QUANTITATIVE STUDY OF CLAY MINERALS IN SOME RECENT MARINE SEDIMENTS AND SEDIMENTARY ROCKS FROM JAPAN

by

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ABSTRACT

CLAY mineral compositions in specimens of Recent marine sediments in the neighbourhood of Japan and sedimentary rocks from Japan were studied. Identification of the clay minerals was made chiefly by X-ray, differential thermal and infrared analyses. Their relative amounts were determined by comparing the intensities of their basal X-ray reflections.

Recent marine sediments: Specimens from the bottom of the Northwestern Pacific Ocean contain montmorillonite, illite, chlorite and kaolinite. The amount of montmorillonite varies locally, but that of kaolinite is generally small. Clay mineral compositions of the Eastern Sea specimens are different from those of the Northwestern Pacific Ocean and the crystallinity of clay minerals seems to be better in the Eastern Sea. It is most likely that the clay minerals in the Eastern Sea sediments were supplied from the continent. Specimens of the bottom sediments at the entrance of Tokyo Bay contain montmorillonite, illite, chlorite and a lesser amount of kaolinite. The amount of montmorillonite varies markedly with locality and tends to be low where the movement of sea water is relatively intense. This may be due to a finer particle size of montmorillonite in comparison with the other clay minerals.

Sedimentary rocks: The relationship between sedimentary formations and clay mineral compositions shows that the compositions are characteristic of the depositional environments. For example, kaolinite is abundant in the formations deposited in fresh water but its content is low in the formations deposited under marine environment. The Paleozoic formations from the Tohoku region contain chlorite and illite without exception; however, the Mesozoic formations contain them in various proportions according to localities.

INTRODUCTION

A CONSIDERABLE number of papers on clay sedimentary petrology have been published, and much information on clay minerals in sedimentary rocks has been accumulated. Nevertheless the available information is still insufficient to clarify the agents controlling the clay mineral composition in sediments

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and sedimentary rocks. One of the main factors is the lack of reliable quantitative analysis of clay minerals.

By the application of the X-ray method for quantitative analysis proposed by Sudo, Oinuma and Kobayashi (1961), a detailed study of clay minerals has been made on the Recent marine sediments near Japan, and Tertiary, Mesozoic and Paleozoic sedimentary rocks from Japan. The purpose of the present paper is to give a summary of the results.

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MATERIAL AND METHOD

The samples discussed in this paper are listed below.

Recent marine sediments

Deep sea sediments:

A. Northwestern Pacific Ocean (sample nos. 1–17).
Collected at water depths of 1800 to 8450 m.

Shallow sea sediments:

B. Eastern Sea (sample nos. E1–E6).
Collected at water depths of 70 to 130 m.

C. Entrance of Tokyo Bay (87 samples).
Collected at water depths of 10 to 400 m.

Sedimentary rocks

Tertiary sediments:

D. Chichibu basin, Saitama Prefecture (22 samples).

Cretaceous–Tertiary sediments:

E. Ashibetsu district, Hokkaido (153 samples).

Devonian–Cretaceous sediments:

F. Tohoku region.

1. Nagasaka district, Iwate Prefecture (27 samples).
2. Hikoroichi district, Iwate Prefecture (25 samples).
3. Ōhunato district, Iwate Prefecture (30 samples).
4. Karakuwa district, Miyagi Prefecture (19 samples).
5. Shizukawa district, Miyagi Prefecture (30 samples).

The samples from the Northwestern Pacific Ocean and the Eastern Sea were collected by the Japanese Deep Sea Expedition in which the writers participated. The samples of the Tokyo Bay bottom sediments were collected also by the writers as members of the Tokyo Bay Research Group. Figure 1 shows the outline of their location.

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The clay fractions (less 2 μ) of the Recent sediments were separated, whereas analyses of the sedimentary rock samples were made on either the pulverized samples or the less than 2 μ clay fraction. These prepared specimens were subjected to various analyses for the identification and quantitative estimation of clay minerals. The general scheme of the identification is summarized in Table 1. For the quantitative estimation, the X-ray method proposed by Sudo, Oinuma and Kobayashi (1961) was applied. In this method, basal reflection intensity ratios derived from standard mixtures are used to estimate the composition of unknown samples.

TABLE 1.—PROCEDURE OF IDENTIFICATION OF CLAY MINERALS

	Procedure	Kinds of clay minerals identified
Clay fraction—	——X.R.A.	(M,I,Ch,V,K)
	—Higher resolution scans of 24–26°(2θ)——X.R.A.(1)	K,Ch
	—Heated to 150°C, 300°C, 450°C, 600°C and 750°C—X.R.A.	M.I.Ch,V,H
	—Treated with ethylene glycol or glycerol—X.R.A.	M.H,(Ch,I)
	—Treated with hydrochloric acid——X.R.A.(2)	Ch,K
	—Treated with ammonium nitrate solution–X.R.A.(3)	V
	—Treated with sodium nitrate, calcium chloride and glycerol——X.R.A.(4)	Al–V
	——D.T.A.	K,(Ch,M,I,V)
	—Treated with piperidine——D.T.A.(5)	M
	——Infrared spectra analysis(6)	K
	——Electron micrographic observation	(K)
	——Chemical analysis	
	——Staining test(7)	M
	——Cation exchange capacity	(M,V)

X.R.A.; X-ray analysis
D.T.A.; Differential thermal analysis
M: montmorillonite, I: illite, Ch: chlorite,
V: vermiculite, K: kaolinite, H: hydrated halloysite,
Al–V; aluminian vermiculite.
(1) Biscaye (1964), (2) Sudo, Oinuma and Kobayashi (1961), (3) Walker (1949), (4) Tamura (1958), (5) Allaways (1948), (6) Kodama and Oinuma (1963), (7) Oinuma (1964).

RESULTS AND DISCUSSION

Northwestern Pacific Ocean

Figure 1 shows the clay mineral compositions of the specimens from the surface layer (0 to 10 cm deep) of the sea bottom in this region. As seen in the figure, the variations in the amount of montmorillonite are remarkable. The amount of kaolinite is small in all of the samples.

Montmorillonite tends to be abundant in the specimens from the northern part of this area. Specimens from the bottom sediments of the Japan, Kuril

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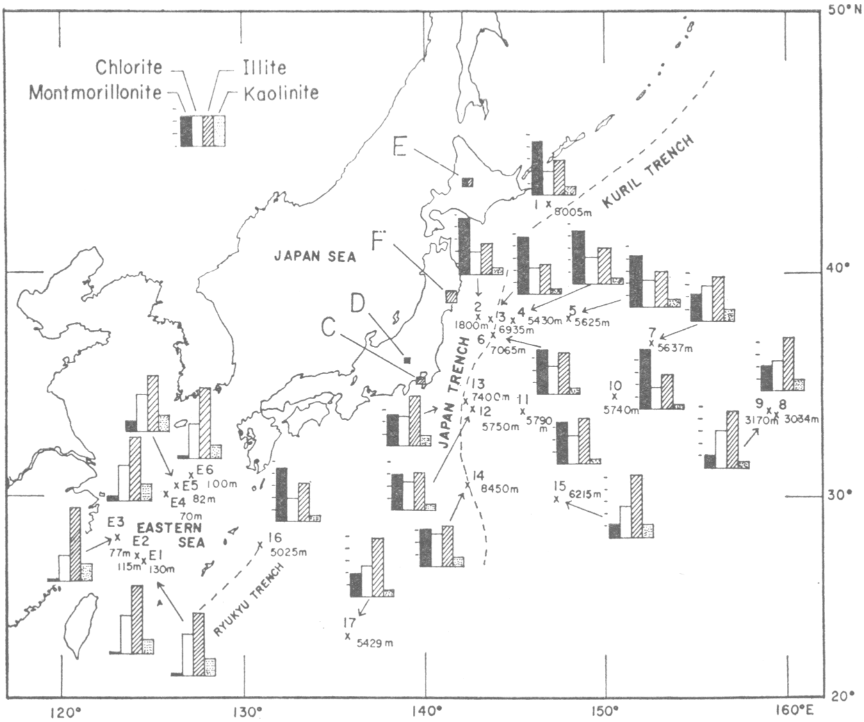

FIG. 1. Areas and localities of specimens and clay mineral compositions of the specimens from the Northwestern Pacific Ocean and the Eastern Sea.

and Ryukyu Trenches are generally rich in montmorillonite, but the specimens (nos. 8, 9, 15 and 17) of the ocean floor sediments are low. Because *in situ* formation of kaolinite under marine environments is unlikely, the kaolinite seems to be inherited and distributed in small amounts over a wide area. Movements of ocean currents which transport clay materials over the vast area may explain the fact that the kinds of clay minerals in this region are rather constant. Although a reasonable explanation cannot be made from a clay mineralogical point of view, it is interesting that the montmorillonite concentration of the trench bottom specimens is higher than that of the ocean floor specimens.

In the specimens from several core samples, no vertical variation in clay mineral composition was observed. Within several meters below the surface of the deep sea bottom, diagenetic changes were not apparent, a result which indicates that the clay minerals were already in a considerably stable form when they were deposited. In a few instances, however, the crystallinity of clay minerals of the very superficial layer of the core tended to be relatively low.

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Eastern Sea

The shallow sea sediments contain illite, chlorite and kaolinite as principal clay minerals and some of them contain a small amount of montmorillonite. The general trend of composition is also given in Fig. 1. Generally, the Eastern Sea sediments include clay minerals having a good crystallinity. The reflections of these clay minerals are sharper than those of the deep sea sediments and are as sharp as those of the Paleozoic or Mesozoic sedimentary rocks. The sediments contain pebbles and sand grains of black clay slate and shale. The evidence may suggest that the clay minerals in this region have been transported by streams from the continent.

Entrance of Tokyo Bay

The principal clay minerals in the specimens collected from the bottom of Tokyo Bay are montmorillonite, illite and chlorite with an occasional occurrence of a very small amount of kaolinite. Figures 2 and 3 show the localities of the samples and cross-sectional distribution of clay minerals respectively. The quantitative variation of montmorillonite is most remarkable. The amount of montmorillonite is generally larger offshore than near the coast. The amount is small in the area where the depth is shallower than 20 m and increases with increase of depth. However, in the deep central area, which is called the Tokyo submarine canyon, montmorillonite shows a

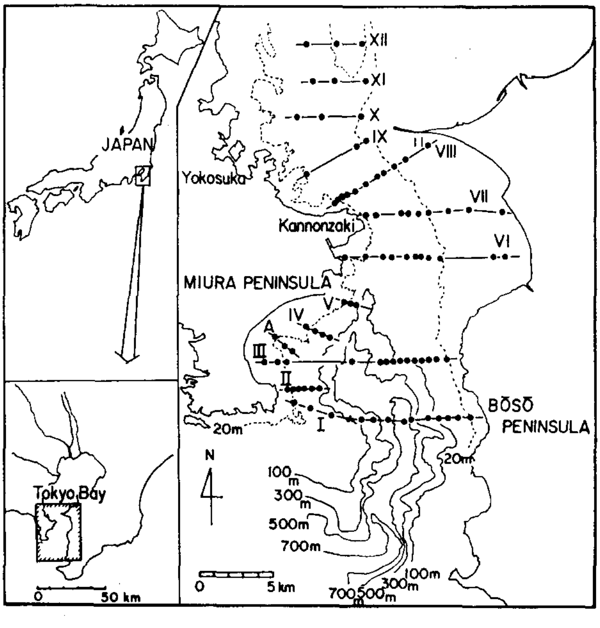

FIG. 2. Localities of the specimens from the entrance of Tokyo Bay.

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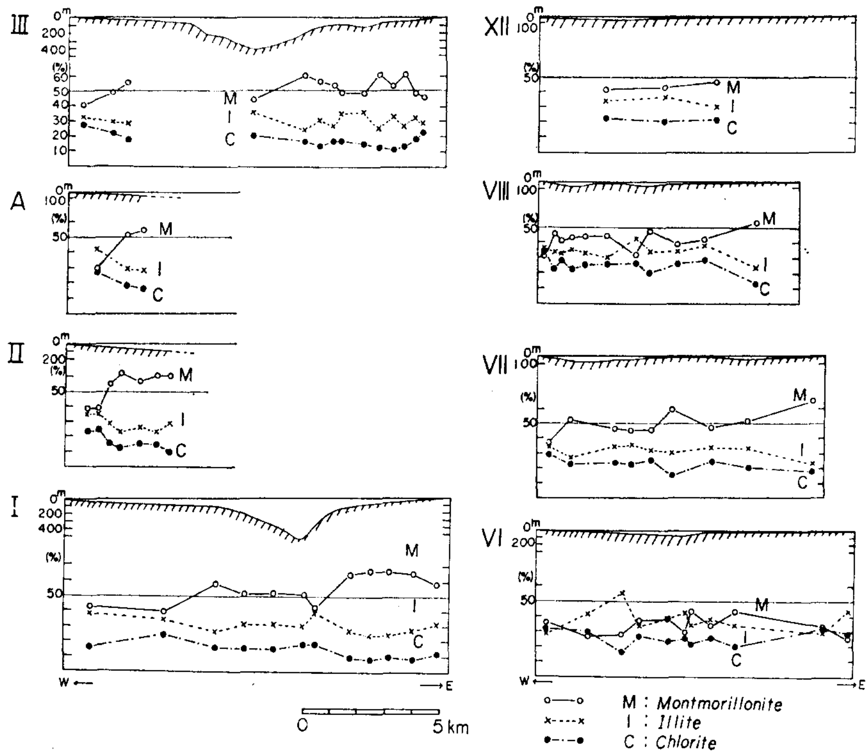

FIG. 3. Some cross sections and clay mineral compositions of the specimens from the entrance of Tokyo Bay.

tendency to decrease. Chlorite and illite have a similar distribution and the amounts are inversely related to that of montmorillonite.

The small amount of montmorillonite in the sediments of the shallow sea (less than 20 m in depth) near the coast suggests that the agitation action of waves causes the fine-grained materials such as montmorillonite to be removed. The quantitative distribution of montmorillonite is shown in Fig. 4a, for comparison with the circulation and tidal currents of Tokyo Bay (Fig. 4b, c). It is obvious that the amount of montmorillonite is smaller where the movements of sea water are more active. This is probably due to the nature of montmorillonite, i.e. it is more easily dispersed in sea water than chlorite and illite. Therefore it is selectively removed from areas of active movement and sedimented in areas of relative calm, thus enriching the sediments in the quiet areas.

Tertiary Formations of Chichibu Basin, Kanto Region

The Chichibu basin, Kanto region, comprises Tertiary formations (Oligocene to Lower Miocene) that are unconformable with the underlying Mesozoic

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and Paleozoic formations. The Tertiary formations, principally conglomerate, sandstone and siltstone, were deposited in a marine environment. Specimens used in the present study were taken from the siltstone because the rock is relatively rich in clay minerals.

Examination of these specimens revealed that illite and chlorite are contained in all horizons. Montmorillonite is present in the upper horizon only, and increases in amount upward. No kaolinite was detected.

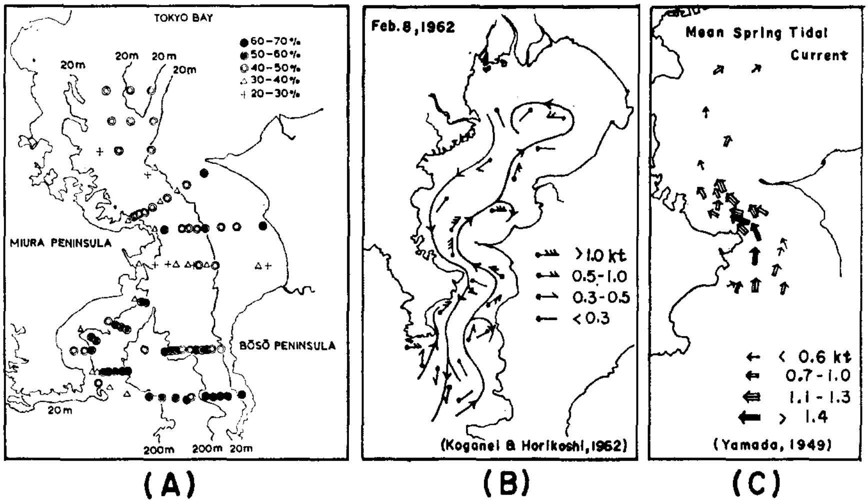

FIG. 4. (a) Distribution of montmorillonite in the sediments of the entrance of Tokyo Bay. (b) Circulations in Tokyo Bay (after Koganei and Horikoshi, 1962). (c) Tidal currents in the entrance of Tokyo Bay (after Yamada, 1949).

According to Arai and Kanno (1960), the source of the Chichibu basin deposits was the Mesozoic and Paleozoic formations surrounding the basin, which from the present study consisted of chlorite and illite. The upper beds of these deposits contain more weathered materials than the lower beds. Therefore, the abandance of montmorillonite in the upper beds of the deposits of the Chichibu basin may imply that the montmorillonite was produced by weathering of chlorite and illite, during erosion and redeposition of the parent material.

Mesozoic and Tertiary Formations of Ashibetsu District, Hokkaido

In Ashibetsu district, the Paleogene Ishikari group unconformably overlies the Cretaceous Hakobuchi formation. The Ishikari group including several coal-bearing formations is unconformably overlain by the Poronai formation. Figure 5 shows the stratigraphic succession with the summary of depositional environments. All of the specimens studied were shale.

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The Ishikari group contain kaolinite, chlorite, illite and hydrous illite, and the marine Hakobuchi, Poronai and Kawabata formations contain chlorite, illite and hydrous illite. In this paper, the term "hydrous illite" is applied to materials such as complicated non-regularly interstratified sequences of illite and montmorillonite layers.

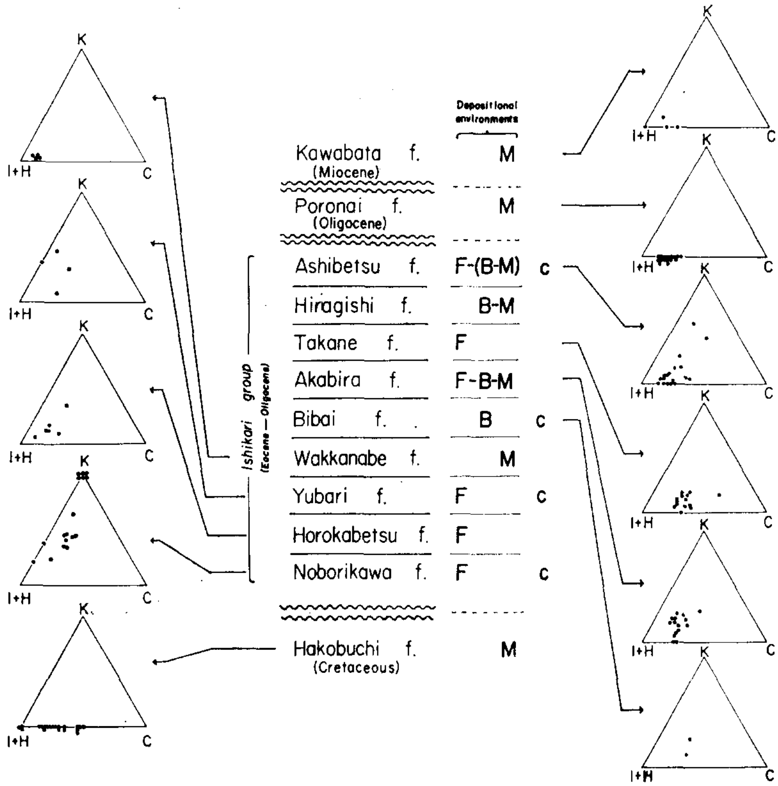

FIG. 5. Stratigraphy and depositional environments of formations in Ashibetsu district and triangular diagrams of kaolinite, chlorite and illite plus hydrous illite, in which the specimens were plotted. Depositional environments—F: fresh water, B: brackish water, M: marine. c: coal-bearing. Depositional environments of Ishikari group were reported by Tsutsumi (1963). Triangular diagrams—K: kaolinite, C: chlorite, I: illite, HI: hydrous illite.

The clay mineral composition of the specimens is represented in Fig. 5 in the form of triangular diagrams of kaolinite, chlorite and illite + hydrous illite. Marine deposits, such as the Hakobuchi, Poronai and Kawabata formations, do not contain kaolinite, whereas the Ishikari group does. Although its depositional environment varies, ranging from fresh water to marine,

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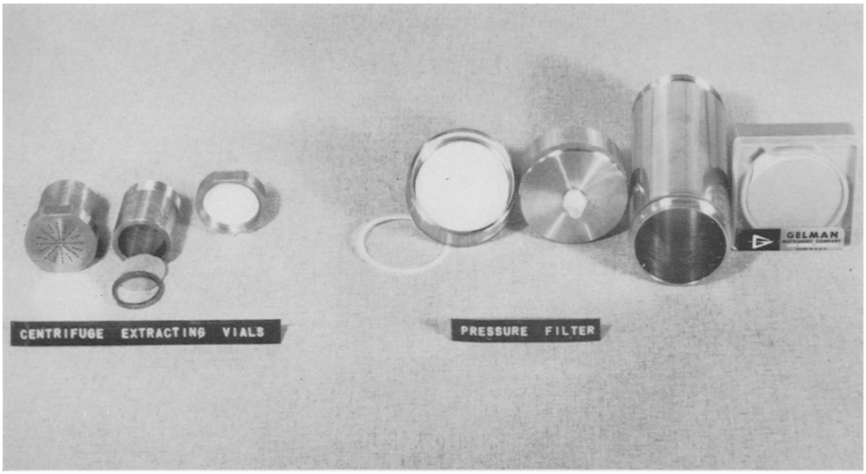

PLATE 1. Extracting vials and pressure filter.

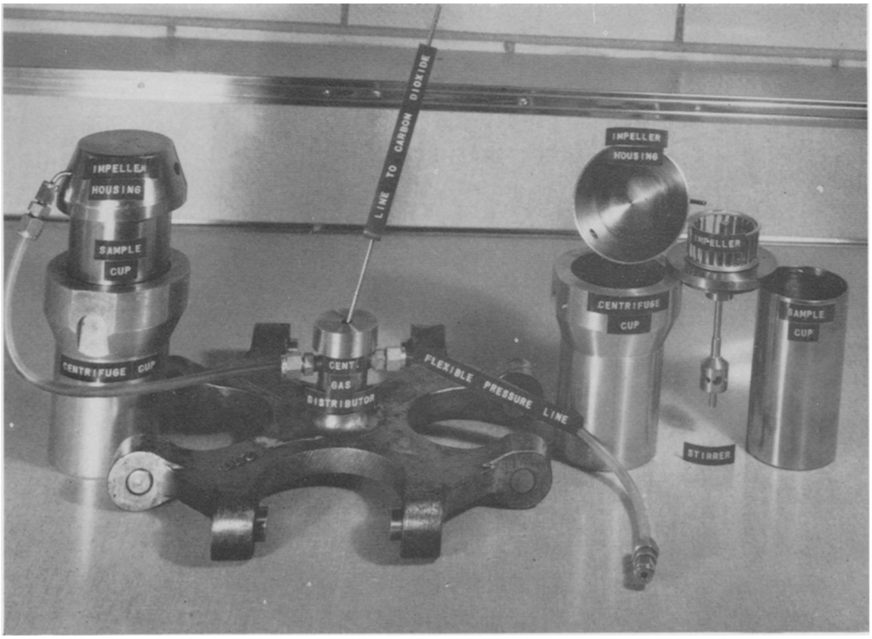

PLATE 2. Stirring assembly and gas power distributing system.

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PLATE 3. Centrifuge and gas power source.

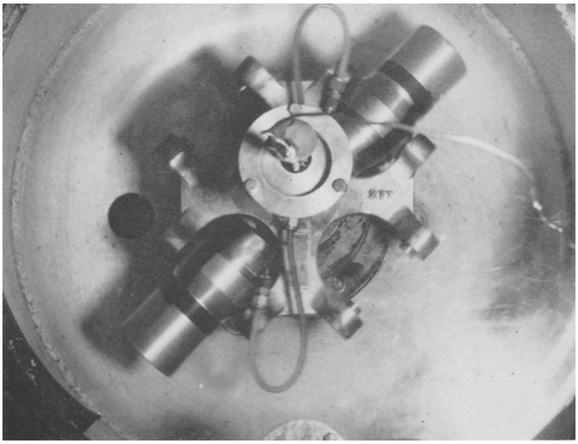

PLATE 4. Centrifuge in operation, 2200 rpm.

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there is the tendency for a marine formation to contain a smaller amount of kaolinite. A typical example is the Wakkanabe formation, whereas the specimens of the Noborikawa, Yubari and Ashibetsu coal-bearing formations contain relatively abundant kaolinite. In the lower part of the Noborikawa coal-bearing formation, just above the unconformity, there was a bed of so-called fire clay which consists almost entirely of kaolinite. There seems to be an enrichment of kaolinite in the beds near the coal seams.

The clay mineral composition in this district seems to have been strongly influenced by chemical factors such as salinity and pH of the depositional environments. An example of the effect of salinity can be demonstrated by the fact that kaolinite is rich in the formations deposited under a fresh-water, or nearly fresh-water, environment. Moreover, the amount of kaolinite is particularly large in the coal-bearing beds of fresh-water origin. This is very reasonable because the environment becomes considerably acidic as the result of the deposition of peat, and generally the acidic condition is suitable for the existence or *in situ* formation of kaolinite. On the contrary, the amount of kaolinite in the formation deposited under the marine environment is very small or negligible. This may be interpreted as the preference of the marine environment for illite and chlorite over kaolinite.

Clay minerals in these specimens show a higher grade of crystallinity than those in the Recent sediments. Consequently, it is inferred that their crystallinity has become better with lapse of time after their deposition.

Mesozoic and Paleozoic Formations of Tohoku Region

The specimens of Mesozoic and Paleozoic formations in the Tohoku region were collected from localities at intervals of 20 to 30 km. These formations consisted of marine sedimentary rocks.

All the specimens from Paleozoic formations (mostly Permian) at five localities contain chlorite and illite. The Triassic specimens from two localities contain also chlorite and illite. The Jurassic specimens from one locality contain illite only, but those from the other localities have chlorite and illite. In the Cretaceous specimens from two localities, chlorite and illite are found in one locality and chlorite only in the other.

As mentioned in the foregoing paragraph, the clay mineral composition of Paleozoic beds are characterized by chlorite and illite, and do not show any conspicuous local variations. The clay mineral compositions of Mesozoic beds are represented also by chlorite and illite, but their amounts vary with locality to a considerable extent. This difference between the Mesozoic and Paleozoic beds might be due to difference in their depositional conditions. In the Paleozoic era, the sediments were probably deposited homogeneously in a relatively wide area. Later, in the period of deposition of the Mesozoic beds, some of the sedimentary basins came to have regional characteristics and might have received material from a different source. This would cause a local variation in the clay mineral composition of the Mesozoic beds. Another

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explanation for the similarity of the clay mineral compositions among Paleozoic beds, is that the present materials were gradually altered to chlorite and illite. In fact, the clay mineral compositions are probably related to both depositional conditions and time.

CONCLUSIONS

The clay minerals contained in some of the sedimentary rocks of Japan and in the Recent sediments of Northwestern Pacific Ocean, Eastern Sea and at the entrance of Tokyo Bay, are shown to be montmorillonite, chlorite, illite, hydrous illite and kaolinite. The Tertiary sedimentary rocks and the Recent sediments show complex combinations of clay minerals, whereas the Mesozoic and Paleozoic sedimentary rocks have simple compositions with generally good crystallinity. In the specimens of Paleozoic sedimentary rocks, especially, the local variation in the composition is small. The hinterland of the area of deposition, mineral composition of the source material, the chemical condition of the depositional environments, the physical conditions such as the ocean and tidal currents and the topography of the area of deposition can all be listed as factors affecting the clay mineral composition. Also post-depositional diagenesis would naturally influence the clay mineral composition of sedimentary rocks.

Since all these factors determine the clay mineral composition of sediments and sedimentary rocks, it is difficult to point out the strongest agent controlling clay minerals. In some cases, however, a certain factor may predominate. For example, the specimens from the Eastern Sea seem to show a strong influence of the source. In the specimens from Tokyo Bay, the amount of montmorillonite characteristically shows local variations suggesting the influence of sea water movements. In the sedimentary rocks of the Ashibetsu district, an influence of chemical conditions of the depositional environment is evident. A somewhat uniform composition of clay minerals in the Paleozoic rocks may be ascribed either to similar supply source and chemical conditions during deposition, or to diagenesis which led to the formation of illite and chlorite.

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