19. CLAY SEDIMENTATION AND PALEOENVIRONMENT IN THE AREA OF DAITO RIDGE (NORTHWEST PHILIPPINE SEA) SINCE THE EARLY EOCENE

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ABSTRACT

The Cenozoic history of the Daito Ridge area, as expressed by clay-mineral assemblages recovered from Deep Sea Drilling Project Sites 445 and 446, appears to have been influenced mainly by three factors: volcanism, tectonics, and climate. Volcanism was strong during the early Eocene; it decreased after the late Eocene and even more after the late Oligocene, with a probable temporary recurrence during the early Pliocene. The ridge was considerably emergent at the beginning of the Eocene. Uplift during the middle Eocene was followed by irregular subsidence from the late Eocene and to the present. They are now small emergent remnants (Daito Islands). The climate was probably warm and conducive to hydrolysis during the Paleogene; irregular cooling began in the latest Oligocene, with probable major stages of cooling during the middle Miocene and, chiefly, since the late Miocene.

INTRODUCTION

During DSDP Leg 58 in the north Philippine Sea, two sites were drilled in the area of Daito Ridge, in the western part of the basin (Figure 1). Site 445 (water depth 3377 m) is in a small basin on the Daito Ridge; the oldest cored sediments are middle-Eocene sandstone and conglomerate. The cored sedimentary sequence is subcontinuous, with only one marked hiatus, which represents the early Oligocene. The sequence consists of very coarse-grained to fine-grained resedimented Eocene clastic sediments (slumps, turbidites, debris flows); latest-Eocene to early-Miocene chalks and cherts; and early-Miocene to Plio-Pleistocene clayey nannofossil and foraminifer ooze.

Site 446 (Holes 446 and 446A; water depth 4952 m) is south of the Daito Ridge, in the Daito Basin. The oldest cored rocks consist of an alternation of early-Eocene mudstone and claystone and dark-gray, aphyric basalt (Site 446 report, this volume). The entire sequence comprises thick Eocene claystone to coarse sandstone, interrupted in the lower part by ash and 26 weakly altered basalt sills; Eocene to Pliocene pelagic clay and mud; and Pliocene terrigenous clay and mud. Numerous hiatuses occur, especially at the Eocene/Oligocene and Pliocene/Pleistocene boundaries.

METHODS

All samples (124 from Site 445, 93 from Site 446) were subjected to X-ray-diffraction analysis on decarbonated particles smaller than 2 μm. We observed some fractions smaller than 8 μm by transmission electron microscopy.

The X-ray-diffraction method is as follows. The samples are dissociated in water, then decarbonated in 5*N* hydrochloric acid. Excess acid is removed by successive centrifugations. Microhomogenization effects deflocculation. The fraction smaller than 2μm is collected by decantation, using Stokes' law; then oriented aggregates are made on glass slides. A GGR θ 60 diffractometer (copper radiation focused by a quartz curved-crystal monochromator) is used to run the X-ray-diffraction scans at 1°/min. The 1.25-mm receiving slit allows better determination of mixed-layer minerals. Four passages are carried out: (1) from 1° to 15° θ on natural samples; (2) from 1° to 7° θ on glycolated samples; (3) from 1° to 7° θ on samples heated for 2 hours at 490°C and (4) from 12° to 14° θ on hydrazine-hydrated samples. Semi-quantitative evaluations are based on peak heights and areas (Chamley, 1971). The height of illite and chlorite 001 peaks (diagram for glycolated sample) are taken as references. By comparison with these values, values for smectite, attapulgite, sepiolite, vermiculite, and irregular mixed-layer clays are corrected by addition of peak height, whereas values for well-crystallized kaolinite are corrected by subtraction. The relative proportions of chlorite and kaolinite are determined from the ratio of peak heights (respectively 3.54 and 3.58 Å): when this ratio is 1, the amount of chlorite is assumed to be twice that of kaolinite. Data are given in percentages, the relative error being about ±5 per cent.

Illite crystallinity values correspond to the 1/10°-θ breadth of the 10-Å peak at half-height. Relative abundance of smectite corresponds to the ratio of 18 and 10 Å peak heights. Both these measurements are made on the X-ray diagram for the glycolated sample.

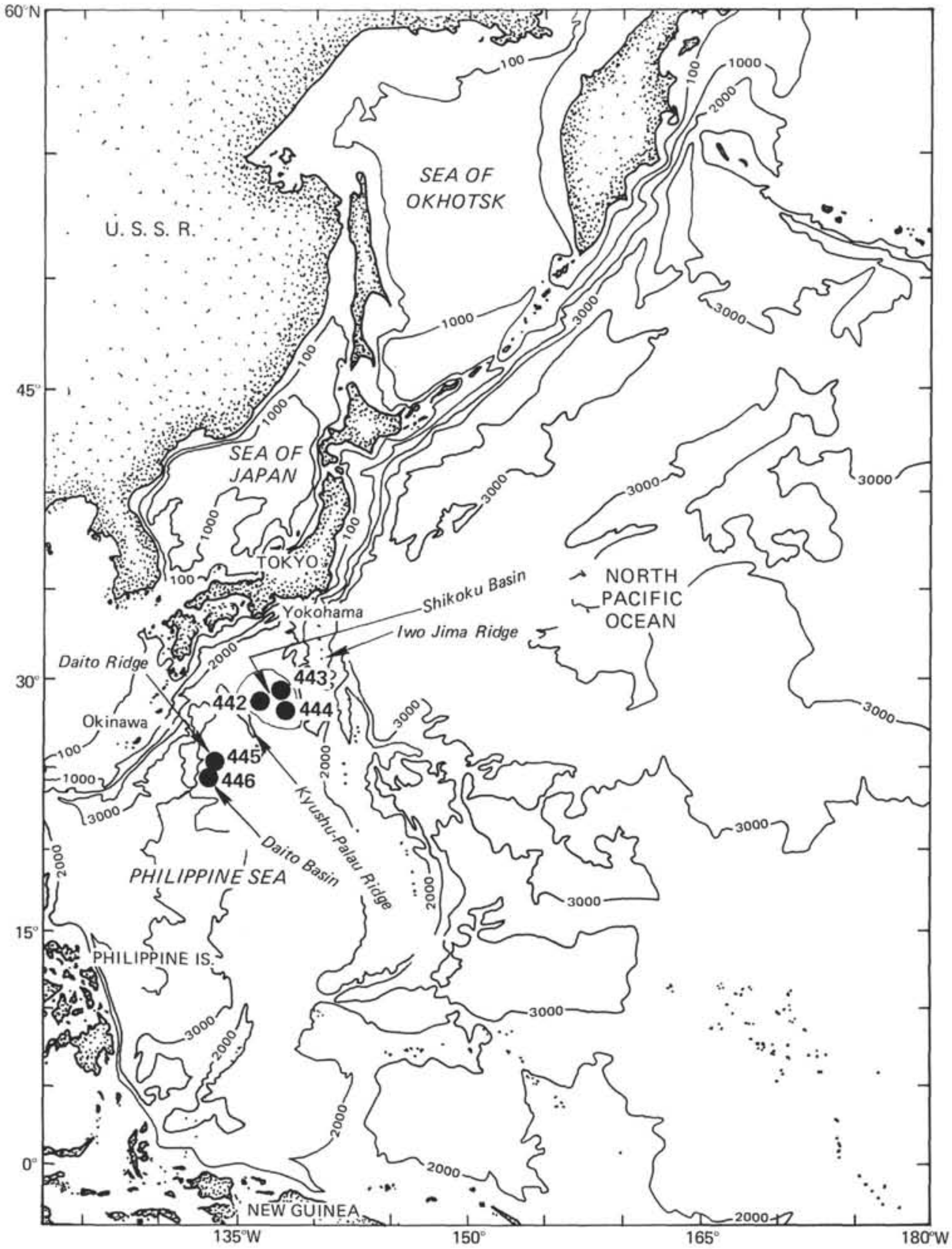

Figure 1. *Location of DSDP Leg 58 sites* (Daito Ridge sites underlined).

RESULTS

Site 445

Three main mineralogical zones are recognized for Site 445 (Figure 2)

1. Middle-Eocene greenish-gray to greenish-brown graded and reworked sequences, cherts, and siliceous ooze (Cores 445-94 to 445-65). Abundant well-crystallized smectite (80–90% of clay minerals) is accompanied by chlorite (maximum 15%) and serpentine of antigorite type (maximum 20%) (Plate 1, Figures 2–4), the amounts of chlorite and serpentine varying independently. Illite is absent or rare (except in Sample 445-69-2, 138 cm, where it forms 15% of clays). Associated minerals are ubiquitous plagioclase, opal-CT in siliceous sediments and mud (Cores 445-74 to 445-65), and zeolites of the heulandite type (Cores 445-79 to 445-68).

2. Late-Eocene to middle-Oligocene gray to yellowish, in part clayey nannofossil chalk (Cores 445-64 to 445-48). Well-crystallized smectite is very abundant (up to 100%), associated with minor amounts of illite and (or) chlorite, and kaolinite from Core 445-56 upward. Non-clay minerals are absent, except rare to common feldspar.

3. Late-Oligocene to Pleistocene brown to yellowish-gray, clayey nannofossil chalk and calcareous ooze (Cores 445-47 to 445-1). The clay assemblage is a mixture of various species, dominated by moderately well-crystallized smectite or illite (Plate 1, Figure 1). The characteristic minerals of this section are irregular

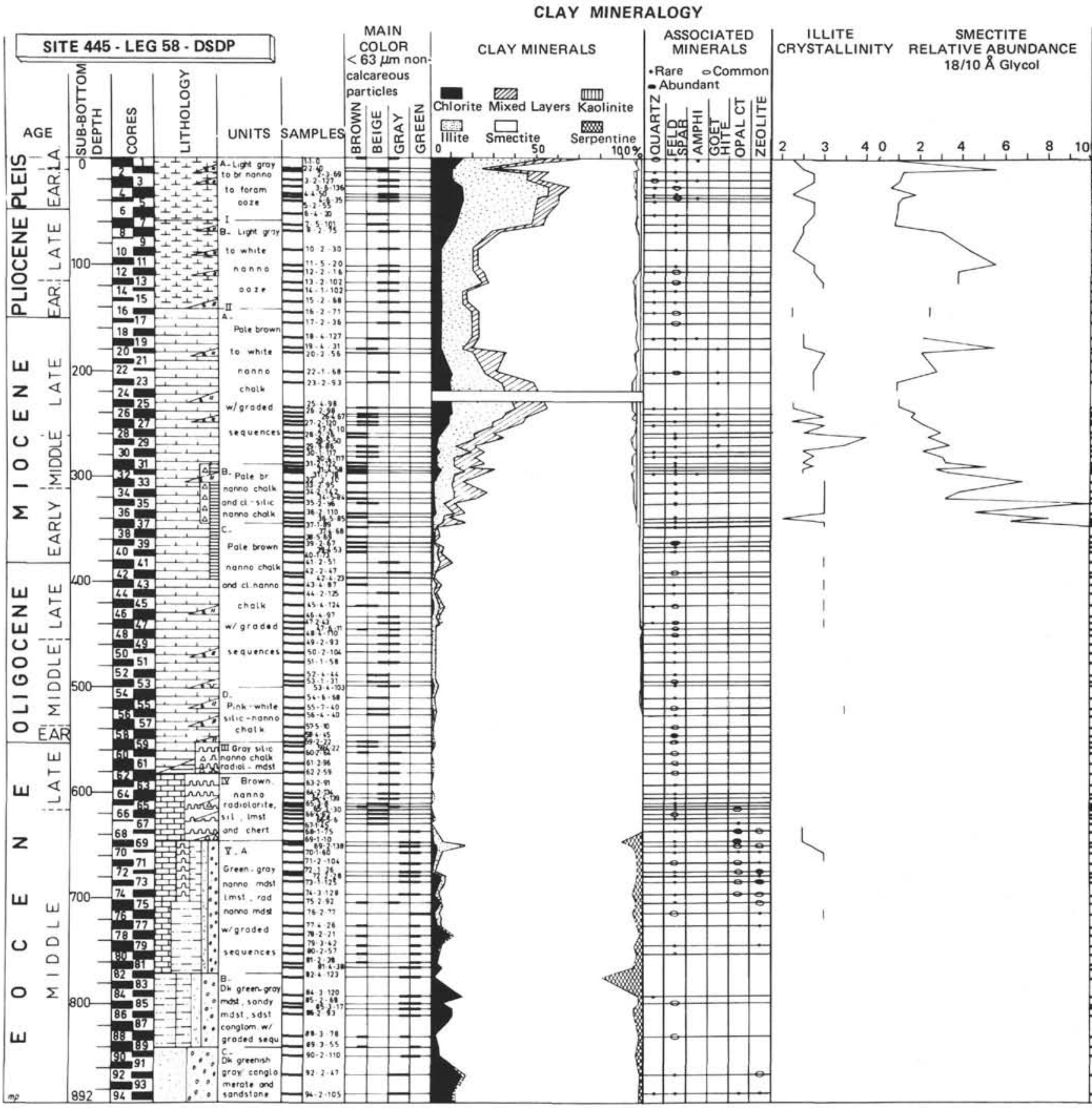

Figure 2. *Clay mineralogy of sediments at Site 445, Daito Ridge area.*

mixed-layer clays which include illite–smectite and chlorite–smectite types. Chlorite, kaolinite, and feldspar occur throughout; accessory minerals are quartz, amphiboles, and goethite. This third mineralogical zone is noteworthy in several respects.

First, there is no clay-mineral distinction between the fine-grained and coarse-grained sediments, which indicates the absence of mineralogic grain sorting.

Second, mixed-layer clays, chlorite, illite, and quartz appear in the latest Oligocene (Core 445-47) and increase upward; intervals of rapid increase occur near the end of the early Miocene (Core 445-37), in the middle Miocene, and chiefly in the late Miocene (from Core 445-28). The maximum contents of clays and associated minerals correspond to the late Miocene (Cores 445-27 to 445-20) and to the late Pliocene and Pleistocene (Cores 445-8 to 445-1). Late-Miocene sediments locally contain very small amounts of vermiculitic mixed-layer clays (illite–vermiculite).

Third, the Pliocene includes a rather long interval of smectite-rich sediments, slightly extending into the latest Miocene (Cores 445-20 to 445-10). More-restricted increases in smectite content exist in ash layers, especially in Sample 445-2-2, 40 cm, where gray, fine pumice fragments show an exceptional enrichment in smectite.

Site 446

Three main mineralogical zones are likewise recognized for Site 446 (Figure 3).

1. Early-Eocene calcareous, nannofossil-bearing, glauconitic, ashy, brownish to bluish-gray claystone and mudstone, interbedded with basalt sills (Cores 446A-28 to 446A-6, and Cores 446-44 to 446-40). Smectite is very abundant and well crystallized, often the only clay mineral identified. Chlorite and illite are present in variable amounts. Biotite from a vesicular basalt of kersantite type was found in Sample 446A-10-5, 86 cm. Mixed-layer clays (probably from glauconite granules) are rare, and serpentine occurs at some levels, in sediments or basalts. Associated non-clay minerals include common to abundant feldspar, frequent zeolites (increasing at sandy levels), rare quartz, and opal CT.

There is no systematic dependence of clay mineralogy on rock type (sediment or basalt) (Figure 4). The two rock types, a few centimeters apart, show slightly different clay-mineral assemblages. Baked sediments do not show any sign of clay change or homogenization. The mineralogical range for sediments is wider than that for basalts, and sediments are characterized by greater amounts of non-smectite clay minerals.

2. Middle-Eocene to late-Eocene terrigenous to hemipelagic greenish-gray claystone and mudstone, with sporadic siliceous fossils and ash (Cores 446-39 to 446-16). Well-crystallized smectite is very abundant (often more than 95% of clay minerals) and accompanied by small quantities of chlorite or serpentine, feldspar, and heulandite. The main part of this zone corresponds closely chronologically and mineralogically, to the lowest zone of Hole 445. However, these reworked sediments at Site 446 are finer grained than the sediments at Site 445, which explains the smaller proportion of coarse-grained minerals such as serpentine and chlorite (Figures 2 and 3). The late-Eocene to middle-Oligocene sediments of the middle mineralogical zone of Site 445 can hardly be compared to contemporaneous sediments of Site 446, because of the large number of hiatuses at Site 446.

3. Oligocene to Pliocene, hemipelagic brown and yellow mud and clay with local ashy or siliceous levels (Cores 446-15 to 446-1). The clay assemblage becomes diversified in this interval, although smectite is still predominant, probably because of the absence of Pleistocene sediment. Illite is fairly abundant to abundant in the youngest sediments, and chlorite and kaolinite are present. As in the upper mineralogical zone of Site 445, irregular mixed-layer clays (chiefly illite–smectite, small amounts of chlorite–smectite) are the typical species, accompanied by quartz and feldspar.

As at Site 445, an irregular increase in primary minerals and little-degraded species (Mixed-layer clays) occurs upward at Site 446, and sporadic well-crystallized smectite appears in pumiceous, fine-grained, white ash layers (Sample 446-13-4, 39 cm). The smectite of these ash layers is authigenic and grows as "tufts" at the periphery of pumice fragments (Plate 2, Figures 3 and 4). Hydrous manganese oxides (todorokite) and serpentine (antigorite) occur sporadically in early-Miocene sediments, and there are possibly small amounts of sepiolite (Plate 2, Figures 1 and 2).

INTERPRETATIONS

The main inferences from vertical mineralogical changes observed at Sites 445 and 446 concern the origin of the clayey sediments and the history of the Daito Ridge area.

Early Eocene

Smectite is very abundant in the clay fraction of alternating basalt sills and early-Eocene sediments cored at Site 446. The smectite in the basalts is obviously authigenic as it is in numerous submarine volcanic rocks (see, for instance, Peterson and Griffin, 1964; Seyfried et al., 1978). The smectite is formed by alteration of the subvitric rock, especially in its most vesicular and fissured parts; similar processes locally produce chlorite, zeolites, or serpentine. The origin of the smectite of the interbedded sediments is a problem. This probably results also from an alteration of volcanic materials, as suggested by the fact that abundance and crystallinity are similar in the sediments and in the basalts, and by its general environment (strong volcanic activity). However, if the smectite is derived from volcanic rocks, is it authigenic or allogenic? These observations are pertinent:

1. The basalts are not contemporaneous with the sediments; they are younger intrusives, as shown by baked sediments both above and below the sills (Site 446 report, this volume).

2. The basalts generally show little or moderate clay alteration. Considering the great extention of the clay-rich sediments, smectite from the basalt sills is clearly insufficient to supply surrounding sediments. Further evidence against such a source are the very low porosity of the clay-rich sediments and the excellent preservation of primary sedimentary structures (Klein et al., this volume).

3. At basalt contacts the sediments are baked only through a few centimeters, as a result of fast cooling of the sills and the very high resistance of the clay-rich materials to thermal and chemical changes. The clay mineralogy of these baked sediments is not significantly different from that of the non-baked sediments.

4. Except in a few pumiceous layers, the clay mineralogy does not depend on the presence or the preservation state of volcanic glass or ash; thus, diffusion of smectite from altered-ash layers to sediments is very improbable, considering the low porosity of the clay-rich sediments and the preservation of primary structure.

5. Accessory minerals associated with smectite are more diverse and more abundant in sediments than in interbedded basalts, which obviously indicates the existence of an allogenic component in the sediments (Figure 5).

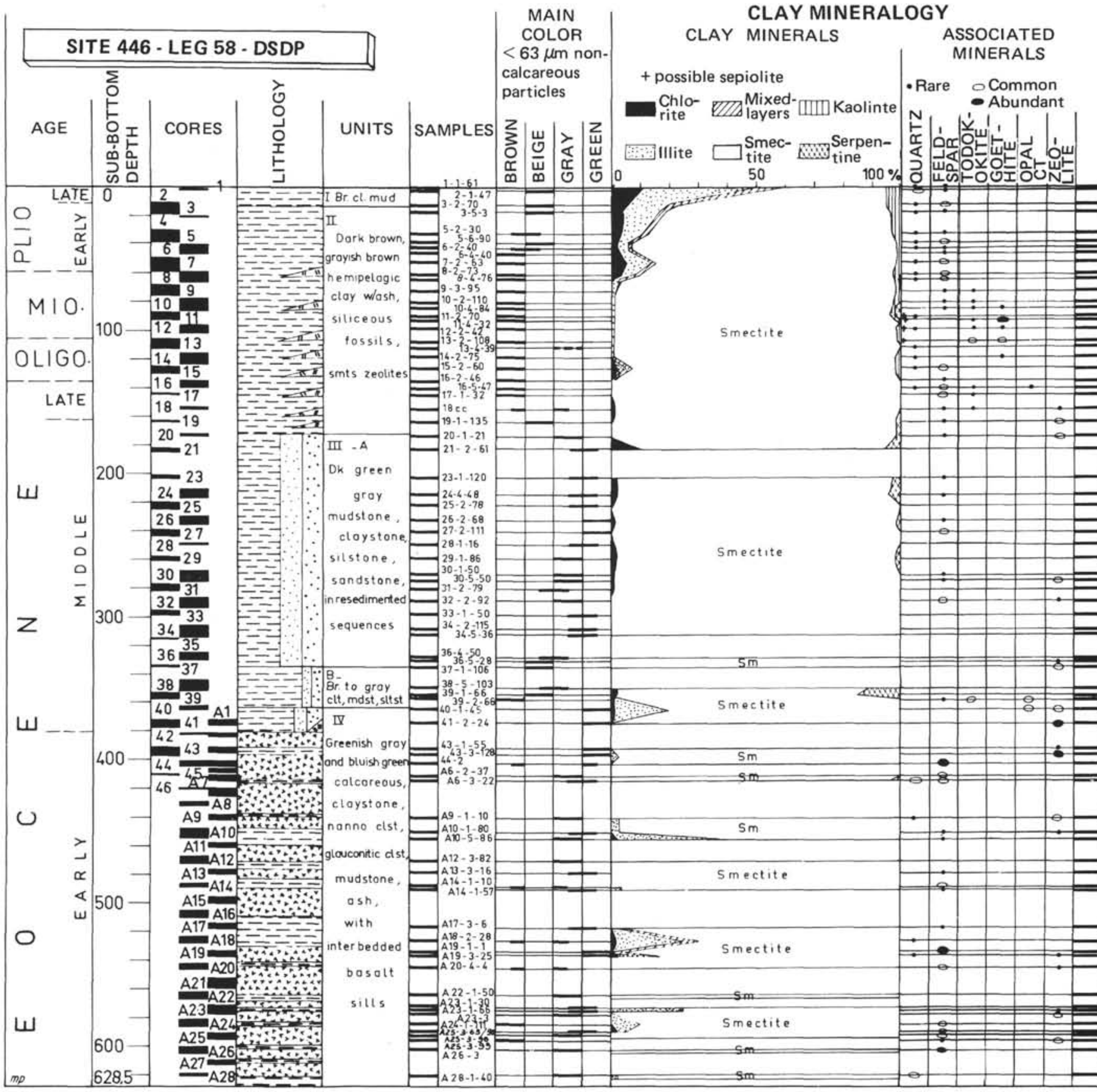

Figure 3. *Clay mineralogy of sediments at Site 446, Daito Ridge area.*

6. The sediments interlayered with the basalt sills contain typically resedimented minerals such as quartz, glauconite, and heavy minerals, and they show various structures of strong currents which do not favor *in situ* chemical evolution: ripples, laminae, slumps, graded-beds (Klein et al., this volume). Evidences of resedimentation are present also in detrital post-early-Eocene sediments, whose clay mineralogy is very similar to that of the early-Eocene sediments.

7. The high sedimentation rate suggested by active resedimentation argues against surficial chemical exchanges between sediment and sea water, which could induce mineralogical changes. A lack of early diagenetic transformation is supported by the disequilibrium of rare-earth elements with sea water (C. Courtois, pers. comm.), which strongly differs from the *in situ* smectite formation in typical pelagic sediments of the Pacific (Courtois and Hoffert, 1977).

Therefore, whereas the clay fraction of the basalt sills was formed by *in situ* alteration, the clay fraction of interbedded sediments chiefly represents detritus eroded from volcanic rocks of neighboring island arcs during early-Eocene time. This assumes strong volcanic activity and widespread emergence of volcanic rocks in the Daito Ridge area, probably contemporaneous with the development of the Philippine back-arc basin (Klein et

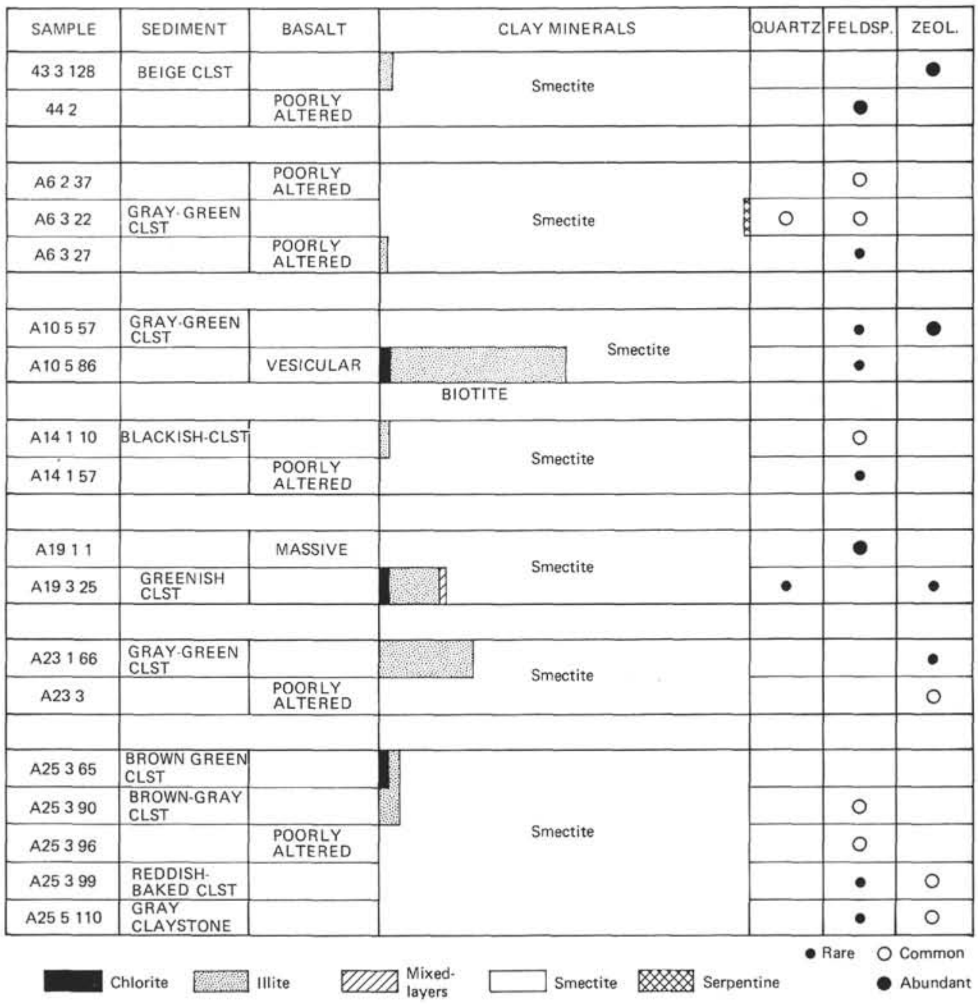

Figure 4. *Clay mineralogy of selected samples from Eocene sediments and basalt interlayers of Site 446.*

al., this volume; Karig and Moore, 1975) The warm climate prevailing during Eocene time in many areas of the world (Furon, 1972) favored degradation of emergent rocks and formation of smectite; this would have been intensified in the Daito Ridge area, because it was situated at subequatorial latitudes (Kinoshita et al., this volume).

Middle Eocene To Middle Oligocene

Sediments of the middle Eocene to the middle Oligocene are marked by very large amounts of smectite, which occurs in reworked sediments. As in the early-Eocene sediments, smectite here was transported from adjacent island arcs, where it formed by subaerial alteration of volcanic rocks under a warm climate. The high sedimentation rate, the detrital and reworked sediments, the absence of basalt intrusions and of progressive change in mineralogy with depth of burial, and the independence of total smectite and the quantity of volcaniclastics indicate that the mineral assemblages are mainly allogenic. The following history is suggested by the clay succession:

From the middle Eocene to the middle Oligocene, the Daito Ridge area still was influenced strongly by island-arc volcanic activity.

In the middle Eocene, the association of smectite with noticeable amounts of minerals such as chlorite, serpentine and, occasionally, quartz indicates source rocks other than basalts. This is particularly evident at Site 445, which is closer to Daito Ridge than Site 446 and characterized by coarser sediments. Rock fragments dredged in this area (Mizuno et al., 1977) and recovered from Site 445 middle-Eocene deposits include various schists, ultrabasic rocks, greenstones, and basalts and microdolerites of alkaline affinity (Tokuyama et al., this volume) which incorporate the aforementioned minerals in the clay size fraction. Thus, compared to the early Eocene, there was either a slight decrease in volcanic activity, permitting significant erosion of older, non-basaltic rocks on the Daito Ridge structure, or uplift of the whole area, leading to subaerial erosion of the old basement of the ridge. The abundance of coarse rock fragments in the middle Eocene favors the second hypothesis.

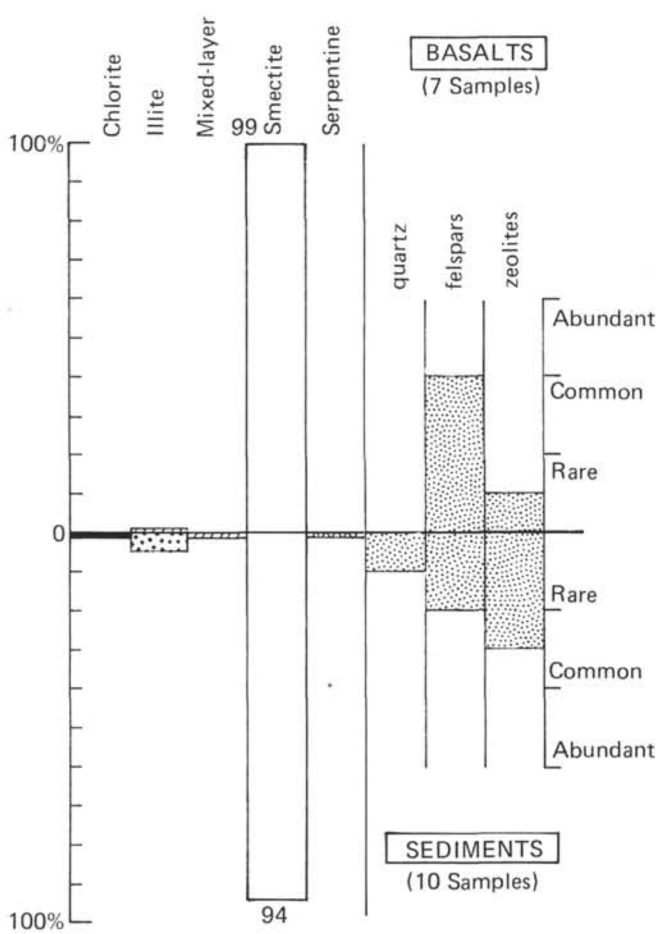

Figure 5. *Clay minerals and accessory minerals of the clay fractions of sediments and interlayered basalts, Site 446.*

From the late Eocene to the middle Oligocene, a recurrence of almost exclusively smectitic clay supplies indicates either recrudescence of volcanic activity or partial subsidence of the Daito Ridge area. The transition from a coarse sediments (angular lithic fragments, large fragments of *Nummulites*) to finer deposits suggests progressive subsidence of the Daito Ridge, as described for many aseismic ridges (Thiede et al., 1975). Mizuno (1978) indicates a subsidence of the Daito Ridge of about 1.5 km from the middle Eocene to the Quaternary. Volcanic activity was probably still great, as shown by the relative abundance of glass particles and ash beds in this interval (Klein et al., this volume).

The development of cherts, radiolarian sediments, and zeolites in the middle Eocene does not correspond to any peculiar characteristics in clay mineralogy. Early diagenetic processes, including those which produced todorokite, were unable to modify the assemblage of clay minerals.

Late Oligocene to Pleistocene

The upper mineralogical zone at Site 445 and contemporaneous sediments at Site 446 correspond to a general diversification of the clay-mineral assemblage (Figure 2 and 3). Chlorite, illite, mixed-layer clays, and kaolinite originated from southeastern Asia and island arcs, as did a noticeable part of the medium to poorly crystallized smectite (Kobayashi et al., 1964; Aoki and Oinuma, 1974; Aoki et al., 1974; Chamley, this volume). This mineralogical diversification has two main implications:

1. The arrival into the Daito Ridge area of sediments from distant sources implies the exchange of large masses of water. This supposes a general subsidence of the aseismic ridge, probably accompanied by a decrease in local volcanic activity. This subsidence, suspected to have begun during the middle Eocene, corresponds to finer-grained, more-pelagic, and less-disturbed deposits and may have been caused by crustal cooling during northward migration of the ridge (Kinoshita et al., this volume; Klein et al., this volume). The decrease in volcanic detritus is conspicuous in this interval and chiefly resulted from submergence of the ridge. However, during the early Pliocene, the increased supply of well-crystallized smectite, which is recorded neither in the north Philippine Sea nor elsewhere in hemipelagic sediments (Chamley, this volume; Chamley, in press), could correspond to a temporary recrudescence of volcanic activity. This hypothesis is supported by relatively higher amounts of ash in this interval (Klein et al., this volume).

2. The increased detrital contribution (chlorite, illite, quartz, amphiboles) from crystalline and old sedimentary rocks and from moderately altered continental deposits (irregular mixed-layer clays, degraded smectite) is a world-wide phenomenon corresponding to general Cenozoic cooling (Chamley, in press). Conditions favoring hydrolysis decline with time after the late Paleogene, operating against formation of smectitic and kaolinitic soils and favoring both moderate alteration and direct erosion of rocks. Both mineralogical and palynological evidences of continental climatic changes in marine sediments are found in the Shikoku Basin as well as in the Daito Ridge area (Chamley this volume, Tokunaga, this volume). The first such evidence appears in the middle Miocene as elsewhere; (e.g., Curtis and Echols, 1975) and is very important in deposits younger than late Miocene (vermiculitic mixed-layer clays) which are influenced by development of the Antarctic ice cap (Shackleton and Kennett, 1975).

Figure 6 schematically summarizes the tentative deciphering of volcanic, tectonic, and climatic effects since the early Cenozoic, as deduced from changes in clay-mineral assemblages in the Daito Ridge area.

ACKNOWLEDGMENTS

I acknowledge the U. S. National Science Foundation and the IPOD-France organization for permitting me to participate on Leg 58 of the Deep Sea Drilling Project. I am grateful to the entire Leg 58 scientific and technical shipboard party for the good work done on board. Financial support of this study was provided by CNEXO (France) (grants 77/5489 and 78/5708). I gratefully acknowledge M. Acquaviva, J. Carpentier, F. Dujardin, C.-H. Froget, M. Picart, J.-P. Ripert, and M. Vieille for technical assistance, and P. Debrabant and C. Latouche for useful review and comments.

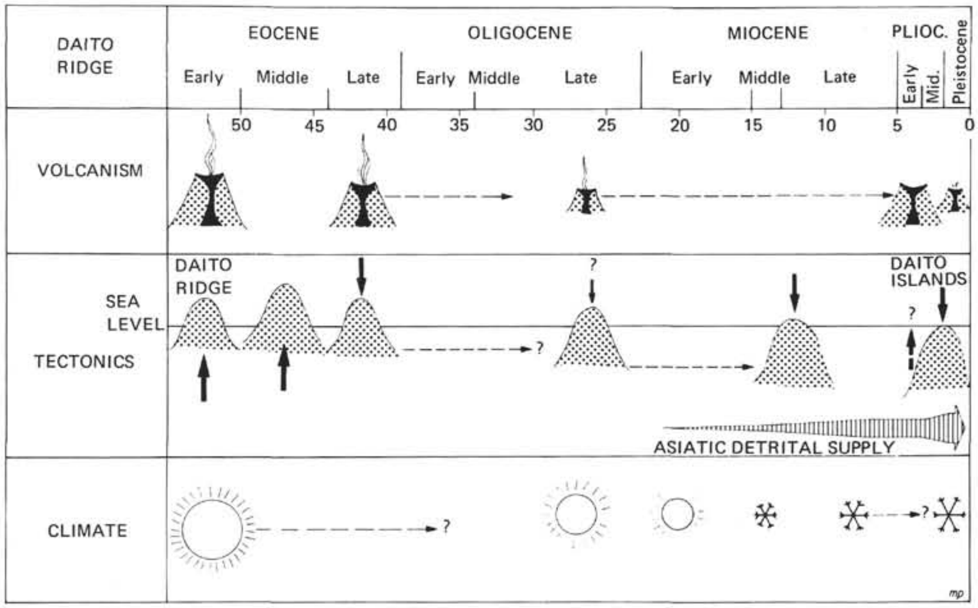

Figure 6. *Paleoenvironmental interpretation of changes in Cenozoic clay-mineral assemblages in the Daito Ridge area. Symbols are self-explanatory.*

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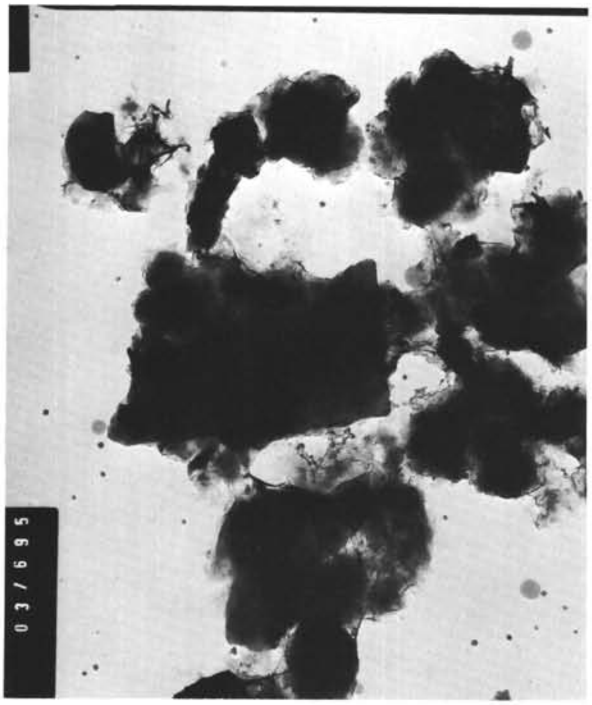
PLATE 1
Transmission Electron Photomicrographs of Samples from Site 445

Figure 1	Sample 445-11-5, 20 cm. Pliocene nannofossil ooze. Smectite, illite, chlorite.
Figures 2–4	Sample 445-69-2, 138 cm. Middle-Eocene silty claystone. Smectite, serpentine, illite.

PLATE 1



1 µm

2 µm

3 µm

4 µm

PLATE 2
Transmission Electron Photomicrographs of
Samples from Site 446

Figures 1, 2	Sample 446-11-2, 70 cm. Miocene brown clay. Smectite, serpentine, sepiolite (?).
Figures 3, 4	Sample 446-13-4, 39 cm. Oligocene white ash. Authigenic smectite as tufts at the periphery of fine pumice fragments.

PLATE 2