# 37. CRETACEOUS CLAY MINERALOGY OF THE CONTINENTAL RISE OFF THE EAST COAST OF THE UNITED STATES, SITE 603, DEEP SEA DRILLING PROJECT LEG 93<sup>1</sup>

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#### ABSTRACT

Cretaceous sediments encountered in Hole 603B contain a 17 Å smectite as the principal clay mineral, with lesser amounts of illite, kaolinite, and randomly interstratified illite/smectite (I/S). No evidence of smectite to I/S transformation with depth is seen in the interval between 1040 and 1570 m below the sediment surface. Although the presence of some volcanic ash suggests a volcanic origin for the smectite, the nature of associated organic matter together with prevailing sedimentological models for Cretaceous sedimentation on the lower continental rise argue more strongly for a dominantly terrestrial origin for the clays.

# INTRODUCTION

Site 603 is one of the three sites drilled on Leg 93 of the International Phase of Ocean Drilling of the Deep Sea Drilling Project. Three holes were drilled at this site in 4634-4639 m of water on the lower continental rise, 450 km east of Cape Hatteras, North Carolina (Fig. 1). Hole 603B was a reentry hole which terminated in Valanginian limestone at 1585.2 m below the sediment surface and permitted the recovery of 486.42 m of Lower Cretaceous to upper Pliocene sediment. Total sediment recovery at Site 603 was 1025.26 m, a record for any one DSDP site. Cretaceous sediments are encountered in lithologic Units III, IV, and V (Fig. 2) and are described as follows (after Site 603 chapter, this volume):

Plantagenet Formation: 97 m of Coniacian-Maestrichtian (?) variegated claystones. The unit is divided into three subunits by the presence or absence of dark gray carbonaceous claystones. Subunit IIIB contains organic-matter-rich claystones considered to be turbiditic in origin, and contains the uppermost Cretaceous sediments studied in this chapter (Section 603B-22-2 to 603B-33-1).

Hatteras Formation: 106 m of Aptian-Turonian black carbonaceous claystones. This unit is subdivided by the relative occurrence of pelagic, reddish brown claystones and black, carbonaceous turbiditic claystones (Sections 603B-33-1 to 603B-44-1).

Blake-Bahama Formation: 261 m of Berriasian-Aptian interbedded nannofossil clays and limestones with sandstone to claystone turbidites. This unit is subdivided by the presence or absence of turbidites. Subunit VA contains abundant claystone, siltstone, and sandstone turbidites. Subunit VB is composed of pelagic carbonates containing laminated and bioturbated nannofossil chalks and limestones (Sections 603B-44-1 to 603B-82-3).

In all, 33 clay samples representing Berriasian to Maestrichtian (?) sediments were analyzed by powder X-ray diffraction to identify and determine semiquantitatively the clay mineral composition.

## ANALYTICAL METHODS

Samples were treated with warm 1M acetic acid solution for 24 hr. to remove CaCO<sub>3</sub>. Three to four successive decantations and resuspensions were needed to prevent further flocculation. Dispersed samples were then allowed to settle according to Stokes' Law. The  $<2-\mu m$  fraction was removed by pipette and concentrated by high-speed centrifugation. The resultant paste was smeared on petrographic "well" slides with a 30- $\mu m$  etched depression in the center. This insures that all samples are the same size and thickness when presented to the X-ray beam.

The analyses were performed on a GE/Diano XRD-5 powder diffractometer upgraded with NIM electronics. CuKa radiation, Ni filtration, and 1° Soller slits were used with a scan rate of both 2°/min. and 0.4°/min. The slower scan rate is necessary to distinguish between the chlorite and kaolinite peaks near 25° 20. Samples were run from 2° to  $35^{\circ} 2\theta$  under three conditions: air-dried, ethylene glycol solvated, and heated to 375°C. Interpretations followed the methods of Brown and Brindley (1980). Illite is a nonexpanding mica with a 10.1 Å periodicity; kaolinite has a 7 Å periodicity and is distinguished from chlorite by its 002 maximum at 3.58 Å; and smectite expands to ~17 Å with glycolation and collapses upon heating. A typical clay mineral diffractogram is shown in Figure 3. Some of the material identified as smectite may included some randomly interstratified (RO) illite/smectite (I/S). Reynolds (1980) has shown that the differences in diffraction patterns between pure smectite and random I/S with small quantities of illite are quite subtle. X-ray patterns of randomly interstratified I/S with high expandabilities characteristically included a broadened 17 Å peak, high background, and a slight shift in the peak near 5.6 Å.

Semiquantitative estimates of clay abundance were made following the method of Biscaye (1965). Smectite, illite, and kaolinite peak areas were multiplied by appropriate weighting factors and summed to represent the total clay fraction. Each component was then divided by the total to compute its representative portion. The accuracy of this method is probably no greater that 10%.

#### RESULTS

Clay-rich Cretaceous sediments from Hole 603B contain a clay mineral suite dominated by smectite with lesser amounts of kaolinite and illite. Smectite is present in all samples except one and, where present, varies between 41 and 92% of the total clay fraction (Table 1 and Fig. 2). Illite is present in all samples and varies between 7 and 31%, and kaolinite is present in most samples, in proportions varying between 1 and 70%, although typically it is less than 20%. Smectite is relatively well crys-

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Figure 1. Location of Site 603 on the lower continental rise east of Cape Hatteras, North Carolina.

tallized and gives a strong diffraction maximum on the glycolated tracing in the range 16.9-17.3 Å. In some tracings the 17 Å peak is broadened and poorly defined, and peak at 5.6 Å is very slightly shifted toward lower spacings, suggesting the presence of a small amount of randomly interstratified I/S (Reynolds, 1980). There does not appear to be any systematic variation in smectite versus I/S with depth, however, and for purposes of this chapter, I/S will be included under the heading of smectite. Previous studies of burial diagenesis of clayey sediments (Perry and Hower, 1970; Hower et al., 1976) have shown that some smectites will convert through randomly interstratified mixed-layer I/S to a regularly interstratified I/S at elevated temperatures if sufficient potassium is available in the system. No such trends were seen in this study, perhaps because the depth of burial was insufficient to drive the reaction beyond its very earliest stage.

The downhole variation in illite shown in Figure 2 is nonsystematic and shows no evidence of association with any *in situ* formation mechanism among the clay mineral suite. Chamley et al. (1983) noted the appearance of certain "illite" events in Mesozoic clays of the Blake-Bahama Basin and in a general way they seem to be reflected in the sediments of Hole 603B, but without the accompanying chlorite. The most prominent events occur in the middle Hauterivian, the middle Barremian, and the Campanian; they have been interpreted as related to renewed erosion along continental margins during episodes of seafloor spreading. They apparently are correlated with high sedimentation rates and abundant resedimentation structures and suggest a morphological rejuvenation of the continental margins that is possibly caused by aperiodic tectonic modifications, especially in connection with the major seafloor spreading stages (Chamley et al., 1983).

Moderately to well-crystallized kaolinite occurs in varying amounts in samples of early Hauterivian through Late Cretaceous age. Its greatest abundance is in the Plantagenet Formation (Fig. 2), where it reaches a maximum of 70% of the clay mineral suite, and is accompanied by a relatively high illite concentration. It is most likely terrestrial in origin and would also appear to coincide with an Atlantic-wide event described by previous authors (Chamley et al., 1983; Chamley and Robert, 1979). It is interpreted as the consequence of the principal spreading stage responsible for the development of deep-water circulation and initial contact between the North and South Atlantic.

## DISCUSSION AND CONCLUSIONS

The Cretaceous sedimentary column in Hole 603B preserves both continental and pelagic sediments. The former are represented by thick, massive, distal turbidites,



Figure 2. Stratigraphy and  ${<}2\text{-}\mu\text{m}$  clay mineralogy of Cretaceous core samples from Hole 603B.

and the latter by laminated marls, bioturbated chalks, and limestones. Earlier interpretation of clays of similar age in the Blake-Bahama Basin (Chamley et al., 1983) assign a terrestrial origin to them, based on both sedimentologic and mineralogic considerations. Hole 603B clays are also for the most part probably land-derived. The frequent occurrence of mud-rich distal turbidites (Sarti and von Rad, this volume), commonly without evidence of bioturbation, together with an abundance of terrigenous-type organic matter (Herbin et al., this volume) strongly suggests that much of the clay mineral suite originated through erosion of soils developed along the continental margin. Although smectite commonly occurs as a byproduct of the submarine alteration of volcanic ash, only a single, thin ash bed of late Albian age was recognized in the section studied. Occasional traces of zeolites may also indicate volcanic contributions but



Figure 3. X-ray powder diffraction tracing of the  $<2-\mu m$  fraction of Sample 603B-26-3, 30-34 cm, saturated with ethylene glycol. Peaks for smectite (S), illite (1), and kaolinite (K) are typical of those encountered in this study.

Table 1. Clay minerals in Hole 603B.

Core-Section (interval in cm)	Depth (m)	$<2 \ \mu m$ clay fraction (%)		
		Smectite	Illite	Kaolinit
24-2, 30-34	1040.42	68	10	22
26-3, 30-34	1059.92	72	11	17
30-2, 81-85	1092.73	_	30	70
32-1, 130-133	1110.82	41	31	28
34-2, 5-8	1129.07	77	23	
34-4, 125-127	1133.26	86	14	-
34-5, 119-122	1134.71	82	16	2
34-5, 133-136	1134.85	78	11	11
36-2, 100-105	1148.03	77	12	11
38-3, 45-50	1166.98	87	7	6
40-2, 105-110	1184.08	85	9	6
42-3, 50-55	1200.73	84	16	-
44-3, 71-74	1218.13	69	13	18
48-2, 105-109	1255.37	82	11	7
50-1, 91-95	1272.93	85	3	7
52-1, 83-85	1292.04	75	3	2
52-1, 123-126	1292.45	92	6	2
52-2, 21-23	1292.92	64	20	16
54-2, 96-100	1312.88	76	21	3
56-3, 22-27	1332.85	86	5	9
60-4, 36-40	1372.88	83	10	7
62-3, 70-73	1390.92	83	15	2
64-3, 16-19	1408.88	80	19	1
66-5, 70-75	1430.43	77	21	2
68-4, 57-59	1446.78	76	23	1
68-4, 98-100	1447.19	72	28	-
68-4, 113-115	1447.34	Insufficient data		
70-2, 62-73	1461.91	76	14	10
74-1, 53-58	1492.85	88	12	_
76-1, 77-80	1512.28	76	24	-
78-4, 2-6	1535.24	86	13	1
80-2, 35-40	1551.08	90	10	-
82-3, 47-52	1570.70	82	18	-

Note: Dash indicates mineral is absent from sample.

they occur infrequently in the clay fraction, and could possibly be diagenetic. Chamley (1979) proposes that the Cretaceous climate was characterized by high temperature and contrasting humidity in areas of low relief. The absence of chlorite is consistent with that interpretation.

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#### REFERENCES

- Biscaye, P. E., 1965. Mineralogy and sedimentation of recent deep-sea clay in the Atlantic Ocean and adjacent seas and oceans. *Geol. Soc. Am. Bull.*, 76:803–832
- Brown, G., and Brindley, G. W., 1980. X-ray diffraction procedures for clay mineral identification. *In Brindley*, G. W., and Brown, G. (Eds.), *Crystal Structures of Clay Minerals and Their X-ray Identification:* London (Min. Soc. London), pp. 305–359.
- Chamley, H., 1979. North Atlantic clay sedimentation and paleoenvironment since the Late Jurassic. In Talwani, M., Hay, W., and Ryan, W. B. F. (Eds.), Deep Drilling Research in the Atlantic Ocean: Continental Margins and Paleoenvironment. Am. Geophys. Un., Maurice Ewing Ser., 3:342-361.

- Chamley, H., Debrabant, P., 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., Gradstein, F. M., et al., *Init. Repts. DSDP*, 76: Washington (U.S. Govt. Printing Office), 437-448.
- Chamley, H., and Robert, C., 1979. Late Cretaceous to early Paleogene environmental evolution expressed by the Atlantic clay sedimentation. In Christensen, W. K., and Birkelung, T. (Eds.), Proc. Cretaceous-Tertiary Boundary Events Symp. Copenhagen (Vol. 2): 71-77.
- Hower, J., Eslinger, E. V., Hower, M. E., and Perry, E. A., 1976. Mechanism of burial metamorphism of argillaceous sediment: 1. Mineralogical and chemical evidence. *Geol. Soc. Am. Bull.*, 87: 727-737.
- Perry, E. A., and Hower, J., 1970. Burial diagenesis in Gulf Coast pelitic sediments. Clays Clay Min., 18:165-177.
- Reynolds, R. C., 1980. Interstratified clay minerals. In Brindley, G. W., and Brown, G. (Eds.), Crystal Structures of Clay Minerals and Their X-ray Identification: London (Min. Soc. London), pp. 249-303.

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