A semiquantitative study of the surficial clay mineral distribution from 79 cores in the Recent sediments in the Gulf of Mexico shows a regional pattern which can be related to source and environment. The dominant factor controlling distribution of these surficial sediments appears to be source. Montmorillonite is the most abundant clay mineral, and illite, chlorite, kaolinite and mixed-layer minerals are present in almost every sample.

Alteration of the clay minerals is influenced by (a) change of environment, and (b) length of exposure. Initial ion exchange, which is caused by a tendency to reach equilibrium with a new environment, occurs so rapidly in the clay minerals after they enter the marine environment that rate of sedimentation is a minor factor in comparison to environmental change. Slow alteration and adjustment toward a stable end state are most complete in areas of slow deposition and extended exposure of the clay minerals to the environment.

INTRODUCTION

This report presents the results of a clay mineral study of the surficial sediments from 79 cores collected in the Gulf of Mexico. The purpose of the study was to identify and determine semiquantitatively the clay minerals present and to relate them to provenance and environment. It was felt that a regional approach to the problem would be more practical than a detailed study as the numerous small-scale variables are minimized and the major chemical changes that the clay minerals undergo are clarified.

In recent years the clay minerals have received special attention by mineralogists and other scientists in the hope that detailed studies would help solve some vexing problems concerning the relation of clays to their physical and chemical environment (Grim, 1958; Keller, 1956; Powers, 1957; Johns and Grim, 1958; Milne and Earley, 1958; Weaver, 1958; Murray and Harrison, 1956; and many others). Because the Gulf of Mexico is essentially a closed basin receiving tremendous amounts of sediments, including montmorillonites from both bentonitic sources and weathered materials, illites, chlorites, mixed-layer clays and some kaolinite, distribution of these clays was studied in an attempt to obtain more information concerning the controversial subject of the relative importance of source area, diagenesis, and environment of accumulation in determining the ultimate clay mineral assemblage.
ACKNOWLEDGMENTS

The cores were collected by the Lamont Geological Observatory and the Woods Hole Oceanographic Institute from the floor of the Gulf of Mexico as a part of a geophysical and oceanographic study under the direction of Dr. M. B. Ewing. Special thanks are due Dr. Ewing, Dr. D. Erierson, and Dr. B. Heezen of the Lamont Geological Observatory for their interest and cooperation in obtaining the samples for this study.

METHOD OF STUDY

Only the sediment from the upper 10 cm of each core was included in this study. Sediments from depths more than 10 cm were not included because of significant differences in rate of deposition from place to place. The deeper samples could range in age from Recent to Tertiary and thereby would introduce some undesirable variables which would make long-range correlation of any interval other than the tops of the cores hazardous and misleading. Fig. 1 shows the location of samples used in this study. Sample designations are the same as those used by the Lamont Geological Observatory. The numbers above the sample locations are depths (in fathoms) of the cores below the water surface. The map shows the generalized bottom topography of the Gulf of Mexico and major geographic locations. The boundary between the slope and continental shelf is placed at 100 fathoms and the boundary between slope and abyssal plain is placed at 1500 fathoms.

Care was taken during preparation of the samples to maintain their original composition as nearly as possible. The core samples were not exposed to temperatures greater than room temperature. They were dispersed in distilled water with ammonium hydroxide rather than washed, in order to prevent the possibility of leaching and alteration of the clay minerals. The minus 2μ clay fraction was sedimented on a glass slide according to a method outlined by Bradley, Grim and Clark (1937, p. 216). X-Ray diffraction traces were run on the untreated slides, again after treating them with ethylene glycol, after heating the slides to 200°C for 1 hr, and after heating at 450°C for 45 min.

Because the Recent clay minerals exhibit such a low order of crystallinity, most of the traces were made with the recorder range increased from the normal 100 to the 200 range. The increased range coupled with a change in time constant to decrease sensitivity of the diffractometer greatly increases intensity of peak reflections, a desirable factor because magnitude of error in semiquantitative estimations is reduced.

Montmorillonite, illite, chlorite, kaolinite and mixed-layer minerals were identified in the samples. Ethylene glycol treatment was used to identify montmorillonite. Samples were heated to 200°C to differentiate montmorillonite and partially collapsible mixed-layer minerals. The treatment is also valuable in identification of partially expandable chlorite. A split 7Å peak was observed on some glycolated traces. After heating to 200°C, the 7Å peak resolves itself into one peak with magnitude of intensity approximately the
same as the sum of the two peaks on the glycolated trace. This expandable chlorite apparently is the same type of material described by Johns, Grim and Bradley (1954 pp. 243–244). They refer to the material as diagenetic chlorite and ascribe this ability to expand and collapse to incomplete development of interlayer brucite; an intermediate stage between montmorillonite and well-crystallized chlorite end members, which may be similar to material classified as vermiculite.

The slides that had been heated to 450°C were used to differentiate kaolinite...
from chlorite. Heating to 450°C causes breakdown and partial collapse of
the structure of sedimentary chlorite (Bradley, 1953) but does not affect
kaolinite. Any residual peaks that remain at 7 Å after heating to 450°C are
attributed to kaolinite. In addition, the 3.5 Å peak of kaolinite is displaced
from an angle about 25° 2θ to about 24.8° 2θ while that of well-crystallized
chlorite is displaced to about 25.2° 2θ. Presence of kaolinite or residual
chlorite or both can be ascertained by direction of displacement of the
3.5 Å peak. The well-crystallized residual chlorite is considered to be allo-
genic.

Basal reflections of well-crystallized clay minerals are sharp whereas poorly
crystalline clay minerals give diffuse low-intensity reflections. Increased
scattering relative to the well-crystallized minerals is due to incomplete
structures and skewed or randomly stacked layers. Much of this material
in the Gulf of Mexico is mixed-layer clay minerals. Obviously much of the
material could be fitted into one of the major clay mineral types. However,
it is structurally and compositionally incomplete and is considered to repre-
sent some intermediate stage of diagenesis. It is felt that estimates of mixed-
layer material can be used as a significant indicator of the degree of crystal-
linity of the clay minerals, and perhaps indicate rapidity of deposition and
relative intensity of diagenesis.

METHOD OF ESTIMATING ABUNDANCE
OF CLAY MINERALS

Scattering of diffracted x-ray beams along various basal planes of a mineral
is not constant. Scattering is related to a form factor function and for any
theoretical structure of a given composition a scattering sequence can be
plotted as a function of sin θ/λ or of the angle 2θ. Theoretical scattering
sequences can be plotted for the basic clay mineral structures in which
composition and population concentration of cations is known. Basically,
the scattering sequences for the three-layer minerals have the same general
configuration. However, slopes on the curves are modified by compositional
changes or by intermixing of clay mineral species.

In estimating relative quantities of specific clay minerals, the scattering
power of various 00l planes must be considered in order that direct comparison
may be made of peak intensities. In view of the many uncontrollable factors
that must be considered, the quantitative estimates of individual clay
mineral components can only be generalized. Assumptions have been made
in the estimates and accuracy of the method is not great; but it is felt, in
view of similar analyses that have been made, that magnitude of error is
not so great as the amount of allowable error in estimation of relative abund-
ance of clay minerals in any specific sample.

The background curve on a diffractometer trace of a relatively pure clay
mineral has the same shape as the theoretically determined scattering se-
quence and it is altered by introduction of various mineral species. In view
of the similarity, and because the form factor function cannot reasonably
be determined for multiply mixed clay minerals with numerous differences in composition and crystallinity, the expediency of a practical scattering sequence constructed from the traces themselves was considered.

A composite scattering sequence was made by taking an average of 14 samples distributed in different environments throughout the area being studied (Fig. 2). Relative reflecting power of basal planes that were used for comparative estimates was measured from this composite scattering sequence. Relative peak intensities were compared within a small angular range because of changes in reflectivity and x-ray absorption over wide angles.

Quantitative estimates are all relative and illite was used as the internal standard (Johns, Grim and Bradley, 1954). Differences in reflecting power were all measured from the composite scattering sequence. Relative abund-

\[\text{Figure 2.} \quad \text{Composite scattering sequence of relative diffraction intensities of clay minerals.}\]

\[\begin{array}{c}
\text{Intensity} \\
0 & 20 & 40 & 60 & 80 & 100 & 120 & 140 & 160 & 180 \\
\end{array}\]

\[\begin{array}{c}
2.0^\circ & 4 & 6 & 8 & 10 & 12 & 14 & 16 & 18 & 20 & 22 & 24 & 26 & 28 & 30 & 32 & 34 & 36 \\
\end{array}\]

\[\begin{array}{c}
\text{Relative abundance of mixed-layer material in each sample was estimated}
\end{array}\]
by comparing the area of the 001 illite peak with the area between the 001 illite and 001 montmorillonite peaks on the trace of the glycolated sample. This latter area includes the 001 chlorite reflection so estimated percentage of chlorite in each sample was subtracted from the relative abundance estimate. Estimate of absolute abundance of mixed-layer material may be

Table 1.—Relative Abundance of Clay Minerals in Each Sample
(Calculated in parts per ten)

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<th>Sample No.</th>
<th>Illite</th>
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<th>Kaolinite</th>
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</table>

tr—trace; less than 0.25.
high or low as a consequence of the method used. However, relative amount of mixed-layer material, when compared between samples, will remain constant independent of any factor used to increase or decrease abundance.

Quantitative estimates of each clay mineral are given in parts per ten (Johns, Grim and Bradley, 1954). Actual amount of clay material is not inferred; some samples contain considerable amounts of calcite and all samples contain some quartz. These impurities are not considered in the estimates.

Relative abundance of clay minerals is shown in Table 1. Location of the samples is indicated in Fig. 1.

CLAY MINERAL DISTRIBUTION PATTERNS

Because of the chemical mobility of certain elements in clay minerals, these minerals have been considered very likely to be good indicators of environment of deposition. Ion exchange or replacement eventually can lead to structural changes or alteration of one mineral to another.

Probably a major factor in distribution of clay minerals is the source. Some workers recently have questioned the plausibility of the depositional factor and have considered influence of source material as a primary factor in distribution of clay minerals (Weaver, 1958; Grim, 1958; Milne and Earley, 1958).

A series of maps showing distribution of the major clay mineral types and their relationships have been prepared from the semiquantitative estimates (Table 1). Distribution cannot be interpreted as absolute because values on which distribution is based are only relative. That is, they show the relative abundance of a particular clay mineral type as a fraction of the total clay minerals present at a given sample location. For example, clay mineral abundance is minor on the Campeche and West Florida Banks, but the ratio of one clay mineral to the others can still be compared with areas containing a high percentage of clay.

Because of the sparse control and character of the data, the maps show only trends as related to source and environment. Contour intervals on each map differ according to relative abundance of the clay under discussion.

Montmorillonite

Regionally, the greatest concentration of montmorillonite is on the abyssal plain with a slight decrease in abundance in the Sigsbee Deep (Fig. 3). Abundance of this mineral is highest in the area receiving sediments directly from the Mississippi Delta; this feature verifies the observation that a considerable amount of montmorillonite is being supplied by the Mississippi drainage area (Johns and Grim, 1958). Sedimentation off the Delta is rapid. Because of rapid burial an area of accelerated deposition is not conducive to diagenetic changes; consequently the montmorillonite would have to be transported from a source or formed directly upon entry into the marine environment.
Another major source of montmorillonite is the Trinity or Brazos River (Grim and Johns, 1954). Influx is apparent in the area southeast of Galveston Bay, but because of lack of control the exact source cannot be defined. The areas that contain the least montmorillonite are the West Florida Bank and the Campeche Bank.

Fig. 3 shows that the prime control over montmorillonite distribution in the Gulf of Mexico is source and this regional study indicates no alteration in the change from shelf to abyssal environment.
Concentration of montmorillonite on the abyssal plain apparently is a result of transportation of this mineral into the environment rather than a result of diagenetic alteration. Particle size of montmorillonite is finer than that of other clay minerals, so the relatively greater abundance might be due to differential settling from suspension. The Recent sediment cover on the western abyssal plain is relatively thin, indicating retarded deposition and accumulation of fine-grained material. The fine-grained montmorillonite should accumulate here if it is primarily an allogetic mineral. However, a
slight decrease in abundance is indicated. Abundance of mixed-layer material complements that of montmorillonite, suggesting that the montmorillonite structure may be slowly undergoing alteration, and an intermediate stage in the diagenesis is represented by presence of mixed-layer material.

**Illite**

Relative abundance of illite in the Gulf of Mexico is fairly uniform (Fig. 4). The striking feature on the map is the relative abundance of illite on the continental slope and adjacent abyssal plain around the periphery of the calcareous Campeche Bank and in the Florida Straits. Weaver (1958, pp. 259–260) maintains that illite is the predominant clay mineral in all sediments, is a detrital mineral, and is a product of the source. The fact that illite surrounds the Campeche Bank does suggest a high illite source on the Yucatan Peninsula.

Illite abundance increases relative to montmorillonite on the north-central and northwest continental slopes, areas with a thick Recent sediment accumulation. Another area of relative increase in abundance is south of the Sigsbee Deep. Mechanical separation due to particle size difference rather than preferential ion exchange seems the most plausible explanation. Illite is most common in areas that are receiving thick accumulations of sediment. This relation suggests that if direct alteration from montmorillonite to illite takes place, it is accomplished shortly after the material enters the marine environment.

**Kaolinite**

Kaolinite is a minor constituent of the clay mineral assemblage in the Gulf of Mexico and over-all distribution is relatively uniform (Fig. 5). Relative abundance of kaolinite is higher on the continental shelf seaward from the Mississippi drainage system and Galveston Bay than on other shelf areas. The abundance suggests that these two distributaries are major source areas. The regional pattern suggests that concentration of kaolinite is lowest on the continental slopes and is highest on the floor of the abyssal plain.

The very low exchange capacity of kaolinite indicates that it has the most stable structure of the clay minerals (Grim, 1953, p. 129). The mineral is relatively unstable in the basic marine environment. However, high energy contribution or very high pH is required for degradation or alteration. The kaolinite distribution pattern appears to substantiate the thesis that kaolinite in the Gulf of Mexico is largely detrital in origin. Deposition occurs throughout the Gulf. Accumulation on the continental slopes is minor, because the clay is carried across in a suspended sediment load, and is high on the abyssal plain where the fine-grained sediment settles out of suspension.

**Chlorite**

The northern continental shelf west of the Mississippi Delta and the northwestern continental shelf contain relatively little chlorite (Fig. 6). The continental slopes east and west of the Mississippi drainage system contain the
greatest relative abundance of chlorite, and a slight decrease is noted on the abyssal plain. Relative abundance of chlorite on the Campeche Bank suggests either a source of chlorite south of this area or possibly a con-

centration of magnesium in solution causing alteration of montmorillonite or mixed-layer material to chlorite. Drainage from the Paleozoic Appalachian entering the Gulf of Mexico through Mobile and Apalachicola Bays could be carrying chlorite and would account for the relative concentration in the northeast part of the Gulf of Mexico.
Locations of samples that contain a chlorite with an expandable 7 Å x-ray reflection are underlined (Fig. 6). This chlorite is considered to be diagenetic. Samples containing the expandable chlorite are distributed generally on the continental slopes, areas of relatively rapid and turbulent transport and deposition. Distribution suggests that diagenesis or readjustment takes place rapidly. Bradley (1953, p. 729) suggests that formation of the brucite layer is a function of pH or solubility of the magnesium; although magnesium chemistry is not well understood, pressure and temperature undoubtedly...
affect solubility and could explain the alteration in the environment intermediate between shelf and abyssal plain.

The expandable chlorite is present in areas in which order of crystallinity of the clay minerals is relatively low. Increase in crystallinity indicates adjustment toward a stable end-state and this rudimentary chloritic material occurs in an environment intermediate between areas of low and high crystallinity. Presence of the incomplete brucite layer may be evidence of a tendency of the clay mineral structure to attain a stable state.
Mixed-layer material is most abundant on the Campeche and West Florida Banks and in the Yucatan Channel and Florida Straits (Fig. 7) and is least abundant on the floor of the abyssal plain. A direct relationship exists between relative amount of mixed-layer material and order of crystallinity of the clay minerals. A visual estimate of the order of crystallinity of the clay minerals based on an arbitrary range from 1 to 3 was made from the x-ray traces; 1 is lowest and 3 is highest order of crystallinity. Changes in crystallinity are indicated by the dashed contour lines (Fig. 7).

The continental slope and abyssal plain seaward from the Mississippi Delta and the continental slope in the northwest Gulf of Mexico have relatively minor amounts of mixed-layer material whereas this material is common in the Sigsbee Deep. Sedimentation is slow in the Sigsbee Deep compared with the other two areas. A logical conclusion, considering rate of deposition vs. diagenesis, is that clay minerals on the northwest and Mississippi Delta slopes were not exposed long enough to be altered, whereas the slow sedimentation in the Sigsbee Deep allows for alteration or adjustment of the clay mineral assemblage to the environment. However, marked differences in the x-ray traces demonstrate that crystallinity of the clay minerals in areas of rapid deposition is generally of higher order than that in the Sigsbee Deep. This indicates that adjustment of the clay minerals to the marine environment takes place almost immediately; complete adjustment to a stable state is a long-term process.

Distribution patterns of montmorillonite and of the mixed-layer material are complementary. Cation exchange capacity of montmorillonite averages from four to thirty times greater than that of other clay minerals. Therefore, if a rapid adjustment of clays from continental to marine environment takes place, montmorillonite would be the first clay mineral to form, and subsequent alteration to other clay mineral types depends on length of time of exposure to the new environment. The good correlation of mixed-layer material and montmorillonite distribution indicates that montmorillonite is undergoing change to a mixed-layer material through some diagenetic process.

DISCUSSION OF RESULTS

Consideration of the distribution patterns of the clay minerals reveals several factors that could affect the relative abundance and distribution of the clay minerals. Among the more important are source, diagenesis, environment of deposition (including chemistry of the sea-water) and surficial and deep currents in the Gulf.

From the study of the distribution patterns and the literature available, it must be concluded that the effect of source area is of prime importance. The patterns reveal two major sources of contribution: (a) drainage into the northwest Gulf, and (b) the Mississippi drainage system with a direct sphere of influence including the eastern half of the Gulf to the escarpment at the West Florida Bank, and extending as far south as the foot of the slope at the
base of the Campeche Bank. As Johns and Grim (1958) have pointed out, the Mississippi River gets its sediment load largely from the drainage basins of the Missouri and Ohio Rivers and these sources contain significantly different clay mineral suites. The Missouri River brings in large amounts of montmorillonite of bentonitic origin whereas the Ohio River contributes sediments that are illitic and chloritic, and contain montmorillonite formed as a result of weathering of illite and chlorite (Murray and Leininger, 1956). The distribution pattern of montmorillonite certainly suggests that source is the primary factor controlling its distribution. Because of the small particle size of the montmorillonite, large quantities of this material probably remain in suspension and eventually are deposited on the abyssal plain. This differential settling accounts for the high montmorillonite content in the deeper portions of the Gulf.

The distribution of mixed-layer material indicates that diagenetic changes are also important factors. The complementary patterns of montmorillonite and mixed-layer materials certainly are not coincidental but indicate a change of montmorillonite to a mixed-layer complex. Most areas that contain the preponderance of montmorillonite are the areas that are receiving substantial quantities of sediment. Most areas that are high in mixed-layer material are areas of slow sedimentation. This may mean that in areas of slow sedimentation some of the montmorillonite is altered. As suggested by Johns and Grim (1958), perhaps the montmorillonite that originated by weathering of other clay minerals is changed whereas the montmorillonite of bentonitic origin tends to be more resistant to diagenetic change. In areas of slow sedimentation some of the montmorillonite derived from weathering of illite and chlorite may be changed back to the original mineral.

Other evidence that diagenetic change is taking place is the formation of an expandable type of chlorite. The expandable chlorite is found in areas of rapid deposition; hence the alteration is rapid. It is suggested that the change in environment from the continental shelf to the abyssal plain with resultant change in the solubility of magnesium is an important factor.

Diagenesis is apparently influenced by two major factors: (a) change in environment, and (b) rate of sedimentation or length of exposure time. Initial ion exchange, caused by a tendency to reach equilibrium with a new environment, takes place very rapidly, so that this type of change would be expected in areas of rapid deposition. Slow alteration and adjustment toward a stable end state is most complete in areas of slow deposition and extended exposure of the clay minerals to the environment.

Other factors certainly have some effect on the distribution patterns, and as more is learned perhaps the distribution patterns of illite, chlorite and kaolinite can be better interpreted.

In summary: (1) The distribution pattern of montmorillonite reflects the effect of the source. (2) The complementary distribution patterns of montmorillonite and mixed-layer clays suggest a diagenetic change. (3) Illite and chlorite distribution patterns suggest that source is of prime importance and that in certain areas diagenetic change is influential.
REFERENCES


