# 6. MIOCENE-PLEISTOCENE SEDIMENTARY RECORD OF CARBON BURIAL UNDER THE BENGUELA CURRENT UPWELLING SYSTEM, SOUTHWESTERN MARGIN OF AFRICA<sup>1</sup>

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# ABSTRACT

The amounts of inorganic and organic carbon in Miocene–Pleistocene sediments from Ocean Drilling Program Sites 1081, 1082, 1084, 1085, and 1087 and Deep Sea Drilling Project Site 532 on the margin of southwest Africa are summarized. Concentrations of CaCO<sub>3</sub> are consistently ~75 wt% at Sites 1085 and 1087 in the Cape Basin but drop below 25 wt% near the Pliocene/Pleistocene boundary at Sites 532, 1081, 1082, and 1084 in Walvis Basin and on Walvis Ridge because of greater opal production during this period. Total organic carbon concentrations range between 0.1 and 18 wt% and are highest in sediments deposited at Site 1084 in Walvis Basin. Organic matter is predominantly marine in origin at all locations, as indicated by a combination of Rock-Eval and C/N measurements. Both the production and preservation of organic matter appear to have fluctuated regularly during the evolution of this upwelling system.

# INTRODUCTION

The upwelling system associated with the Benguela Current, the eastern boundary current offshore southwest Africa, is one of the most productive areas of the modern ocean and is consequently a major locus of carbon burial. Sediments from Deep Sea Drilling Project (DSDP) Sites 362 and 532, which are at virtually the same location on Walvis Ridge,

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have provided a preliminary record of the evolution of upwelling and enhanced biological productivity of this system. Siesser (1980) concluded from concentrations of organic carbon and diatom assemblages in rotary-drilled cores from Site 362 that upwelling-enhanced productivity has gradually increased since its onset ~10 Ma. Hydraulic piston coring (HPC) at Site 532 yielded a less disturbed and more complete record of the upwelling history (Hay et al., 1982). Maxima in concentrations of organic carbon and diatom microfossils indicate that productivity peaked at this location in the late Pliocene to early Pleistocene (Meyers et al., 1983; Dean et al., 1984). The HPC record also revealed light-dark alternations that correspond to cycles in concentrations of organic carbon, calcium carbonate, and clay minerals. Diester-Haass et al. (1986, 1990, 1992) explain the color cycles as resulting from a combination of glacial-interglacial shifts in the axis of the Benguela Current, sea-level changes that modified the delivery of organic matter, and climate changes that changed the delivery of clastic sediment components. Moreover, these authors conclude that carbonate dissolution fueled by the oxidation of organic matter is a major factor in the cause of the cycles.

One of the goals of Leg 175 was to improve the record of the evolution of the Benguela Current by obtaining sediment from multiple locations along the margin of southwestern Africa. An important part of this record is to describe the history of burial of inorganic and organic carbon at these different locations. This report provides a summary of measurements of the carbonate and organic carbon contents of sediment from five Leg 175 sites and from Site 532. These six sites comprise a transect along the axis of the Benguela Current. Organic geochemical parameters are used to evaluate the sources of the total organic matter and the degree of its preservation in sediment from the six sites.

# SAMPLING AND ANALYSIS

# **Depositional Settings**

The sedimentary sequences at the six drill sites provide a composite late Miocene to present history of the upwelling system that currently extends from the mid-Namib margin to north of Walvis Ridge. The drill sites (Fig. F1) are seaward of the main upwelling core, but they contain second-order signals of the enhanced marine productivity associated with the upwelling that are transported to their respective locations by the Benguela Current and its meanders and eddies (cf. Lutjeharms and Stockton, 1987). The cored locations (Table T1) and their sedimentary sequences are briefly described here in the south-north direction of the current flow.

Site 1087 (31°28′S, 15°19′E) is in 1372 m of water on South Africa slope. Its sediments are greenish gray nannofossil oozes with varying amounts of foraminifers and clay. Site 1085 (29°22′S, 13°59′E) is at a water depth of 1713 m on Namibia slope. Sediments at this site are greenish gray foraminifer-bearing nannofossil oozes. Site 1084 (25°31′S, 13°02′E) is in 1992 m of water in the northern landward part of Cape Basin. The sediments contain alternations of bioturbated olive-gray to black clays rich in nannofossils and diatoms that overlie an olive-gray clayey nannofossil ooze. Site 1082 (21°06′S, 11°49′E; water depth 1279 m) is in Walvis Basin on the southern flank of Walvis Ridge. Its sediments also exhibit alternations of olive-gray to black bioturbated clays

**F1.** Locations of the six drill sites on the southwest Africa margin, p. 12.



**T1**. Core sites and depositional parameters, p. 19.

rich in nannofossils and diatoms. Sites 532 and 1081 are on the continental shoulder of Walvis Ridge. Site 532 (19°45′S, 10°31′E) is on a ridge terrace in 1331 m of water. Sediments here consist of alternations of olive-to-brown nannofossil ooze with varying amounts of foramini-fers, diatoms, and clay. Site 1081 (19°37′S, 11°19′E) is more landward of Site 532 in water 794 m deep. Site 1081 sediments have alternations of olive-gray to black clays with varying amounts of nannofossils and diatoms that overlie an olive-gray clayey nannofossil ooze.

# Inorganic and Organic Carbon Measurements and C/N Ratios

The total organic carbon (TOC) contents of sediment samples from Sites 1081, 1082, 1084, and 1085 were determined onboard the *JOIDES Resolution* by the difference between total carbon concentrations as measured by a Carlo Erba NA 1500 NCS analyzer (Verardo et al., 1990) and the carbonate-carbon concentrations as measured by a Coulometrics 5011 inorganic carbon analyzer (Engleman et al., 1985). Freeze-dried samples were combusted at 1000°C in an oxygen atmosphere in the NCS analyzer, and the resulting combustion products were chromatographically separated and quantified to yield the concentrations of total carbon and nitrogen. The TOC and total nitrogen values were used to calculate atomic C/N ratios of the samples. These C/N ratios are assumed to represent those of sediment organic matter in this report.

Carbon analyses of sediments from Site 1087 were done onshore (Meyers and Robinson, Chap. 2, this volume) and those from Site 532 were done onboard the *Glomar Challenger* by procedures different from JOIDES Resolution shipboard procedures. For samples from both sites, calcium carbonate concentrations were determined using the carbonate bomb technique of Müller and Gastner (1971), which yields results comparable to the coulometrics procedure (P. Meyers, unpubl. data). Weighed samples were reacted with 3-N HCl, and the volume of CO<sub>2</sub> released from each sample was measured and compared to the volumes released from known amounts of pure CaCO<sub>3</sub> to determine the percentage in the sample. The carbonate-free residue remaining after acid treatment was collected, rinsed, and dried. Concentrations of TOC and total residual nitrogen (TN) in the carbonate-free residues were directly measured with a CHNS analyzer. This procedure involves heating the carbonate-bomb residue at 1020°C and measuring the combustion products by gas chromatography (e.g., Verardo et al., 1990). TOC concentrations are reported on a whole-sediment basis, adjusting for the carbonate concentrations determined from the bomb technique. Atomic C/N ratios have been calculated from the TOC and TN concentrations.

Meyers and Silliman (1996) compared the results of the direct TOC procedure with those calculated by difference. The direct procedure is more reliable at TOC concentrations <0.1 wt%, but agreement between the two procedures is good above this value. Because most of the sediments from beneath the Benguela Current upwelling system are rich in organic matter, the results of the two procedures are considered equivalent.

# **Rock-Eval Pyrolysis**

Rock-Eval pyrolysis of organic matter consists of heating samples at a rate of 25°C/min between 300° and 600°C to yield the amount of volatile hydrocarbons present in the sediment (S<sub>1</sub>), the amount of hydrocarbons released during pyrolysis (S<sub>2</sub>), and the amount of CO<sub>2</sub> released during heating to 390°C (S<sub>3</sub>). These values are combined with TOC values to provide the information necessary to calculate the hydrogen index (HI =  $100 \times S_2/TOC$  in milligrams hydrocarbon per gram of TOC) and the oxygen index (OI =  $100 \times S_3/TOC$  in milligrams CO<sub>2</sub> per gram of TOC). The temperature of maximum hydrocarbon release during pyrolysis ( $T_{max}$ ) is also obtained and provides a measure of organic matter thermal maturity (Espitalié et al., 1977).

# **RESULTS AND DISCUSSION**

The inorganic and organic carbon results are compiled from Leg 175 shipboard analyses for Sites 1081, 1082, 1084, and 1085 (Wefer, Berger, Richter, et al., 1998), from shore-based analyses for Site 1087 (Meyers and Robinson, Chap. 2, this volume), and from shipboard (Shipboard Scientific Party, 1984) and shore-based analyses (Meyers et al., 1984; Rullkötter et al., 1984) for Site 532. Subbottom depths have been converted to approximate sediment ages using the shipboard biostratigraphic summaries (Shipboard Scientific Party, 1984; Giraudeau et al., 1998). As more refined postcruise chronostratigraphies become available for the Leg 175 results, the shipboard age assignments for the sites of this drilling leg may undergo minor adjustments. Moreover, as determinations of mass accumulation rates become available, more sophisticated interpretations of the paleoproductivity records than attempted in this summary will become possible.

# **CaCO<sub>3</sub> Concentrations**

Three important patterns in the magnitude and variability of CaCO<sub>3</sub> concentrations in sediment deposited over the past 10 m.y. emerge from comparison of the results from the six sites. First, concentrations are notably higher in sediments at Sites 1087 and 1085 than at the more northerly locations (Fig. F2). Second, strong minima in concentrations occur in the upper Pliocene and lower Quaternary sections of sediments from Sites 1084, 1082, 532, and 1081 but are weakly expressed or absent in time-equivalent sections of Sites 1087 and 1085. These minima correspond to the early Matuyama Diatom Maximum described by Lange et al. (1999) and indicate an excursion from coccolith-dominated production to one dominated by diatoms. Third, welldeveloped alternations between high and low CaCO<sub>3</sub> concentrations are present at all sites and especially at Sites 1084, 1082, and 532. These variations in carbonate content correspond to light-dark color cycles that are described at these locations (Dean et al., 1984; Diester-Haass et al., 1986; Vidal et al., 1998; Wefer, Berger, Richter, et al., 1998), with the lighter portions of the cycles having higher CaCO<sub>3</sub> concentrations.

The different patterns indicate that different combinations of changes in delivery of calcareous material, dilution by noncalcareous components, and carbonate dissolution fueled by oxidation of organic **F2**. Concentrations of calcium carbonate, p. 13.



matter or corrosiveness of subsurface water masses occurred at the six locations.

### **Organic Carbon Concentrations**

The pattern of increasing TOC concentration since the Miocene that was documented at DSDP Sites 362 and 532 on Walvis Ridge (Siesser, 1980; Meyers et al., 1983) also appears at the sites in Walvis and Cape Basins (Fig. F3). However, this trend is not as well developed at Sites 1087 and 1085 as at the more northerly locations, where TOC concentrations range between 5 and 10 wt% in Pliocene and Pleistocene sediments (Fig. F3). Such concentrations are notably higher than in most deep-sea sediments from the South Atlantic (~0.3 wt%) (Premuzic et al., 1982; Keswani et al., 1984). The elevated TOC concentrations under the Benguela Current reflect the combination of elevated paleoproductivities, which deliver abundant organic matter to the sediments, and the high accumulation rates of sediments, which favors preservation of the organic matter.

Well-developed alternations between higher and lower TOC concentrations exist at Sites 1084, 1082, 532, and 1081 (Fig. F3). Like the similar alternations in CaCO<sub>3</sub> concentrations, these accompany light-dark color cycles in the sediments. However, changes in sediment color are not as closely linked to TOC concentrations as they are to CaCO<sub>3</sub> concentrations (Dean et al., 1984; Diester-Haass et al., 1986; Vidal et al., 1998). Nonetheless, good correspondence exists between higher TOC concentrations and lower CaCO<sub>3</sub> concentrations in sediment from Sites 1084, 1082, 532, and 1081 (Fig. F4). Some of this correspondence is probably related to the Matuyama Diatom Maximum (Lange et al., 1999) and associated elevated production of organic matter, yet some of it may indicate dissolution of CaCO<sub>3</sub> as a result of oxidation of organic matter (e.g., Diester-Haass et al., 1986, 1992). An additional factor that is probably involved is downslope transport of organic carbon-rich sediments from the shelf edge that is related to glacial-interglacial changes in sea level (Diester-Haass et al., 1986, 1990, 1992; Summerhayes, et al., 1995).

# **Organic Carbon/Nitrogen Values**

Organic C/N values calculated for sediment samples using TOC and total nitrogen concentrations vary from 1 to 22 (Fig. F5). Most of these atomic ratios are intermediate between unaltered algal organic matter (5–8) and fresh land-plant material (25–35) (e.g., Emerson and Hedges, 1988; Meyers, 1994). The low C/N values occur in sediment that is poor in organic carbon; these values may be biased by the tendency of clay minerals to absorb ammonium ions generated during the degradation of organic matter (Müller, 1977). Because of their setting offshore of a coastal desert, it is likely that delivery of land-derived organic matter to these sediments has been minimal since the late Miocene initiation of the Namib Desert, which has been linked to the onset of coastal upwelling by pollen contents of Site 532 sediment (van Zinderen Bakker, 1984).

C/N values are higher in sediments richer in TOC than in those lean in TOC (Fig. F5). Many of the values are higher than in fresh algal organic matter. C/N values that are elevated above algal values are common in organic carbon–rich marine sediments (Suess and Müller, 1980;

**F3.** Concentrations of organic carbon, p. 14.



**F4**. Comparison of concentrations of organic carbon and calcium carbonate, p. 15.



**F5**. Comparison of C/N ratios to organic carbon, p. 16.



Meyers, 1997). They evidently result from the selective loss of nitrogen as organic matter settles from the photic zone, because nitrogen-bearing proteins are more labile than other organic matter components such as carbohydrates and lipids (Verardo and McIntyre, 1994). This type of preferential nitrogen depletion and consequent carbon enrichment is recognized in other organic carbon-rich sediments, such as those present in the Mediterranean Sea (Meyers and Doose, 1999; Nijenhuis and de Lange, 2000), the equatorial Atlantic (Verardo and McIntyre, 1994), and the northwest Mexican slope (Ganeshram et al., 1999). The C/N elevations are most pronounced when TOC concentrations are highest, suggesting that a higher rate of organic matter delivery leads to diminished organic matter degradation. The existence of the diagenetically elevated C/N values is further evidence that organic matter preservation was enhanced during accumulation of the Pliocene-Pleistocene sediments under the Benguela Current, presumably because of the presence of a strongly developed oxygen minimum zone along the continental margin of Cape and Walvis Basins.

# **Results of Rock-Eval Pyrolysis**

Rock-Eval pyrolysis was originally developed to characterize the organic matter present in oil source rocks, which typically is more thermally mature and present at higher concentrations than commonly found in nonlithified sediments obtained by scientific ocean drilling. Rock-Eval analyses have nonetheless proved valuable in helping to determine organic matter sources in DSDP and Ocean Drilling Program (ODP) samples. The hydrogen index (HI) and the oxygen index (OI) relate to the origin of the total organic matter and are commonly plotted against each other in a Van Krevelen-type diagram in which a comparison of elemental H/C and O/C ratios is approximated. In the HI-OI plot, three main types of organic matter and their thermal alteration pathways are defined. Type I organic matter is especially rich in aliphatic hydrocarbons and hydrocarbon-like components and is derived from algae and microbial biomass. Type II organic matter is moderately rich in these aliphatic components and originates from the waxy coatings of land plants and partially degraded algae. Type III organic matter is poor in hydrocarbon-like materials but rich in cellulose and lignin. Land-plant organic matter is usually rich in these woody components and consequently has lower HI and higher OI than found in lipid-rich and cellulose-poor algal organic matter. Type III organic matter, therefore, usually typifies woody land-plant matter, but it may also represent poorly preserved algal organic matter.

The source distinction between continental and marine organic matter that can be made from the results of Rock-Eval pyrolysis becomes blurred by diagenesis. Oxidation of organic matter affects both HI and OI values. As hydrocarbon-rich organic matter (Type I or II) is oxidized, its hydrogen content decreases and its oxygen content increases and it takes on the HI-OI characteristics of Type III vascular plant organic matter. A further constraint on the use of Rock-Eval pyrolysis for determination of organic matter source is that samples should contain at least 0.5 wt% TOC to yield meaningful results (Katz, 1983; Peters, 1986).

The results of Rock-Eval analyses of Leg 175 organic carbon–rich sediments show that their organic matter content appears to be dominated by varying mixtures of Type II algal material and Type III land-plant material (Fig. F6). The indication of large proportions of land-derived organic matter in many of the sapropels conflicts, however, with C/N **F6.** Rock-Eval pyrolysis diagrams for organic matter, p. 17.



ratios in these sediments that are too low (<20) for land-plant organic matter (Wefer, Berger, Richter, et al., 1998). The contradiction between the Rock-Eval source characterization and the elemental source characterization is evidence that the marine organic matter has been moder-ately oxidized, because well-preserved Type II organic matter has high HI values (Espitalié et al., 1977; Peters, 1986).

Comparison of TOC concentration with Rock-Eval HI values shows that sediment samples with higher TOC concentrations also have higher HI values (Fig. F7), which is a pattern also found in sapropels in the Mediterranean (Bouloubassi et al., 1999; Meyers and Doose, 1999). Higher TOC concentrations also correspond to higher C/N ratios (Fig. F5). As a consequence, sapropels having higher HI values also have higher C/N ratios, which is an indication of partial but not extensive alteration of marine organic matter during sinking and incorporation into bottom sediments. The correspondence between higher Rock-Eval HI values, higher elemental C/N ratios, and higher TOC concentrations suggests that the rate of export production and the degree of preservation of marine organic matter in bottom sediments are related and have increased as algal productivity has increased since the Miocene.

### **Thermal Maturity of Organic Matter**

Rock-Eval  $T_{max}$  values are generally <425°C in the Miocene–Pleistocene sediments from the locations sampled during Leg 175 (Wefer, Berger, Richter, et al., 1998; A. Rosell-Melé, unpubl. data). These values indicate that the organic matter in sediments from Cape and Walvis Basins and Walvis Ridge is thermally immature with respect to petroleum generation (Espitalié et al, 1977; Peters, 1986). Because organic matter is sensitive to temperatures that are only slightly elevated (>60°C), the thermal immaturity of the Miocene–Pleistocene organic matter is evidence of low heat flows along the margin of the southeastern South Atlantic Ocean since the times these sediments were deposited. Moreover, the thermal immaturity also is evidence of minimal contributions of recycled detrital organic matter derived from erosion of ancient sedimentary rocks present on the African landmass.

# Paleoceanographic Significance of Organic Carbon–Rich Sediments

TOC concentrations in sediments younger than the latest Miocene are larger and more variable than those in older sediments (Fig. F3). Maximum TOC concentrations appear within the last 2 m.y. at most sites but not in the most recent sediment, suggesting that upwelling once induced greater biological productivity than at present. Moreover, Pliocene–Pleistocene sediments at Sites 1087 and 1085 are not nearly as rich in organic carbon as those closer to Walvis Ridge (Wefer, Berger, Richter, et al., 1998), indicating that upwelling in most of Cape Basin has never created similarly elevated levels of productivity as at the more northerly locations. The greater concentrations of CaCO<sub>3</sub> present in sediments at Sites 1087 and 1085 (Fig. F2), and by implication less opal, is further evidence of the absence of similarly elevated productivity at these locations over the past 10 m.y.

The well-developed variations in TOC concentrations that are present in Pliocene–Pleistocene sediments at Sites 1084, 1082, 532, and 1081 have been postulated to represent glacial–interglacial changes in

**F7.** Relation of Rock-Eval pyrolysis hydrogen indices and organic carbon concentrations, p. 18.



the intensity and locus of the Benguela Current (Diester-Haass et al., 1990, 1992). This interpretation was necessarily constrained to essentially a single location (Site 362/532) by availability of cored sequences. The expanded geographic coverage provided by the multisite comparison in this summary shows that upwelling has not migrated up the coast as the Benguela Current has evolved but has instead remained in the region of Walvis Basin and Walvis Ridge, where the trade winds are able to transport surface water offshore. The TOC variations consequently are probably the result of glacial–interglacial variations in trade wind intensity and in sea level rather than reorientation of the Benguela Current.

# **SUMMARY**

Comparison of the amounts of inorganic and organic carbon in Miocene-Pleistocene sediments from ODP Sites 1081, 1082, 1084, 1085, and 1087 and DSDP Site 532 on the margin of southwest Africa provides insights into the evolution and oscillations of enhanced paleoproductivity associated with the Benguela Current upwelling system. Concentrations of CaCO<sub>3</sub>, which record coccolith production in this system, are consistently ~75 wt% at Sites 1085 and 1087 in the Cape Basin. In contrast, concentrations in sediments near the Pliocene/Pleistocene boundary drop to 25 wt% at Sites 532, 1082, and 1084 in Walvis Basin and on Walvis Ridge because of greater opal production at these areas during this period. Organic matter is predominantly marine in origin at all locations; its accumulation started to increase in the late Miocene and peaked in the early Pleistocene. TOC concentrations range between 0.1 and 18 wt% and are highest in sediments deposited at Site 1084 in Walvis Basin. TOC concentrations are especially elevated in dark parts of prominent light-dark color alternations that record varying combinations of elevated productivity, downslope transport of organic carbon-rich sediments from the shelf edge, and dissolution of CaCO<sub>3</sub> in the seafloor. Both the production and preservation of organic matter appear to have fluctuated regularly during the evolution of this upwelling system. Moreover, the correspondence of higher Rock-Eval hydrogen index values, higher elemental C/N ratios, and higher TOC concentrations suggests that the rate of export production and the degree of preservation of marine organic matter in bottom sediments are related and have increased since the Miocene.

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**Figure F1.** Locations of the six drill sites that provide the histories of carbon burial on the southwest Africa margin discussed in this report. Areas of perennial and episodic upwelling as summarized by Lutjeharms and Stockton (1987) are indicated by lighter and darker shading. Bathymetry is given in 1000-m increments.



**Figure F2.** Concentrations of calcium carbonate in Miocene, Pliocene, and Pleistocene sediments from Sites 1087, 1085, 1084, and 1082 in Cape and Walvis Basins and Sites 532 and 1081 on Walvis Ridge. Age profiles for the six sites are ordered from south to north to show temporal and geographical differences in calcium carbonate burial.



**Figure F3.** Concentrations of organic carbon in Miocene, Pliocene, and Pleistocene sediments from Sites 1087, 1085, 1084, and 1082 in Cape and Walvis Basins and Sites 532 and 1081 on Walvis Ridge. Age profiles for the six sites are ordered from south to north to show temporal and geographical differences in organic carbon burial.



**Figure F4.** Comparison of concentrations of organic carbon and calcium carbonate in Miocene, Pliocene, and Pleistocene sediments from Sites 1087, 1085, 1084, and 1082 in Cape and Walvis Basins and Sites 532 and 1081 on Walvis Ridge. Variable dilution of organic matter by carbonates contributes to the well-developed color cycles in sediments from Sites 532, 1081, 1082, and 1084.



**Figure F5.** Comparison of C/N (organic carbon; total nitrogen) ratios to organic carbon concentrations in Miocene, Pliocene, and Pleistocene sediments from Sites 1087, 1085, 1084, and 1082 in Cape and Walvis Basins and Sites 532 and 1081 on Walvis Ridge. Improved preservation of marine organic matter is accompanied by diagenetically elevated C/N values.



**Figure F6.** Rock-Eval Van Krevelen–type diagrams for organic matter in Miocene, Pliocene, and Pleistocene sediments from Sites 1085, 1084, and 1082 in Cape and Walvis Basins and Sites 532 and 1081 on Walvis Ridge. Thermal alteration pathways of organic matter Types I, II, and III from source material to graphite are indicated by dashed lines.



**Figure F7.** Relation of Rock-Eval hydrogen indices and organic carbon concentrations in Miocene, Pliocene, and Pleistocene sediments from Sites 1087, 1085, 1084, and 1082 in Cape and Walvis Basins and Sites 532 and 1081 on Walvis Ridge. Improved preservation of marine organic matter is accompanied by higher hydrogen index values.



**Table T1.** Core sites and depositional parameters of sedimentarysequences under the Benguela Current.

Depositional parameter	Site 1087	Site 1085	Site 1084	Site 1082	Site 532	Site 1081
Water depth (m):	1372	1713	1992	1279	1331	794
Distance from land (km):	200	200	155	140	190	120
Sedimentation rate (cm/k.y.):	2–7	2–13	10–27	7–21	2–20	4–15
Subbottom depth (mbsf):	423	604	605	600	267	453
Sediment ages (Ma):	0–9	0–15	0–4.7	0-5.8	0–7.5	0–9