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## Sensitivity of the North Atlantic Basin to Cyclic Climatic Forcing During the Early Cretaceous

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## SENSITIVITY OF THE NORTH ATLANTIC BASIN TO CYCLIC CLIMATIC FORCING DURING THE EARLY CRETACEOUS

<sup>1</sup>WALTER E. DEAN AND <sup>2</sup>MICHAEL A. ARTHUR

### ABSTRACT

Striking cyclic interbeds of laminated dark-olive to black marlstone and bioturbated white to light-gray limestone of Neocomian (Early Cretaceous) age have been recovered at Deep Sea Drilling Project (DSDP) and Ocean Drilling Project (ODP) sites in the North Atlantic. These Neocomian sequences are equivalent to the Maïolica Formation that outcrops in the Tethyan regions of the Mediterranean and to thick limestone sequences of the Vocontian Trough of France. This lithologic unit marks the widespread deposition of biogenic carbonate over much of the North Atlantic and Tethyan seafloor during a time of overall low sealevel and a deep carbonate compensation depth. The dark clay-rich interbeds typically are rich in organic carbon (OC) with up to 5.5% OC in sequences in the eastern North Atlantic. These eastern North Atlantic sequences off northwest Africa, contain more abundant and better preserved hydrogen-rich, algal organic matter (type II kerogen) relative to the western North Atlantic, probably in response to coastal upwelling induced by an eastern boundary current in the young North Atlantic Ocean. The more abundant algal organic matter in sequences in the eastern North Atlantic is also expressed in the isotopic composition of the carbon in that organic matter. In contrast, organic matter in Neocomian sequences in the western North Atlantic along the continental margin of North America has geochemical and optical characteristics of herbaceous, woody, hydrogen-poor, humic, type III kerogen. The inorganic geochemical characteristics of the dark clay-rich (<60% CaCO<sub>3</sub>) interbeds in Neocomian sequences in the North American Basin and the Cape Verde Basin off northwest Africa suggest that most of the detrital clastic material was derived from deep-sea fans off North America and Morocco during relatively wet intervals to dilute pelagic biogenic carbonate. Inorganic geochemical characteristics of the clastic material in the bioturbated, white, carbonate-rich (>80% CaCO<sub>3</sub>) interbeds in both the eastern and western basins of the North Atlantic suggest that they contain minor amounts of relatively unweathered eolian dust derived from northwest Africa during dry intervals.

### INTRODUCTION

Most of the Jurassic to Pleistocene sections recovered at Deep Sea Drilling Project (DSDP) sites in the North Atlantic consist of cyclically interbedded strata in which the main differences between the interbeds are color, the amount of organic matter, and (or), for carbonate strata, the amount of CaCO<sub>3</sub> (Dean and others, 1978; Jansa and others, 1978 and

1979; McCave 1979a; Dean and Gardner, 1982; Arthur and Dean, 1986; Dean and Arthur, 1987). The primary causes of the cyclicity were variations in one or more of the following variables: the position of the calcite compensation depth (CCD), influx of terrigenous clastic material, influx of terrestrial and marine organic matter, and redox conditions in the bottom waters and sediments. The ultimate driving force for the cyclic depositional patterns, however, probably was external to the depositional basins and related to cyclic variations in global climate. Because estimates of the periodicities of these cyclic interbeds generally fall within the range of 20–100 ky (e.g., Arthur and Fischer, 1977; Dean and others, 1978; Arthur, 1979; Arthur and Natland, 1979; McCave, 1979a; Dean and Gardner, 1982; Robertson and Bliefnick, 1983; Arthur and others, 1984; Cotillon, 1984; Cotillon and Rio, 1984; Arthur and Dean, 1991), it has been suggested that this cyclicity may be caused by cyclic variations in one or more climate-related forcing mechanism that, in turn, are related to changes in the Earth's orbital parameters (so called Milankovitch cycles) that have periodicities within the same range (e.g., Schwarzscher and Fischer, 1982; Arthur and others, 1984; ROCC Group, 1986). Arthur and others (1984) suggested that periodic changes in insolation, evaporation, wind stress, and (or) rainfall in a wide variety of environments caused changes in the input of terrigenous detritus, water-mass stratification, surface productivity, deep-water oxygen content, and rates of carbonate dissolution. Superimposed on these cyclic patterns, and sometimes accentuating them, are strata of reworked material introduced by slumping, turbidity currents, and debris flows (Dean and others, 1978; Arthur and von Rad, 1979; McCave, 1979 a and b; Kelts and Arthur, 1981; Summerhayes, 1981; Dean and Gardner, 1982; Summerhayes and Masran, 1983; Robertson and Bliefnick, 1983; Arthur, Dean, and Stow, 1984; Sarti and von Rad, 1987). The cyclicity in the supply and (or) preservation of organic matter and CaCO<sub>3</sub> had a pronounced influence on subsequent diagenetic events and on geochemical partitioning in adjacent beds (Arthur and Dean, 1991; Frank and others, this volume).

The most striking of the cyclic depositional sequences are the interbeds of dark olive to black claystone (<30% CaCO<sub>3</sub>) or marlstone (30–60% CaCO<sub>3</sub>), and white to light-gray limestone (>60% CaCO<sub>3</sub>) (Fig. 1) of Neocomian age (Berriasian, Valanginian, Hauterivian, and Barremian Stages of the Lower Cretaceous). This sequence has been recovered at DSDP Sites 101, 105, 367, 387, 391, 534, and 603 (Fig. 2) in the North Atlantic, and at Sites 535 and 540 in the Gulf of Mexico (Büfler, Schlager, and others, 1984). This unit has been called the Blake-Bahama Formation by Jansa and others (1979), and apparently is equivalent to the upper Tithonian (latest Jurassic to Aptian (mid-Cretaceous) Maïolica Formation of Garrison (1967) that is exposed in the Tethyan regions of the Mediterranean (Bernoulli, 1972; Jan-

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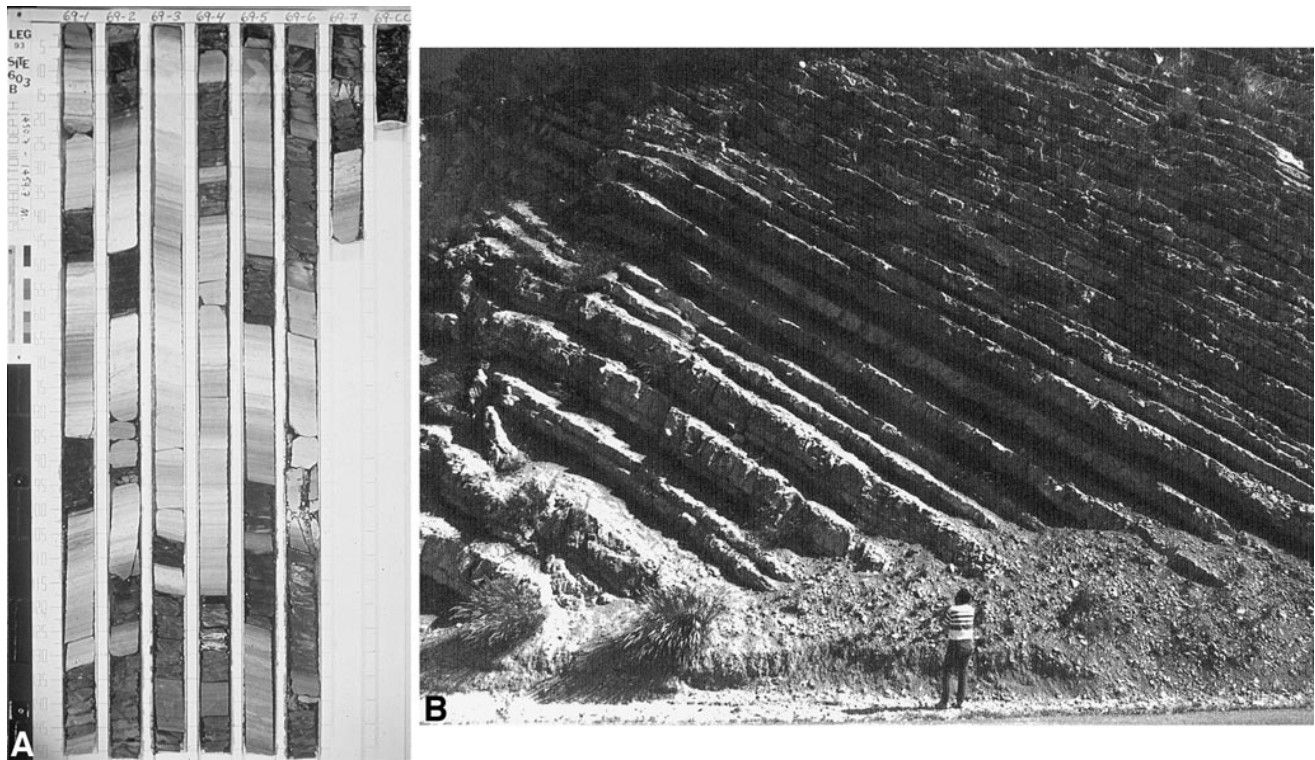


FIGURE 1. (A) Typical interbedded limestones (light) and marlstones (dark) in Neocomian sections from DSDP Hole 603B, Core 69. (B) Outcrop of Neocomian limestone/marlstone cycles in southern France.

sa and others, 1979), and to thick limestone sequences of the Vocontian trough in France (Cotillon, 1984; Cotillon and Rio, 1984; Fig. 1B). This lithologic unit marks the widespread deposition of biogenic carbonate over much of the North Atlantic and Tethyan seafloor during a time of overall low sealevel and a deep CCD (Tucholke and Vogt, 1979; Thierstein, 1979; Arthur and Dean, 1986). An abrupt rise of the CCD in the Barremian-Aptian marked the end of major pelagic carbonate deposition in the deep sea during the Cretaceous and resulted in the widespread deposition of non-

calcareous black shales of the Hatteras Formation (Jansa and others, 1979; Arthur and Dean, 1986; Dean and Arthur, 1987) over much of the deep North Atlantic.

Much of the Blake-Bahama Formation consists of interbedded white to light gray limestones and dark-olive to black marlstones or claystones (Figs. 1 and 3). The carbonate-rich beds consist of two distinct types: (1) bioturbated white to light gray limestone, and (2) finely to coarsely laminated, streaked or microburrowed, medium brown to light gray limestone or marlstone (Dean and Arthur, 1987). In most sections, beds of laminated limestone are more abundant than beds of bioturbated limestone (Fig. 1A). In the western North Atlantic Basin, the interbedded carbonate-poor beds usually are massive, dark brown to black claystone or calcareous claystone that average about 2.35% organic carbon (OC) on a carbonate-free basis (maximum, 3.2%  $n = 27$ ; Dean and Arthur, 1987). In DSDP Hole 367 in the eastern North Atlantic Basin, the dark colored beds are finely laminated, and contain more carbonate (average of about 61% as  $\text{CaCO}_3$ ;  $n = 53$ ) and OC (average of about 6.5% on a carbonate-free basis; Dean and Gardner, 1982). The distinct microlaminations in the dark beds in the eastern North Atlantic Basin suggest that these beds were deposited under anoxic or near-anoxic conditions without burrowing organisms. The laminae in many of the limestone and marlstone beds, however, typically are thick and indistinct, and it is not clear whether or not they indicate deposition under anoxic bottom water as commonly has been assumed. The origin of the laminated Neocomian carbonates with high OC contents and accumulation rates (e.g., Arthur and Dean, 1986 and 1991) remains an important question. Of equal

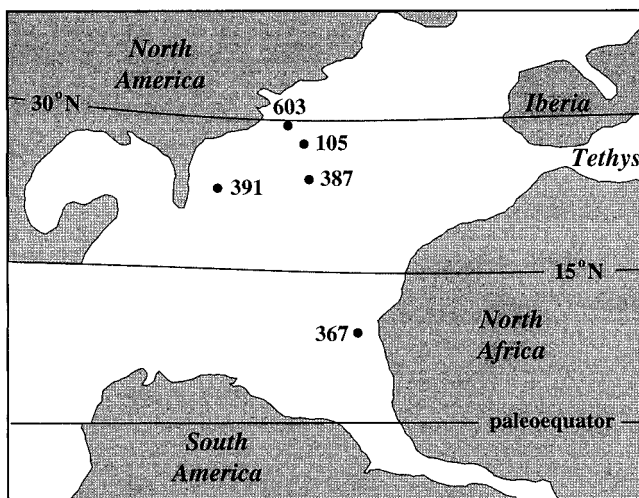


FIGURE 2. Index map showing locations of DSDP sites in the North Atlantic discussed in this report plotted on a continental reconstruction of 100 million years ago.



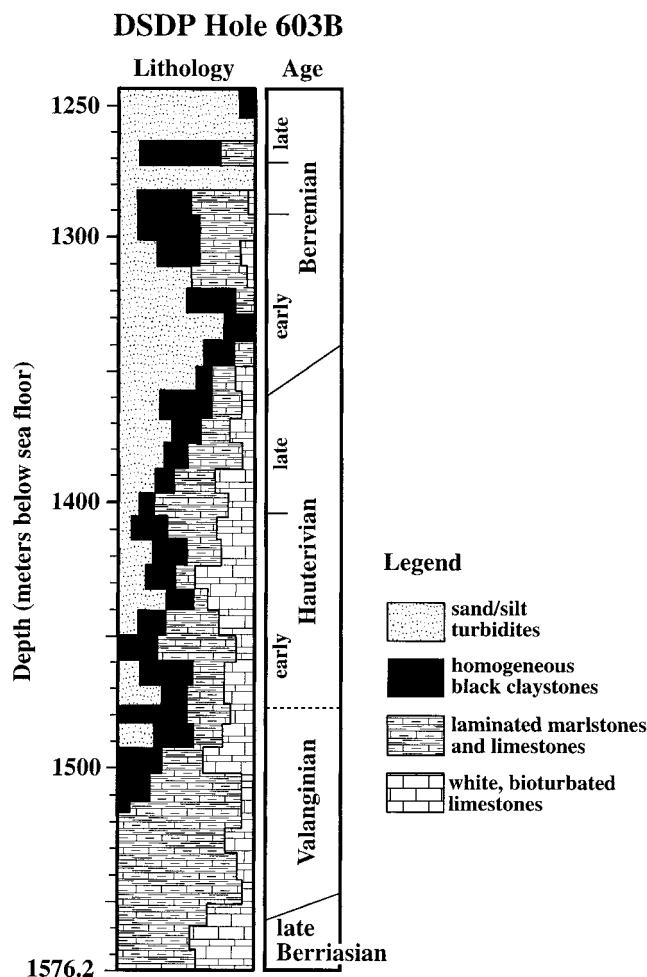


FIGURE 3. Age and lithology of the Blake-Bahama Formation recovered in DSDP Hole 603B.

importance is whether the change from limestone deposition during the Neocomian to black shale deposition of the Hatteras Formation of Aptian to Turonian age was simply due to a shoaling of the CCD (e.g., Thierstein 1979; Tucholke and Vogt, 1979), or to a more complex set of factors such as changes in climate, sediment inputs, carbonate productivity, and (or) bottom-water circulation during the middle Cretaceous.

This study is part of our on-going investigation of cyclic sedimentation in the oceans through time, particularly cycles of OC accumulation in Mesozoic strata in the Atlantic, Tethyan, and Western Interior Basins. For this investigation, we have used measurements of type, abundance, and isotopic composition of organic matter, and elemental concentrations in Neocomian sequences from the eastern and western basins of the North Atlantic. The data sets for this investigation were collected over a period of more than 12 years. The DSDP holes for which we have the most data are Hole 367 in the Cape Verde Basin off northwest Africa, and Hole 603B in the North American Basin off eastern North America (Fig. 1). A large number of samples of Neocomian carbonates and other lithologic units in Hole 603B were collected during DSDP Leg 93 in the North American Basin, and geochemical results and preliminary interpretations are

presented in the Leg 93 Initial Reports volume (Dean and Arthur, 1987). For that study, the Neocomian carbonates in Hole 603B were grouped into homogeneous, black claystones, bioturbated, white limestones, and transitional laminated limestones (Fig. 3), and those groupings will be retained in this paper. Results of analyses of Neocomian carbonates from DSDP Holes 367 and 603B indicated that there is a marked difference in the composition of these cyclic carbonate units that otherwise have similar appearances. This prompted us to obtain additional samples from those two holes, and from Holes 105, 387, and 391C in the western North Atlantic Basin (Fig. 2).

We thought it would be a fitting tribute to Bill Sliter and the many contributions he has made to the study of Cretaceous marine carbonate rocks to include results and interpretation of analyses of all of these samples in this volume. In particular, we address the issue of possible variation in deep-water oxygen concentration and its role, if any, in sediment geochemical variations. Bill Sliter, of course, loved to debate the merits of "oceanic anoxic events" as causes of enrichments in sedimentary organic carbon.

## METHODS

Total carbon and inorganic (carbonate) carbon in most samples were determined by coulometry (Engleman and others, 1985). Values of organic carbon (OC) were determined by difference between total carbon and carbonate carbon. Percent  $\text{CaCO}_3$  was calculated by dividing percent carbonate carbon by 0.12, which is the fraction of carbon in  $\text{CaCO}_3$ . Total carbon and organic carbon on samples collected prior to 1985 were determined with a LECO carbon analyzer. The Rock-Eval pyrolysis method (discussed below) also measures total organic carbon. Percent  $\text{CaCO}_3$  also was calculated by assuming that all of the calcium in a whole-rock sample was from  $\text{CaCO}_3$ . For data sets for which we have values of percent  $\text{CaCO}_3$  calculated by both methods (most), there is usually excellent agreement between the two methods. Percentages of organic carbon and total nitrogen were measured on some samples using a Carlo Erba CHN analyzer. See Arthur and others (1987) for a comparison of methods of carbon analyses by different methods on samples from DSDP Hole 603B.

Pyrolysis of organic matter was performed using a Rock-Eval pyrolysis instrument for comparison with similar analyses commonly used in the study of ancient black shales. The Rock-Eval pyrolysis method provides a rapid estimate of the hydrocarbon-generation potential, and the abundance and type of organic matter in sediments and sedimentary rocks (Tissot and Welte, 1984; Peters, 1986). The yield of free or adsorbed hydrocarbons is determined by heating the sample in flowing helium to  $250^\circ\text{C}$ , and is recorded as the first characteristic peak on a pyrogram ( $S_1$ , mg HC/g sample). The second peak is composed of hydrocarbons generated by thermal breakdown of kerogen as the sample is heated from  $250^\circ\text{C}$  to  $550^\circ\text{C}$  ( $S_2$ , mg HC/g sample).  $\text{CO}_2$  also is generated by kerogen decomposition and is retained during the heating interval from  $250^\circ\text{C}$  to  $390^\circ\text{C}$  and analyzed as the integral of the third peak on the pyrogram ( $S_3$ , mg  $\text{CO}_2$ /g sample). Our data are expressed in terms of a hydrogen index (HI), in which the  $S_2$  peak is normalized to

the OC content of the sample (mg HC/g OC), and oxygen index (OI), which is the  $S_3$  peak normalized to the OC content of the sample (mg  $CO_2$ /g OC).

Stable-carbon isotope ratios in organic matter were determined using standard techniques (Pratt and Threlkeld, 1984; Dean and others, 1986). Acidified samples were combusted at 1,000°C under oxygen pressure in an induction furnace. The resulting  $CO_2$  was dehydrated and purified in a high-vacuum, gas-transfer system, and isotope ratios were determined using an isotope-ratio mass spectrometer. Results are reported in the standard per mil (‰)  $\delta$ -notation relative to the University of Chicago Pee Dee belemnite (PDB) marine-carbonate standard,

$$\delta^{13}C \text{ ‰} = [(R_{\text{sample}}/R_{\text{PDB}}) - 1] \times 10^3$$

where R is the ratio ( $^{13}C : ^{12}C$ ).

Samples for inorganic geochemical analysis were oven dried at 90°C, then ground in a ceramic mill to pass a 100 mesh (149  $\mu$ m) sieve. Concentrations of 10 major elements (Si, Al, Fe, Mg, Ca, Na, K, Ti, P, and Mn) were determined by wavelength-dispersive X-ray fluorescence spectrometry (XRF) in the U.S. Geological Survey Analytical Laboratories in Denver (Taggart and others, 1987). The same samples were analyzed for 40 major and trace elements by induction-coupled, argon-plasma emission spectrometry (ICP; Lichte and others, 1987). Splits of samples for XRF were homogenized by igniting the sample at 925°C to obtain loss on ignition (LOI), and then fusing the ignited sample into a glass disc with lithium tetraborate (Taggart and others, 1987). Splits of samples for ICP were digested in HF, aqua regia, and  $HClO_4$ , and the resulting solution taken to dryness on a hotplate. The salts were then dissolved in aqua regia for analysis (Lichte and others, 1987). There is excellent agreement between concentrations of major elements determined by XRF and by ICP, with correlation coefficients usually higher than 0.97 (Fig. 4). An extensive analysis of precision and accuracy of determination of major and trace elements by different methods within USGS laboratories, and between USGS laboratories and three other laboratories, is reported for samples of the Monterey Formation by Piper and Isaacs (1995). In general, the precision, determined by analyzing standards and duplicate samples, is better than 10% at a concentration of 10 times the limit of detection. XRF is applicable to a wide variety of sample types, and usually provides higher precision and accuracy than alternative methods for the determination of the major elements (Taggart and others, 1987). After many analyses of standards and duplicate samples over a period of more than 20 years, we feel comfortable reporting results of XRF analyses to three significant figures, and results of ICP analyses to two significant figures. We will use the results of major-element analyses by XRF in this paper. The trace-element results are not the focus of this paper, but they are presented and discussed for Hole 367 by Dean and Gardner (1982) and Dean and Arthur (1987), and for Hole 603B by Dean and Arthur, 1987).

The geochemical data described in this paper and in Dean and others (1987) are available in digital form at the World Data Center-A for Paleoclimatology, NOAA/NGDC, 325 Broadway, Boulder, CO 80303 (phone: 303-497-6280; fax: 303-497-6513), or on the Internet as NOAA/NGDC Paleo-

climatology Data Contribution Series # 1999-031 (<http://www.ngdc.noaa.gov/paleo/contrib-list.html>).

## RESULTS AND INTERPRETATION

### CHARACTERIZATION OF ORGANIC MATTER

#### *Organic Carbon Contents*

Average whole-rock values of %  $CaCO_3$ , % OC, Rock-Eval  $S_2$ , HI, and OI, and  $\delta^{13}C$  for Neocomian carbonate samples from DSDP Holes 105, 367, 387, 391C, and 603B are given in Table 1. Figure 5 illustrates the differences in content of  $CaCO_3$  and OC in the three types of carbonate lithologies in DSDP Hole 603B. Note the distinct cluster of black claystones with relatively high OC and low  $CaCO_3$  values. The white bioturbated limestones fall within an even tighter cluster with low OC and high  $CaCO_3$  values. The transitional laminated carbonates have OC and  $CaCO_3$  values that are intermediate between the other two lithologic types, but closer to, and overlapping with, the bioturbated limestones. Variations in concentrations of  $CaCO_3$  and OC in samples from Holes 105, 387, and 391C from the western North Atlantic Basin, and Hole 367 from the Cape Verde Basin in the eastern North Atlantic are shown in Figure 6. Neocomian carbonates at other sites (105, 387, and 391) in the western North Atlantic Basin (Fig. 6A) have characteristics like those at Site 603, including a tight cluster of high carbonate, low-OC bioturbated limestones, relatively OC-rich marlstones, and limestones and marlstones with intermediate concentrations of both  $CaCO_3$  and OC. Overall, the Neocomian lithologies in the eastern North Atlantic in Hole 367 (Fig. 6B) have higher concentrations of both  $CaCO_3$  and OC than in equivalent lithologies in the western North Atlantic.

#### *Rock-Eval Pyrolysis Results*

Results of Rock-Eval pyrolysis usually are displayed as plots of OI versus HI (Fig. 7) to define types, source, and preservation of kerogen in rocks. Three types of kerogen (I, II, and III) and their thermal maturation pathways are shown by the curves in Figure 7. Type I kerogen consists of lipid-rich, sapropelic, algal organic compounds typified by the most OC-rich Eocene Green River oil shale (Dean and Anders, 1991). Type II kerogen also consists of H-rich compounds such as those found in autochthonous marine organic matter. Marine hydrocarbon source rocks typically contain type II kerogen, as does most of the lacustrine Green River oil shale. Type III kerogen is the other extreme and consists of H-poor organic compounds such as those found in terrestrial organic matter. Type III kerogen also may be formed by oxidation of type II kerogen, either during deposition and early burial or by reworking, lowering the HI and increasing the OI. Therefore, the extreme range of OI values shown for all data sets shown in Figure 7 could result from a mixture of marine and terrestrial organic matter or from oxidation of marine organic matter in the depositional environment. Another possibility is that some of the  $CO_2$  measured in the  $S_3$  peak from which the OI is derived could have come from  $CaCO_3$  as suggested by Katz (1983), although

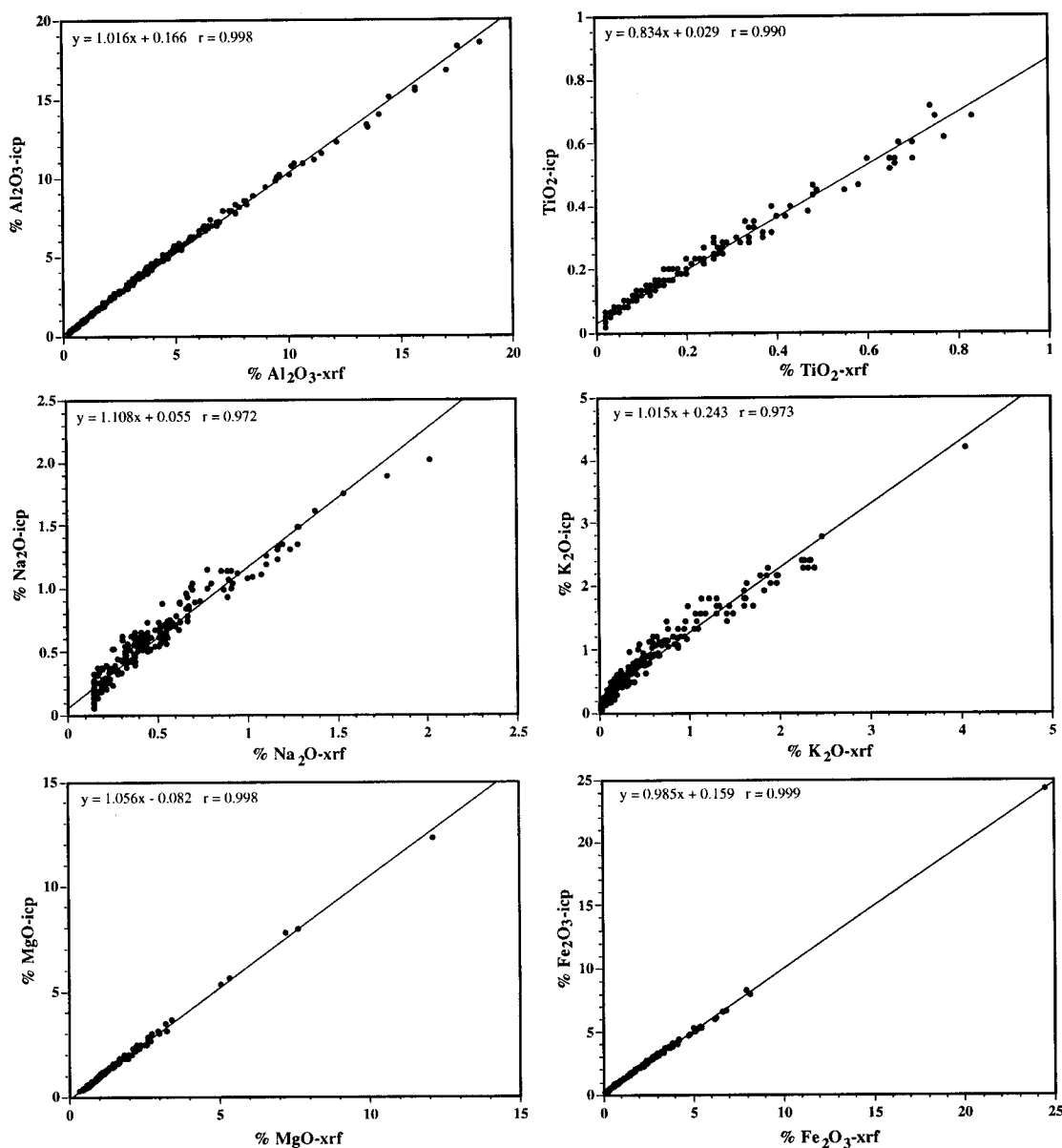


FIGURE 4. Scatter plots comparing concentrations of major-element oxides by XRF and ICP. The solid diagonal line in each plot is the linear regression line, and the equation of each line is given in the upper left of each plot.

the temperature of pyrolysis (550° C) is considerably lower than the thermal dissociation temperature of CaCO<sub>3</sub> (900° C). Figure 7 shows that the Rock-Eval hydrogen indices of all samples analyzed from the western North Atlantic Basin are low (mostly less than about 200 mg HC/g OC; Figs. 7A and B), compared with black marlstones and limestones from Hole 367 (Fig. 7C).

Because the HI is derived from S<sub>2</sub> by normalizing to % OC (HI = [S<sub>2</sub>/%OC] × 100), Langford and Blanc-Valleron (1990) suggested that the average value for HI for a particular data set could be obtained from the slope of a regression equation for a graph of % OC versus S<sub>2</sub> (Fig. 8). The data plotted for samples from the western North Atlantic all have low slope coefficients (0.39 to 2.41; Figs. 8A and B), and the black claystones from Hole 603B have the lowest. Multiplying these slope coefficients by 100 (Langford and

Blanc-Valleron, 1990) yields average values of HI ranging from 39 to 241. In contrast, the slope coefficient for black marlstones and limestones from Hole 367 is much higher (4.55; Fig. 8C), yielding an average value of HI of 455. Langford and Blanc-Valleron (1990) further suggest that lines with zero intercept and slopes of 7 (HI = 700) and 2 (HI = 200) separate the pyrolytic hydrocarbon yields of type I and type II, and type II and type III kerogen, respectively (heavy dashed lines in Fig. 8). From Figures 7 and 8 one can see that only samples from Hole 367 contain predominantly type II kerogen; kerogen in most samples from the western North Atlantic is type III. The magnitude of the positive OC intercept is a measure of the amount of highly refractory organic matter. The plots in Figure 8 suggest that the amount of refractory organic matter is fairly minor except in samples from Hole 603B.

TABLE 1. Average values of selected geochemical parameters in Neocomian carbonates on either a carbonate-free (carb.-free) basis or a bulk-rock basis. [OC, organic carbon; S<sub>2</sub>, Rock-Eval pyrolysis S<sub>2</sub> peak; H-Index, hydrogen index; O-index, oxygen index; n.a., not analyzed]

Variable	Hole							
	603B black	603B trans.	603B white	105	387	391C	367 black	367 white
Average carb.-free								
% Si <sub>2</sub> O <sub>3</sub>	53.2	47.1	53.9	57.9	78.7	54.1	52.2	64.6
% Al <sub>2</sub> O <sub>3</sub>	18.7	13.0	13.1	13.1	12.5	12.6	11.5	9.3
% Fe <sub>2</sub> O <sub>3</sub>	6.8	8.9	7.4	5.3	13.6	6.8	5.1	4.1
% Na <sub>2</sub> O	1.39	1.55	1.40	1.93	3.00	1.77	1.08	1.44
% K <sub>2</sub> O	3.05	1.76	1.48	1.29	1.77	1.69	1.88	0.79
% TiO <sub>2</sub>	0.83	0.45	0.38	0.34	0.48	0.52	0.47	0.27
% P <sub>2</sub> O <sub>5</sub>	0.10	0.42	0.39	0.27	0.94	0.26	0.28	0.50
% MnO	0.03	1.14	1.71	0.38	0.72	0.24	0.14	0.75
% OC	2.35	3.95	1.11	2.40	4.26	2.33	6.46	1.65
% total N	0.08	0.14	0.04	n.a.	n.a.	0.11	n.a.	n.a.
Average bulk sample								
% CaCO <sub>3</sub>	17.0	76.5	83.4	73.9	78.9	66.6	61.2	82.5
% MgO	2.18	1.05	0.78	0.92	2.23	1.44	1.19	0.77
% OC	1.89	1.04	0.23	0.66	0.76	0.78	2.35	0.2
% total N	0.07	0.04	0.01	n.a.	n.a.	0.02	n.a.	n.a.
S <sub>2</sub>	0.70	1.50	0.00	1.18	1.61	1.37	8.11	0.41
H-index	35	86	7	134	186	116	332	172
O-index	64	98	232	124	475	335	151	658
δ <sup>13</sup> C	-25.3	-26.0	-25.0	n.a.	n.a.	n.a.	-28.1	-27.2

### Carbon Isotopic Composition

The stable carbon-isotope composition of OC often has been interpreted in terms of the source of the organic matter. The basis for interpreting the isotopic composition of OC as a source signal in recent marine sediments is the observation that modern terrestrial vegetation is depleted in <sup>13</sup>C relative to modern marine plankton, and generally has δ<sup>13</sup>C values that are about 5–6‰ lighter (more negative) than those of marine plankton (e.g., Deines, 1980). Most organic matter in modern marine sediments, judged by other geochemical and optical criteria to be mainly marine, has δ<sup>13</sup>C values similar to those of marine plankton and particulate organic matter (about -21 ‰). Simoneit (1986) observed that values of δ<sup>13</sup>C of organic matter in most Cretaceous strata from Atlantic DSDP sites are in the range of -24 to -29‰. From this he concluded that "the organic matter of most of the samples throughout the Cretaceous is of terrestrial origin", even though these samples had atomic H/C ratios >1.2 (equivalent to an HI of about 450), which is typical of marine type II kerogen (e.g., Katz and Pheifer, 1986). Figure 9 shows the same relation that Simoneit (1986) observed, that is the more terrestrial, H-depleted organic matter in samples from Hole 603B is isotopically heavier than the more marine, more H-rich organic matter in Hole 367. In fact, the organic matter in the most OC-rich black marlstone beds at Site 367 is most enriched in hydrogen, and is isotopically lightest, whereas the organic matter in the black claystone beds in Hole 603B is the least enriched in hydrogen and is isotopically heaviest.

The δ<sup>13</sup>C values of the OC in samples from Hole 367 (Fig. 9) are similar to those from other Cretaceous marine OC-rich strata (Dean and others, 1986; Simoneit, 1986). The δ<sup>13</sup>C values of the OC in Hole 603B, particularly in the black claystones, are similar to those of other Cretaceous terrestrial OC-rich strata such as coals (-24 to -26‰ Dean and Arthur, 1987; Holmes, 1995). Arthur and others (1985)

and Dean and others (1986) concluded that the isotopic composition of Cretaceous organic matter was indeed recording a source signal, but that Cretaceous marine photosynthesis resulted in greater isotopic fractionation, possibly because of higher CO<sub>2</sub> concentrations in the atmosphere and oceans. Why Cretaceous terrestrial plants did not fractionate carbon isotopes more is not known, but is probably related to differences in physiology between modern and Cretaceous plants (e.g. stomatal frequency, electron transport, etc.; e.g., Luo and others, 1996). The observations are that Cretaceous marine OC had δ<sup>13</sup>C values of about -27 to -29‰, and Cretaceous terrestrial OC had δ<sup>13</sup>C values of about -24 to -26‰. This conclusion is illustrated particularly well in the comparison of δ<sup>13</sup>C and HI values in samples from Holes 603B and 367 in Figure 9. As discussed earlier, values of δ<sup>13</sup>C in samples from Hole 367 are typical of Cretaceous marine organic matter, and those in samples of black claystone from Hole 603B are typical of Cretaceous terrestrial organic matter.

### Nitrogen Isotopic Composition

The nitrogen isotopic composition of the organic matter in the Neocomian samples also indicates that fractionation of nitrogen isotopes by Early Cretaceous marine organisms, like the fractionation of carbon isotopes, was very different than fractionation by modern marine plankton (Fig. 10; Rau and others, 1987). The ellipse in Figure 10 represents an envelope around 55 values of δ<sup>13</sup>C and δ<sup>15</sup>N in near-shore marine samples from the Bering Sea and eastern North Pacific Ocean reported by Peters and others (1978), and the line is the linear regression line through those points (correlation coefficient of 0.81). Samples from the more marine end of the ellipse are from the Bering Sea, and Tanner, Santa Catalina, and Santa Barbara Basins, southern California Borderland. Samples from the more terrestrial end are from the Gulf of Alaska, San Pedro shelf, and Bandaras Bay



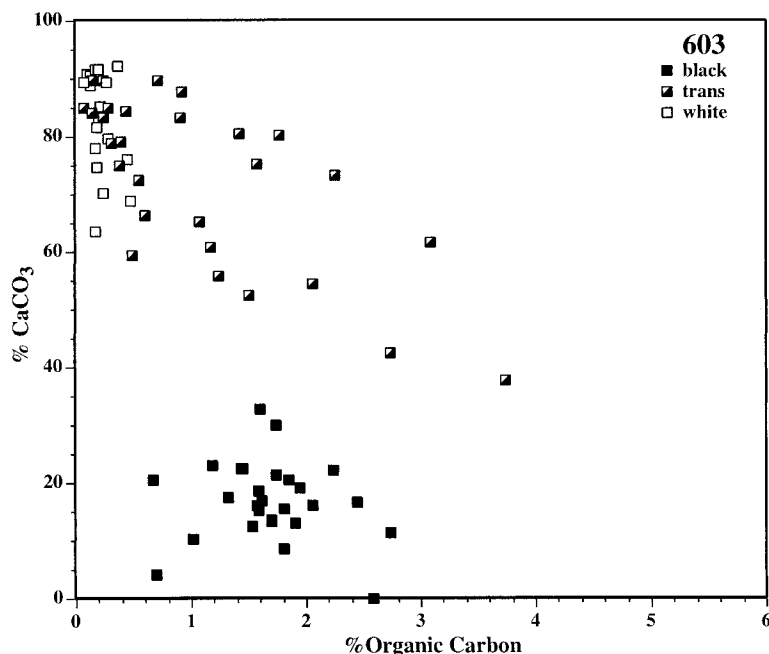


FIGURE 5. Cross plot of percent organic carbon versus percent  $\text{CaCO}_3$  in samples of Neocomian black claystones, white bioturbated limestones, and transitional (trans.) marlstones and limestones from DSDP Hole 603B.

(Puerto Vallarta), Mexico. Peters and others (1978) concluded that mixing of organic matter from terrestrial and marine sources was the dominant process determining the carbon and nitrogen isotopic composition of coastal marine sediments. Values of  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  in the more terrestrial, H-depleted organic matter in the black claystones from Hole 603B, are similar to those in the more terrestrial samples in the Peters and others data. As discussed earlier, values of  $\delta^{13}\text{C}$  in samples from Hole 367 are typical of Cretaceous marine organic matter, and those in samples from Hole 603B, particularly samples of black claystone, are typical of Cretaceous terrestrial organic matter. Values of  $\delta^{15}\text{N}$  for most Cretaceous samples also are considerably lower than those in any organic matter in the modern nitrogen cycle except for nitrogen fixing organisms (Rau and others, 1987), and the values in samples of black marlstones and limestones from Hole 367, the lithologies suspected of containing the most marine organic matter, are the lowest. Such low values of  $\delta^{15}\text{N}$  prompted Rau and others (1987) to suggest that the organic matter in some Cretaceous OC-rich strata may have been produced by nitrogen-fixing phytoplankton that proliferated in phosphate-rich, nitrate-poor surface waters overlying anoxic deepwater in the eastern North Atlantic.

#### Carbon/Nitrogen Ratios

The ratio of organic carbon to total nitrogen (C/N) also has been used as an indicator of the source of organic matter in modern sediments (e.g., Walsh and others, 1985). Terrestrial organic matter tends to be depleted in proteinaceous, N-rich compounds and enriched in humic, C-rich compounds. Consequently, the C/N ratio of terrestrial organic matter tends to be high (ca. 14–30; Müller, 1977; Meyers and others, 1984; Meyers, 1987) compared to that of freshly

deposited, lipid-rich, autochthonous marine organic matter (C/N ratio <10). Usually such analyses are performed on whole-rock samples rather than on kerogen separates. However, interpretations based on C/N ratios determined in this way may be misleading because of selective consumption of N-bearing organic compounds during the decomposition of organic matter, and because a substantial portion of the nitrogen in some samples may be adsorbed inorganic nitrogen (e.g., Waples, 1985). This adsorbed nitrogen usually is in the form of  $\text{NH}_3$  or  $\text{NH}_4^+$  fixed on clays. Consequently, the most serious errors in using C/N ratios as indicators of organic matter source would occur in clay-rich, organic-poor sediments. The fact that the C/N ratios of all Neocomian lithologies from Hole 603B, regardless of clay content, are close to 30 (Fig. 11A) suggests that source of this nitrogen was mainly terrestrial organic matter. The C and N values in samples from Hole 391C show considerably more scatter (Fig. 11B). Most samples have C/N ratios of <10, which suggests that the source of this organic matter was marine.

The pyrolysis results (Figs. 7 and 8), carbon isotope results (Figs. 9 and 10), and C/N ratios (Fig. 11) all suggest that there were differences in the sources and degree of preservation of organic matter in the Neocomian carbonates in the eastern and western basins of the North Atlantic during the Early Cretaceous. The bioturbated white limestones in Hole 603B probably contain a background hemipelagic "rain" of organic matter consisting of a mixture of terrestrial and highly degraded marine organic matter. This is indicated by their low HI and  $\text{S}_2$  values (Figs. 7 and 8), and values of  $\delta^{13}\text{C}$  that are higher than those in samples from the more-marine Neocomian OC-rich strata in Hole 367 (Fig. 9). The only significant amounts of marine organic matter in samples from Hole 603B are preserved in some



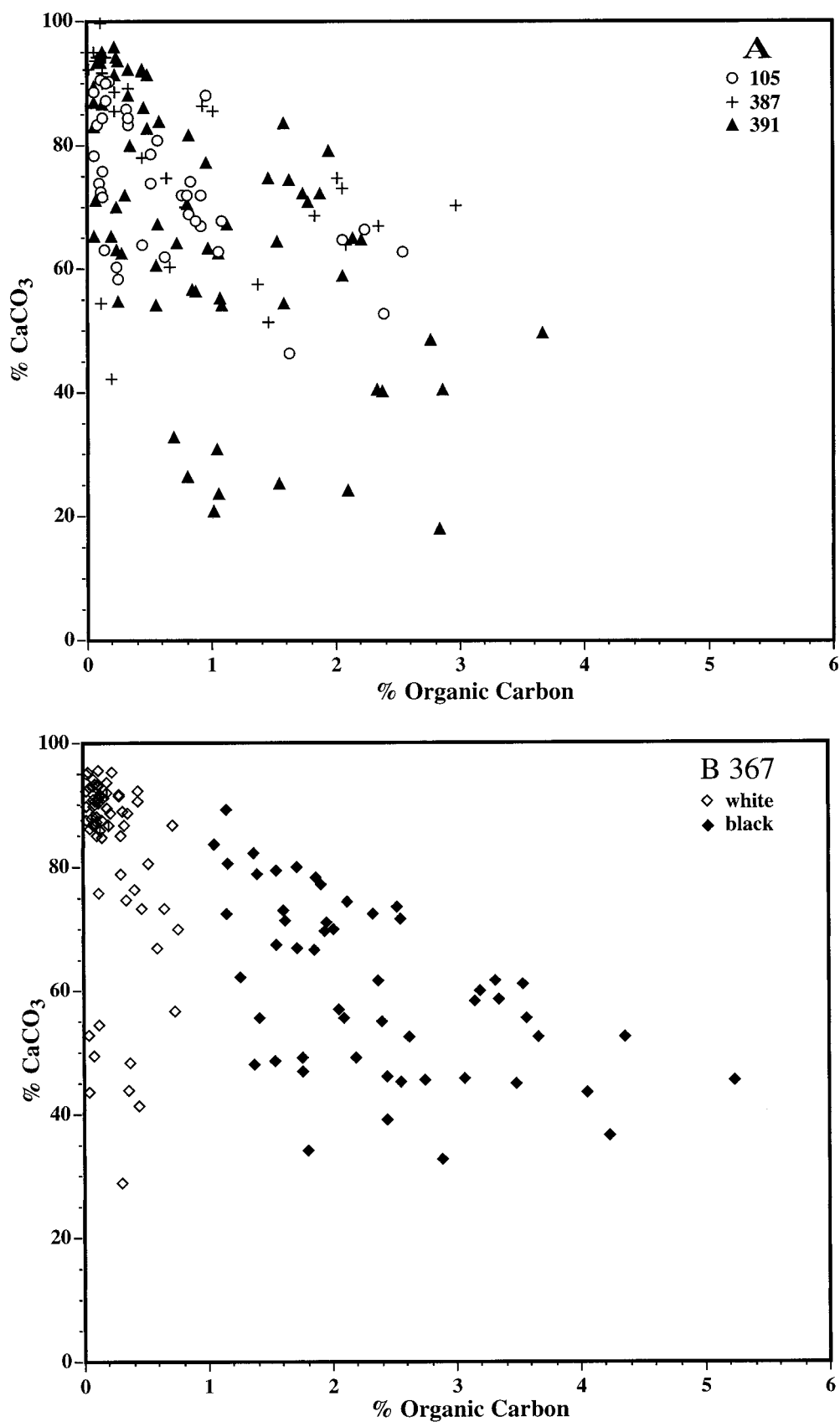
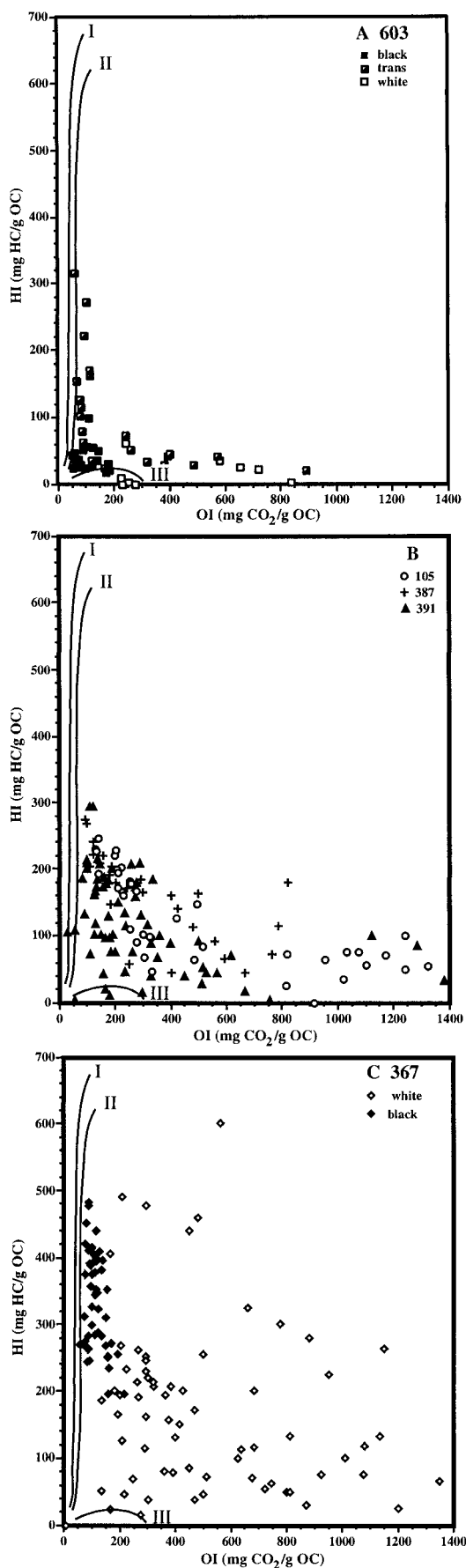


FIGURE 6. Cross plots of percent organic carbon versus percent  $\text{CaCO}_3$  in samples of Neocomian lithologies from DSDP Holes 105, 387, and 391C (A) and 367 (B).



marlstones and limestones with higher values of HI and S<sub>2</sub>, and higher concentrations of OC (Figs. 7 and 8). The limestones and marlstones in Hole 367 also contain mixed terrestrial and marine organic matter with a much larger proportion of marine organic matter than their equivalent lithologies in Hole 603B as indicated by higher values of HI and S<sub>2</sub> (Figs. 7 and 8) and lower values of δ<sup>13</sup>C (Fig. 9).

#### GEOCHEMICAL CHARACTERISTICS OF DETRITAL CLASTIC SOURCES

A Q-mode factor analysis of inorganic geochemical data from Hole 603B (Dean and Arthur, 1987) showed that there was a distinct difference between the black claystones and the carbonate lithologies. In particular, the black claystones had distinctly higher concentrations of Al<sub>2</sub>O<sub>3</sub>, K<sub>2</sub>O, and TiO<sub>2</sub>, and lower concentrations of MnO, all on a carbonate-free basis (Table 1). This suggests that there may have been a different source of detrital clastic material during deposition of the carbonate lithologies than during deposition of the black claystones, which, we assume, derived their detrital clastic material from the North American continent. Although both Al<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> are higher in the black claystones, the difference is greatest for TiO<sub>2</sub> which results in a lower Al<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub> ratio (Fig. 12A). Low-carbonate samples from all sites have a background Al<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub> ratio of about 20, which is about the value of modern deep-sea red clay from the Hatteras abyssal plain (Dean and Arthur, 1987) and average North Atlantic dust (Prospero, 1982). The Al<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub> ratio becomes significantly higher in limestones with >80% CaCO<sub>3</sub> from all sites in both the eastern and western North Atlantic Basins (Figs. 12A, B and C). At first we were concerned that this might be an analytical artifact, but analyses of hundreds of other limestone samples using the same techniques in the same laboratories, such as samples from the Greenhorn and Niobrara Formations of the Cretaceous Western Interior Seaway of North America, do not show the same relations (Arthur and Dean, 1991; Dean and Arthur, 1998). Also, the close agreement between Al<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> determined by both XRF and ICP (Fig. 4) does not suggest that there is an analytical problem. There are very few other published analyses of Neocomian carbonates, particularly limestones, that we are aware of. Chamley and others, 1983, and Kegami and others, 1983 present XRF analyses of major-element oxides in samples from the Blake-Bahama Formation in Hole 534A in the Blake-Bahama Basin off Florida. Most samples had Al<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub> ratios in the 20s which is what we would expect for a site so close to North America. However, one limestone sample (80% CaCO<sub>3</sub>) reported by Chamley and others (1983) had an Al<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub> ratio of 46, and another (74% CaCO<sub>3</sub>) had an Al<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub> ratio of 70. A few analyses of samples from Holes 105 and 387 are reported by Donnelly (1980). Values of Al<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub> in samples of dark colored marlstones from

←

FIGURE 7. Van Krevelen-type plots of Rock-Eval pyrolysis oxygen index versus hydrogen index for samples from DSDP Holes 603B (A), 105, 387, and 391C (B), and 367 (C). Roman numerals I, II, and III refer to the thermal maturation pathways for type I, II, and III kerogen, respectively. See text for descriptions of kerogen types.

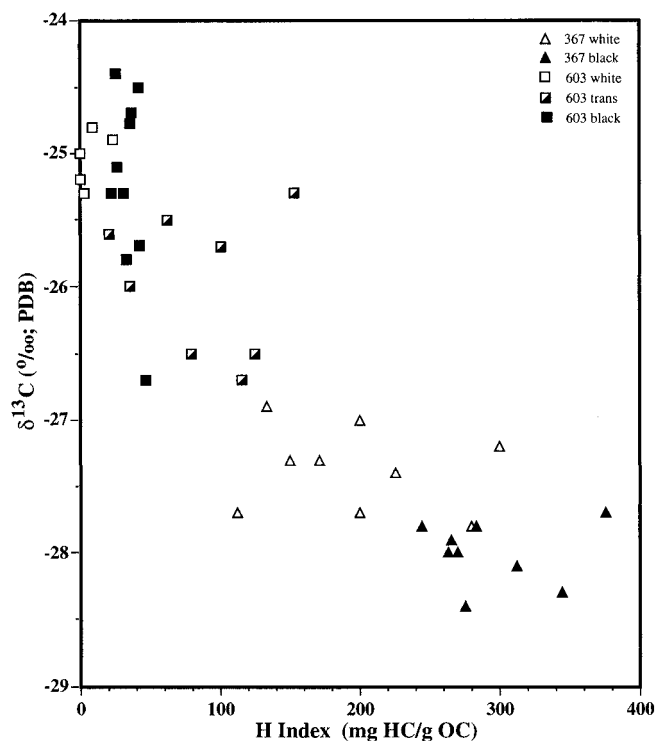
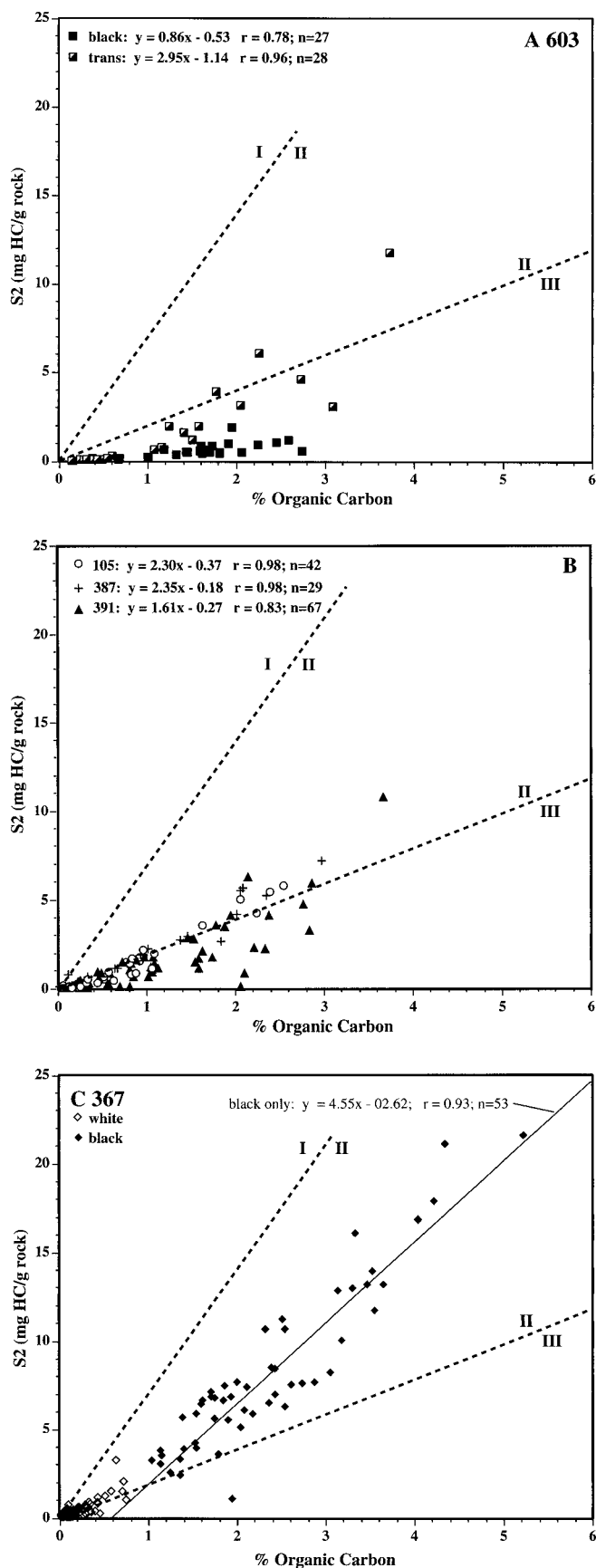


FIGURE 9. Cross plot of Rock-Eval pyrolysis hydrogen index versus  $\delta^{13}\text{C}$  of organic carbon in samples from DSDP Holes 367 and 603B.

both holes are about 25. Values in samples of light colored limestones from Hole 105 are only slightly elevated (average of 31), but the one sample of limestone (97%  $\text{CaCO}_3$ ) from Hole 387 has an  $\text{Al}_2\text{O}_3/\text{TiO}_2$  ratio of 155. We believe that the high-carbonate samples do indeed have elevated values of  $\text{Al}_2\text{O}_3$ , and, particularly,  $\text{TiO}_2$ , are near the detection limit, we do not place much confidence in the absolute numbers.

Samples from all sites with  $<50\%$   $\text{CaCO}_3$  have a background  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio between about 0.5 and 1.0 (Fig. 13). Average upper continental crust has an average  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  value of about 1.0, and modern deep-sea red clay from the Hatteras abyssal plain has a  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  value of 0.5 (Dean and Arthur, 1987). We are assuming that pure pelagic carbonate does not contain significant amounts of titanium, aluminum, or potassium, but this is not true for sodium because of the high Na/Ca ratio in seawater. Shells of planktonic foraminifera contain about 0.32%  $\text{Na}_2\text{O}$  (Delaney and others, 1985). Pure pelagic oozes from the Mid-Pacific Mountains and Hess Rise contain 0.1–0.3%  $\text{Na}_2\text{O}$  and  $<0.03\%$   $\text{K}_2\text{O}$  (Dean, 1981). If we assume that detrital clastic material contains 4.0%  $\text{K}_2\text{O}$  and 2.0%  $\text{Na}_2\text{O}$  ( $\text{Na}_2\text{O}/\text{K}_2\text{O} = 0.5$ ; the concentrations in pelagic red clay from the Hatteras abyssal plain, Dean and Arthur, 1987), and pure pelagic carbonate

FIGURE 8. Cross plots of percent organic carbon versus Rock-Eval pyrolysis  $\text{S}_2$  in samples from DSDP Holes 603B (A), 105, 387, and 391C (B), and 367 (C). Heavy dashed lines labeled I/II and II/III are the boundaries between types I and II, and types II and III kerogen, respectively (see text for discussion).

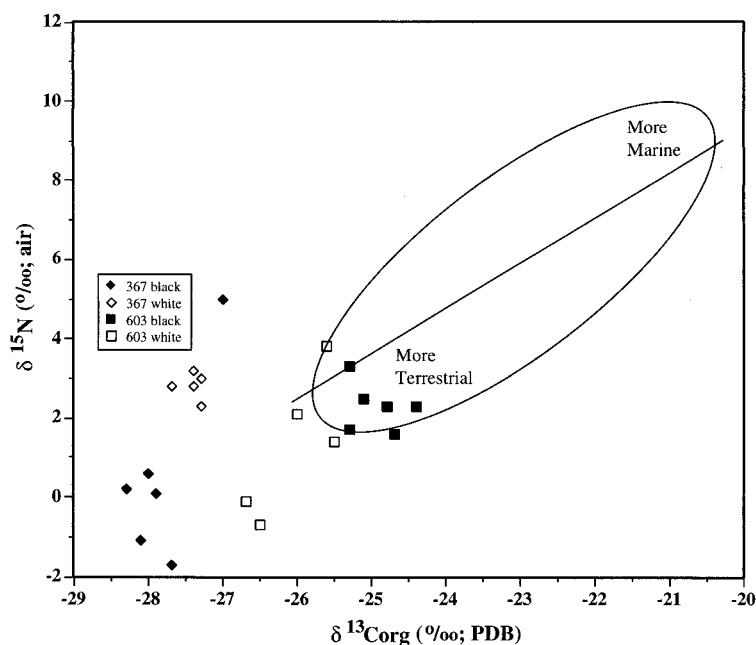


FIGURE 10. Cross plot of  $\delta^{13}\text{C}$  of organic carbon and  $\delta^{15}\text{N}$  of total nitrogen in samples of Neocomian carbonates from DSDP Holes 367 and 603 (Rau and others, 1987). Also shown is the range of values in near-shore marine samples from the eastern North Pacific Ocean reported by Peters and others (1978; ellipse). The line is the linear regression through those data.

contains 0.32%  $\text{Na}_2\text{O}$  (foraminiferal calcite of Delaney and others, 1985), then we can calculate a mixing line of these two materials from 100% pelagic clay to 100% pelagic carbonate. This mixing line is shown on the three  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  plots in Figure 13.

The higher  $\text{K}_2\text{O}$  content of the black claystones results in a lower  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio (Fig. 13A). Like the  $\text{Al}_2\text{O}_3/\text{TiO}_2$  ratio, the difference in the  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio is greatest between the black claystones and limestones with >80%  $\text{CaCO}_3$ . For samples from Hole 603B, the difference in the  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio between black claystones and white limestones can be explained entirely by mixing of pelagic biogenic carbonate and clastic material having a  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio of 0.5. However, the much higher  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio in high-carbonate samples from other sites, particularly in samples from Holes 387 and 391C in the western North Atlantic (Fig. 13B) and 367 in the eastern North Atlantic (Fig. 13C), cannot be explained by mixing of pure pelagic carbonate with clastic material having a  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio of 0.5. The clastic material in the limestones from these three holes must have a  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio of >0.5.

Based on Figures 12 and 13, we consider  $\text{Al}_2\text{O}_3/\text{TiO}_2$  ratios of 20–25, and  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratios of 0.5–0.6 to be typical of terrigenous clastics derived from the eastern margin of North America during the Early Cretaceous. An  $\text{Al}_2\text{O}_3/\text{TiO}_2$  ratio of about 20 also seems to be about average for detrital clastic material reaching the northwest African margin (Site 367) during deposition of the low-carbonate lithologies (Fig. 12C). However, the  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio in samples of low-carbonate lithologies from Hole 367 (0.2–0.5) are even lower than those in low-carbonate lithologies in the western North Atlantic making the contrast between the marlstones and limestones even greater (Fig. 13C). Taking into account possible errors due to calculating ratios using values of  $\text{TiO}_2$

and  $\text{K}_2\text{O}$  that are at or near the detection limits in limestones, it appears that terrigenous clastics delivered to the margin of northwest Africa during deposition of the limestones had an  $\text{Al}_2\text{O}_3/\text{TiO}_2$  ratio of >80, and a  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio of >6 (Figs. 12C and 13C). We suggest that the increase in  $\text{Al}_2\text{O}_3/\text{TiO}_2$  from about 20 to as much as 120, and the increase in  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  from about 0.5 to as much as 9, in the carbonate lithologies at sites in the western North Atlantic are indicative of mixing of North American and African clastic sources in these pelagic carbonates. By this reasoning, the clastic material in the limestone samples from Holes 387 and 391C with  $\text{Al}_2\text{O}_3/\text{TiO}_2$  ratios >60 (Fig. 12B), and  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratios >6 (Fig. 13B) is mostly clastic material from Africa, most likely transported as eolian dust. If this is true, then that Cretaceous clastic material must have had  $\text{Al}_2\text{O}_3/\text{TiO}_2$  and  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratios higher than clastic material from Africa reaching the Atlantic today. For example, the  $\text{Al}_2\text{O}_3/\text{TiO}_2$  ratio of terrigenous clastic material being deposited on the Zaire Fan today is about 26, and has been as high as 30 over the past 200,000 years (Schneider and others, 1997). Another possibility is that the clastic material in the carbonate lithologies in the western North Atlantic was derived from two different North American clastic sources, a dominant source with relatively low concentration of Na and Al, and a second source with relatively high concentrations of Na and Al. However, the trends of the two ratios in the carbonate lithologies toward values that are similar to those in samples of white limestone in Hole 367 would make a North African source more reasonable.

The variability in the  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio in samples from Hole 603B is mainly due to a variable  $\text{K}_2\text{O}$  concentration, with a more or less constant  $\text{Na}_2\text{O}$  concentration on a carbonate-free basis (Table 1). The variability in the  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio in samples from Hole 367 is due to lower  $\text{Na}_2\text{O}$  con-



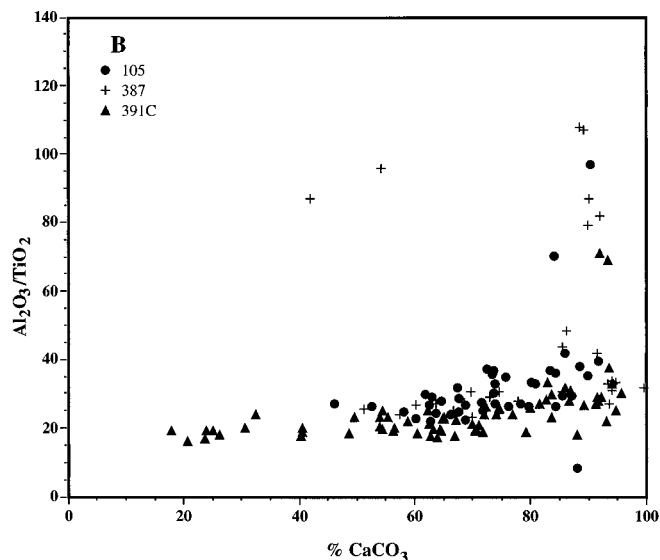
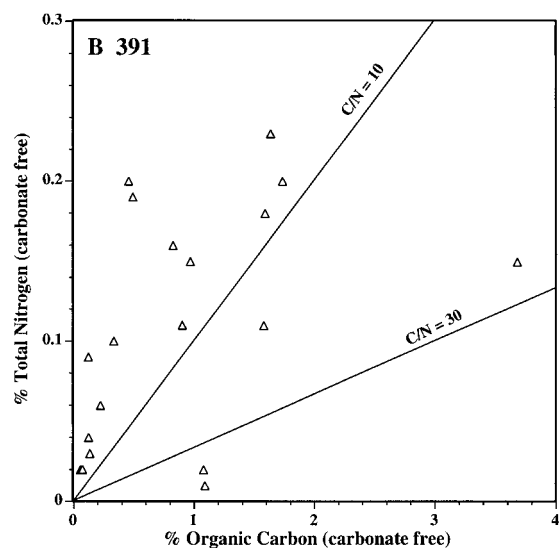
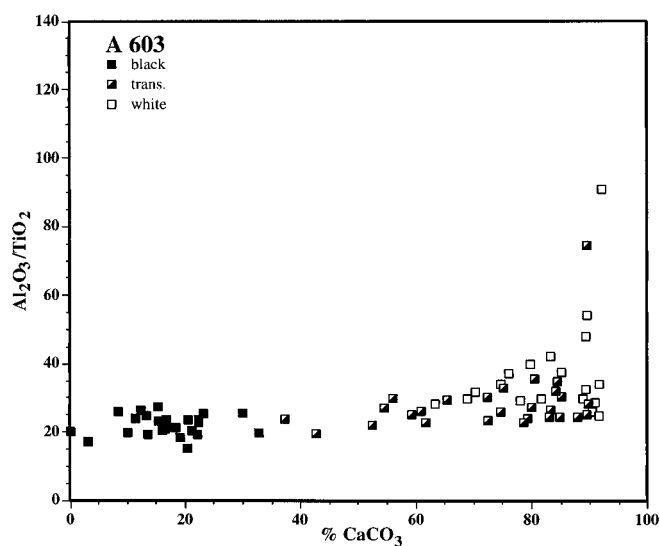
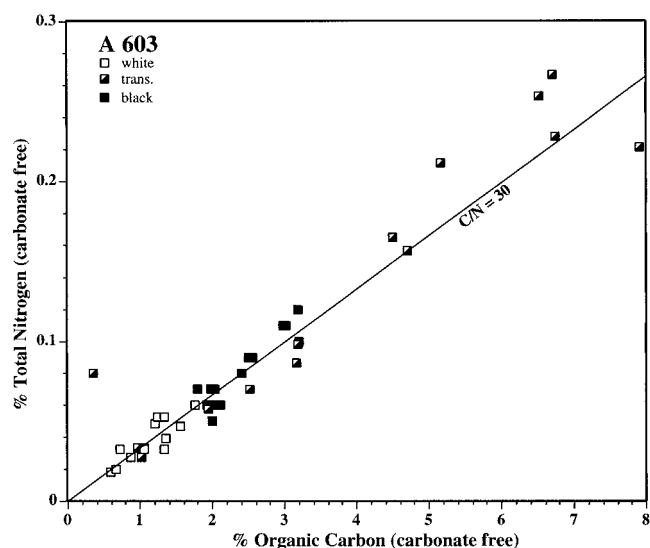


FIGURE 11. Cross plots of percent organic carbon versus percent total nitrogen, both on a carbonate-free basis, in samples from DSDP Holes 603 B(A) and 391C (B).

centrations and higher  $K_2O$  concentrations in the black marlstones and limestones relative to the white limestones (Table 1). One possible explanation for the marked difference in  $Na_2O/K_2O$  ratio is that the Neocomian strata in Hole 367 may have had two clastic sources, a low-sodium, high-potassium source for the black marlstones, and a high-sodium, low-potassium source for the white limestones, and both with about the same  $Al_2O_3/TiO_2$  ratio.

Alternatively, the compositional differences between lithologies at each site may be due to fluctuations in climatic conditions, such as alternating drier and wetter conditions over a generally more arid northwest Africa. Similarly, the compositional differences between the two sites may be the result of regional climatic differences such as more arid conditions on northwest Africa, and more humid conditions on eastern North America (e.g., Parrish and Curtis, 1982; Barron and Washington, 1982; Hallam, 1985). If the compositional differences are due to differences in climate, then the

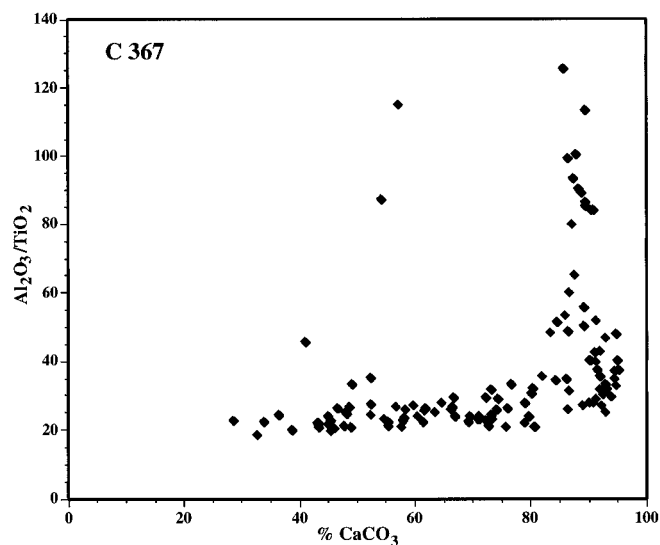


FIGURE 12. Cross plots of percent  $CaCO_3$  versus  $Al_2O_3/TiO_2$  ratio in samples from DSDP Holes 603B (A), 105, 387, and 391C (B), and 367 (C).

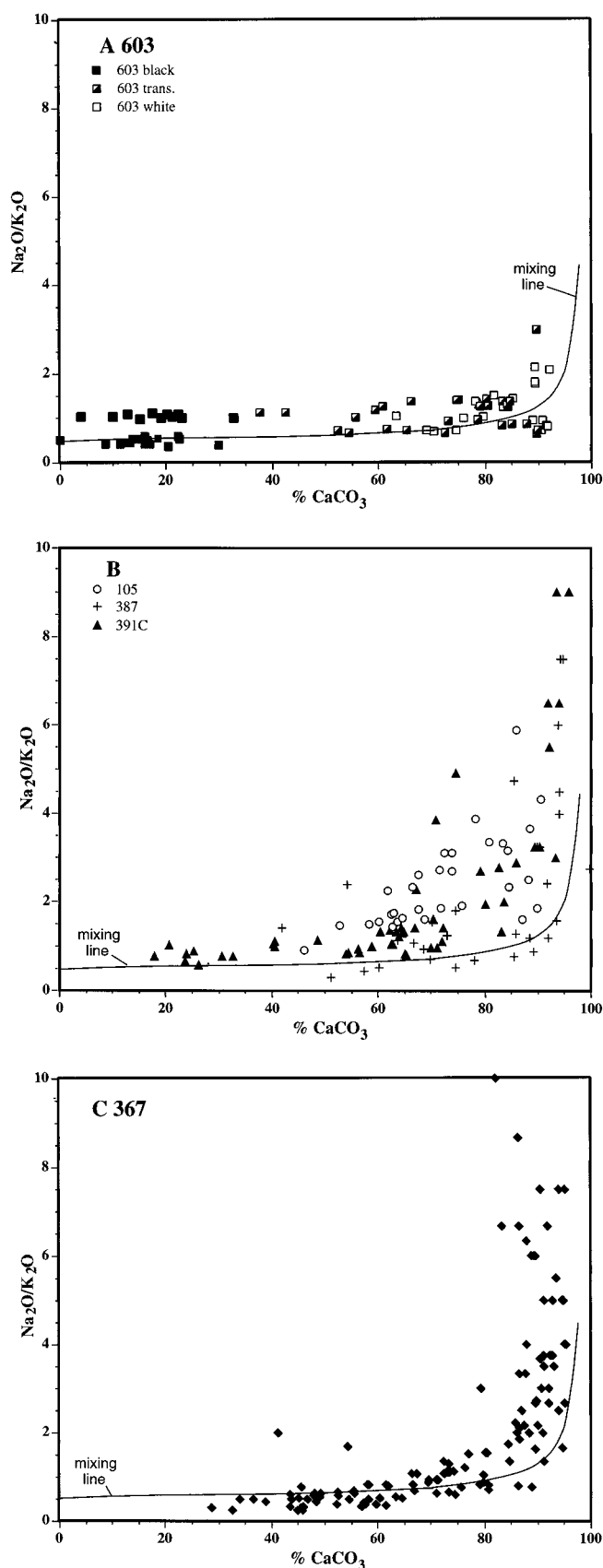


FIGURE 13. Cross plots of percent  $\text{CaCO}_3$  versus  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio in samples from DSDP Holes 603 B (A), 105, 387, and 391C (B), and 367 (C). See text for discussion of the mixing line (solid curve).

low-sodium characteristics of the claystones at Site 603 may reflect highly weathered feldspars in the humid coastal soils of North America. Increased precipitation and runoff are known to increase the chemical weathering of plagioclase feldspars and, consequently, increase the weathering flux of sodium (e.g., Johnson and Meade, 1990; White and Blum, 1995). In contrast, the weathering flux of potassium does not seem to be affected by climate. Thus, humid weathering of feldspars would preferentially increase the proportions of potassium feldspar relative to plagioclase feldspar and hence produce a low  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio. The limestones sampled from Hole 367 have particularly high  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratios (Fig. 13C). The high-sodium characteristics of the white limestones at Site 367 may reflect the eolian supply of less weathered clastic material from the dry African continent (Barron and Washington, 1982; Hallam, 1985). By this analogy, the low-sodium black marlstones and limestones would represent wetter periods with more decomposition of plagioclase feldspar relative to potassium feldspar (lower  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio). The white limestones, therefore, represent slow deposition during dry periods when little, but unweathered, eolian clastic material reached the African continental margin allowing the accumulation of biogenic  $\text{CaCO}_3$  relatively undiluted by terrigenous clastic material. It would appear that Na-rich, eolian clastic material reached the more proximal regions of the eastern North Atlantic, just as it does today (e.g., Prospero, 1982), but not more distal sites such as 603, which always received weathered clastic material from North America. These sediment sources are discussed in greater detail below.

The higher  $\text{Al}_2\text{O}_3/\text{TiO}_2$  ratios in the limestones also may be telegraphing a weathering index. For years scientific dogma held that both aluminum and titanium were immobile in low-temperature environments. However, recent studies have shown that aluminum may not be so immobile (e.g., Michalopoulos and Aller, 1995). Thus the relative enrichment of aluminum in the very small fraction of detrital clastic material, postulated to have been transported by wind from a dry African continent during deposition of the limestone units, may be indicative of unweathered material.

The average  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio in the limestone units in Hole 367 is about 7 (Table 1), but the maximum in samples with highest concentrations of  $\text{CaCO}_3$  is 11. In contrast, the average  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio in most samples of white limestones and transitional laminated marlstones and limestones from Hole 603B is about 4, lower than all samples of black marlstones and limestones from Hole 367. Thus, the Neocomian lithologic units in Hole 603B are never enriched in  $\text{SiO}_2$ . The  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratios at other western North Atlantic sites more proximal to Africa are elevated in the  $\text{CaCO}_3$ -rich samples, with values as high as 10 in some samples from Hole 387. This suggests that the excess  $\text{SiO}_2$  probably is from eolian quartz derived from northwest Africa just as it is today (Windom, 1975; Prospero, 1982). X-ray diffraction analyses of whole-rock samples of Neocomian limestones (<80 calcite) from Hole 367 by Mélières (1978) usually show 3–4% quartz. However, biogenic silica also is a likely source of excess  $\text{SiO}_2$ . Three samples from Hole 367 and two from Hole 387 contain about 98%  $\text{SiO}_2$  plus  $\text{CaCO}_3$ , in about equal proportions. The visual core descriptions in the Initial Reports volumes indicate that these sam-

ples were associated with chert and/or porcellanite. Therefore, the most likely source of the excess  $\text{SiO}_2$  in these samples is biogenic silica.

## DISCUSSION

### CYCLICITY

As mentioned earlier, the Neocomian carbonate cycles have periodicities typical of cycles of the Earth's orbital parameters (Milankovitch cycles). The Neocomian North Atlantic Ocean, therefore, appears to have been sensitive to variations in orbital forcing. The average uncompacted sedimentation rate of the Neocomian carbonates in Hole 603B is 2 cm/ky or 2 m/100 ky (van Hinte, Wise, and others, 1987). This means that an average limestone/marlstone cycle 2 m thick would correspond to a periodicity of about 100,000 years, equivalent to the Earth's eccentricity cycle, thought to be the "pacemaker of the ice ages" (Hays and others, 1976; Imbrie and others, 1984). However, the cycles are decimeter-scale not meter-scale in thickness, with an average thickness of about 40–50 cm (Fig. 1A). Therefore, the cycles are closer in periodicity to the Earth's 21,000-year orbital precession cycle or 41,000-year obliquity cycle, which are common in Cretaceous pelagic carbonate cycles (e.g., Fischer, 1980; Arthur and others, 1984; Fischer and others, 1985; Bottjer and others., 1986; ROCC Group, 1986; Oglesby and Park, 1991; Pratt and others, 1993; Savrda and Bottjer, 1994; Sageman and others, 1997). Cotillon and Rio (1984) estimated that Neocomian carbonate cycles at DSDP Site 535 in the western Tethys (Gulf of Mexico) had average periodicities of 16,000 to 26,000 years; those at Site 534 in the western North Atlantic off Florida had average periodicities of about 16,000 years; and those in the Vocontian Basin of southern France (Fig. 1B) had average periodicities of 16,000 to 20,000 years.

Orbital forcing is external to the basins of deposition, and, therefore, the direct internal cause or causes must be determined and links established between the external and internal causes. For pelagic carbonate cycles, the usual suspects for internal direct causes of the cycles are carbonate production by calcareous plankton, carbonate dissolution, and (or) carbonate dilution, usually by terrigenous clastic material (e.g., Dean and others, 1978; Arthur and others, 1984; ROCC Group, 1986; Dean and Gardner, 1986; Arthur and Dean, 1991). Preservation of calcareous nannofossils in Neocomian carbonate sequences varies from excellent to poor (e.g., Covington and Wise, 1987). In Hole 603B, calcareous nannoplankton are poorly preserved in the bioturbated limestones and best preserved in the black claystones, the opposite of what one would suspect if the clay-rich beds were produced by dissolution. Covington and Wise (1987) attributed this to a combination of inhibition of carbonate dissolution in clay-rich sediments (e.g., Schlanger and Douglas, 1974; Frank and others, this volume), and the fact that the black claystones are turbidites, redeposited from shallower depths. All of the above observations suggest that dissolution can probably be eliminated as the primary cause of the cycles, and that dilution by terrigenous clastic material is the primary cause, particularly in the western basin.

Supply of terrigenous clastic material by rivers to the narrow early North Atlantic on the one hand, and supply of

aeolian dust on the other hand, requires climatic periods that were wet and dry, respectively. Thus the limestone/marlstone cycles must be the result of alternating wet-dry climate cycles on both the North American and African continents. However, during a supposed ice-free Cretaceous period what are the links between these climatic cycles and the Earth's orbital cycles, particularly the 21-ky precessional and 41-ky obliquity cycles? The prominence, ubiquity, and high amplitude of these cycles suggest that there must have been a highly dynamic climate-ocean-atmosphere system. Climate-model simulations by Barron and others (1985) suggested that orbital variations in the Cretaceous may have enhanced the effect of low-latitude solar insolation and produced regions of intense precipitation. In particular, they found that the northern margin of the Tethys Ocean had all of the climatic characteristics necessary to be the most sensitive to orbital variations. Large land-ocean temperature contrasts in this region would have resulted in greater monsoonal circulation and intense winter precipitation. This is in line with model results by Glancy and others (1986) that showed that insolation changes had the largest effect on mid-Cretaceous climate in the Northern hemisphere winter in the form of increased storminess along the northern coast of Tethys. Oglesby and Park (1991) modeled the precession cycle at 100 Ma (Albian) and found that variance in surface winds was two to three times larger in winter than in summer. They also concluded that Cretaceous bedding cycles were deposited at precessional periods of 19 and 23 ky and not at the 100-ky eccentricity period. Eccentricity variation only has a small direct effect on insolation, but does modulate the precession cycle. However, Herbert and Fischer (1986) found that the short (100-ky) eccentricity period dominated the carbonate cycles in the Scisti a Fucoidi at Piobbico in the central Apennines of Italy. Sageman and others (1997) found that the 41-ky obliquity periodicity, and the short and long (400-ky) eccentricity periodicities dominated the carbonate cycles in the Cenomanian/Turonian Bridge Creek Limestone of the Western Interior Basin of the United States. Park and Oglesby (1991) extended the modeling reported by Oglesby and Park (1991) to examine the Cretaceous climatic response to combined precession and obliquity variations in solar insolation. They found that, for most regions, the modeled responses due to precessional changes were considerably larger than those due to obliquity changes. It appears likely, therefore, that the cyclicity in the North Atlantic-Tethys Neocomian strata was induced by orbital variations. As yet it is not clear exactly how the climate and ocean circulation responded to the forcing. However, we outline a speculative scenario below based on inferences from our data.

### A DEPOSITIONAL MODEL

Although we have emphasized the pelagic carbonate lithologies that dominate the Berriasian and Valanginian sections at Site 603, the fact is that sandstone to claystone turbidites, and even conglomerates, are abundant in the Neocomian section, and dominate in the Barremian section (Fig. 3; Sarti and von Rad, 1987). The importance of turbidites in the sediment history of continental margin settings was recognized early in the Deep Sea Drilling Project (e. g. Kelts

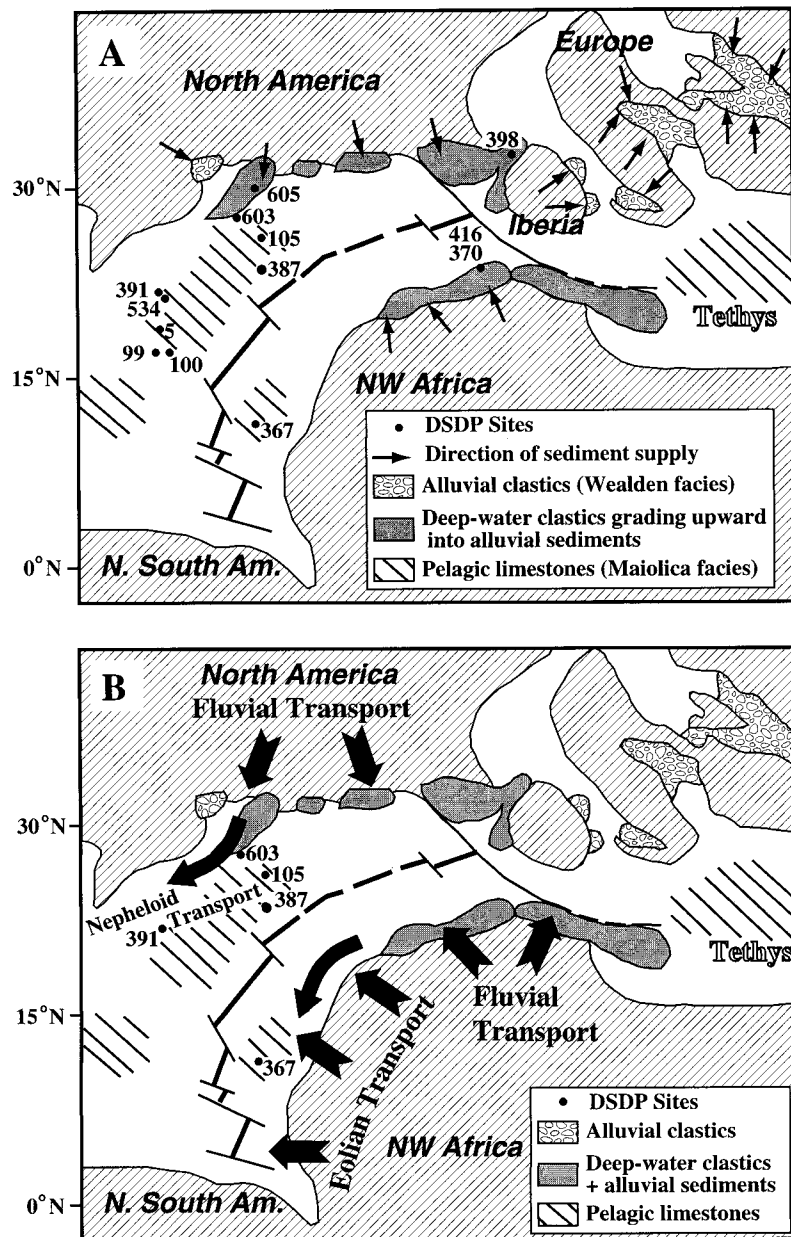


FIGURE 14. Maps showing the distributions of (A) clastic and pelagic-carbonate facies of Neocomian age recovered from the indicated DSDP Sites and land outcrops, and inferred from seismic reflection characteristics; and (B) inferred sediment transport mechanisms, plotted on a Neocomian continental reconstruction (modified from Sarti and von Rad, 1987).

and Arthur, 1981). Deep-water turbidite fan facies were recognized in seismic profiles off the east coast of North America (e.g., Jansa and McQueen, 1978; Tucholke and Mountain, 1979), but the lithologic characteristics were not known until the thick sequences of turbidites were cored at Site 603 (Sarti and von Rad, 1987). Deep-water (up to 4500 m?) clastics have been cored off Morocco (Arthur and von Rad, 1979; Lancelot and Winterer, 1980; Fig. 14A) in an environment interpreted to have been a distal fan. "Wealden-type" deltaic deposits of Neocomian age, similar to those originally described by Allen (1959), have been recognized at numerous places in Europe (e.g., von Rad and Arthur, 1979; Fig. 14A). Therefore, the deep-sea fan facies that have been documented off North America and Morocco are po-

tentially the dominant sources of terrigenous clastic material, probably delivered by nepheloid transport, to dilute pelagic carbonates in both the eastern and western North Atlantic basins (Fig. 14A), with  $\text{Al}_2\text{O}_3/\text{TiO}_2$  and  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratios of about 20 and 0.5, respectively.

The high-Al, high-Na clastic material, probably blown by the dominant easterly winds across the narrow North Atlantic (Barron and Washington, 1982), contributed varying proportions of detrital clastic material to sites in the western North Atlantic. In some samples from Holes 387, 391C, and 367 apparently most, if not all, of the detrital clastic material is high-Al, high-Na eolian dust from Africa (Figs. 12B and C and 13B and C). We suggest that this material came from relatively unweathered eolian dust from northwest Africa



(Fig. 14B), as is occurring today (e.g., Prospero, 1982). Dust transported off northwest Africa by the Saharan air layer (SAL) can be identified as a discrete atmospheric component at least 5000 km from the coast of Africa (Carlson and Prospero, 1972). Estimated deposition rates of dust within the North Atlantic trade-wind belt are  $100\text{--}400 \times 10^{12}$  g/yr, in contrast to an estimated rate of  $12 \times 10^{12}$  g/yr in the North Atlantic north of the trades (Prospero, 1982). Today, outbreaks of Saharan dust are associated with the northward advance of the Inter-Tropical Convergence Zone (ITCZ) during the summer. At those times, the northeasterly meridional trade winds that drive the Canary Current upwelling system are strongest, and coastal rivers dry up (Tiedeman and others, 1989). Therefore, dry, dusty, windy conditions on northwest Africa are coincident with strongest upwelling, which may or may not translate into increased productivity depending upon the availability of nutrients. Even if productivity was higher during these dry intervals, well-oxygenated waters may not have allowed preservation of produced organic matter. Such periods would correspond to periods of deposition of relatively undiluted pelagic carbonate sediments during the Neocomian. Wetter periods, represented by the OC-rich marlstones in the Neocomian sequences, would have higher runoff with higher nutrient loadings, and, therefore the potential for higher productivity, even though wind-driven upwelling may have been less. Also, greater runoff may have lowered surface water salinity, created a more stable stratification of the water column, longer deep-water residence time, and oxygen-deficient bottom waters with the potential for increased preservation of organic matter. For example, glacial-age sediments at Ocean Drilling Project (ODP) Site 658 in the upwelling area off Cape Blanc, northwest Africa, record dry conditions with no fluvial influx, but with strong trade winds and enhanced upwelling (Stein and others, 1989; Tiedeman and others, 1989). Interglacial sediments record wet conditions with greater fluvial influx of clay, weaker trade winds and weaker upwelling. In spite of the differences in upwelling between glacial and interglacial times, high accumulation rates of OC occurred during both periods (Stein and others, 1989). However, high abundances of marine diatoms, interpreted as high productivity, only occur during interglacials (Stabell, 1989), suggesting that productivity was higher during the wetter interglacials than during the dryer glacials, even though upwelling was weaker.

We constructed several mixing models to try to explain the patterns of  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  versus %  $\text{CaCO}_3$  shown in Fig. 13. We start with the assumptions that: 1. the mass accumulation rate (MAR) of  $\text{CaCO}_3$  was constant, and we chose a value of  $1 \text{ g/cm}^2/\text{yr}$ ; 2. variations in  $\text{CaCO}_3$  are due to variations in dilution by terrigenous clastic material; 3. clastic material is derived from eolian dust from northwest Africa, and from alluvial detrital clastic material in extensive deep-sea fans, but ultimately derived from rivers draining the North American continent and the rising Atlas Mountains of Morocco. For a first approximation (solid curve in Fig. 15), we assumed that in a sample with 90%  $\text{CaCO}_3$ , all of the detrital clastic material was derived from northwest African dust with a  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio of 9 (Fig. 13) and with a MAR of  $0.1 \text{ g/cm}^2/\text{yr}$  (1/10 of the  $\text{CaCO}_3$  MAR). We assumed that the eolian MAR remained constant, and that

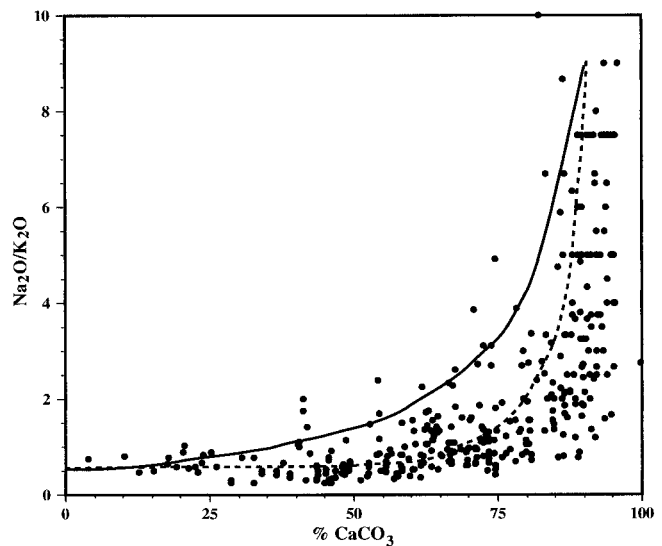


FIGURE 15. Mixing models (solid and dashed curves) for dilution of pelagic carbonate, assumed to be deposited at a constant rate, by eolian dust from northwest Africa, with a  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio of about 9, and reworked deep-sea fan material from the margins of eastern North America and Morocco, with a  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio of about 0.5. Solid circles represent all observed data. See text for descriptions of assumptions and methods of calculations.

any additional dilution of  $\text{CaCO}_3$  to values lower than 90% was due to addition of deep-sea clastics, probably delivered by nepheloid transport (Fig. 14B), characterized by a  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio of 0.5 (Fig. 12). Using this mixing model, the  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio did not decrease with decreasing  $\text{CaCO}_3$  content as rapidly as it does in the data (Fig. 15). We conclude, therefore, that the eolian contribution to the clastic fraction was not constant, and decreased with increasing influx of deep-sea clastics. As a second approximation (dashed curve in Fig. 15), we assumed that the eolian contribution decreased as deep-sea clastic sources increased, with the eolian contribution decreasing by 50% for each 10% increase in deep-sea clastics (10% decrease in %  $\text{CaCO}_3$ ). Using this second mixing model, the  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio still did not decrease as rapidly as in most of the data (Fig. 15). Therefore, the eolian clastic flux must decrease more rapidly, perhaps exponentially, with increased influx of clastics from deep-sea fans. The point is that influx of eolian clastic material was important during dry intervals when influx of clastic material from deep-sea fans essentially shut down, but, overall, the fan-derived clastics were much more important. During the Jurassic, the proto-Atlantic/Tethys region was characterized by dry climates. In the Early Cretaceous, with the opening of the Atlantic Ocean, the Atlantic/Tethys region became much more humid (Hallam, 1985); only northwest Africa remained dry. Rainfall and runoff in the humid regions around the margins of the young North Atlantic Ocean contributed to the development of extensive deep-sea fans (the Wealden facies).

The predominance of carbonate-dominated Neocomian sequences at DSDP Sites such as 99, 100, 367, 387, 391, and 534, and in Tethyan sections (the so-called Maiolica facies; Fig. 14A), is in marked contrast to the coarse-clastic-dominated Neocomian sequences recovered at DSDP Sites

370, 398, and 603. Certainly the biggest difference between the Neocomian sections recovered on the northwestern Africa margin at Sites 367 and 370 is the dominant pelagic-carbonate (Maiolica) facies at 367 and the siliciclastic, terrigenous, deep-sea fan facies at 370 (Fig. 14A), presumably derived from the rising Atlas Mountains (Lancelot and Seibold, 1978).

We do not have mineralogical data that might help to explain the geochemical data, particularly for the limestones. One of the reasons for the lack of mineralogical data on the clastic fraction in limestones is that a large sample needs to be dissolved to obtain enough insoluble residue for X-ray diffraction analyses, and large samples are not available. However, reconnaissance mineralogical studies of samples of Jurassic to Pleistocene age from other Atlantic DSDP sites by Chamley and Debrabant do provide some information (e.g., Chamley, 1979; Chamley and Debrabant, 1984; Chamley and others, 1983; Debrabant and Chamley, 1982; Debrabant and others, 1984). These studies have established that smectite is the dominant clay mineral in the clastic fraction of the Neocomian carbonates in the eastern and western basins of the North Atlantic as well as the Gulf of Mexico. Of particular significance is the low abundance of kaolinite, which is a common clay mineral in many continental and shallow-water sequences of the same age. For example, kaolinite generally comprises <10% of the clay fraction in 20 samples of the Blake Bahama Formation recovered from Hole 603B, whereas smectite comprises >80% (Huff, 1987). The most abundant clay minerals in modern African dust are kaolinite and illite. Therefore, these minerals might be expected to be most abundant in the clastic fraction of limestones. Analysis of the clay-mineral fraction of one sample of a Neocomian carbonate (lithology unknown) from Hole 367 by Timofeev and others (1978) shows that illite and kaolinite are the dominant minerals. Therefore, the apparent lack of kaolinite may be a sampling problem—nobody samples the limestones for clay mineralogy. Chamley and Debrabant (1984) explained the apparent low abundance of kaolinite to preferential entrapment of kaolinite relative to smectite in marginal basins. Abundant illite was found only in samples from Hole 416 off northwest Africa and Hole 387 in the western North Atlantic. The abundance of illite (as well as mixed-layer clays and kaolinite) off northwest Africa (Site 416) may reflect the uplift of the Atlas Mountains beginning in the Jurassic. The abundance of illite at Site 387 and its low abundance at every other site in the western North Atlantic is difficult to explain. Chamley and Debrabant (1984) suggested that the clastic fraction of the Neocomian carbonates at Site 387 was derived from an eastern North Atlantic source and transported by bottom currents. The low  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio in samples of Neocomian black marlstones and limestones from Hole 367 also may be due to an increase in the supply of illite to this site during wetter conditions that also resulted in greater decomposition of Na-feldspars in the source clastic debris.

During dry phases, deposition of unweathered detrital clastic material was much reduced, and relatively pure (>85%  $\text{CaCO}_3$ ), undiluted pelagic carbonate sediment accumulated. During wet phases, bottom currents appear to have been active, with a strong nepheloid layer that distrib-

uted reworked, highly weathered terrigenous clastic material from deep-sea fans (Fig. 14B) that diluted the carbonate sediment resulting in marlstones of highly variable  $\text{CaCO}_3$  content. Many of the laminations in laminated marlstones and limestones in the western basin are thick and not very distinct, and it is not clear whether they indicate deposition in quiet, anoxic waters as has been assumed, or by bottom currents (Sarti and von Rad, 1987). Bottom waters probably were not anoxic because of the presence of epibenthic macrofossils and infaunal burrows (Haggerty, 1987). However, the very small sizes of the burrows suggests that oxygen levels were very low (Savrdá and Bottjer, 1986). Haggerty (1987) concluded that the laminated limestones recovered in Hole 603B were deposited from the suspended loads of nepheloid layers associated with weak bottom-current activity. Robertson and Bliefnick (1983) ruled out nepheloid deposition because of coarse radiolarian tests, and suggested that multi-annual pulses of surface productivity seemed to be the best way to explain the regularity and widespread nature of the laminations throughout the central North Atlantic. The inorganic geochemical composition of the wet-phase clastics at all western basin sites suggests that the diluting clastic material had a rather uniform composition whether proximal to the source, as at Site 603, or more distal, as at Site 387. Dark-colored, laminated marlstones and limestones in Hole 603B have the same geochemical signature as subadjacent or superjacent redeposited black claystones (Dean and Arthur, 1987). This supports the concept of redistribution of clastic material from point sources on deep-sea fans by nepheloid transport.

The characteristics of organic matter in the laminated marlstones and limestones in the western basin also suggest dominantly oxic conditions. Although these transitional lithologies are relatively enriched in organic carbon in comparison to bioturbated limestones (on a carbonate-free basis; Table 1), the organic matter is primarily of terrestrial derivation with some additional, oxidized marine material, as indicated by the low  $\text{S}_2$  yields, indicating that the organic matter was poorly preserved, probably under oxic conditions. The organic carbon enrichments probably occurred because of higher rates of sedimentation during deposition of these transitional carbonates as well as by preferential transport of less dense organic matter in nepheloid layers. Much oxidation of organic matter probably occurred during transport. This increased flux of more labile organic matter was sufficient to consume pore-water oxygen and allow sulfate reduction and pyrite formation at a few centimeters depth in the sediment. Manganese, iron and other redox-sensitive metals were reduced. However, much of the iron was fixed in pyrite, whereas the manganese probably migrated to the sediment/water interface where it was oxidized and precipitated as minor oxyhydroxides or oxide coatings on carbonate particles. Subsequently, this Mn was re-reduced as the next cycle accumulated above it and was incorporated in carbonate cements that formed preferentially in the limestones (Table 1; Frank and others, this volume). Thus, there is little or no evidence for significant oxygen deficiency in the western basin during deposition of the laminated transitional carbonates, although dissolved oxygen contents of deep-water masses probably were lower than

during the bioturbated limestone depositional phase of each cycle.

Bioturbated limestones formed because of a lack of dilution by clastic detritus and more highly oxygenated deep waters, and probably not because of higher productivity. Bioturbation was probably more effective because of the overall lower sedimentation rates of the carbonate-rich intervals and the lack of current reworking of sediment. This interpretation is based, in part, on the geochemical signal of a probable African dust source that becomes prominent in intervals with  $>85\%$   $\text{CaCO}_3$ . At lower carbonate contents, the dust signal is masked by terrigenous clastic material reworked from deep-sea fans. Thus, it is the lack of dilution by this clastic source that allowed nearly pure biogenic carbonate sediment (the Maiolica facies) to accumulate. The dearth of clastic material in the dry phase of a limestone-marlstone cycle must indicate both a waning clastic source, even near the source as at Site 603, and diminishing strength of bottom currents and nepheloid transport. This suggests that lower rates of fluvial transport also occurred during the dry phases. This appears to coincide with stronger easterly winds that transported a greater flux of dust from Africa to the eastern and western basins. As discussed earlier, modeling of the geochemical data indicates that the inferred African dust must have had greater source strength during the dry phase of the cycle, and must have waned rapidly as the climate became wetter. Thus, it appears that stronger winds and drier climates prevailed around the entire North Atlantic during dry phases, and that deep waters in the western basin were more oxygenated but bottom currents less vigorous than during the wet phases.

Deep waters in the eastern basin behaved similarly to those in the western basin during the dry part of the cycle. The inorganic geochemical composition of non-carbonate material in the limestones is interpreted as indicating an African dust source. Interestingly, the strong similarity of geochemical compositions of non-carbonate material in high-carbonate ( $>85\%$   $\text{CaCO}_3$ ) intervals in both the eastern and western basins provides us with a way to tie the cycles in both basins to one another. The limestone part of each cycle must be coeval in the eastern and western basins because the eolian source appears only in the carbonate-rich beds. Thus, during the limestone depositional phase, deep waters in the entire North Atlantic appear to have been well oxygenated as evidenced by the intense bioturbation of limestone beds and the lack of preserved organic matter in those intervals. The greater strength of the easterly winds during the dry phase must have produced greater rates of upwelling along the North African margin, but lack of nutrients prevented increased productivity and (or) strongly oxidizing conditions reduced the preservation of produced organic matter. Higher primary productivity during deposition of the OC-poor limestones may be indicated by the higher Si/Al ratios from siliceous plankton, although higher quartz contents would also be expected from stronger winds. The greatest indication of increased productivity and (or) preservation of organic matter in the eastern basin is in the laminated marlstones and limestones there.

Evidence of enhanced preservation of marine-derived organic matter at Site 367 in the eastern basin is the much higher hydrogen content of the organic matter in marlstones

(Fig. 8C). The carbon-isotopic composition of the organic matter in samples from Hole 367, in comparison to that in samples from Hole 603B in the western basin, also confirms the marine origin of the organic matter in marlstones within the eastern basin (Fig. 9). However, the nitrogen isotopic composition suggests that the organic matter was derived mainly from nitrogen-fixing cyanobacteria (Rau and others, 1987; Fig. 10). This may be an indication that nitrogen was limiting to primary production as might be expected if shallow intermediate waters were anoxic and characterized by intense bacterial nitrate reduction. If so, the data would suggest that dysoxia or anoxia characterized at least part of the water column during the wet phase of a cycle, in agreement with the finely laminated character of many black marlstones and limestones in Hole 367. This is in contrast to the western basin, which remained mostly oxic. Therefore, organic-matter preservation was enhanced under anoxic conditions in the eastern North Atlantic during periods of lower wind strength and, presumably, lower rates of upwelling. The oxygen-depleted conditions must have arisen initially from more sluggish deep-water turnover rate (increased residence time) rather than an increase in productivity and rate of oxygen consumption. As deep-water oxygen concentrations decreased, and phosphate concentrations increased through time (1000's of yrs.), primary productivity could have increased in eastern-basin surface waters even though rates of wind-driven upwelling were slower.

One possible explanation for greater oxygenation of deep water in the tectonically isolated North Atlantic Basin during each orbitally induced dry phase is that the water column was characterized by haline-dominated deep convection. Such convection would have been driven by relatively high rates of evaporation from the sea surface and low rates of precipitation and runoff into the basin. During the wet phase of each cycle, however, overall deep convection was inhibited by the stronger vertical salinity gradients that resulted from increased precipitation and runoff. Although deep-water oxygen concentrations appear to have been lower than during the dry phase, the deep water remained oxygenated. Deep boundary currents were probably present as suggested by the redeposited clastic material and sedimentary structures in laminated marlstones and limestones, particularly in the western North Atlantic Basin. These currents could have been generated by deep water entering the basin through portals to the eastern or western Tethys by an estuarine-like circulation. Export of somewhat freshened surface water masses from the North Atlantic may have induced inflow of deeper waters external to the basin (e.g., see Slingerland and others, 1996, for a model of estuarine circulation in the Western Interior Seaway, the northwestern arm of the Cretaceous Tethys Ocean).

## CONCLUSIONS

Of the Neocomian carbonate units investigated, the organic matter in samples of black marlstones and limestones from DSDP Hole 367 in the Cape Verde Basin off northwest Africa contain predominantly marine organic matter as indicated by higher values of pyrolysis  $\text{S}_2$  and HI, and lower values of  $\delta^{13}\text{C}$ . The other end member, terrestrial organic matter, predominates in samples of homogeneous black



claystones, deposited by turbidity currents, from Hole 603B in the western North American Basin off Cape Hatteras. The organic matter in these samples, although low in abundance, has the geochemical characteristics of Cretaceous coals. Most samples of marlstones (<60%  $\text{CaCO}_3$  from Hole 603B, and from Holes 105, 387, and 391C, contain mixtures of organic matter from marine and, predominantly, terrestrial sources. Bioturbated white limestones (>60%  $\text{CaCO}_3$ ) in all holes sampled contain very low concentrations of highly oxidized organic matter.

Ratios of  $\text{Al}_2\text{O}_3/\text{TiO}_2$  and  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  suggest that there were at least two sources of detrital clastic material to the narrow Atlantic Ocean during the Early Cretaceous. Highly weathered clastic material was delivered to near-shore fans off North America and Morocco by turbidity currents during wet climatic intervals, and thence to deep-sea sites by nepheloid transport. This material mixed with pelagic carbonate to form dark colored marlstones and limestones with low  $\text{Al}_2\text{O}_3/\text{TiO}_2$  and  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratios. Higher runoff during wetter intervals probably provided higher nutrient loadings, and the potential for higher productivity, even though wind-driven upwelling may have been less. Also, greater runoff may have lowered surface-water salinity, created a more stable stratification of the water column, longer residence time, and oxygen-deficient bottom waters with the potential for increased preservation of organic matter.

Relatively unweathered eolian dust with high  $\text{Al}_2\text{O}_3/\text{TiO}_2$  and  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratios, derived from northwest Africa, is found in low concentrations in bioturbated white limestones that accumulated under highly oxidizing, sediment-starved conditions in both the eastern and western basins of the North Atlantic during dry climatic conditions. Today, dry, dusty, windy conditions on northwest Africa are coincident with strongest upwelling, which may or may not translate into increased productivity depending upon the availability of nutrients. Even if productivity was higher during these dry intervals, well-oxygenated waters may not have allowed preservation of produced organic matter, resulting in the accumulation of bioturbated, organic-poor carbonate sediments. During wetter intervals, represented by marlstones, the eolian dust signal was masked by the diluting terrigenous clastic material reworked from deep-sea fans. It is the lack of dilution by this reworked terrigenous clastic source, rather than increased carbonate production, that allowed the nearly pure biogenic carbonate sediment (the Maiolica facies) to accumulate.

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#### REFERENCES

- ALLEN, P., 1959, The Wealden environment, Anglo-Paris Basin: *Philosophical Transactions of the Royal Society of London, Series B*, v. 242, p. 283–346.
- ARTHUR, M. A., 1979, North Atlantic Cretaceous black shales: The record at Site 398 and a brief comparison with other occurrences, in Sibuet, J.-C. Ryan, W. B. F., and others, *Initial Reports of the Deep Sea Drilling Project: U. S. Government Printing Office, Washington*, v. 47, Part 2, p. 719–751.
- , and DEAN, W. E., 1986, Cretaceous paleoceanography of the western North Atlantic Ocean, in Vogt, P. R., and Tucholke, B. E. (eds.), *The Geology of North America*, v. M: Geological Society of America, Boulder, Colorado, p. 617–630.
- , and DEAN, W. E., 1991, A holistic geochemical approach to cyclomania—Examples from Cretaceous pelagic limestone sequences, in Einsele, G., Ricken W., and Seilacher, A. (eds.), *Cycles and events in stratigraphy: Springer-Verlag, Berlin*, p. 126–166.
- , and FISCHER, A. G., 1977, Upper Cretaceous-Paleogene magnetic stratigraphy at Gubbio, Italy: *Lithostratigraphy and sedimentology: Geological Society of America Bulletin*, v. 88, p. 367–371.
- , and NATLAND, J. H., 1979, Carbonaceous sediments in the North and South Atlantic: The role of salinity in strable stratification of Early Cretaceous basins, in Talwani, M., Hay, W. W., and Ryan, W. B. F. (eds.), *Deep Sea Drilling Results in the Atlantic Ocean: Continental Margins, and Paleoenvironment: American Geophysical Union, Washington, Maurice Ewing Series*, v. 3, p. 375–401.
- , and VON RAD, U., 1979, Early Neogene base-of-slope sedimentation at Site 397, DSDP Leg 47A: Sequential evolution of gravitative mass transport processes and redeposition along the Northwest African passive margin, in von Rad, U., Ryan, W. B. F., and others, *Initial Reports of the Deep Sea Drilling Project: U. S. Government Printing Office, Washington*, v. 47, Part 2, p. 603–639.
- , DEAN, W. E., and CLAYPOOL, G. E., 1985, Anomalous  $^{13}\text{C}$  enrichment in modern organic carbon: *Nature*, v. 315, p. 216–218.
- , DEAN, W. E., and STOW, D. A. V., 1984, Models for the deposition of Mesozoic-Cenozoic fine-grained organic-carbon-rich sediment in the deep sea, in Stow, D. A. V., and Piper, D. J. W. (eds.), *Fine-Grained Sediments: Processes and Products: Geological Society of London Special Publication 15*, p. 527–559.
- , DEAN, W. E., BOTTJER, D. J., and SCHOLLE, P. A., 1984, Rhythmic bedding in Mesozoic-Cenozoic pelagic carbonate sequences: The primary and diagenetic origin of Milankovitch-like cycles, in Berger, A., Imbrie, J., Hays, J. D., and others (eds.), *Milankovitch and climate, Part 1: Riedel Publishing Company, Amsterdam*, p. 191–222.
- , HAGERTY, S., DEAN, W. E., CLAYPOOL, G. E., DAWS, T., MCNAMAN, D., MEYERS, P. A., and DUNHAM, K., 1987, A geochemical note: comparison of techniques for obtaining  $\text{CaCO}_3$ , organic carbon, and total nitrogen in limestones and shales, in van Hinte, J. E., Wise, S. W., Jr., and others, *Initial Reports of the Deep Sea Drilling Project: U. S. Government Printing Office, Washington*, v. 93, p. 1263–1268.
- BARRON, E. J., and WASHINGTON, W. M., 1982, Cretaceous climate: A comparison of atmospheric simulations with the geologic record: *Palaeogeography, Palaeoclimatology, and Palaeoecology*, v. 40, p. 103–133.
- , ARTHUR, M. A., and KAUFFMAN, E. G., 1985, Cretaceous rhythmic bedding sequences: A plausible link between orbital variations and climate: *Earth and Planetary Science Letters*, v. 72, p. 327–340.
- BERNOULLI, D., 1972, North Atlantic and Mediterranean Mesozoic facies: A comparison, in Hollister, C. D., Ewing, J. I., and others, *Initial Reports of the Deep Sea Drilling Project: U. S. Government Printing Office, Washington*, v. 11, p. 801–871.
- BOTTJER, D. J., ARTHUR, M. A., DEAN, W. E., HATTIN, D. E., and SAVRDA, C. E., 1986, Rhythmic bedding produced in Cretaceous pelagic carbonate environments: Sensitive recorders of climatic cycles: *Paleoceanography*, v. 1, p. 467–481.
- BUFFLER, R. T., SCHLAGER, W., and others, 1984, *Initial Reports of*



- the Deep Sea Drilling Project: U. S. Government Printing Office, Washington, v. 77.
- CARLSON T. N., and PROSPERO, J. M., 1972, The large-scale movement of Saharan air outbreaks over the equatorial North Atlantic: *Journal of Applied Meteorology*, v. 11, p. 283–297.
- CHALMEY, H., 1979, North Atlantic clay sedimentation and paleoenvironments since the Late Jurassic, in Talwani, M., Hay, W. W., and Ryan, W. B. F. (eds.), *Deep Sea Drilling Results in the Atlantic Ocean: Continental Margins, and Paleoenvironment*: American Geophysical Union, Washington, Maurice Ewing Series, v. 3, p. 342–361.
- , and DEBRABONT, P., 1984, Paleoenvironmental history of the North Atlantic region from mineralogical and geochemical data: *Sedimentary Geology*, v. 40, p. 151–167.
- , CANDILLIER, A. M., and FOULON, J., 1983, Clay mineralogical and inorganic geochemical stratigraphy of Blake-Bahama Basin since the Callovian, Site 534, Deep Sea Drilling Project Leg 76, in Sheridan, R. E., and Gradstein, F. M., and others, *Initial Reports of the Deep Sea Drilling Project*: U.S. Government Printing Office, Washington, v. 76, p. 437–451.
- COTILLON, P., 1984, Tentative world-wide correlation of Early Cretaceous strata by limestone-marl cyclicities in pelagic deposits: *Geological Society of Denmark Bulletin*, v. 33 p. 91–102.
- , and RIO, M., 1984, Cyclic sedimentation in the Cretaceous of Deep Sea Drilling Project Sites 535 and 540 (Gulf of Mexico), 534 (central Atlantic), and the Vocontian Basin (France), in Buffer, R. T., Schlager, W., and others, *Initial Reports of the Deep Sea Drilling Project*: U. S. Government Printing Office, Washington, v. 77, p. 339–376.
- COVINGTON, J. M., and WISE, S. W., JR., 1987, Calcareous nannofossil biostratigraphy of a Lower Cretaceous deep-sea complex: Deep Sea Drilling Project Leg 93, Site 603, lower continental rise off Cape Hatteras, in van Hinte, J. E., Wise, S. W., Jr., and others, 1987, *Initial Reports of the Deep Sea Drilling Project*: U.S. Government Printing Office, Washington, v. 93, Pt. 2, p. 617–660.
- DEAN, W. E., 1981, Inorganic geochemistry of sediments and rocks from the Mid-Pacific Mountains and Hess Rise, Deep Sea Drilling Project Leg 62, in Thiede, J., Vallier, T. L., and others, *Initial Reports of the Deep Sea Drilling Project*, v. 62, Washington, U.S. Government Printing Office, p. 685–710.
- , and ANDERS, D. E., 1991, Effects of source, depositional environment, and diagenesis on characteristics of organic matter in oil shale from the Green River Formation, Wyoming, Utah, and Colorado: *U.S. Geological Survey Bulletin* 1973-F, 16 p.
- , and ARTHUR, M. A., 1987, Inorganic and organic geochemistry of Eocene to Cretaceous strata recovered from the lower continental rise, North American Basin, Site 603, Deep Sea Drilling Project Leg 93, in van Hinte, J. E., Wise, S. W., Jr., and others, 1987, *Initial Reports of the Deep Sea Drilling Project*: U. S. Government Printing Office, Washington, v. 93, Pt. 2, p. 1093–1137.
- , and ———, 1998, Geochemical expressions of cyclicity in Cretaceous pelagic limestone sequences: Niobrara Formation, Western Interior Seaway, in Dean, W. E., and Arthur, M. A., eds., *Stratigraphy and Paleoenvironments of the Cretaceous Western Interior Seaway*: SEPM (Society for Sedimentary Geology) Concepts in Sedimentology and Paleontology, No. 6, p. 227–255.
- , and GARDNER, J. V., 1982, Origin and geochemistry of redox cycles of Jurassic to Eocene age, Cape Verde Basin (DSDP Site 367), continental margin of North-west Africa, in Schlager, S. O., and Cita, M. B. (eds.), *Nature and Origin of Cretaceous Carbonate-rich Facies*: Academic Press, New York, p. 55–78.
- , and GARDNER, J. V., 1986, Milankovitch cycles in Neogene deep-sea sediment: *Paleoceanography*, v. 1, p. 539–553.
- , JANSÁ, L. F., CEPEK, P., and SEIBOLD, E., 1978, Cyclic sedimentation along the continental margin of Northwest Africa, in Lancelot, Y., and Seibold, E., and others, *Initial Reports of the Deep Sea Drilling Project*: U. S. Government Printing Office, Washington, v. 41, p. 965–986.
- , ARTHUR, M. A., and CLAYPOOL, G. E., 1986, Depletion of  $^{13}\text{C}$  in Cretaceous marine organic matter: Source, diagenetic, or environmental signal?: *Marine Geology*, v. 70, p. 119–157.
- DEBRABONT, P., and CHAMLEY, H., 1982, Influences océaniques et continentales dans les premières dépôts de l'Atlantique Nord: *Geological Society of France Bulletin*, v. 7, p. 473–486.
- , ———, and FOULON, J., 1984, Paleoenvironmental implications of mineralogical and geochemical data in the western Florida Straits (Leg 77, Deep Sea Drilling Project), in Buffer, R. T., Schlager, W., and others, *Initial Reports of the Deep Sea Drilling Project*: U.S. Government Printing Office, Washington, v. 77, p. 377–396.
- DEINES, P., 1980, The isotopic composition of reduced organic carbon, in Fritz, P., and Fontes, J. C. (eds.), *Handbook of Environmental Isotope Geochemistry*: Elsevier, Amsterdam, v. 1, p. 329–406.
- DELANEY, M. L., BÉ, A. W. H., and BOYLE, E. A., 1985, Li, Sr, Mg, and Na in foraminiferal calcite shells from laboratory culture, sediment traps, and sediment cores: *Geochimica et Cosmochimica Acta*, v. 49, p. 1327–1341.
- DONNELLY, T. W., 1980, Appendix. Chemical composition of deep sea sediments—Sites 9 through 425, Legs 2 through 54, Deep Sea Drilling Project, in Rosendahl, B. R., Hekinian, R., and others, *Initial Reports of the Deep Sea Drilling Project*: U. S. Government Printing Office, Washington, v. 54, p. 899–949.
- ENGLEMAN, E. E., JACKSON, L. L., NORTON, D. R., and FISCHER, A. G., 1985, Determinations of carbonate carbon in geological materials by coulometric titration: *Chemical Geology*, v. 53, p. 125–128.
- FISCHER, A. G., 1980, Gilbert-bedding rhythms and geochronology, in Yochelson, E. I. (ed.), *The Scientific Ideas of G. K. Gilbert*: Geological Society of America Special Paper 183, p. 93–104.
- , HERBERT, T., and PRIMOLI-SILVA, I., 1985, Carbonate bedding cycles in Cretaceous pelagic and hemipelagic sediments, in Pratt, L. M., Kauffman, E. G., and Zelt, F. B. (eds.), *Fine-grained deposits and biofacies of the Western Interior Seaway: Evidence of cyclic sedimentary processes*: Society of Economic Paleontologists and Mineralogists, 1985 Midyear Meeting, Golden, Colorado, Field Trip Guidebook No. 4, p. 1–10.
- FRANK, T. D., ARTHUR, M. A., and DEAN, W. E., 1999, Diagenesis of Lower Cretaceous pelagic carbonates, North Atlantic: Paleocyanographic signals obscured: *Journal of Foraminiferal Research*, v. 29, p. 340–351.
- GARRISON, R. E., 1967, Pelagic limestones of the Oberalm Beds (Upper Jurassic-Lower Cretaceous), Austrian Alps: *Canadian Petroleum Geology Bulletin*, v. 15, p. 21–49.
- GLANCY, T. J., BARRON, E. J., and ARTHUR, M. A., 1986, An initial study of the sensitivity of modeled Cretaceous climate to cyclic insolation forcing: *Paleoceanography*, v. 1, p. 523–537.
- HALLAM, A., 1985, A review of Mesozoic climates: *Journal of the Geological Society of London*, v. 142, p. 433–445.
- HAGGERTY, J. A., 1987, Petrology and carbon and oxygen stable isotopic composition of macrofossils and sediments from the Blake-Bahama Formation, Deep Sea Drilling Project Site 603, western North Atlantic lower continental rise, in van Hinte, J. E., Wise, S. W., Jr., and others, 1987, *Initial Reports of the Deep Sea Drilling Project*: U. S. Government Printing Office, Washington, v. 93, Pt. 2, p. 1003–11021.
- HAYS, J. D., IMBRIE, J., and SHACKLETON, N. J., 1976, Variations in the earth's orbit: Pacemaker of the ice ages: *Science*, v. 36, p. 622–625.
- HERBERT, T. D., and FISCHER, A. G., 1986, Milankovitch climatic origin of mid-Cretaceous black shale rhythms in central Italy: *Nature*, v. 321, p. 739–743.
- HOLMES, C. W., 1995, Is increasing atmospheric  $\text{CO}_2$  really a harbinger of global warming—a perspective through geologic time, in Carter, L. M., (ed.), *Energy and the environment—application of geosciences to decision making*: U.S. Geological Survey Circular 1108, p. 79–81.
- HUFF, W. D., 1987, Cretaceous clay mineralogy of the continental rise off the east coast of the United States, Site 603, Deep Sea Drilling Project Leg 93, in van Hinte, J. E., Wise, S. W., Jr., and others, 1987, *Initial Reports of the Deep Sea Drilling Project*: U. S. Government Printing Office, Washington, v. 93, Pt. 2, p. 985–988.
- IMBRIE, J., HAYS, J. D., MARTINSON, D. G., MCINTYRE, A., MIX, A. C., MORLEY, J. J., PISIAS, N. G., PRELL, W. L., and SHACKLETON, N. J., 1984, The orbital theory of Pleistocene climate: Support from a revised chronology of the marine  $\delta^{18}\text{O}$  record, in Berger, A., Imbrie, J., Hays, J. D., and others (eds.), *Milankovitch and climate, Part 1*: Riedel Publishing Company, Amsterdam, p. 269–306.

- JANSA, L. F., and MCQUEEN, R. W., 1978, Stratigraphy and hydrocarbon potential of the central North Atlantic Basin: *Geoscience Canada*, v. 5, p. 176–183.
- , GARDNER, J. V., and DEAN, W. E., 1978, Mesozoic sequences of the central North Atlantic, in Lancelot, Y., Seibold, E., and others, Initial Reports of the Deep Sea Drilling Project: U.S. Government Printing Office, Washington, v. 41, p. 991–1010.
- , ENOS, P., TUCHOLKE, B. E., GRADSTEIN, F. M. and SHERIDAN, R. E., 1979, Mesozoic-Cenozoic sedimentary formations of the North American Basin, western North Atlantic, in Talwani, M., Hay, W. W., and Ryan, W. B. F. (eds.), *Deep Sea Drilling Results in the Atlantic Ocean: Continental Margins, and Paleoenvironment*: American Geophysical Union, Washington, Maurice Ewing Series, v. 3, p. 1–48.
- JOHNSON, M. J., and MEADE, R. H., 1990, Chemical weathering of fluvial sediments during alluvial storage: The Macuapanim Island point bar, Solimões River, Brazil: *Journal of Sedimentary Petrology*, v. 60, p. 827–842.
- KATZ, B. J., 1983, Limitations of Rock-Eval pyrolysis for typing organic matter: *Organic Geochemistry*, v. 4, p. 195–199.
- , and PHEIFER, R. N., 1986, Organic geochemical characteristics of Atlantic Ocean Cretaceous and Jurassic black shales: *Marine Geology*, v. 70, p. 43–66.
- KEGAMI, H., ISHIZUKA, T., and AOKI, S., 1983, Geochemistry and mineralogy of selected carbonaceous claystones in the lower Cretaceous from the Blake-Bahama Basin, North Atlantic, in Sheridan, R. E., and Gradstein, F. M., and others, Initial Reports of the Deep Sea Drilling Project: U.S. Government Printing Office, Washington, v. 76, p. 429–436.
- KELTS, K., and ARTHUR, M. A., 1981, Turbidites after ten years of Deep Sea Drilling—wringing out the mop?, in Warme, J. E., Douglas, R. G., and Winterer, E. L. (eds.), *The Deep Sea Drilling Project: A Decade of Progress*: Society of Economic Paleontologists and Mineralogists Special Publication 32, p. 91–127.
- LANCELOT, Y. and SEIBOLD, E., 1978, The evolution of the central northeastern Atlantic—summary of results of DSDP Leg 41, in Lancelot, Y., Seibold, E., and others, Initial Reports of the Deep Sea Drilling Project, v. 41: U.S. Government Printing Office, Washington, p. 1215–1245.
- , and WINTERER, E. L., 1980, Evolution of the Moroccan oceanic basin and adjacent continental margin—a synthesis, in Lancelot, Y., Winterer, E. L., and others, Initial Reports of the Deep Sea Drilling Project: U.S. Government Printing Office, Washington, v. 50, p. 801–821.
- LANGFORD, F. R., and BLANC-VALLERON, M.-M., 1990, Interpreting Rock-Eval pyrolysis data using graphs of pyrolyzable hydrocarbons vs. total organic carbon: *American Association of Petroleum Geologists Bulletin*, v. 74, p. 799–804.
- LICHTE, F. E., GOLIGHTLY, D. W., and LAMOTHE, P. J., 1987, Inductively coupled plasma-atomic emission spectrometry, in Baedeker, P. A. (ed.), *Methods for Geochemical Analysis*: U.S. Geological Survey Bulletin 1770, p. B1–B10.
- LUO, Y., SIMS, D. A., BALL, J. T., THOMAS, R. B., and TISSUE, D. T., 1996, Sensitivity of leaf photosynthesis to CO<sub>2</sub> concentrations is an invariant function for C<sub>3</sub> plants: A test with experimental data and global applications: *Global Biogeochemical Cycles*, v. 10, p. 209–222.
- MCCAVE, I. N., 1979a, Depositional features of organic black and green mudstones at DSDP Sites 386 and 387, western North Atlantic, in Tucholke, B. E., Vogt, P. R., and others, Initial Reports of the Deep Sea Drilling Project: U.S. Government Printing Office, Washington, v. 43, p. 411–416.
- , 1979b, Diagnosis of turbidites at Site 386 and 387 by partial-counter size analysis of the silt (2–40 µm) fraction, in Tucholke, B. E., Vogt, P. R., and others, Initial Reports of the Deep Sea Drilling Project: U.S. Government Printing Office, Washington, v. 43, p. 395–405.
- MÉLIERES, F., 1978, X-ray mineralogy studies, Leg 41, Deep Sea Drilling Project, eastern North Atlantic Ocean, in Lancelot, Y., and Seibold, E., and others, Initial Reports of the Deep Sea Drilling Project: U.S. Government Printing Office, Washington, v. 41, p. 1065–1086.
- MEYERS, P. A., 1987, Organic-carbon content of sediments and rocks from Deep Sea Drilling Project Sites 603, 604, and 605, western margin of North America, in van Hinte, J. E., Wise, S. W., Jr., and others, Initial Reports of the Deep Sea Drilling Project: U.S. Government Printing Office, Washington, v. 93, Pt. 2, p. 1187–1193.
- , TRULL, T. W., and KAWKA, O. E., 1984, Organic geochemical comparison of Cretaceous green and black claystones from Hole 530A in the Angola Basin, in Hay, W. W., Sibuet, J.-C., and others, Initial Reports of the Deep Sea Drilling Project: U.S. Government Printing Office, Washington, v. 75, p. 1009–1018.
- MICHALOPOULOS, P., and ALLER, R. C., 1995, Rapid clay mineral formation in Amazon delta sediments: Reverse weathering and oceanic elemental cycles: *Science*, v. 270, p. 614–617.
- MÜLLER, P. J., 1977, C/N ratios in Pacific deep-sea sediments: Effects of inorganic ammonium and organic nitrogen compounds sorbed by clays: *Geochimica et Cosmochimica Acta*, v. 41, p. 765–776.
- OGLESBY, R. J., and PARK, J., 1991, The effect of precessional insolation changes on Cretaceous climate and cyclic sedimentation: *Journal of Geophysical Research*, v. 94, p. 14,793–14,816.
- PARK, J., and OGLESBY, R. J., 1991, Milankovitch rhythms in the Cretaceous: A GCM modelling study: *Palaeogeography, Palaeoclimatology, and Palaeoecology*, v. 90, p. 329–355.
- PARRISH, J. T., and CURTIS, R. L., 1982, Atmospheric circulation, upwelling, and organic-rich rocks in the Mesozoic and Cenozoic Eras: *Palaeogeography, Palaeoclimatology, and Palaeoecology*, v. 40, p. 31–66.
- PETERS, K. E., 1986, Guidelines for evaluating petroleum source rock using programmed pyrolysis: *American Association of Petroleum Geologists Bulletin*, v. 70, p. 318–329.
- , SWEENEY, R. E., and KAPLAN, I. R., 1978, Correlation of carbon and nitrogen stable isotope ratios in sedimentary organic matter, *Limnology and Oceanography*, v. 23, p. 598–604.
- PIPER, D. Z., and ISAACS, C. M., 1995, Geochemistry of minor elements in the Monterey Formation, CA: seawater chemistry of deposition, U.S. Geological Survey Professional Paper 1566, 41 p.
- PRATT, L. M., and THRELKELD, C. N., 1984, Stratigraphic significance of <sup>13</sup>C/<sup>12</sup>C ratios in mid-Cretaceous rocks of the Western Interior, U.S.A., in Stott, D. F., and Glass, D. J., (eds.), *Mesozoic of Middle North America*: Canadian Society of Petroleum Geology, Memoir 9, p. 305–312.
- , ARTHUR, M. A., DEAN, W. E., and SCHOLLE, P. A., 1993, Paleo-oceanographic cycles and events during the Late Cretaceous in the Western Interior Seaway of North America, in Caldwell, W. G. E. and Kauffman, E. G. (eds.), *Evolution of the Western Interior Basin*: Geological Association of Canada, Special Paper 39, p. 333–354.
- PROSPERO, J. M., 1982, Eolian transport to the World Ocean, in Emiliani, C. (ed.), *The Sea*, v. 7: New York, Wiley, p. 801–874.
- RAU, G. H., ARTHUR, M. A., and DEAN, W. E., 1987, <sup>15</sup>N/<sup>14</sup>N variations in Cretaceous Atlantic sedimentary sequences—Implications for historic changes in marine nitrogen biogeochemistry: *Earth and Planetary Science Letters*, v. 82, p. 269–279.
- ROBERTSON, A. H. F., and BLIEFNICK, D. M., 1983, Sedimentology and origin of Lower Cretaceous pelagic carbonates and redeposited clastics, Blake-Bahama Formation, Deep Sea Drilling Project Site 534, western equatorial Atlantic, in Sheridan, R. E., Gradstein, F. M., and others, Initial Reports of the Deep Sea Drilling Project: U.S. Government Printing Office, Washington, v. 76, p. 795–828.
- ROCC (Research on Cretaceous Cycles) Group, 1986, Rhythmic bedding in Upper Cretaceous pelagic carbonate sequences—Varying sedimentary response to climatic forcing: *Geology*, v. 14, p. 153–156.
- SAGEMAN, B. B., RICH, J., ARTHUR, M. A., DEAN, W. E., 1997, Evidence for Milankovitch periodicities in Cenomanian-Turonian lithologic and geochemical cycles, Western Interior U.S.A.: *Journal of Sedimentary Research*, v. 67, p. 286–302.
- SARTI, M., and VON RAD, U., 1987, Early Cretaceous sedimentation at Deep Sea Drilling Project Site 603, off Cape Hatteras, in van Hinte, J. E., Wise, S. W., Jr., and others, Initial Reports of the Deep Sea Drilling Project: U.S. Government Printing Office, Washington, v. 93, Pt. 2, p. 891–940.
- SAVRDA, C. E., and BOTTJER, D. J., 1986, Trace-fossil model for reconstruction of paleo-oxygenation in bottom waters: *Geology*, v. 14, p. 3–6.
- , and ———, 1994, Ichnofossils and ichnofabrics in rhythmic-

- cally bedded pelagic/hemi-pelagic carbonates: Recognition and evaluation of benthic redox and scour cycles, *in* de Boer, P. L., and Smith, D. G. (eds.), *Orbital Forcing and Cyclic Sequences: International Association of Sedimentologists, Special Publication 19*, p. 195–210.
- SCHLANGER, S. O., and DOUGLAS, R. G., 1974, The pelagic-ooze-chalk-limestone transition and its implication for marine stratigraphy, *in* Hsü, K. J., and Jenkyns H. (eds.), *Pelagic Sediments on Land and Under the Sea: International Association of Sedimentologists, Special Publication 1*, Blackwell, Oxford, p. 117–148.
- SCHNEIDER, R. R., PRICE, B., MÜLLER, P. J., KROON, D., and ALEXANDER, I., 1997, Monsoon related variations in Zaire (Congo) sediment load and influence of fluvial silicate supply on marine productivity in the east equatorial Atlantic during the last 200,000 years: *Paleoceanography*, v. 12, p. 463–481.
- SCHWARZACHER, W., and FISCHER, A. G., 1982, Limestone-shale bedding, *in* Einsele, G., and Seilacher, A. (eds.), *Cyclic and Event Stratification: Springer-Verlag, Berlin*, p. 72–95.
- SIMONEIT, B. R. T., 1986, Biomarker geochemistry of black shales from Cretaceous oceans—An overview: *Marine Geology*, v. 70, p. 9–42.
- SLINGERLAND, R., KUMP, L. R., ARTHUR, M. A., FAWCETT, P., SAGEMAN, B. B., and BARRON, E. J., 1996, Estuarine circulation in the Turonian Western Interior Seaway of North America: *Geological Society of America Bulletin*, v. 108, p. 941–952.
- STABELL, B., 1989, Initial diatom record of Sites 657 and 658: On the history of upwelling and continental aridity, *in* Ruddiman, W., Sarnthein, M., and others, *Proceedings of the Ocean Drilling Program, Scientific Results: Ocean Drilling Program, College Station, Texas*, v. 108, p. 149–156.
- STEIN, R., TEN HAVEN, H. L., LITKE, R., RULLKÖTTER, J., and WELTE, D. H., 1989, Accumulation of marine and terrigenous organic carbon at upwelling Site 658 and nonupwelling Sites 657 and 659: Implications for the reconstruction of paleoenvironments in the eastern subtropical Atlantic through late Cenozoic times: *in* Ruddiman, W., Sarnthein, M., and others, *Proceedings of the Ocean Drilling Program, Scientific Results: Ocean Drilling Program, College Station, Texas*, v. 108, p. 361–385.
- SUMMERHAYES, C. P., 1981, Organic facies of middle Cretaceous black shales in the deep North Atlantic: *American Association of Petroleum Geologists*, v. 65, p. 2364–2380.
- , and MASRAN, T. C., 1983, Organic facies of Cretaceous and Jurassic sediments from DSDP Site 534 in the Blake-Bahama Basin, western North Atlantic, *in* Sheridan, R. E., and Gradstein, F. M., and others, *Initial Reports of the Deep Sea Drilling Project: U.S. Government Printing Office, Washington*, v. 76, p. 469–480.
- TAGGERT, J. E., JR., LINDSAY, L. R., SCOTT, B. A., VIVIT, D. V., BARTEL, A. J., and STEWART, K. C., 1987, Analysis of geologic materials by wavelength-dispersive X-ray fluorescence spectrometry, *in* Baedeker, P. A. (ed.), *Methods for Geochemical Analysis: U.S. Geological Survey Bulletin 1770*, p. E1–E19.
- THIERSTEIN, H. R., 1979, Paleooceanographic implications of organic carbon and carbonate distribution in Mesozoic deep-sea sediments, *in* Talwani, M., Hay, W. W., and Ryan, W. B. F. (ed.), *Deep Sea Drilling Results in the Atlantic Ocean: Continental Margins, and Paleoenvironment: American Geophysical Union, Washington, Maurice Ewing Series*, v. 3, p. 249–274.
- TIEDEMANN, R., SARNTHEIN, M., and STEIN, R., 1989, Climatic changes in the western Sahara: Paleo-marine sediment record of the last 8 million years (Sites 657–661), *in* Ruddiman, W., Sarnthein, M., and others, *Proceedings of the Ocean Drilling Program, Scientific Results: Ocean Drilling Program, College Station, Texas*, v. 108, p. 241–261.
- TIMOFEEV, P. P., EREMEEV, V. V., and RATEEV, M. A., 1978, Palygorskite, Sepiolite, and other clay minerals in Leg 41 oceanic sediments: Mineralogy, facies, and genesis, *in* Lancelot, Y., and Seibold, E., and others, *Initial Reports of the Deep Sea Drilling Project: U.S. Government Printing Office, Washington*, v. 41, p. 1087–1101.
- TISSOT, B. P., and WELTE, D. H., 1984, *Petroleum Formation and Occurrence*, 2nd ed.: New York, Springer-Verlag, 538 p.
- TUCHOLKE, B. E., and MOUNTAIN, G. S., 1979, Seismic stratigraphy, lithostratigraphy, and paleosedimentation patterns in the North American Basin, *in* Talwani, M., Hay, W. W., and Ryan, W. B. F. (eds.), *Deep Sea Drilling Results in the Atlantic Ocean: Continental Margins, and Paleoenvironment: American Geophysical Union, Washington, Maurice Ewing Series*, v. 3, p. 58–86.
- , and VOGT, P. R., 1979, Western North Atlantic: Sedimentary evolution and aspects of tectonic history, *in* Tucholke, B. E., Vogt, P. R., and others, *Initial Reports of the Deep Sea Drilling Project: U.S. Government Printing Office, Washington*, v. 43, p. 791–825.
- VAN HINTE, J. E., WISE, S. W., JR., and others, 1987, Initial Reports of the Deep Sea Drilling Project, v. 93: U.S. Government Printing Office, Washington, 1423 p.
- VON RAD, U., and ARTHUR, M. A., 1979, Geodynamic, sedimentary, and volcanic evolution of the Cape Bojador continental margin (NW Africa), *in* Talwani, M., Hay, W. W., and Ryan, W. B. F. (eds.), *Deep Sea Drilling Results in the Atlantic Ocean: Continental Margins, and Paleoenvironment: American Geophysical Union, Washington, Maurice Ewing Series*, v. 3, p. 187–203.
- WALSH, J., PREMUIX, E. T., GAFFNEY, J. S., ROWE, G. T., HARBOTTLE, G., STOENNER, R. W., BALSAM, W. L., BETZER, P. R., and MACKO, S. A., 1985, Organic storage of CO<sub>2</sub> on the continental slope off the Mid-Atlantic bight, the southern Bering Sea, and the Peru coast: *Deep-Sea Research*, v. 32, p. 853–883.
- WAPLES, D. W., 1985, Organic and inorganic nitrogen in sediment from Leg 80, Deep Sea Drilling Project, *in* de Graciansky, P. C., Poag, C. W., and others, *Initial Reports of the Deep Sea Drilling Project: U.S. Government Printing Office, Washington*, v. 80, p. 993–998.
- WHITE, A. F., and BLUM, A. E., 1995, Effects of climate on chemical weathering in watersheds: *Geochimica et Cosmochimica Acta*, v. 59, p. 1729–1747.
- WINDOM, H. L., 1975, Eolian contributions to marine sediments: *Journal of Sedimentary Petrology*, v. 45, p. 520–529.

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