36. CLAY MINERALOGY OF CRETACEOUS AND CENOZOIC SEDIMENTS OFF THE MOROCCAN MARGIN, DEEP SEA DRILLING PROJECT SITES 415 AND 416

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ABSTRACT

The Upper Jurassic to Pleistocene, mainly turbiditic sediments recovered at Sites 415 and 416 on the Moroccan margin contain diversified and variegated clay assemblages. Most of the clay species and associated non-clay minerals are considered to be detrital materials, including smectite, fibrous clays, and perhaps part of the zeolite (clinoptilolite) and cristobalite. The Tithonian and Lower Cretaceous turbidites are chiefly marked by the presence of micaceous and chloritic minerals, resulting from tectonic rejuvenations during the oceanic widening. Minor variations in the abundance of illite, kaolinite, and mixed-layer clays could have been caused by climatic variations. Most of the Upper Cretaceous and Paleogene sediments contain abundant well-crystallized smectite, mainly inherited from continental soils developed in poorly drained areas. This suggests that the climate was hot, with strong variations in seasonal humidity. Fibrous clays developed during two periods, attapulgite (= palygorskite) during the early Aptian, attapulgite and sepiolite during the early Eocene. Both minerals indicate the existence of hot and seasonally humid climates which changed northward and southward. Their occurrence or increase in abundance shows the existence of marginal, closed to semi-closed basins in which they formed in confined chemical conditions and were reworked as a result of the margin instability. During the late Cenozoic, the increase of primary minerals and the reappearance of mixed-layer clays and associated non-clay minerals reflect a progressive and irregular worldwide cooling, unfavorable for establishment of soils and favoring direct erosion of rocks and the development of oceanic currents. This change chiefly began in the late Miocene, but it began earlier off Morocco than off southern Europe, suggesting an arid trend along northwest Africa as long ago as the Oligocene.

INTRODUCTION

Leg 50 of the Deep Sea Drilling Project drilled two sites off the coast of Morocco (see Site Chapters, this volume). Site 415 was located in the southern part of the Moroccan Basin (31°01.72'N, 11°39.11'W) in a water depth of 2794 meters; Hole 415 was spot cored down to the lowermost Miocene at 283 meters beneath the sea floor. Hole 415A bottomed in the upper Albian at 1079.5 meters, and Hole 415B reached the lower Pliocene at 64 meters. Six lithologic units were recognized, mainly comprising marly ooze, marlstone, claystone, and chalk. Site 416 was drilled in the northern part of the Moroccan Basin, near Site 370 (Leg 41) at 32° 50.18'N and 10°48.06'W, in 4191 meters water depth. The pilot hole, Hole 416, penetrated less than 20 meters of upper Cenozoic sediments (upper Pliocene at base). Hole 416A reached the Tithonian (Upper Jurassic) at 1624 meters beneath the sea floor. The lithology, combining both Site 370 and 416 data, is divided into seven lithologic units characterized by numerous mudstone to sandstone turbidites, especially in the Cretaceous sequence, which is about 900 meters thick.

METHODS

We studied the clay mineralogy of 21 samples from Site 415, and 76 samples from mainly the Cretaceous series of Site 416. The less than 2 μ m decalcified particles of all samples were analyzed by X-ray diffraction. We also studied some of the less than 8 μ m fraction by transmission electron microscopy.

In the X-ray-diffraction method the samples were disaggregated in water, then decarbonated in 5N hydrochloric acid. The excess acid was removed by successive centrifugations. The samples were deflocculated by microhomogenization and the 2 μ m fraction was collected by decantation on the basis of Stokes Law.

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Oriented aggregates were made on glass slides. A CGR θ 60 diffractometer (copper radiation focused by a quartz curved-crystal monochromator) was used to run the X-ray diffraction scans at 1°/min. A receiving slit of 1.25 mm allows a better determination of mixed-layer minerals. Four passages were carried out as follows: (1) from 1° to 15° θ on natural sample; (2) from 1° to 7° θ on glycolated sample; (3) from 1° to 7° θ on sample heated for two hours at 490°C; (4) from 12° to 14° θ on hydrazine-hydrated sample. Semiquantitative evaluations were made on the basis of the peak heights and areas (Chamley, 1971). The height of 001 illite and chlorite peaks (diagram of glycolated sample) were taken as references. Kaolinite, smectite, attapulgite, sepiolite, and irregular mixed-layer mineral contents were corrected by comparison with this value (by addition to a subtraction from the peak height). The relative abundance of chlorite and kaolinite was obtained from the ratio of their peak heights (respectively at 3.54 and 3.58 Å): when this ratio is 1, the amount of chlorite was assumed to be twice as much as that of kaolinite. Final data are given in percentages, the relative error is about ± 5 per cent.

Note that the same samples were used also for geochemical studies so that we could establish some correlation between mineralogy and geochemistry (Debrabant et al., this volume).

RESULTS

Site 415 (Figure 1)

Upper Albian to Cenomanian, Cores 415A-14 to 7: The shale and calcarenite of Unit VIc, the marlstone with interbeds of shale, dolomite, and limestone of Unit VIb, and the pelagic clay of Unit VIa are characterized by very abundant and well-crystallized smectite and small amounts of illite and kaolinite. The presence of at-tapulgite (= palygorskite) in very small amounts is clearly revealed by electro-microscopy. This fibrous mineral is fairly abundant in Sample 415A-10-1, 78 cm. The mineralogical composition of the upper Albian to Cenomanian sediments is not related to the lithological zonation.

Paleocene, Cores 415A-6 to 5: The marlstone of Unit V and mudstone of Unit IV were very poorly sampled. The clay fraction contains, in addition to abundant smectite, about 10 per cent attapulgite accompanied by cristobalite and clinoptilolite. The early Eocene mudstone of Core 415A-1 was not analyzed.

Lower to middle Miocene, Cores 415-5 to 3: The alternating nannofossil chalk and marl of units III and II have comparable amounts of illite and smectite. Chlorite and irregular mixed-layered illite-smectite and chlorite-smectite minerals occur in the clay fraction in addition to quartz and feldspars and amphiboles. Attapulgite occurs in small amounts along with traces of sepiolite in Sample 415-5-6, 27cm. Kaolinite occurs throughout but is very rare.

Upper Miocene, Pliocene, Pleistocene, Cores 415-2 and 1, Core 415B-2: The uppermost sequence of the hole (nannofossil marl and ooze, Unit I) contains abundant illite and fairly abundant chlorite. Smectite is relatively poorly represented (15-20%) of the clay fraction). Accessory minerals are irregular mixed-layer clays (5%), kaolinite (5%), attapulgite (traces to 5%), along with quartz, feldspars, amphiboles and, in some places, goethite.

Site 416 (Figure 2; back pocket, this volume)

Tithonian, Berriasian, Valanginian, Cores 416A-57 to 416A-33: About 225 meters of turbidites were sampled in Unit VII (alternating quartzose siltstone and mudstone sequences with hard calciturbidites) and in the lower part of Unit VI (graded calcareous and quartzose sequences of siltstone or fine sandstone to mudstone). This highly variable lithology is marked by a moderately variable clay mineralogy. Generally, illite and smectite are the dominant species, the former often predominating. Chlorite, irregular mixed-layer clays (chiefly illite-smectite, and in places chlorite-smectite), kaolinite and quartz occur throughout, whereas feldspars, amphiboles, and goethite are less common. Chlorite is generally more abundant than kaolinite. Moderate variations in the relative abundances of the clay minerals occur throughout the interval, but they do not relate to the color of non-calcareous sediments, which ranges from red to greenish gray. Minor mineralogical variations appear to relate to the general stratigraphy: illite is more common in the Upper Jurassic sediment (up to 70% of the clay fraction), kaolinite increases in the Berriasian (from traces to 5%), and both kaolinite and irregular mixed-layer clays increase in the lower part of the Valanginian section (10% each).

Valanginian, Hauterivian, Cores 416A-32 to 7: The turbidites of Unit VI are more than 500 meters thick. They were well sampled from the upper Valanginian, and poorly sampled from the Hauterivian samples. They contain abundant illite, degraded chlorite, mixed-layer chlorite-smectite and illite-smectite, and kaolinite. Chlorite and kaolinite are present in similar proportions (10–20%). The smectite of the sequence is the most poorly crystallized and least abundant of the whole site. Numerous moderate variations in relative abundance of the minerals occur as in the underlying layers, favoring, in places, illite and chlorite (i.e., Sample 416A-11-3, 9 cm; Plate 1, Figure 5), and, in places, smectite and kaolinite (i.e., Sample 416A-12-1, 147 cm; Plate 1, Figure 6).

Aptian, Albian-Cenomanian lithological Unit V (claystone with some siltstone and fine sandstone): One sample in Core 416A-6 contains about 10 per cent attapulgite in the clay fraction, in addition to the minerals identified below. Chlorite is present in traces only, illite being the dominant mineral.

Lower Eocene, lower part of Unit III (turbidites as graded sequences of mudstone, siltstone, sandstone, and gravel with porcellanite): The uppermost part of Core 416A-5 (5-1, 32 cm) radically differs from underlying and overlying sediments. Nearly the whole clay fraction consists of very long, electron-transparent and sometimes flexuous fibers of sepiolite and attapulgite. Other clay minerals are very sparse, comprising traces of illite and chlorite (Plate 1, Figures 3 and 4). Dolomite is probably present in idiomorphic rhombohedrons.

Upper Oligocene to Pliocene-Pleistocene, Core 416-1 and Cores 416A-3 to -1: Rather poorly sampled, the uppermost parts of Holes 416 and 416A consist of turbidites of sandstones, nannofossil marl and chalk (Unit II, upper Oligocene to upper Miocene), and nannofossil marl and nannofossil ooze (Unit I, Pliocene to Pleistocene). The clay fraction is characterized by the reappearance of abundant smectite in the Oligocene sediments, accompanied by small amounts of various other clay minerals: illite, chlorite, mixed-layer clays, kaolinite, attapulgite, and sepiolite. Toward the top of the series illite and chlorite irregularly increase, whereas smectite and fibrous clays decrease or even disappear (sepiolite) (Plate 1, Figures 1 and 2). Quartz and feldspars are common; amphiboles and goethite occur in some of the more recent sediments.

DISCUSSION

Our purpose is to compare the results of Sites 415 and 416, and to discuss their significance relative to previous data obtained from Legs 47A, 47B, and 48, also located on the northeast Atlantic margin (Chamley and Giroud d'Argoud, in press; Chamley and Diester-Haass, in press; Chamley et al., in press; Debrabant et al., in press).

Smectite is the prevailing clay mineral in the middle Upper Cretaceous, sampled at Site 415 only. The same mineral is dominant in the clay fraction of the middle to Upper Cretaceous sediments drilled on outcrops on the northeast Atlantic margin (i.e., Aubry and Pomerol, 1975; Estéoule-Choux and Louail, 1976) and on the northwest Atlantic margin (i.e., Zemmels, et al., 1972; Pastouret et al., 1978). Smectite also prevails in Paleogene sediments which were poorly sampled during Leg 50, but are well represented in cores from other DSDP sites on the African margin (i.e., Berger and von Rad, 1972; Mélières, 1978). The abundance and the crystallinity of smectite are not related to the gross lithology of the rock and the mineral is not associated with the presence of volcanic glass or ash, except on the Rockall Plateau south of the Iceland Sea (Debrabant et al., in press). The chemical data show abundant Al, Fe, Ti, V, Ca, and Li and do not indicate that the smectite was derived from volcanic sources. At most sites, including those of Leg 50, the suite of rare-earth elements is typical of continental shale or soil (Courtois and Chamley) and does not indicate marine or volcano-hydrothermal intervention during the genesis of the smectite and other clay minerals. The relationships among smectite, zeolites, and cristobalite are very poor; zeolite minerals are generally of a clinoptilolite type, which frequently formed during early diagenesis of volcanic materials (Sommer, 1972; see also Chamley et al., in press). Thus smectite is chiefly thought to be contributed in the detrital supply from adjacent landmasses, especially from continental soils and perhaps sediments formed in the poorly drained areas widespread during Cretaceous times. Such an origin supposed a rather hot and arid climate

with strong seasonal variation in humidity, as is present today in some warm Mediterranean to tropical African areas (Paquet, 1969).

Two periods are marked by a large supply of illite, accompanied by chlorite, irregular mixed-layer clays, quartz, feldspars and, at times, amphiboles. The most recent period is the late Cenozoic. At both Sites 415 and 416, illite and associated minerals greatly increase in upper Miocene sediments, as they do in other North Atlantic sites. This general phenomenon is attributed to world cooling during the Cenozoic, beginning in the late Eocene or early Oligocene and progressing irregularly until the late Pleistocene. Major stages of cooling occurred in the late Miocene during significant enlargement of the Antarctic ice cap (Kennett, Houtz, et al., 1975), and in the Pliocene and Pleistocene during the development of the Arctic ice cap (Berggren, 1972). The effect of such a cooling on the continental clay mineral assemblages is the reduced formation of smectite- or kaolinite-rich soils, and increased erosion and transport toward the ocean of primary minerals which had been partly and moderately transformed into irregular mixed-layer clays (Millot, 1964; Chamley, 1971). Indirectly, the world cooling favored the development of meridianal marine currents responsible for the influx of illite and associated primary minerals from the northern latitudes into middle and low latitudes (Griffin et al., 1968). Note that the pre-late-Miocene increase of primary minerals off the Moroccan margin is greater than in the sites drilled off the European margin (Legs 47B and 48). For example, the average percentages of illite and chlorite in lower and middle Miocene are as follows:

Site	Location	Illite (%)	Chlorite (%)
400	Northern Bay of Biscay	14	2
398	Western Iberian margin	15	3
415	West Moroccan margin	32	6

This variation could be caused by a lithologic variation in the outcrops furnishing the detrital material. The Pyrenean and Armorican rocks, however, are as rich in micas and chlorites as the North African rocks. Another possible cause is local tectonic rejuvenation (Dillon and Sougy, 1974), but similar events probably would have affected the detrital sedimentation on the Iberian and the North African margins. A more probable cause is a transition to a desert-type climate in northwest Africa beginning in the Paleogene in northwest Africa. Comparable climatic trends have occurred in this area since at least the late Miocene (Diester-Haass, 1976; Chamley et al., 1977; Chamley and Diester-Haass, in press; Diester-Haass and Chamley, 1978). Their effect could have been to regionally inhibit alteration of primary clay minerals and favor their preservation and transport toward the ocean.

The second sequence, characterized by primary minerals, occurs in the uppermost Jurassic and the Lower Cretaceous of Site 416 and corresponds to rapid and highly variable turbiditic sedimentation. Such a large







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and lasting supply of micaceous and chloritic material associated with quartz, feldspars, and amphiboles could have had several causes. Climatic cooling, whose intensity was similar to that of the late Cenozoic, is one possible cause. The climatic data available for the Early Cretaceous, however, do not support this possibility. On the contrary, the average temperature during this period was probably rather high. Furthermore, the extreme climatic changes necessary to produce the great abundance of primary minerals also should have caused similar mineralogical changes in deposits in the large oceanic areas. Such changes do not exist. For example, the abundance of primary minerals does not increase off the Iberian peninsula, in the uppermost Jurassic to lowermost Cretaceous sediments of Site 398, or at most DSDP sites drilled at (Cretaceous) latitudes comparable to that of Site 416 on the western North Atlantic (i.e., those drilled during Legs 11 and 44; Zemmels et al., 1972; Chamley, unpublished; Pastouret et al., 1978). Thus the event which caused the marked mineralogical peculiarities of Site 416 must have been relatively restricted. A tectonic rejuvenation of the northwestern African margin would have produced such an effect, for instance, by marginal physiographic compensation owing to major phases of oceanic rifting (Montadert et al., in press). Note that the minor quantitative variations of clay species and associated minerals within the Upper Jurassic and Lower Cretaceous series were caused either by slight changes in the depositional environment, clearly demonstrated by the highly variable lithology, or by minor changes in the continental climate (i.e., humidity increasing from Tithonian until Hauterivian, as suggested by the increase in kaolinite).

The amount of fibrous clays within two intervals of the Hole 416A sequence markedly increases. The first is marked by the occurrence of attapulgite. Considered to be lower Aptian (Core 416A-6; Site 416 Report, this volume), it also corresponds to Cores 415A-10 to 415A-5 (Cenomanian to Miocene). The attapulgite appears in the Albian sediments of Sites 398 and 400 (Legs 47B and 48) in the northeast Atlantic. The Leg 50 attapulgitebearing Cretaceous sediments are probably the same age. The second fibrous-clay interval, assigned to the lower Eocene (Site 416 report, this volume, Sample 416A-5-l, 32 cm), is characterized by attapulgite and sepiolite. Both fibrous minerals are very abundant in the Eocene sediments of the African emerged basins (Millot, 1964; Chamley, 1971) and they have also been found in other DSDP sites of the east Atlantic margin (Berger and von Rad, 1972; Mélières, 1978). In both intervals the fibrous clays occur in reworked and frequently turbiditic sediments. They are widely dispersed among the typical detrital species and are more or less broken. Thus we consider them to be detrital minerals, similar to the other clay species, deposited under the same conditions as were the post-Eocene fibrous minerals present off the African continent (i.e., Section 416A-3-1 and Core 416-1; see also Chamley and Millot, 1975; Froget and Chamley, 1977). Attapulgite and sepiolite were probably inherited from marginal basins, where they had been formed, perhaps together with

clinoptilolite and cristobalite, in alkaline chemical environments. They were then redeposited during periods of marginal instability and subsidence (see also Weaver and Beck, 1977) to their present sites. The formation of the fibrous minerals on the periphery of the African shield would have required very hydrolyzing climates with distinct dry seasons (Millot, 1967) and the existence of closed or semi-closed marginal basins. The relative abundance of fibrous clays decreases northward from the Moroccan margin in lower-Eocene sediments, as shown below:

Site	Location	Attapulgite (%)	Sepiolite (%)
400	Northern Bay of Biscay	4	1
398	Western Iberian margin	7	32
416	Western Moroccan margin	43	53

We found also the same trend south of the Moroccan margin. This suggests that during the Eocene the latitude of North Africa corresponded to a continental zone marked by very strong hydrolysis and thus defined the approximate location of the Eocene equatorial zone. Such an opinion agrees with global tectonic reconstructions (Pautot and LePichon, 1973) and suggests the possibility of using detrital clay minerals as indicators to locate and track the migration of former latitudes.

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PLATE 1 Site 416, Electron Photomicrographs

- Figure 1 Sample 416-1-1, 33 cm (15,900 ×), Pliocene/Pleistocene. Rather well-shaped illite and chlorite lamellae. Rare attapulgite in short and broken fibers.
- Figure 2 Sample 416A-3-3, 39 cm $(20,500 \times)$, Oligocene. Abundant cloudy smectite. Common attapulgite and sepiolite in broken fibers.
- Figures 3, 4 Samples 416A-5-1, 32 cm, lower Eocene. Very abundant fibrous clays (undifferentiated attapulgite and sepiolite) in long, electron-transparent and sometimes flexible fibers. Small amounts of phyllites (illite, chlorite), rhombohedrons (dolomite?), and coccoliths (non-decalcified material). Figure 3, 12,500×; Figure 4, 25,000×.
- Figure 5 Sample 416A-11-3, 9 cm $(10,000 \times)$, Hauterivian. Sample rich in illite and chlorite.
- Figure 6 Sample 416A-12-1, 147 cm $(10,000 \times)$, Hauterivian. Sample rich in cloudy, small smectite sheets and in imbricated hexagons of kaolinite.

CLAY MINERALOGY



PLATE 1