~700 ng/g) as compared with cores 17954-2 and 17961-2 are not associated, as would be expected, with lower $\delta^{13}C_{org}$ values in this core. Finally, taking water depth as a first order approximation of distance from the coast, the absence of any correlation of the $\delta^{13}C_{org}$ surface sediment values in the SCS with water depth (Figure 6) again suggests that there is no overwhelming importance of terrigenous organic matter on the distribution of modern bulk sedimentary $\delta^{13}C_{org}$. In the absence of a reliable quantitative proxy for terrestrial input to marine sediments, for example, an n–alkane/C_{org} ratio for the terrigenous end-member [e.g., Prahl et al., 1994], however, it is not possible to conclusively quantify the subdued, time-varying influence of terrestrial organic matter on the $\delta^{13}C_{org}$

signal, most importantly the increased input of terrigenous organic matter during glacials (see implications and further discussion in section 5.1).

Keil et al. [1994] and Goni et al. [1998] reported a grain size dependence of the carbon isotopic composition of surficial sediment particles on the Washington Margin and in the Gulf of Mexico. Both studies showed heavier $\delta^{13}C_{org}$ values associated with finer-grained particles. In the two SCS core records where grain size data are available, however, lower $\delta^{13}C_{org}$ values are associated with higher % clay (core 17940-2; Figure 2), or there is no correlation between $\delta^{13}C_{org}$ and % clay (core 17961-2; Figure 4), excluding the possibility that the carbon isotopic signal in the SCS is governed by effects
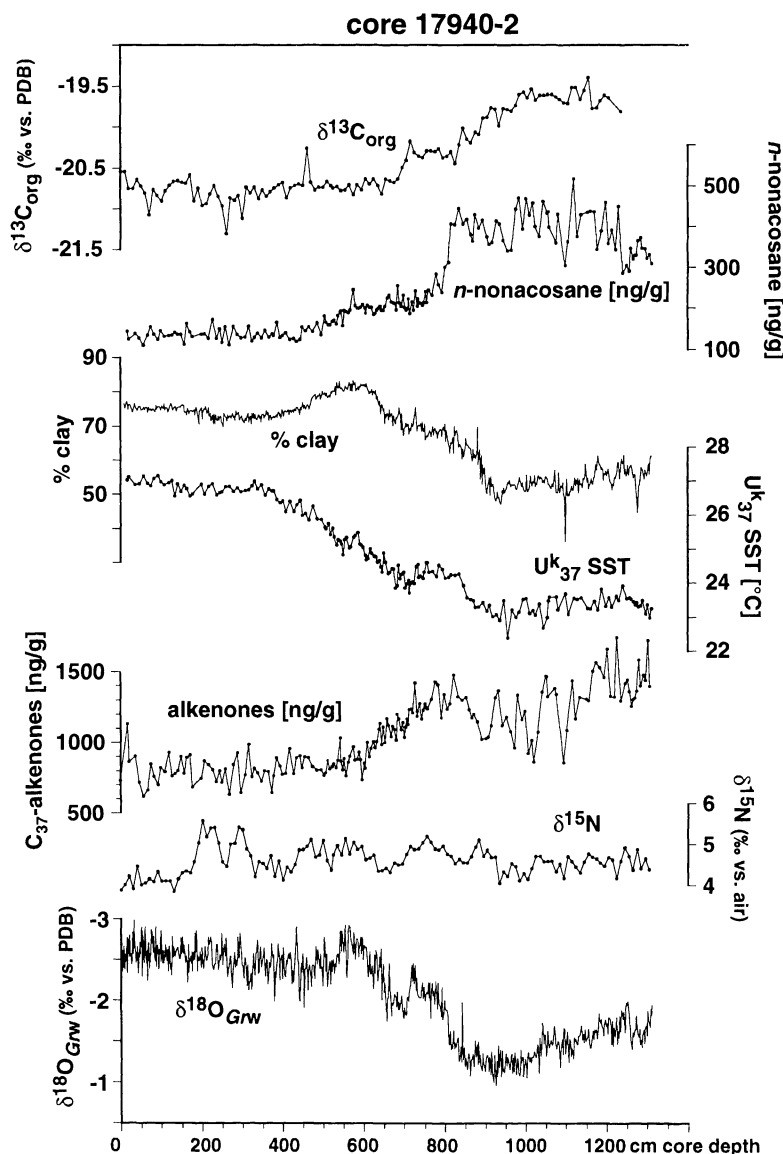

Figure 2. Bulk sedimentary $\delta^{13}C_{org}$, concentration of n-nonacosane in ng/g, percent clay of the siliciclastic fraction [from Wang et al., 1999], U^k_{37} SST estimates [from Pelejero et al., 1999], concentration of C_{37}-alkenones in ng/g, bulk sedimentary $\delta^{15}N$ [from Kienast, 2000], and $\delta^{18}O_{G.ruber white}$ [from Wang et al., 1999] of core 17940-2 in the northern SCS versus core depth.

core 17954-2
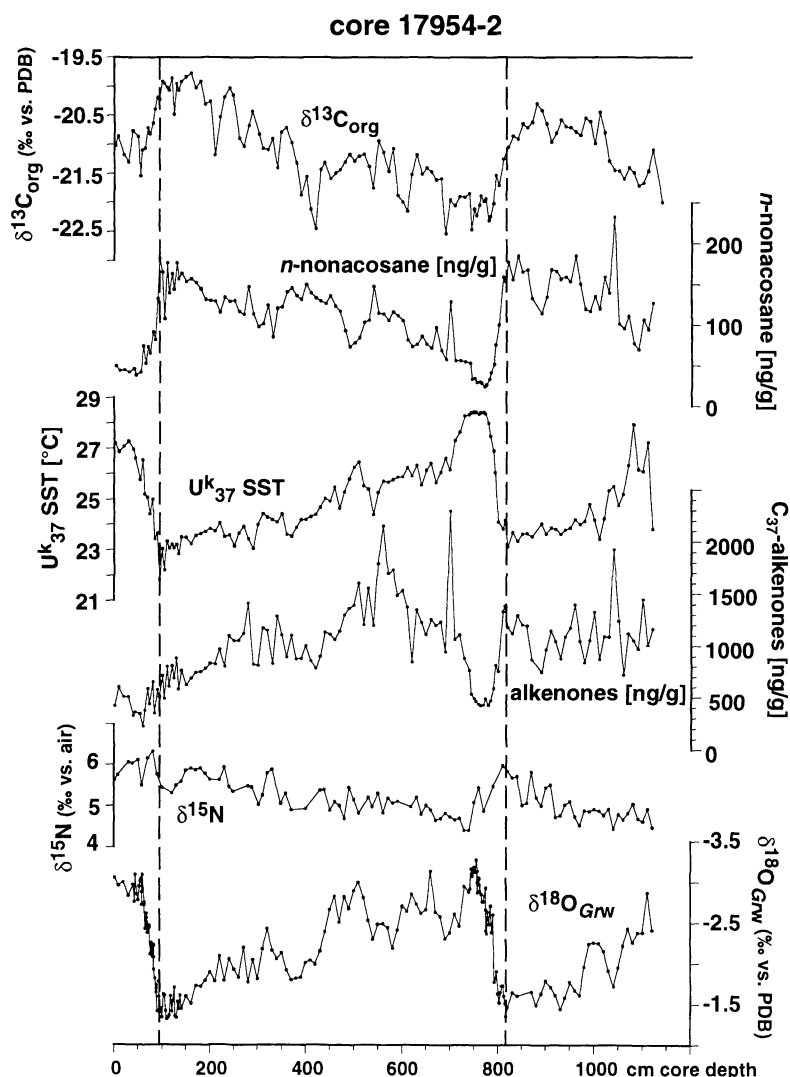

Figure 3. Bulk sedimentary $\delta^{13}C_{org}$, concentration of n-nonacosane in ng/g, U^k_{37} SST estimates [from *Pelejero et al.*, 1999], concentration of C_{37}-alkenones in ng/g, bulk sedimentary $\delta^{15}N$ [from *Kienast*, 2000], and $\delta^{18}O_{G.ruber white}$ [from *Wang et al.*, 1999] of core 17954-2 in the western central SCS versus core depth. Vertical stippled lines denote the onset of terminations I and II, respectively, as defined by the $\delta^{18}O_{G.ruber}$ and SST records.

associated with the hydrodynamic sorting of particles with a different, grain size dependent, carbon isotopic composition.

The distance from continental sources and the similarity in the $\delta^{13}C_{org}$ record of core 17924-3 (Figure 5) suggest that the interpretation of a subordinate influence of terrigenous organic matter on the carbon isotopic composition holds true also for this site, where no biomarker and grain size data are available.

3.2. Temperature Effect

Fontugne and Duplessy [1981] observed a positive correlation between increasing sea surface temperature (SST) and increasing $\delta^{13}C_{org}$ values in marine plankton from the Atlantic and Indian Oceans. This trend is opposite to that observed for the SCS sediment records (Figures 2-4).

Accordingly, $\delta^{13}C_{org}$ in the SCS is not affected by any possible thermal effect on C isotope fractionation as postulated by *Fontugne and Duplessy* [1981]. Moreover, *Fontugne and Duplessy* [1981] state that in fact a correlation (between SST and $\delta^{13}C_{org}$) does not exist for the temperature range 15-31°C, the range of SST observed in the SCS. In light of recent understanding [*Rau et al.*, 1989, 1996, 1997; *Freeman and Hayes*, 1992; *Raven et al.*, 1993; *Goericke and Fry*, 1994] the SST dependency of $\delta^{13}C_{org}$ claimed by these authors is more likely to be indirect, such as the SST-dependent solubility of CO$_2$ in sea water, the temperature dependence of $\delta^{13}C$ of CO$_2$(aq), changes in the diffusivity of CO$_2$(aq) with SST, variable average growth rates as a function of SST, the induction of a carbon concentrating mechanism, or any combination of these factors.

3.3. Marine Production, Upwelling and Biological Factors

In order to evaluate any possible influence of changes in marine production on $\delta^{13}C_{org}$, we use the sedimentary concentration of C_{37}-alkenones as production tracer (Figures 2-4). This selection is justified by the fact that bulk C_{org} in the SCS is affected by time-varying changes in the admixture of terrigenous organic matter (see section 3.1). Given the overriding control of linear sedimentation rates on accumulation rates of sedimentary components [Middelburg et al., 1997], we chose not to calculate alkenone accumulation rates but rather incorporate changes in sedimentation rate in a

more qualitative way into the discussion. Thus, the higher glacial concentrations of alkenones in core 17940-2 (Figure 2) are offset by significantly lower sedimentation rates during this period (see Table 1), suggesting unchanged levels of haptophyte and, by inference, overall marine production on a glacial-interglacial timescale at this site, in agreement with unchanged nutrient levels/utilization (see discussion below in this section). In contrast, the alkenone record of core 17954-2 (Figure 3) shows highest alkenone concentrations paralleled by higher sedimentation rates during the later part of marine isotope stage (MIS) 5, and during MIS 6, suggesting elevated marine production during these periods, decoupled
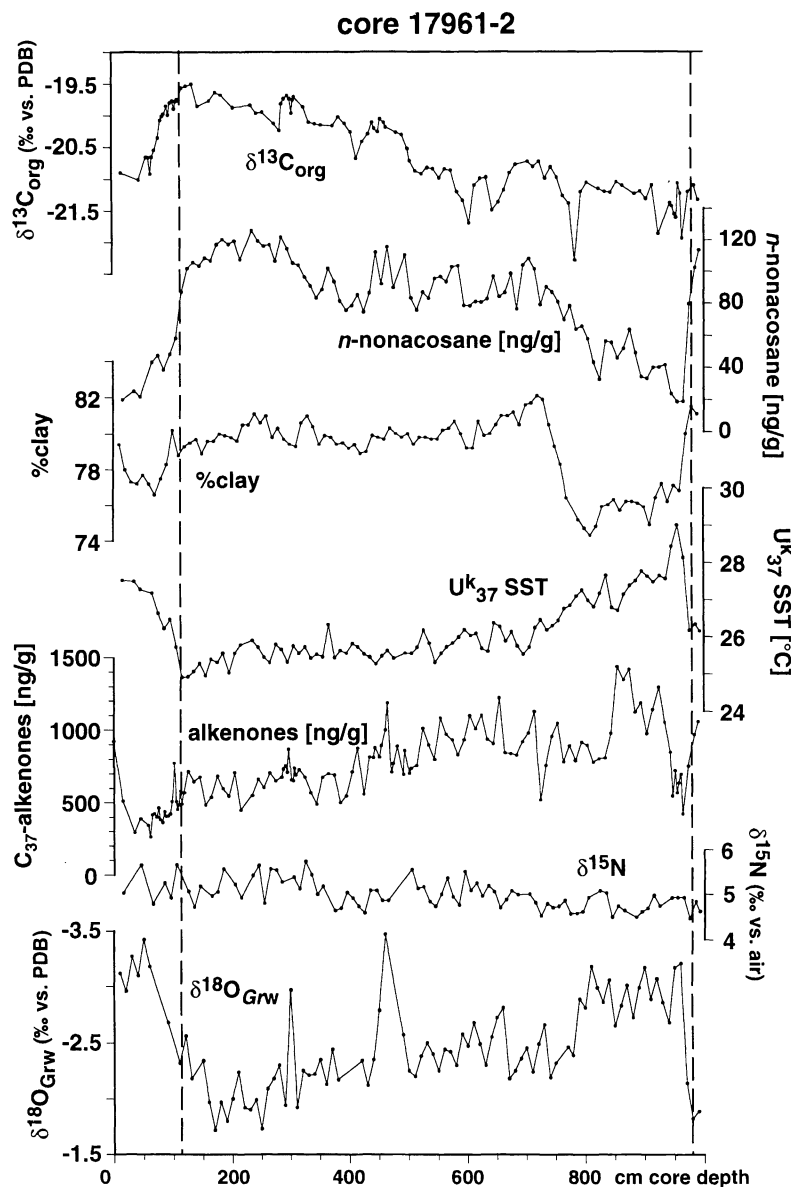

Figure 4. Bulk sedimentary $\delta^{13}C_{org}$, concentration of n-nonacosane in ng/g, percent clay of the siliciclastic fraction [from Wang et al., 1999], U^k_{37} SST estimates [from Pelejero et al., 1999], concentration of C_{37}-alkenones in ng/g, bulk sedimentary $\delta^{15}N$ [from Kienast, 2000], and $\delta^{18}O_{G.ruber white}$ [from Wang et al., 1999] of core 17961-2 in the southern SCS versus core depth. Vertical stippled lines denote the onset of terminations I and II, respectively, as defined by the SST and benthic $\delta^{18}O$ (not shown [see Wang et al., 1999]) records.
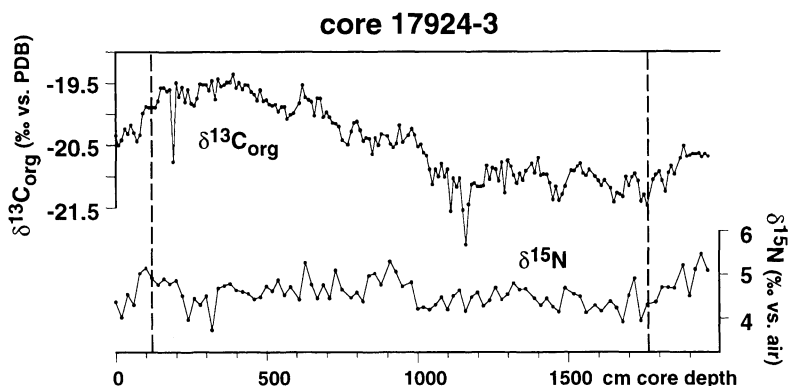
Figure 5. Bulk sedimentary $\delta^{13}C_{org}$ and bulk sedimentary $\delta^{15}N$ [from *Kienast*, 2000] of core 17924-3 in the northeastern SCS versus core depth. Vertical stippled lines denote the onset of terminations I and II, respectively, as defined by the $\delta^{18}O_{G.ruber}$ record (M. Sarnthein, unpublished data, 1999).

from glacial or interglacial boundary conditions. This reconstruction of temporal changes in marine production at site 17954-2 is in agreement with benthic foraminiferal evidence from the same core (Z. Jian et al., manuscript in preparation, 2000). In core 17961-2 (Figure 4), lower alkenone concentrations occur during both glacial terminations, paralleled by decreased sedimentation rates [*Wang et al.*, 1999, Table 1]. Throughout the rest of the core record, alkenone-derived marine production is more or less uniform. Given the similarity of the $\delta^{13}C_{org}$ records throughout the SCS and the absence of an obvious correlation between $\delta^{13}C_{org}$ and variable marine production at any site, $\delta^{13}C_{org}$ in the SCS seems not to be significantly affected by changes in marine production, as inferred from the concentration of alkenones.

Recent surface seawater CO_2 measurements throughout the SCS [*Chen et al.*, 1998] and maps of global air-sea pCO_2 differences [*Takahashi et al.*, 1997] show that the SCS surface waters are in equilibrium with atmospheric CO_2. Similarly, palaeoceanographic studies to date do not show any indication of significant upwelling at any site in the SCS during the last 220 kyr, including the locales presented in this study [see *Wang et al.*, 1999, and references therein]. An exception to this general observation might be an area off Vietnam, marginally including site 17954 in the present study. Here, upwelling is predicted based on a climatology-driven circulation model in response to summer monsoonal winds [*Wiesner et al.*, 1996], and planktonic foraminiferal $\delta^{13}C$ data have been interpreted to reflect changes in upwelling intensity with time at site 17954 [*Wang et al.*, 1999]. However, the postulated upwelling at site 17954 is neither reflected in the nitrogen isotopic record (Figure 3 [*Kienast*, 2000]) nor is the $\delta^{13}C_{org}$ record of core 17954-2 significantly different from the other records presented here. Moreover, if site 17954 was influenced by changes in upwelling induced by variable summer monsoonal winds, one would expect a consistent glacial-interglacial contrast in marine production in accord with glacial-interglacial changes in the intensity of the summer monsoons as reconstructed from various sedimentary proxies [*Wang et al.*, 1999]. The alkenone-based reconstruction of marine production at this site (see above in this section) does not conclusively support this scenario.

Accordingly, current understanding of SCS (palaeo) oceanography suggests that the SCS surface ocean pCO_2 at the sites presented is more or less in thermodynamic equilibrium with the atmosphere both today and during the last 220 kyr.

A constant nitrogen isotopic composition of bulk sedimentary organic matter (4-6‰) at all sites and throughout the records (Figures 2-5) has been interpreted to reflect complete consumption of nitrate during primary production [*Kienast*, 2000], suggesting nitrate-limited production in the SCS throughout the last 220 kyr. Similarly, foraminiferal Cd/Ca ratios of cores 17940-2 and 17950-2 [*Lin et al.*, 1999] (see Figure 1 for location of core 17950-2) indicate no systematic glacial-interglacial variability, which is interpreted to reflect relatively unchanged HPO_4^{2-} concentrations in SCS surface waters on a glacial-interglacial timescale. Although this evidence is not direct, such records of nutrient availability/utilization strongly suggest that $\delta^{13}C_{org}$ in the SCS has not been significantly affected by changes in growth rate, cell volume/geometry, and/or nutritional status as observed by *Laws et al.* [1995], *Bidigare et al.* [1997], *Burkhardt et al.* [1999], *Pancost et al.* [1997], *Popp et al.* [1998], and *Kukert and Riebesell* [1998]. In addition, invariably low opal concentrations (<5%, i.e., close to the precision of the method [*Mortlock and Froelich*, 1989]) throughout the SCS core records (not shown) could be interpreted to indicate a relatively unchanged phytoplankton community structure, suggesting a negligible effect of changes in species composition on the $\delta^{13}C_{org}$ signal [*Pancost et al.*, 1999; *Popp et al.*, 1999] in the SCS. It is noted, however, that the low opal concentrations could also be due to opal solution in the sediments.

3.4. Diagenesis

The preferential degradation of ^{12}C relative to ^{13}C in the water column inferred from the $\delta^{13}C_{org}$ distribution in plankton samples, sediment trap material, and surface sediments in the equatorial Atlantic [*Westerhausen et al.*, 1993] and the Angola Basin [*Müller et al.*, 1994] is not evident from the $\delta^{13}C_{org}$ surface sediment distribution with respect to water depth in the SCS (Figure 6). This implies that

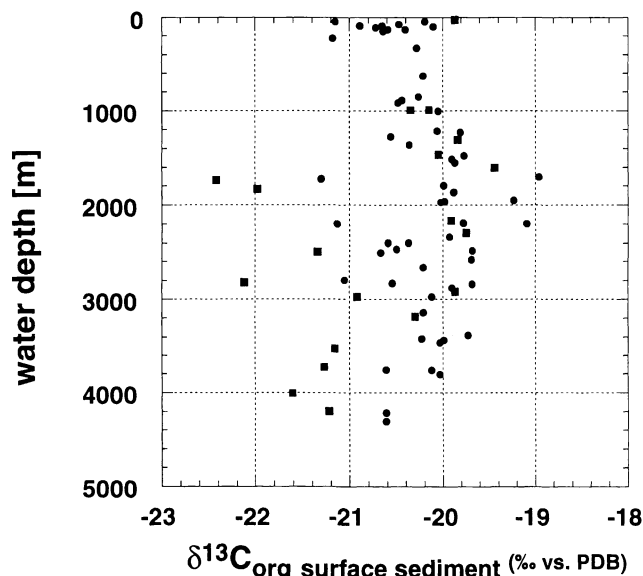**Figure 6.** Bulk sedimentary $\delta^{13}C_{org}$ of surface sediments versus water depth (n=76, filled circles: this study, filled squares: from *Calvert et al.* [1993]) in the SCS. Note the absence of any consistent trend of $\delta^{13}C_{org}$ with increasing water depth.

there is no discernible differential preburial diagenesis of the C isotopes in the SCS water column.

A comparison of the $\delta^{13}C_{org}$ signatures of fluff versus surface sediment samples shows a constant, albeit significant, ^{12}C depletion of around 1‰ at the water-sediment interface (Figure 7). This could be interpreted as a reflection of early diagenetic modification of the $\delta^{13}C_{org}$ signal. However, it is more likely that the offset is due to the anthropogenic CO$_2$ invasion into surface waters. A $\delta^{13}C_{org}$ decrease of several permil owing to anthropogenic CO$_2$ emission has been postulated by *Bentaleb and Fontugne* [1996] and is substantiated by a comparison of sinking organic matter and surface sediments by *Fischer et al.* [1997]. This interpretation is corroborated by a similar [see *Quay et al.*, 1992; *Beveridge and Shackleton*, 1994] decrease of $\delta^{13}C_{G.ruber}$ values in the uppermost samples (n=5; 0-5 cm) of box core 17940-1 compared to the Holocene average (see *Wang et al.* [1999] for $\delta^{13}C_{G.ruber}$ record). In light of these new findings, previous interpretations of pre-burial diagenetic modification of the $\delta^{13}C_{org}$ signal (see discussion above in this section) probably need to be reinterpreted.

The $\delta^{13}C_{org}$ down-core records do not display any consistent secular trend with depth which would be indicative of diagenesis [cf. *Fontugne and Calvert*, 1992]. Moreover, given the different linear sedimentation rates (Table 1), and therefore different C$_{org}$ burial rates at the four coring sites, and the different bulk sediment compositions, the similarity of the $\delta^{13}C_{org}$ records at all sites suggests that the carbon isotopic composition has not been significantly modified by diagenesis.

It is noted that *Thunell et al.* [1992] reported $\delta^{13}C_{org}$ records from two stations in the eastern SCS off Palawan (GGC 4 and GGC 11; see Figure 1 for location) showing glacial values that are consistently heavier by up to 2‰ than in the four records

presented in the present study. Replications of the analyses on the 1992 study material in our laboratory indicate that this deviation is not due to any analytical offset or differences in sample treatment. These records could indicate decreased terrigenous input to the SCS off Palawan during sea level low stand compared to the records presented here.

4. Estimating pCO$_2$ From Sedimentary $\delta^{13}C_{org}$

We have applied the approach of *Rau et al.* [1991a] to convert bulk sedimentary $\delta^{13}C_{org}$ values to pCO$_2$ estimates. This is justified by the narrow range of CO$_2$(aq) values in SCS surface waters [*Chen et al.*, 1998] that obviates the possibility of using a site-specific $\delta^{13}C_{org}$-CO$_2$(aq) relationship and, even though there is no a priori reason to assume a linear relationship [e.g., *François et al.*, 1993], allows intercomparison with other low-latitude $\delta^{13}C_{org}$-pCO$_2$ estimates (see section 5). Using the approach of *Popp et al.* [1989], moreover, yields almost identical estimates, as has been noted earlier by *Müller et al.* [1994]. The $\delta^{13}C_{org}$ records have been corrected for changes in $\delta^{13}C$ of dissolved inorganic carbon (DIC) in seawater by subtracting the difference between the $\delta^{13}C_{G.ruber}$ downcore values and the average latest Holocene $\delta^{13}C_{G.ruber}$ value of 1.5‰ ($\delta^{13}C_{G.ruber}$ data from *Wang et al.* [1999]). The corrected $\delta^{13}C_{org}$ records were converted to CO$_2$(aq) values according to the linear relationship between $\delta^{13}C_{org}$ and CO$_2$(aq) derived by *Rau et al.* [1989]:

$$[CO_2(aq)] = (\delta^{13}C_{org} + 12.6)/-0.8, \qquad (1)$$

and the CO$_2$(aq) values were converted to partial pressure of CO$_2$ (pCO$_2$) using Henry's Law:

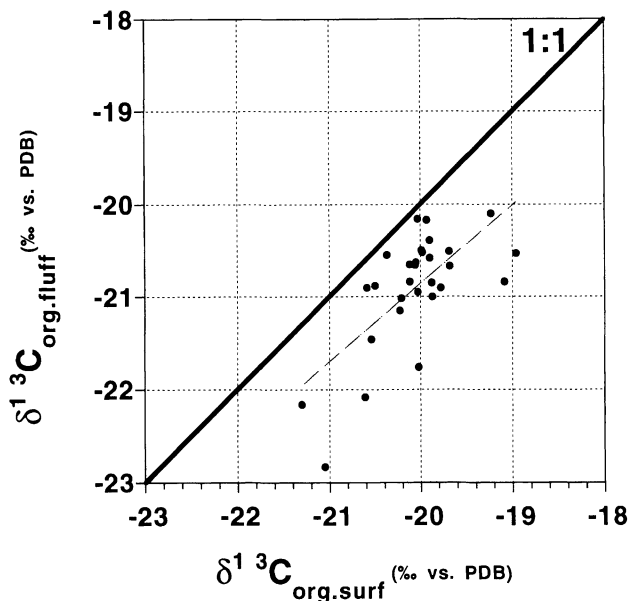**Figure 7.** Bulk sedimentary $\delta^{13}C_{org}$ of fluff samples versus surface sediments. Note that the fluff samples are consistently lighter by 1‰ on average compared to the surface sediment. Thick black line shows 1:1 correlation; the thin line is the actual correlation of the data (n=31), with gradient = 0.85 and r=0.68.

$$pCO_2 = CO_2(aq)/\ \alpha, \qquad (2)$$

where α is a temperature dependent solubility constant. This constant was calculated according to *Weiss* [1974] using the U^k_{37} SST estimates (Figures 2-4; *Pelejero et al.,* 1999). In cases where $\delta^{13}C_{G.ruber}$ and U^k_{37} SST estimates are not available at the same depths as the $\delta^{13}C_{org}$ data, the former are linearly interpolated (never exceeding distances of 10 cm) to the depth of the $\delta^{13}C_{org}$ values.

Our selection of $\delta^{13}C_{G.ruber}$ and U^k_{37} SST estimates to correct for temporal changes in $\delta^{13}C_{DIC}$, and to calculate α, respectively, makes the SCS records most closely comparable to a similar record from the Angola current region reported by *Müller et al.* [1994] (Figure 9) who used an identical approach. It is noted, however, that *G. ruber,* a shallow-dwelling foraminifera, might not optimally monitor temporal changes in $\delta^{13}C_{DIC}$ at the depth where the majority of the marine organic matter is produced, i.e., at the chlorophyll maximum.

5. Discussion

If $\delta^{13}C_{org}$ were a reliable proxy for local pCO_2, i.e., a measurable descriptor for the desired variable [*Wefer et al.,* 1999], it should meet three key criteria: (1) $\delta^{13}C_{org}$-pCO_2 estimates should yield reliable absolute values; (2) the temporal evolution of these estimates should correlate with reconstructions of processes that have been shown to influence local pCO_2 in the present ocean, i.e., upwelling intensity; and finally, (3) the temporal evolution of local estimates should be consistent with atmospheric CO_2 concentrations as recorded in ice cores, specifically in regions of the ocean where equilibrium between atmospheric and oceanic CO_2 is expected. In sections 5.1 - 5.3 we will test these three premises.

5.1. Absolute $\delta^{13}C_{org}$-pCO_2 Values

Taken at face value, i.e., based on the assumption of 0% terrigenous organic matter contribution to the bulk C_{org} throughout the records, the SCS pCO_2 estimates presented in Figure 8 (see also Figure 12) suggest that sea surface pCO_2 values were persistently higher than the ice core CO_2 record by ~100 ppm, and, accordingly, that the SCS has been a constant, significant source of CO_2 to the atmosphere for the last 220 kyr, irrespective of glacial or interglacial boundary conditions. Given the time-varying presence of terrigenous organic matter in SCS sediments throughout the records, these SCS pCO_2 estimates are maximum values. In addition, given the evidence from n-nonacosane abundances of significantly increased terrigenous organic matter input during glacials as compared to interglacials (Figures 2-4), the reconstructed glacial-interglacial difference at each core site is a minimum estimate. An admixture of only 10-15% terrigenous C_{org} ($\delta^{13}C_{org}$ ca. -26‰) to the bulk C_{org} would be enough to decrease the concentration estimates from supersaturated to equilibrium values. The increased glacial contribution of terrigenous organic matter, however, implies even lower glacial pCO_2 estimates, i.e., an even larger glacial-interglacial contrast in excess of the ~ 80 ppm observed in ice cores. In turn, this implies that the SCS at the sites presented above was

either a considerable CO_2 sink during glacial times or, alternatively, that it was a strong source during interglacials. Neither of these scenarios, however, is compatible with the current understanding of SCS palaeoceanography.

All $\delta^{13}C_{org}$-pCO_2 estimates published to date are based on the (tacit) assumption of 0% terrigenous C_{org} contribution throughout the records, seriously qualifying the absolute values of the pCO_2 estimates. Thus, the inability to quantify the terrigenous fraction of the bulk C_{org} precludes a quantitative CO_2 estimate based on $\delta^{13}C$ of bulk organic matter. This pitfall can be overcome, however, by using the $\delta^{13}C$ composition of a single, uniquely marine biomarker, e.g., alkenones [*Jasper and Hayes,* 1990, 1994; *Andersen et al.,* 1999] (see further discussion below in section 5.2).

5.2. The pCO_2 Estimates and Upwelling Intensity

In today's ocean, low latitude upwelling regions are the major sources of CO_2 from the ocean to the atmosphere, with supersaturation ranging up to 140 μatm [*Tans et al.,* 1990; *Takahashi et al.,* 1997] (see also Figure 1). Accordingly, $\delta^{13}C_{org}$-pCO_2 records from upwelling regions, which all show Holocene pCO_2 values 40-100 ppm higher than preindustrial atmospheric CO_2 values, have been attributed to this supersaturation [*Pedersen et al.,* 1991; *Müller et al.,* 1994; *Westerhausen et al.,* 1994; *Raymo et al.,* 1996]. As discussed in section 5.1, the reconstructed supersaturation could also be due to the admixture of terrigenous C_{org}. However, the pCO_2
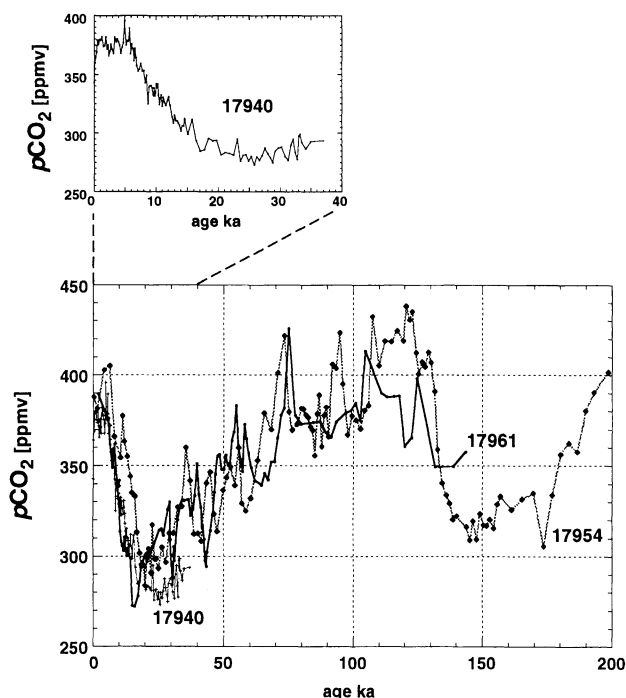

Figure 8. The pCO_2 estimates of cores 17940-2, 17954-2, and 17961-2 from the northern, western central, and southern SCS versus age. The pCO_2 of core 17940-2, that has the best age control of the three records [see *Wang et al.,* 1999], is enlarged to highlight the timing of changes during the last deglaciation.
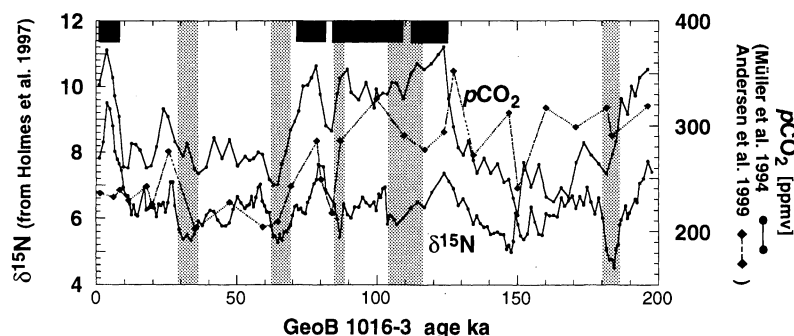
Figure 9. Bulk sedimentary $\delta^{15}N$ data from *Holmes et al.* [1997] and pCO_2 reconstruction based on bulk $\delta^{13}C_{org}$ [*Müller et al.*, 1994] and on $\delta^{13}C_{alkenone}$ [*Andersen et al.*, 1999] of core GeoB 1016-3 from the Angola Basin (8 in Figure 1). Shaded vertical bars: $\delta^{15}N$ minima are upwelling maxima labeled according to *Holmes et al.* [1997]; black bars on upper x axis denote periods of maximum air-sea CO_2 difference (ΔpCO_2) based on $\delta^{13}C_{org}$-pCO_2 estimates, according to *Müller et al.* [1994]. Note the lack of an expected consistent correlation between upwelling maxima and ΔpCO_2 maxima.

estimates from bulk sedimentary $\delta^{13}C_{org}$ records from major upwelling sites do not appear to reflect changes in upwelling intensity with time. For example, off Angola, times of maximal differences between the local pCO_2 and the Vostok ice core record, which imply periods of maximum upwelling [*Müller et al.*, 1994], do not coincide with minima in sedimentary $\delta^{15}N$ [*Holmes et al.*, 1997] (Figure 9). This relationship should be expected if the interpretation of the $\delta^{15}N$ variations from the same core is correct. The interpretation of the $\delta^{15}N$ records as reflecting upwelling strength is corroborated by synchronous changes in % C_{org} and SST [*Holmes et al.*, 1997], biogenic Ba [*Schneider et al.*, 1997], and organic biomarker compound abundances [*Hinrichs et al.*, 1999] from the same core, all pointing to lower $\delta^{15}N$ during periods of high production, i.e., intense upwelling. A similar independence of $\delta^{13}C_{org}$ variations and changes in upwelling intensity can be observed in records from the NW Mexican Margin (core NH22P [*Ganeshram et al.*, 1999]), the Panama Basin (core P7GC [*Pedersen et al.*, 1991]) and the western Indian Margin (core MD76-131 [*Ganeshram et al.* 2000). In all these cases, sedimentary nitrogen isotope data (*Ganeshram et al.*, 1999; *Farrell et al.*, 1995; and *Ganeshram et al.*, 2000, respectively), in agreement with C_{org} concentrations and various other indicators of marine production, indicate significant changes in upwelling intensity during the last glacial-interglacial cycle that are not reflected in the sedimentary $\delta^{13}C_{org}$ records and the pCO_2 values derived from them.

A similar conclusion can be tentatively drawn from a set of sediment surface $\delta^{13}C_{org}$ values from the eastern equatorial Pacific (Figure 10) (T.F. Pedersen, unpublished data, 2000). Here, $\delta^{15}N$ values (Figure 10) [*Farrell et al.*, 1995] show a clear response to equatorial upwelling, with lowest values centered on the equator and increasing $\delta^{15}N$ values toward the north, reflecting increasing relative nutrient utilization [cf. *Farrell et al.*, 1995]. Recent studies clearly demonstrate that the equatorial Pacific upwelling is not only associated with increased levels of macronutrients but also with high CO_2 concentrations in surface waters (up to 500 μatm), i.e., with fluxes of CO_2 from the ocean to the atmosphere that rapidly

diminish to the north and south of the equator where ocean-atmosphere equilibrium is reached with respect to CO_2 [*Chavez et al.*, 1999; *Feely et al.*, 1999]. However, the surface sediment $\delta^{13}C_{org}$ distribution determined on the same set of samples as $\delta^{15}N$ does not mirror the ~100 μatm gradient in $CO_2(aq)$ governed by equatorial upwelling (Figure 10). Given the lower SSTs associated with the core of equatorial upwelling, pCO_2 estimates derived from these bulk sedimentary $\delta^{13}C_{org}$ values would be, if anything, inversely correlated with pCO_2.

In view of these considerations, we suggest that the postulation of an inverse coupling between pCO_2 and upwelling, i.e., a decreased local pCO_2 due to increased upwelling-driven production in the eastern Atlantic equatorial upwelling belt [*Struck et al.*, 1993; *Westerhausen et al.*, 1994] (7 in Figure 1) should be re-evaluated. The
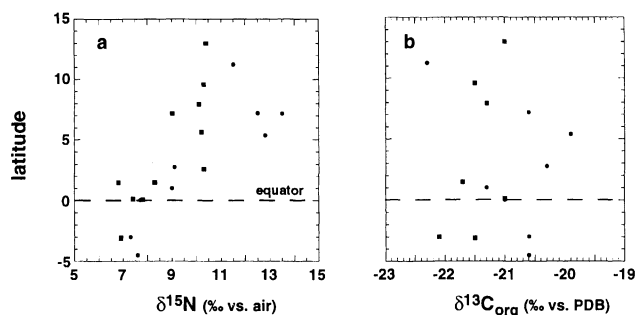

Figure 10. (a) Bulk sedimentary $\delta^{15}N$ [from *Farrell et al.*, 1995], and (b) bulk sedimentary $\delta^{13}C_{org}$ surface sediment distribution ($\delta^{13}C_{org}$ data of T.F. Pedersen, unpublished data, 2000) along two transects (~ 110°W, filled circles; and ~ 90-95°W, filled squares) across the eastern equatorial Pacifc upwelling zone. Note the clear reflection of increasing relative nitrate utilization, i.e., the decreasing supply of nitrate by upwelling from the equator toward the north and the absence in the $\delta^{13}C_{org}$ distribution of any obvious reflection of the equatorial upwelling.

correlation between decreased pCO_2 estimates with increased productivity of this equatorial Atlantic record in these studies was caused, to a significant extent, by the correction of the $\delta^{13}C_{org}$ values for winter SST variations that are, in turn, coupled to upwelling intensity [*Westerhausen et al.*, 1994]. The annual average SSTs (U^k_{37} estimates; 23.2-27.5°C; [*Westerhausen et al.*, 1994]) and even the foraminiferal transfer function winter SSTs in this region are higher than 15°C throughout the record (except for a few single values during stage 8), where, as *Fontugne and Duplessy* [1981] and *Freeman and Hayes* [1992] have shown, there is very little dependency of $\delta^{13}C_{org}$ on SST. Thus, the SST correction seems unjustified. Without the SST correction, there is no significant correlation ($r=0.32$) between $\delta^{13}C_{org}$ and the concentration of C_{org} or derived export productivity (Figure 11), in accord with the records summarized above. In addition, the postulation of an inverse coupling between upwelling intensity and marine productivity is at odds with all observations on modern equatorial upwelling systems that clearly demonstrate that changes in the direction and intensity of CO_2 exchange between ocean and atmosphere on interannual and longer timescales are governed by physical, rather than biological, processes [*Feely et al.*, 1987, 1999; *Chavez et al.*, 1999].

It is possible that some of the observed down-core and surface sediment variability of $\delta^{13}C_{org}$ in upwelling areas is obscured by changes in various biological factors (see references in section 3.3), most importantly phytoplankton growth rate [*Laws et al.*, 1995; *Bidigare et al.*, 1997]. Increased average phytoplankton growth rates are associated with decreased C isotope fractionation [*Laws et al.*, 1995;

Figure 11. Bulk sedimentary $\delta^{13}C_{org}$ (corrected for $\delta^{13}C_{DIC}$ using $\delta^{13}C_{G.ruber}$) versus estimated export production (in gCm^{-2} yr^{-1}) of core 16772 in the eastern equatorial Atlantic ($n=153$, $r=0.32$; all data from *Westerhausen et al.* [1994]).

Bidigare et al., 1997]. Thus, the influence of upwelling-related higher concentrations of $CO_2(aq)$ on the $\delta^{13}C_{org}$ of phytoplankton is potentially counteracted by increased growth rate associated with higher supply of nutrients due to the same upwelling. To overcome any potential bias due to this counteracting control on $\delta^{13}C_{org}$, future studies of sedimentary $\delta^{13}C_{org}$ should include some measure of average phytoplankton growth rate [e.g., *Andersen et al.*, 1999; *Stoll and Schrag*, 2000]. The first attempt to correct for the effect of variable growth rates on $\delta^{13}C_{org}$-pCO_2 estimates [*Andersen et al.*, 1999], however, does not lead to an improved correlation of temporal changes in upwelling intensity and pCO_2 estimates either (Figure 9). Moreover, if recent findings by *Riebesell et al.* [2000] are corroborated, in addition to growth rate and CO_2 concentration, carbon isotope fractionation may be affected by the growth-limiting resource (e.g., light levels, NO_3- versus NH_4+ N-source, N versus P or Fe as the growth-limiting nutrient), making it even more difficult to reliably extract the influence of changes in $CO_2(aq)$ on the sedimentary $\delta^{13}C_{org}$.

5.3. Marine $\delta^{13}C_{org}$-pCO_2 Estimates in Comparison to the Ice Core CO_2 Record

Ignoring for the moment the uncertainty in absolute values of the low-latitude marine isotopic pCO_2 estimates and the apparent decoupling of upwelling intensity and pCO_2, most of the marine $\delta^{13}C_{org}$ records that we have discussed here record a ~2‰ glacial-interglacial difference, irrespective of the local physico-environmental setting. This is precisely the amplitude expected from the ~80 ppm change in atmospheric CO_2 observed in ice cores [*Rau*, 1994]. Two of these records, however, deviate significantly from this general trend, possibly due to the effect of changes in phytoplankton growth rate discussed in section 5.2. *Pedersen et al.* [1991] [see also *Pedersen and Bertrand*, 2000] report a negligible deglacial $\delta^{13}C_{org}$ change in the Panama Basin, and the (uncorrected) $\delta^{13}C_{org}$ data of *Westerhausen et al.* [1994] from the equatorial Atlantic seem to indicate, if at all, a reversed trend, i.e., a $\delta^{13}C_{org}$ increase associated with temination I. The other low-latitude marine $\delta^{13}C_{org}$ records (Figure 1) show a very similar change to lighter values after termination I, suggesting a common driving mechanism.

We evaluate the possible role of $CO_2(aq)$ in determining these changes in $\delta^{13}C_{org}$ by examining the phasing of changes in the Vostok CO_2 record and the low latitude marine $\delta^{13}C_{org}$ records/pCO_2 estimates, focusing on the MIS 5/4 transition and the last deglaciation. Thus, the Vostok CO_2 record shows a more or less steady decrease in atmospheric CO_2 concentration starting immediately after the penultimate (MIS 5e) CO_2 maximum at ~130 to ~115 ka (Figure 12). In sharp contrast, low-latitude marine pCO_2 estimates from a wide range of environmental settings [*Fontugne and Calvert*, 1992; *Müller et al.*, 1994; *Raymo et al.*, 1996; this study], including the molecular record of *Jasper and Hayes* [1994], indicate persistently high pCO_2 values from ~130 ka until the MIS 5/4 transition (~70 ka) when there is a sharp pCO_2 drop (the assertion of synchroneity of these estimates is constrained by the uncertainty in the age models and the low resolution of some of these records). None of the published low-latitude pCO_2 estimates follows the Vostok CO_2 record during this
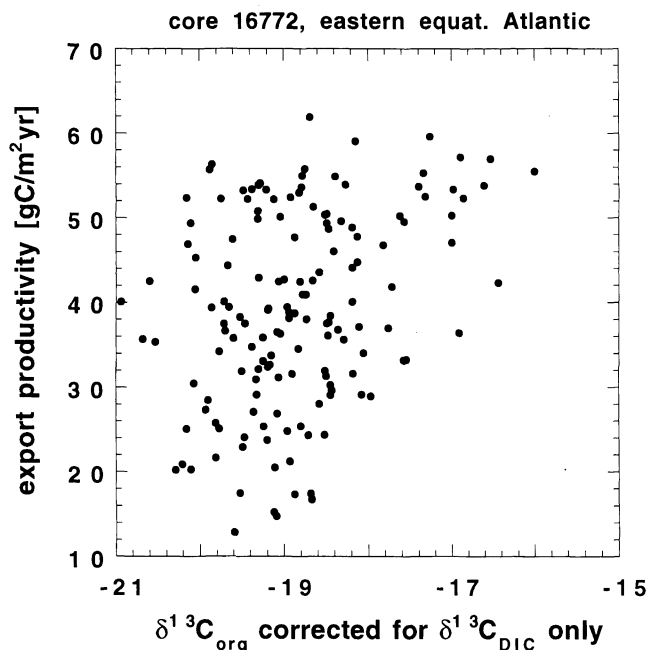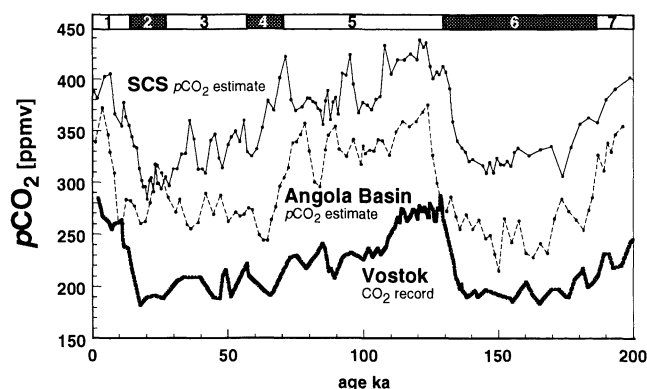
Figure 12. Comparison of the SCS and Angola Basin pCO_2 estimates with the Vostok CO_2 record (Angola Basin data from *Müller et al.* [1994]; Vostok CO_2 record from *Petit et al.* [1999]). All records are plotted on the same CO_2 scale but using their independently derived age models. The record of core 17954-2 from the western central SCS was chosen to represent the SCS estimates because it covers the longest time period.

time period, including the records from the SCS and the eastern Mediterranean where, based on current understanding, CO_2 equilibrium between the ocean and the atmosphere is expected. If $\delta^{13}C_{org}$ is indeed driven by changes in $CO_2(aq)$, then there are two important corollaries: (1) persistently high surface pCO_2 in low-latitude upwelling regions during the later part of MIS 5 would need to be compensated for by even larger sinks elsewhere and (2) there would need to be a mechanism to drive those areas of the ocean that presumably are in equilibrium out of equilibrium synchronously with atmospheric CO_2 during the later part of MIS 5. In the case of the SCS, the high late MIS 5 pCO_2 estimates could be due to the increased admixture of terrigenous C_{org} as indicated by the n-nonacosane records (Figures 3, 4). However, the similarity in most of the low-latitude $\delta^{13}C_{org}$ records during this time interval and the corresponding conclusion from pCO_2 estimates derived from marine biomarkers [*Jasper and Hayes*, 1994] seem to argue against this possibility.

In contrast to the discrepancy during the MIS 5/4 transition, the timing of changes in $\delta^{13}C_{org}$ in the SCS cores during the last (and penultimate) deglaciation (Figures 2-5) seems to parallel findings from the Vostok and Byrd CO_2 records that show increases in atmospheric CO_2 leading global ice volume [*Petit et al.*, 1999] and the Bølling/Allerød warming in the Northern Hemisphere [*Blunier et al.*, 1997; *Stocker*, 2000], respectively. Despite this agreement in timing of the onset of deglacial change, SCS pCO_2 reaches Holocene levels only at ~5 ka following a continuous rise (Figure 8), i.e., significantly later than the ice core records of atmospheric CO_2 that indicate Holocene levels were reached at ~10 ka [e.g., *Neftel et al.*, 1988]. Note that these discrepancies in the timing of deglacial events are much larger than any uncertainty in stratigraphic correlation of ice core and SCS sedimentary records, and in the absolute age control of the SCS cores, particularly of core 17940-2 [*Wang et al.*, 1999]. Moreover, this offset in timing is most likely not due to variable admixture of terrigenous C_{org} at site 17940, given the un-

changed concentrations of n-nonacosanes in core 17940-2 during the last ca. 10 ka (Figure 2).

In summary, the discrepancy in timing of marine $\delta^{13}C_{org}$-pCO_2 estimates and the ice core CO_2 record suggests that $CO_2(aq)$ may not be the main/single driving mechanism of low-latitude $\delta^{13}C_{org}$ variability. The remarkably similar low-latitude $\delta^{13}C_{org}$ records from a wide range of environmental settings, particularly the similar timing of changes during the MIS 5/4 transition, however, suggests a common mechanism governing $\delta^{13}C_{org}$. This similarity in both timing and amplitude is even more surprising given the wide range of biological factors that have been shown to influence $\delta^{13}C_{org}$ (see references in section 3.3), as well as the temporal and spatial variability in the admixture of terrigenous C_{org}.

In this context it is noted that the power spectra of the ice core CO_2 record, on the one hand, and the frequency distribution of variability in low-latitude upwelling intensity/marine production, on the other hand, seem to indicate a negligible role of changes in the low-latitude ocean on atmospheric CO_2. Thus, whereas low-latitude upwelling/marine production is significantly controlled by precessional periodicities [*Lyle*, 1988; *Clemens et al.*, 1991; *Beaufort et al.*, 1997; *Holmes et al.*, 1997; *Reichart et al.*, 1997; *Schubert et al.*, 1998], the Vostok CO_2 record is dominated by the 100-kyr component, with almost no power at lower periodicities [*Petit et al.*, 1999].

6. Conclusions

Sedimentary records from the SCS and other low-latitude locales caution the use of bulk sedimentary $\delta^{13}C_{org}$ records as a proxy indicator of local $CO_2(aq)$. In addition, the weakness/absence of a correlation between $\delta^{13}C_{org}/pCO_2$ estimates and upwelling indicators suggests that previous evaluations of the importance of changes in upwelling-related oceanic degassing on glacial-interglacial atmospheric CO_2 levels need to be reassessed, possibly using sedimentary $\delta^{15}N$ as a more reliable proxy of nutrient utilization and/or supply.

The similarity in timing and absolute values of marine $\delta^{13}C_{org}$ records from very different settings could be taken as evidence for a common driving force for determining $\delta^{13}C_{org}$. Previous studies concluded that all models that try to explain the biological/physiological processes governing the $\delta^{13}C$ of phytoplankton must be able to reproduce the latitudinal trend in $\delta^{13}C_{org}$ observed in the present-day ocean. We further suggest that all these models should be able to reproduce similar (in timing and amplitude) global-scale $\delta^{13}C_{org}$ variations on glacial-interglacial timescales, irrespective of the local setting. This conundrum provides a unique opportunity for combined plankton physiological and palaeoceanographic studies to determine the mechanism(s) governing the isotopic composition of marine organic matter. As with all palaeoceanographic reconstructions, pCO_2 estimates based on $\delta^{13}C_{org}$ and/or $\delta^{13}C_{alkenones}$ should be corroborated by independent evidence, i.e., an independent sedimentary proxy.

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Correction to "A critical review of marine sedimentary $\delta^{13}C_{org}$-pCO$_2$ estimates: New palaeorecords from the South China Sea and a revisit of other low-latitude $\delta^{13}C_{org}$-pCO$_2$ records" by Markus Kienast, Stephen E. Calvert, Carles Pelejero, and Joan O. Grimalt

In the paper "A critical review of marine sedimentary $\delta^{13}C_{org}$-pCO$_2$ estimates: New palaeorecords from the South China Sea and a revisit of other low-latitude $\delta^{13}C_{org}$-pCO$_2$ records" by Markus Kienast, Stephen E. Calvert, Carles Pelejero, and Joan O. Grimalt (*Global Biogeochemical Cycles*, 15(1), 113–127, 2001), an incorrect version of Figure 9 was published. The correct Figure 9 and its caption appear below.
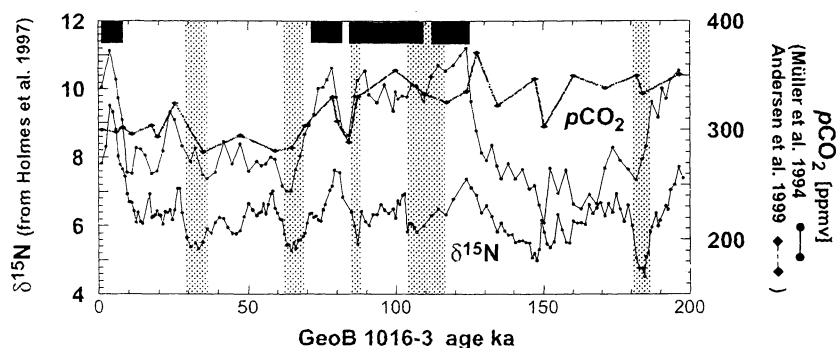

Figure 9. Bulk sedimentary δ^{15}N data from *Holmes et al.* [1997] and pCO$_2$ reconstruction based on bulk $\delta^{13}C_{org}$ [*Müller et al.*, 1994] and on $\delta^{13}C_{alkenone}$ [*Anderson et al.*, 1999] of core GeoB 1016-3 from the Angola Basin (8 in Figure 1). Shaded vertical bars: δ^{15}N minima are upwelling maxima labeled according to *Holmes et al.* [1997]; black bars on upper x axis denote periods of maximum air-sea CO$_2$ difference (ΔpCO$_2$) based on $\delta^{13}C_{org}$-pCO$_2$ estimates, according to *Müller et al.* [1994]. Not the lack of an expected consistent correlation between upwelling maxima and ΔpCO$_2$ maxima.

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