18. CENOZOIC CLAY-MINERAL STRATIGRAPHY IN THE SOUTH PHILIPPINE SEA, DEEP SEA DRILLING PROJECT LEG 59

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ABSTRACT

X-ray-diffraction studies on the <2-μm fractions of clay separates collected from cores of DSDP Leg 59 in the South Philippine Sea (Holes 447A, 449, and 450) indicate that clay components have remained fairly constant since the beginning of the Neogene. They consist primarily of $>80\%$ smectite with minor amounts of chlorite, illite, kaolinite, and possibly vermiculite. Variations in the smectite contents, crystallinity, and compositional interstratification are usually associated with fluctuations in the concentration of volcanic glass, which strongly suggests that the alteration of the glass introduced authigenic smectite into these sediments.

Anomalous biogenic and pelagic sedimentation during the middle Miocene at DSDP Sites 449, 199, and 53 in the South Philippine Sea may indicate a northward shift of the North Equatorial Counter Current over the area, probably related either to increased monsoonal circulation in the vicinity or to intensification of boundary-current flow during this time. A less probable hypothesis would be that these sites moved northward through the equatorial region during the middle Miocene.

INTRODUCTION

The major objectives of this study are to develop a Cenozoic clay-mineral stratigraphy for pelagic-clay units in the South Philippine Sea and from this information to infer the paleo- and modern oceanographic conditions that affected the mineralogic composition of the clays in these units. To accomplish this, 79 clay separates from Cenozoic pelagic clays from the West Philippine Basin and Parece Vela Basin are analyzed using X-ray-diffraction (XRD) methods. Samples are selected from Holes 447A, 449, and 450. A semiquantitative determination of the percentages of the clay minerals is made using a <2-μm fraction from each sample.

Clay mineralogic data gathered from these analyses are employed to build a stratigraphic history of the deposition of Cenozoic pelagic-clay units at the sites mentioned. This stratigraphic history includes discussion of modern and ancient clay-sediment sources and oceanic-circulation patterns affecting sedimentation in this region.

ANALYTIC METHODS

Sample Preparation

Seventy-nine samples, averaging 5 cc each, were suspended in deionized water by ultrasonic probe and chemically treated through a series of steps to remove carbonates, soluble salts, and amorphous material. The first chemical treatment was a solution (*p*H 5) of 0.1*N* sodium acetate and acetic acid, which dissolved any carbonates and soluble salts from the sample. The sediment was then washed and treated with a solution (*p*H 3) of 0.2*M* ammonium oxalate and 0.2*M* oxalic acid to dissolve amorphous Fe-hydroxides and oxyhydroxides. This method, devised by Schwertmann (1964) and discussed by Heath and Dymond (1977), was chosen over the standard sodium-dithionite leach of Mehra and Jackson (1960) because Schwertmann's solution does not attack Fe-rich smectite or crystalline goethite (Landa and Gast, 1973).

The standard hydrogen-peroxide treatment for oxidizing organics was deleted because its reaction with the less crystalline smectites would alter their X-ray properties (J. Butler, personal communication). Also, the samples contained no visible organic matter.

Finally, the <2-μm fraction of each sample was separated by settling through a water column containing deionized water and Calgon to minimize flocculation of the clay particles. The resulting clay slurries were treated with a 1.0*M* solution of MgCl (to saturate the clay exchange sites with Mg ions) and washed a final time. The slurries were then applied to glass slides, dried at room temperature, and X-rayed. The XRD equipment consisted of a Philips diffractometer with a copper tube and a nickel filter.

Mineral Analyses

The slides were X-rayed after air drying and again after treatment with ethylene glycol (to shift the smectite peak from 12.0–14.0Å/18.0Å after glycolation). Illite shows a peak at 10.0Å, and chlorite and kaolinite have coincident peaks at 7.0Å. These peaks are not affected by glycolation. A slow scan across 24° to 26° 2θ on glycolated samples, as outlined by Biscaye (1964), was used to determine the relative proportions of chlorite versus kaolinite. The 002-kaolinite reflection occurs at 3.59Å, whereas the 003-chlorite reflection appears at 3.54Å. If this method was inconclusive, the clay separates were heated to 270°C for 14 hours (Hawkins, 1970); the low temperature and long exposure were necessary because the glass slides used in this study were not Pyrex. The relative percentages of the clay minerals were calculated for each sample using techniques outlined by Biscaye (1965).

RESULTS

Site 447

Located on the western edge of the sedimentary apron west of the Palau-Kyushu Ridge in the West Philippine Basin (18°00.88′N; 133°17.37′E) under 6022 meters of water, Hole 447A penetrates 113.0 meters of sediment including 37.5 meters of pelagic clay that cap the sedimentary column. The clay unit consists of an undated surface layer, approximately 5.0 meters thick, overlying 32.5 meters of lower(?) Miocene and upper middle to lower upper Oligocene clay sediments. A 2.0-meter thick middle Oligocene variegated clay zone occurs below a depth of 85.0 meters within a volcaniclastic breccia unit.

The <2-μm fractions taken from samples at this site contain increasing percentages of smectite down-section (from 89–96%) in the upper 9.0 meters of the hole (Fig. 1). The major glycolated smectite peaks are broad and shifted 0.1° to 0.4° 2θ to the right of their normal position at 18.0Å, indicating compositional interstratification within the mineral lattice. Reflections less than 18.0Å result from the presence of randomly distributed layers of chlorite, illite, or possibly vermiculite within the smectite lattice. The glycolated smectite peaks showing reflections larger than 18.0Å indicate orderly repetition of some of these compositional layers within the smectite lattice. In this case, the major smectite peaks occur between 17.67 and 16.99Å with subpeaks at 21.0, 19.0, and 16.6Å. Chlorite, illite, and kaolinite decrease over this same interval, with chlorite being slightly more abundant than the other two clays. The presence of minor amounts of vermiculite is suspected in these samples but cannot be confirmed by K saturation and heating because of low-peak amplitudes and masking by chlorite and illite peaks.

Very little material was recovered from 9.0 to 28.0 meters. Below this relative void, in the interval from 28.0 to 37.5 meters (upper middle to lower upper Oligocene), smectite averages 90%, with a sharp increase to 98% at 33.0 meters and a subsequent decrease to 86% below this peak; XRD peak characteristics indicate that it also maintains the same interstratified lattice noted in samples from the upper sections.

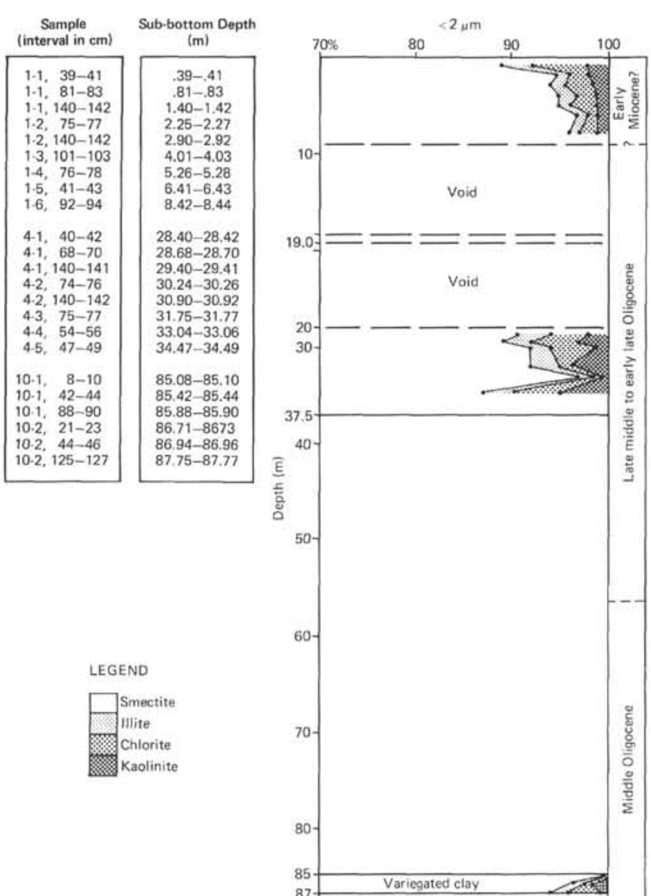

Figure 1. Clay mineralogy of the <2-μm fraction of samples from Hole 447A.

The middle Oligocene variegated clay unit at 85.0 meters is dominantly smectite, ranging from 91% to 94% down-section. Interstratification appears up-section in these lattices, as in the previously described clay units, but to a lesser degree. The major glycolated smectite peaks are narrower and occur at 17.67Å in these samples.

Site 449

This site is located on the western side of the Parece Vela Basin close to the eastern flank of the Palau-Kyushu Ridge (18°01.84′N; 136°32.19′E) in 4712 meters of water. It penetrates 111.0 meters of sediments, the upper 104.5 meters of which are nearly continuous pelagic clay. This sediment is undated from the surface to a depth of approximately 28.5 meters. The pelagic clay from this level down to 57.0 meters is middle Miocene and is interrupted by radiolarian ooze deposits from 40.9 to 47.5 meters. The clays are undated from 57.0 to 76.0 meters and are identified as being lower Miocene below this level down to the base of the unit.

Smectite dominates the <2-μm fraction of the sediment at this site, constituting at least 80% of the clay minerals in all samples (Fig. 2). The percentage of smectite increases fairly steadily down-section, from 81% near the surface to 98% at 29.0 meters. The glycolated smectite peaks are very broad and moderate in amplitude, indicating interstratification and low crystallinity. The interstratified layers (possibly illite or chlorite) cause a major peak at 17.0Å and one subpeak at 19.0Å, though the general peak extends from 16.0 to 20.0Å. Chlorite is slightly more abundant than illite or kaolinite over this same interval of undated sediments. Vermiculite is suspected in these samples as well, but because of low-peak amplitudes and masking of its peaks by chlorite and illite, it cannot be confirmed by K saturation and heating.

From 29 to 38 meters, smectite maintains a maximum of 98%, and chlorite and illite occur in fairly equal proportions with some kaolinite present. The smectite peaks for samples from this area have greater intensity, which indicates an increase in the crystallinity and reduction in the interstratification within the lattice. The major glycolated smectite peak is narrowed to 17.67Å with no subpeaks.

Smectite content decreases to 88% over the interval from 38.0 to 58.0 meters in middle Miocene sediments. Clay deposition is masked by a radiolarian ooze, which extends from 40.9 to 47.5 meters. The general glycolated smectite peaks for samples in or near the radiolarian ooze show fairly discrete subpeaks at 19.2, 18.4, and 16.9Å, with the major peak at 17.7Å. The subpeak at 18.4Å is unique to smectites in this region.

Chlorite is less abundant than illite and kaolinite in the sediments immediately above and below the ooze. Smectite constitutes nearly 100% of the clay component from 58.0 meters to the base of the pelagic clay unit at

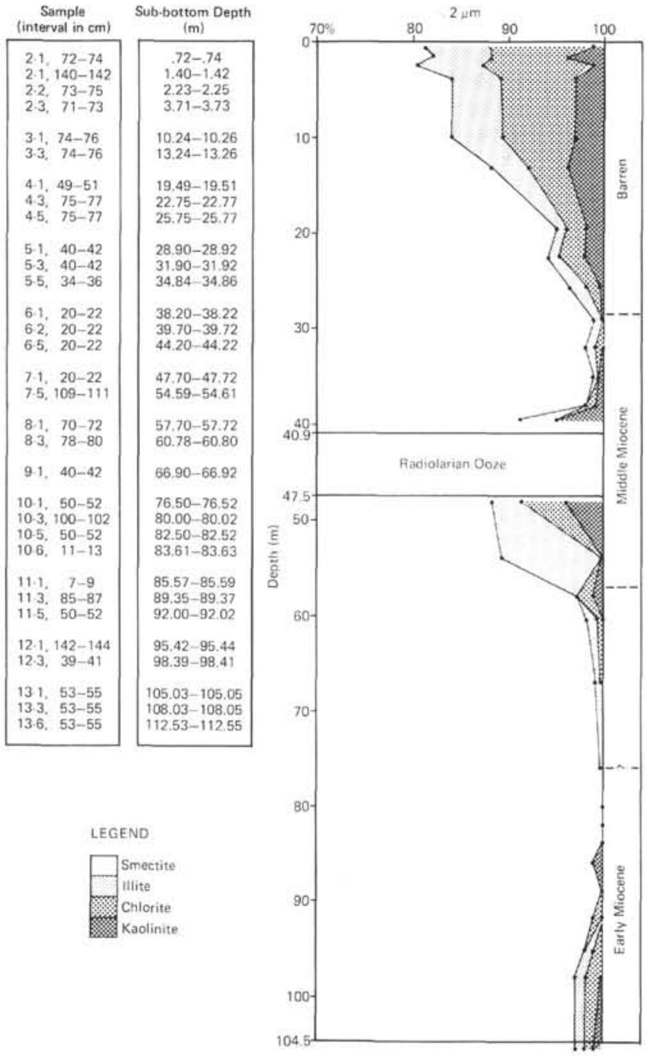

Figure 2. Clay mineralogy of the <2-μm fraction of samples from

104.5 meters in the lower Miocene sediments. Chlorite, illite, and kaolinite appear as a few scattered traces. The glycolated smectite peaks show increased intensity in samples from interval 66.90 to 89.37 meters. The peaks are symmetrical around a "d" spacing of 17.67 Å with no minor peaks, which indicates a decrease in interstratification. Smectites sampled from the base of the pelagic clay between 92.0 and 104.5 meters exhibit broad low-intensity peaks on glycolated samples. This marks a shift to less crystalline, more interstratified lattices. The major peaks occur between 17.67 and 16.9 Å, with distinct minor peaks at 19.2 and 21.5 Å.

Site 450

Located on the east side of the Parece Vela Basin in the sedimentary apron west of the West Mariana Ridge (18°00.02′N; 140°47.34′E) in 4707 meters of water, Hole 450 penetrates 330.0 meters of sediments, including 88.0 meters of near-surface pelagic clay. The lower portion of Core 1 is dated early Pleistocene, but below this zone down to 26.5 meters the sediments are undated. Upper Miocene pelagic clays occur from 26.5 to 45.5 meters; middle Miocene sedimentation continues from below 45.5 meters past the base of the pelagic clay unit to a depth of 88.0 meters. Two large coring gaps occur within the middle Miocene clays from 47.3 to 64.5 meters and from 67.3 to 93.0 meters. Volcanic ash layers become more abundant in the lower two-thirds of the clay unit.

The upper 2.0 meters of near-surface deposits contain the lowest smectite percentages found at this site—76% (Fig. 3). Glycolated samples from this unit show broad low-amplitude smectite peaks with the major reflection at 17.0 Å and strong subpeaks at 18.0 Å and 19.6 Å. This indicates an interstratified, poorly crystallized lattice structure.

In the undated underlying clays, smectite gradually increases to 96% at 11.0 meters and averages 94% from that point down to 22.0 meters. Glycolated smectite peaks from these samples indicate that peak intensity increases with depth, which could reflect both decreasing interstratification and increasing crystallinity in the clay lattice. Minor interstratification is indicated by a major glycolated peak between 17.67 and 17.37 Å with no subpeaks. A 3-meter interval (5.25–8.27 m) contains smectite that gives two equal and discrete glycolated reflections at 18.0 Å and 17.37 Å.

Below this zone to 40.4 meters in upper Miocene clays, the smectite content fluctuates sharply between 88

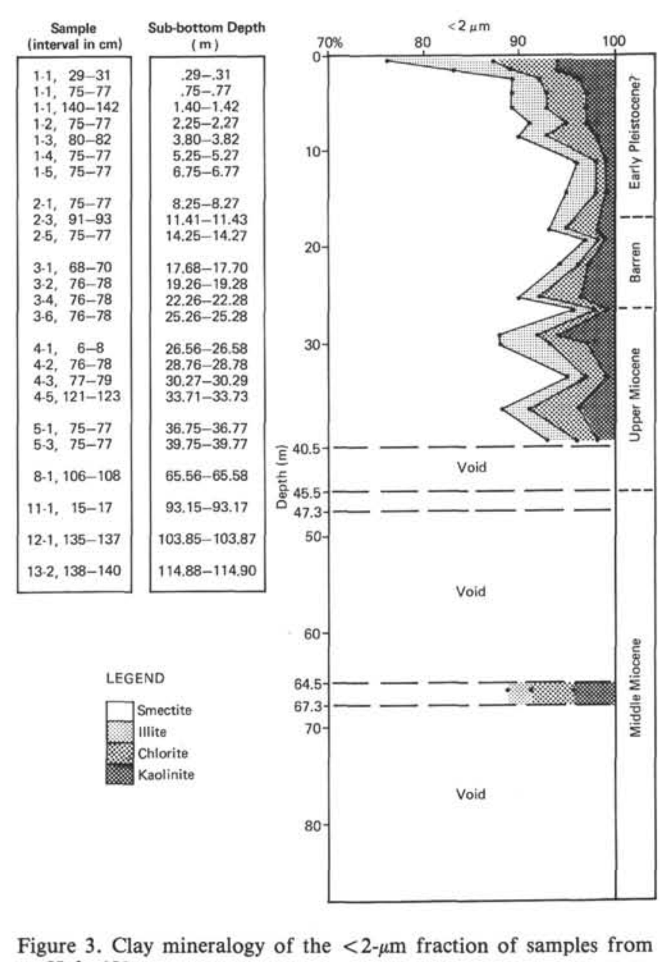

Figure 3. Clay mineralogy of the <2-μm fraction of samples from Hole 450.

and 96% over ranges of 4 to 6 meters. Lower in the section, the middle Miocene clays contain 88% smectite. In the upper and middle Miocene clays, broad glycolated smectite peaks show a major reflection between 17.67 and 16.99 Å, with a subpeak at 19.0 Å for samples with less than 93% smectite. Glycolated peaks for samples with more than 93% smectite show only a major reflection at 17.33 Å (no subpeaks), which indicates less compositional interstratification in these smectites. Chlorite, illite, and kaolinite occur in minor amounts throughout the unit. Also, vermiculite is suspected in samples from this site, as at the previous two sites, but its presence cannot be confirmed by XRD methods because of low-peak amplitudes and masking by chlorite peaks.

SOURCES FOR PELAGIC CLAY MINERALS IN THE SOUTH PHILIPPINE BASIN

Modern and ancient pelagic clays in the Philippine Basin cored at DSDP sites on Leg 59 have smectite as their dominant clay component. Smectite has several authigenic and detrital sources within the Philippine Sea. Both Murdmaa et al. (1977) and Kolla et al. (1978) noted a detrital source for smectite on the island of Mindanao but found that the influence of this clay source was very limited regionally. These authors studied surface sediments in the South Phillippine Basin and concluded that the origin of smectite found in these sediments is primarily authigenic. Murdmaa et al. (1977) suggest *in situ* alteration of fine-grain volcaniclastic sediment as the parental material.

Another authigenic mechanism is the formation of smectite by hydrothermal alteration of basalts from numerous ridge systems in the region. Core descriptions from ridge sites drilled on Leg 59 (Sites 448 and 451) note the presence of chlorite plus green and brown smectites in fractures and on slickensides and reaction rims in ridge basalts.

A third authigenic mechanism of formation could be the low-temperature chemical combination of Fe-hydroxides (from hydrothermal activity on a volcanic ridge) and silica (from biogenic sources). Heath and Dymond (1977) and Hein et al. (1978) have documented this process in surface sediments from separate regions in the Pacific Ocean.

As mentioned, authigenic chlorite was found associated with Philippine Ridge basalts. Therefore the Palau-Kyushu and West Mariana Ridges both are sources of the chlorite found in pelagic sediments of this region. Kolla et al. (1978) note Eastern China as another detrital source area for chlorite. These authors, however, found that because this land area drains into the East China Sea, any material that escapes deposition on the continental shelf is carried northward by the Kuroshio Current to the northern portion of the Philippine Basin.

The loess deposits of eastern China are an abundant source of illite; the majority of this clay is transported, along with chlorite, through the East China Sea to the northern sector of the Philippine Basin (Murdmaa et al., 1977). Kolla et al. (1978) also note that Luzon Island acts as a source of detrital illite and chlorite and that these sediments, accompanying the sediments from the East China Sea, are transported northward along the western margin of the basin in the Kuroshio Current. It is possible that illite is also formed at the volcanic ridges by hydrotheral alteration of the ridge basalts, which could then supply this mineral to pelagic clays in the basin.

Kaolinite has been found as an alteration product in the upper sections of basaltic breccia at Sites 448 and 451 (Aldrich et al., this volume). Because this clay has no other known detrital source in the region, the volcanic arcs are presumed to be the major suppliers of the kaolinite in Philippine Sea sediments. Aldrich et al. (this volume) have also detected vermiculite in the altered ridge basalts. Theoretically, this clay should also find its way into pelagic sediments in the Philippine Basin. Detection of this mineral in samples from Holes 447A, 449, and 450 was hampered by low-peak amplitudes and masking of vermiculite peaks before and after chemical treatment by illite and chlorite in the samples.

DISCUSSION

In Hole 447A, a pulse in smectite content at a depth of 33.0 meters in the late Oligocene is associated with the presence of scattered lumps of partially altered volcanic glass. The increase in smectite may probably be related to its authigenic formation from this glass. The smectite maximum is correlated with a slight change in compositional interstratification in the smectite lattices (the major glycolated XRD peak shifts to 17.32 Å). The possible sources for detrital clay minerals deposited here have been discussed in the previous section.

At Hole 447A the presence of pelagic clays indicate that sedimentation occurred at depths below the CCD during early late Oligocene through early Miocene. The lack of younger clays could be a function of nondeposition at the site after the early Miocene or, more likely, of erosion down to early Miocene levels due to uplift associated with faulting along the Palau-Kyushu Ridge.

This lower Miocene pelagic sedimentation can be correlated with the first pelagic clays deposited at Site 449 in the western Parece Vela Basin. Smectites from samples in these correlated zones in Holes 447A and 449 exhibit similar compositional interstratification in their lattice structures, although the subpeaks in samples from Hole 449 are more pronounced. Smectite content is slightly higher in the lower Miocene clays at Hole 449 than at Hole 447A, which is probably a function of the increased percentage of altered volcanic glass in Site-449 lower Miocene clays, probably resulting from volcanism on the West Mariana Ridge during this period. This could also account for the slight change in smectite interstratification in the Site 449 clays. The distribution of the glass at these two sites indicates that the Palau-Kyushu Ridge acted as a partial barrier to westward transport of the glass.

Factors affecting abyssal sedimentation in Site 449 changed during the middle Miocene, as evidenced by the modification in the clay-mineral suite (chlorite, illite, and kaolinite increased relative to smectite) and the deposition of a radiolarian ooze. The ooze is a result of increased biologic productivity in waters over Site 449. The increased productivity represented by the ooze is documented at two other DSDP sites in the region—(Sites 53 and 199, Fig. 4). The associated change in the clay suite may result from a shift in middle Miocene paleocirculation and/or decreased production of authigenic smectite in these sediments. Volcanic glass is present throughout the middle Miocene section at this site; however, the presence of the siliceous ooze may have inhibited alteration of the glass to smectite in the sediments surrounding the ooze.

Three hypotheses that could account for the anomalous sedimentation are (1) warming of the Asian continent with establishment of a more predominant summer-monsoonal circulation in the Philippine Sea region; (2) a general increase in oceanic boundary current circulation during this period; or (3) movement of the site northward through the equatorial zone.

R. Reynolds (in press) suggests that the establishment of a warmer climate on the Asian continent was associated with prolonged monsoonal circulation in neighboring Western Pacific waters, specifically for Leg 58 sites in the Sea of Japan. His paleotemperature curves show a warming trend in the Sea of Japan sites throughout the middle Miocene. This monsoonal circulation would move warmer, more productive equatorial waters northward through the Philippine Sea to affect the Sea of Japan region.

Intensification of oceanic boundary currents in the Atlantic during the middle Miocene has been suggested by R. E. Casey (1973). He proposes that uplift of the Panama block combined with closing of the Tethys seaway separated the Atlantic and Pacific water masses, which resulted in the intensification of boundary currents (and meridinal circulation in general) in the Atlantic. These conditions can also be assumed to affect Pacific-current regimes. In addition to this phenomenon, the formation of circum-Antarctic circulation (Barker and Burrell, 1977) and the associated initiation or intensification of glaciation on the Antarctic continent during the early Miocene (Tucholke et al., 1976) could also cause or enhance intensification of Pacific boundary currents. These conditions could result in a northward shift of the productive equatorial waters into the latitude of DSDP Sites 53, 199, and 499. R. Reynolds (1978) notes a similar increase in productivity during the Miocene at DSDP Hole 77B in the eastern equatorial Pacific, which could be correlated with the shifting current systems influencing the Philippine Sea circulation to the west. According to the plate-tectonic reconstructions of the South Philippine Basin by Karig (1975) and Hilde et al. (1977), it is improbable that plate motions carried Site 449 (and Sites 53 and 199) northward through the equatorial region. Extrapolation from the locations of older DSDP sites marked on Karig's diagrams places Site 449 near the equator during the late Eocene to early Oligocene. This predates the middle Miocene event considerably. Paleomagnetic data gathered from middle Miocene sediments at Site 449 are inconclusive in establishing the position of the site relative to the equator during this time (Site 449 report, this volume).

Sedimentation above the radiolarian-rich zone is undated but shows a return to pre-middle Miocene sedimentary conditions. The general decrease of smectite relative to illite, chlorite, and kaolinite up-section may

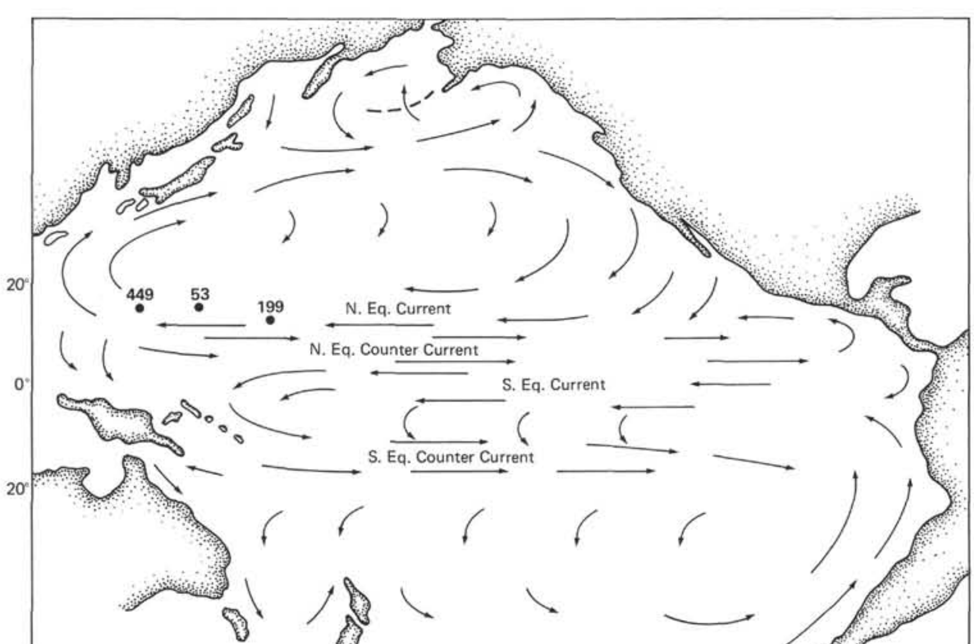

Figure 4. The DSDP sites in the western equatorial Pacific that show increased productivity during the middle Miocene plotted on a diagram of surface-water circulation patterns in the Pacific Ocean (after Pickard, 1975).

be associated with a slight decrease in the abundance of volcanic glass in the sediment; furthermore, there was less time for alteration of the glass in the more recently deposited sediments. In the smectites, compositional interstratification increases and crystallinity decreases in samples taken from older to younger sediments up-section.

At Site 450, recovery of middle Miocene pelagic clays was too poor to allow comparison with the corresponding sediments at Site 449. However, the upper Miocene, overlying undated, and possibly lower Pleistocene pelagic clays at Site 450 show the same general mineralogic trends as the undated upper clays at Site 449. Compositional interstratification increases and crystallinity decreases in younger smectites up-section at both sites. The short fluctuations in smectite content at Site 450 are probably associated with the variable supply of volcanic glass from the West Mariana Ridge.

CONCLUSIONS

The clay components of the pelagic sediments in the South Philippine Sea appear to have remained fairly constant since the beginning of the Neogene. They consist primarily of smectite, generally 80% or greater, with minor proportions of illite, chlorite, and some kaolinite and possibly vermiculite. Variations in the smectite content, compositional interstratification, and crystallinity are generally associated with variations in the concentration of and alteration time for volcanic glass in these pelagic sediments and reflect the importance of authigenic smectite formation in oceanic pelagic-clay units.

Smectite in the Philippine marginal basins has at least four possible sources: (1) *in situ* alteration of volcanic glass derived from ridge volcanism; (2) hydrothermal alteration of ridge basalts; (3) current transport of terrigenous smectite from Indonesia; and (4) low-temperature combination of Fe-hydroxides and biogenic silica.

Chlorite can be directly derived from the volcanic ridges in the area. It has been noted in fractures and slickensides in ridge basalts at Sites 448 and 451 (the Palau-Kyushu and West Mariana ridges). Chlorite also has detrital sources on Luzon Island and eastern China (transported through the East China Sea).

Illite is abundantly supplied by the loess sediments of eastern China, which enter the current system of the Philippine Basin through the East China and Yellow seas. It is also a possible product of hydrothermal alteration of ridge basalts.

Kaolinite lacks known detrital sources in the region but has its major source in the altered basalts of the Palau-Kyushu and West Mariana ridges.

Vermiculite is also suspected in these sediments because it has been found as a hydrothermal-alteration product (along with smectite, chlorite, and kaolinite) in basalts from the Palau-Kyushu and West Mariana ridges.

Anomalous biogenic and pelagic middle Miocene sedimentation at Site 449 (and Sites 53 and 199) could indicate a northward shift of the North Equatorial Counter Current over the site. This shift could have been induced by intensified monsoonal circulation from warming on the Asian continent or intensified boundary-current flow initiated by separation of Atlantic and Pacific water masses plus formation of circum-Antarctic flow. A less probable explanation would be that the site moved northward through the equatorial region during the middle Miocene.

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