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Preliminary Report on the Mineralogical Studies of Bottom Surface Sediments of South Yatsushiro Kai

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Abstract

Mineralogical investigation of bottom surface sediments was conducted in the relatively shallow marine environment of South Yatsushiro Kai. Identification of clay fraction minerals in this study was made chiefly by XRD analysis complimented with SEM method in determining the type of clay and non-clay mineral suites present in the sediments. Based on analysis results, detrital clay minerals are dominated by illite; chlorite is the second most abundant clay mineral type, followed by kaolinite, smectite and the least abundant 10 Å halloysite. This suite occurs in all samples analyzed and closely reflects the type and character of the surrounding source rock and soil sediments. It is unlikely that homogenization of clay mineral suite occurred in this environment since variable transport mechanisms and associated sediment mixing are operative.

Sediment samples also contain considerable proportion of other materials not usually regarded as clay minerals such as quartz, calcite/aragonite, feldspar and subordinate amount of pyrite, hornblende, gypsum and clinoptilolite.

Key words: bottom sediment, mineral suite, clay fraction, South Yatsushiro Kai

Introduction

Yatsushiro Kai is a semi-enclosed, bay-like body of water in the west central margin of Kyushu mainland. It is one of the most thoroughly studied sites of water effluent and environment pollutant in Japan since the discovery of Minamata disease in 1950's. Other significant studies conducted in the area include the ecological investigation of benthic

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ecosystem by Tanaka *et al.* (1987); and the sedimentological report of Rifardi and Ōki (1996) on recent marine sediments of South Yatsushiro Kai which highlights some excellent observations on textural characteristics of bottom sediments and behavior of marine currents.

From the same set of sediment samples analyzed by Rifardi, a supplementary mineralogical investigation was carried out utilizing the XRD and SEM methods of mineral analysis. This paper presents the results of such investigations which aim to obtain a comprehensive summarization of the character, composition and provenance of clay mineral and non-clay mineral constituents in the sediments. It should be noted that clay analysis in this report is not quantitative but merely comparative interpretation of intensities of X-ray reflections and description of clay's morphological characteristics.

Although this paper contains mostly mineralogical data, some relevant information on continental and marine transport / deposition are briefly discussed to support the depositional features and origin of the mineral suite.

Background

A. Geology

Like most of the fine grain size phyllosilicates found in marine environment, the clay and non-clay mineral suites present in the bottom sediments of South Yatsushiro Kai are detrital or allogenic and reflect the composition of the source rocks. Apparently, mineral distribution is closely linked to the vast exposures of sedimentary, volcanic and metamorphic rock formations observed in the adjacent areas as shown in Fig. 1.

The oldest rocks sorrounding the bay are early Paleozoic low grade metamorphic rocks in the northeast coast which consist mostly of biotite slate and some unmetamorphosed mudstone and sandstone. Deposits of limestone, chert and mafic volcanic rocks are of subordinate amounts (Katada and Yamada, 1977).

Most islands in the west side of the bay are underlain by: 1) several late Cretaceous sedimentary formations of the Goshonoura, Onagawa, and Shimanto Groups which are composed mainly of mudstone, shale and conglomerate and by 2) Eocene clastic rocks consisting predominantly of mudstone and sandstone with coal seams. This Paleogene formation was subsequently intruded by quartz diorite and granite porphyry during middle Miocene time.

Pyroclastic materials and lava flows of hornblende andesite cover most part of the area S-SE of Yatsushiro Kai. These rocks are the same volcanic rocks found in Nagasaki prefecture which were repeatedly erupted during Neogene volcanism (Isshiki, 1977).

Recent deposits composed of unconsolidated silt, sand and gravel are commonly found on river beds and floodplain while terrace deposits occupy most of the river banks and adjacent low lying terrane.

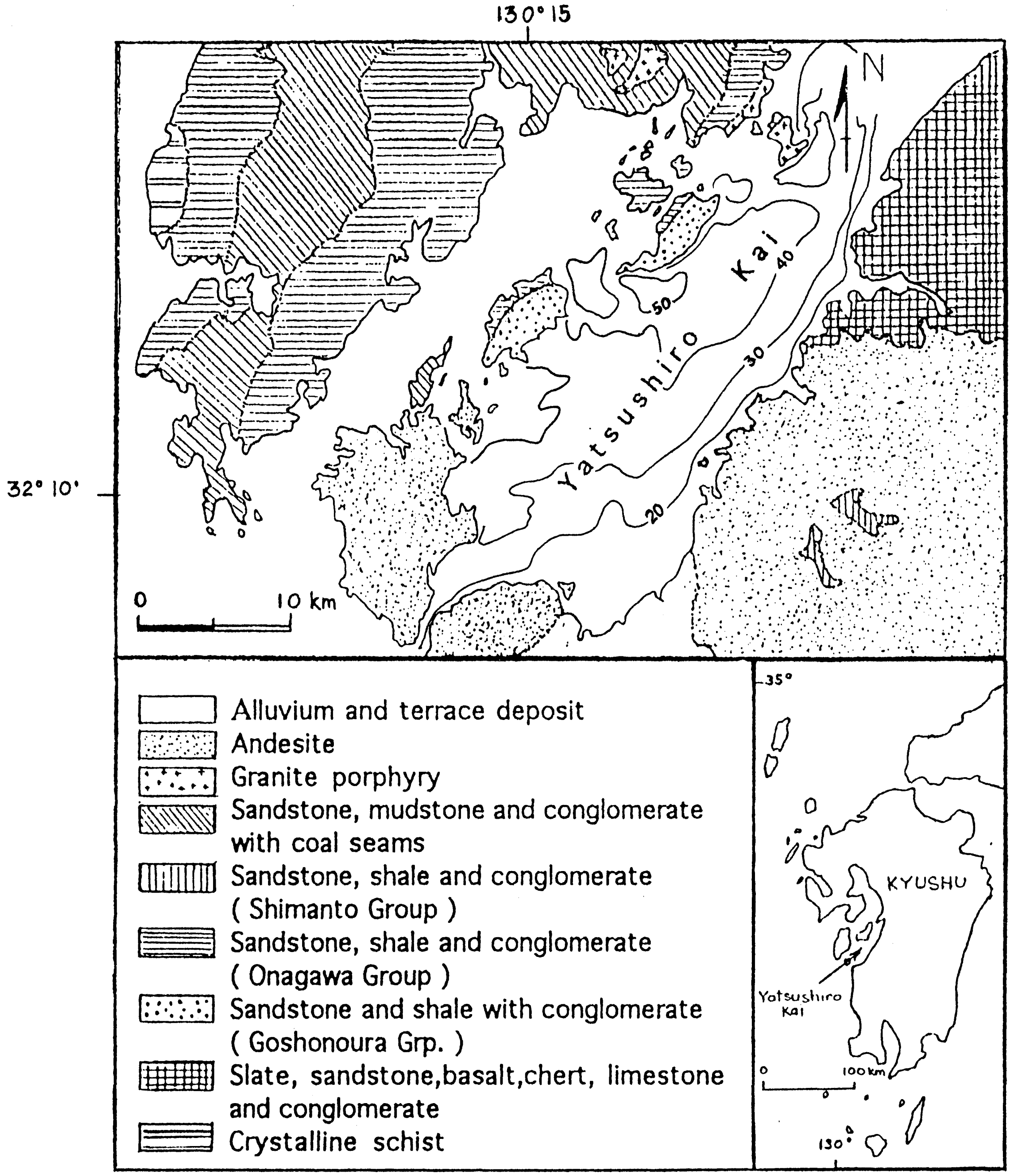

Figure 1 Geological map of the area surrounding South Yatsushiro Kai (after Imai *et al.*, 1979)

B. Environmental Condition of South Yatsushiro Kai

The presence of several islands which nearly enclosed the coastal water of the west central margin of Kyushu mainland has brought the present bay-like configuration of South Yatsushiro Kai. It is connected to the deeper marine environment by three main bay-mouths; two of them are located in the western side of the bay and the other one in the south. Based on the environment's geomorphological outline, Yatsushiro bay might had been formed from valley drowning when sea-water rises during land submergence. This is supported by the existence of several features of submerged shorelines where estuaries have developed in river mouths.

Some characteristic features of the bay are given in Table 1. All data presented in this table were taken from the initial report of Rifardi and Ōki (1996).

Table 1. Characteristic features of South Yatsushiro Kai

Area	Approximately 546 km^2 (lenght - 39 km ; width - 14 km)
Submarine Topography	Characterized by moderately gentle slope except in some portions near the western coast where channel-like topography exists.
Bottom Current	North and south to southeasterly flowing bottom currents are active near bay-mouth areas and becoming less active toward the shallower central area.
Tidal Current	Strong tidal currents persist within the bay-mouth areas and also along or near the shorelines .
Sediment Material	Sand- size materials predominate in the surface sediments but silty sand to muddy sand are common near the east coast.
Sediment Texture	Generally poorly sorted; coarser grained sediments occupy the deeper portion of the bay where strong bottom currents exist; sand to silt size materials are commonly found in coastal areas while silty sand to muddy sand sediment predominate near the center of the bay. Also, some fine sediments are strikingly concentrated in the northeastern shore.
Water Depth	Commonly ranging from 40m to 50m depth near the western coast and progressively decreasing toward the shallower eastern coast.
Bottom Water Temperature	Ranging from 9°C to 11.8°C (during March 1996 sampling)
Bottom Water pH	Ranging from 7.3 to 7.8 (surface water pH is relatively lower toward the near- shore environment particularly in river-mouth estuaries.)
Bottom Water Salinity	Ranging from 33% to 39%

C. Sediment Transport and Deposition

The large bulk of source sediment material surrounding the bay is transported by several continuously flowing rivers. It is later deposited in near-shore environment and only during major storms or floods does much sediment escape the near-shore area. Theoretically, most of the coarse sediments are deposited near the coast while much of the fine grained sediments (silt to mud size fractions) containing clay minerals are distributed seaward to the mudline area at a depth at which the proportions of clayey silt, silty clay and clay reach a near maximum value (Stanley and Wear, 1978). However, some mud sediments are trapped and remain in near-shore environment, most frequently near river estuaries and subaqueous deltaic plain. In most cases, a significant amount of fine clays are held in suspension during river transport and once these suspended materials escape the river and estuaries, they are exposed to various marine currents which determine how much are trapped on the near-shore areas and how much escape to the deep marine environment (Weaver, 1989).

Sediments which are previously deposited on the offshore marine environment can be transported by either active tidal current or bottom current and redeposited them to the near shore environment. The resulting reworked sediments are usually a mixture of material from the two source areas.

Material and Method

The sixty (60) sediment samples used in this study represent the upper two (2) centimeters of the core sediments collected by Rifardi. Figure 2 shows the map indicating the sampling locations of all specimens analyzed.

All samples were examined by X-Ray Diffractometry (XRD) method while only representative specimens were observed using the Scanning Electron Microscope (SEM). Procedures of mineralogical investigation are given in Figure 3.

For X-ray analysis, two sets of sample preparation were made for each raw sample; one for clay mineral ($<2\mu m$) identification and the other for non-clay (bulk) mineral identification. Separation of the clay ($<2\mu m$) from silt fraction was made by sedimentation and centrifugation and the obtained slurry of $<2\mu m$ particles was sedimented and dried on a glass slide (oriented mount sample) for clay analysis. All oriented samples were then subjected to ethylene glycol solvation while some selected $<2\mu m$ specimens were separately prepared for HCl treatment. Untreated, glycolated and HCl treated samples were all subjected to the Rigaku (Geigerflex) X-ray diffractometer under 30 Kv, 15mA operating condition. For non-clay mineral identification, bulk samples preparation include some procedures like air drying and powdering of raw samples prior to mounting on the glass slide.

To determine the morphological characteristics of the clay size fraction minerals, a special high magnification Scanning Electron Microscope (Hitachi S-41005) was used. Specimens for this analysis were prepared by placing raw samples on a double sided adhesive tape sputtered with Au-Pd alloy to ensure electrical conductivity and to prevent

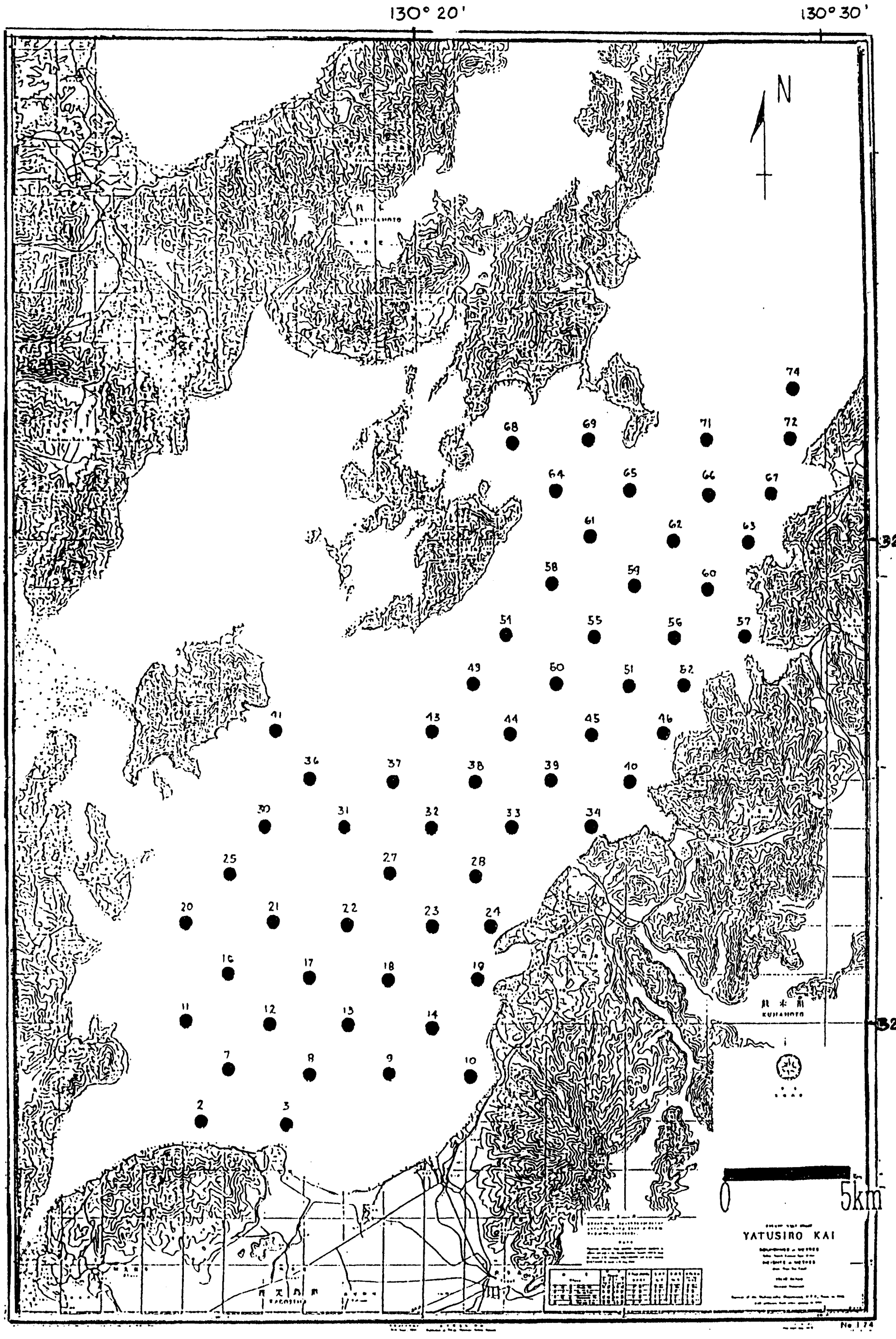

Figure 2. Map of South Yatsushiro Kai showing sampling locations of all bottom sediment samples analyzed.

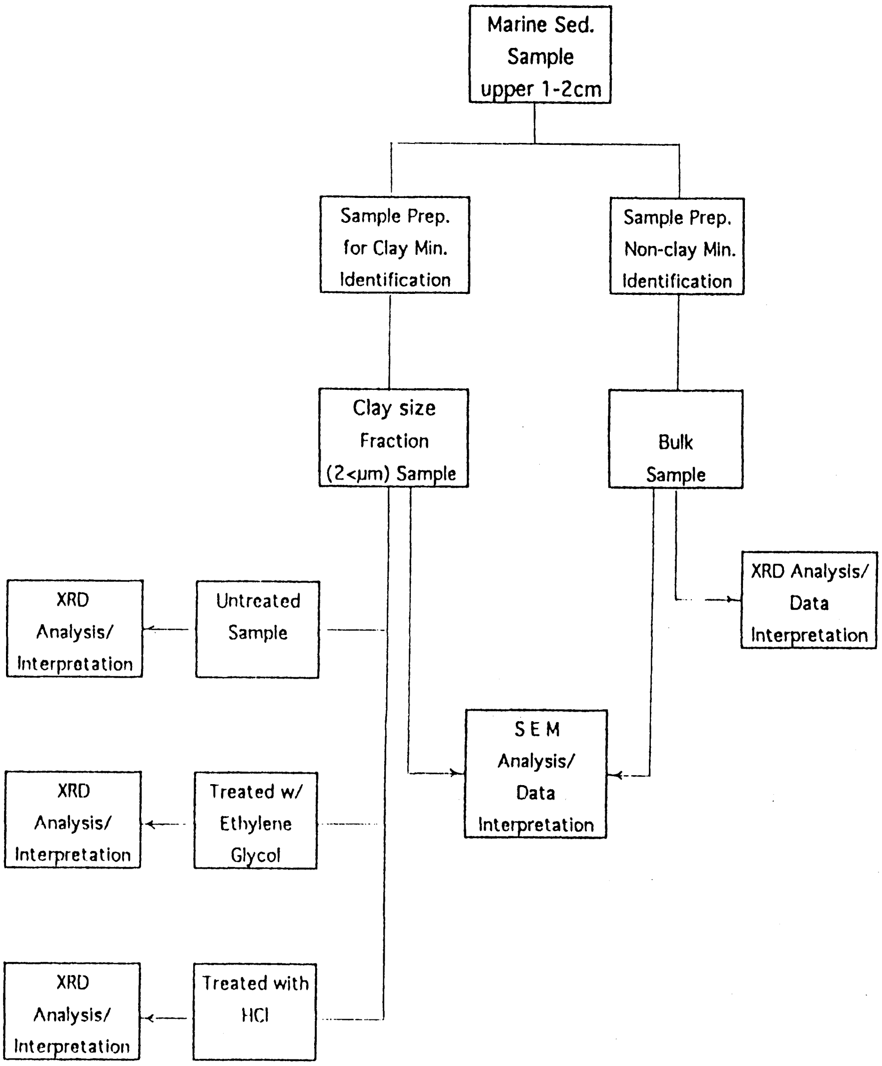

Figure 3. Flow chart showing procedures of mineralogical investigation of bottom sediment samples

charging effects. Images generated by the emission of secondary electrons were examined in the phosphorescent screen of the instrument.

Results and Discussions

Analyses of sand, silty sand and muddy sand samples in recent sediments from various localities within South Yatsushiro Kai indicate similar clay mineral suite of *Illite-Chlorite-Kaolinite-Smectite-10Å Halloysite* type (in decreasing abundance), but relative proportions of these clay species vary from sample to sample. It seems that water currents in this environment are not particularly effective in segregating clay minerals by size. In part, this is because the clay minerals commonly occur as mixed mineral aggregates and even if they are dispersed, they commonly are deposited on and in sands during conditions of slack water after the sand is deposited (Weaver, 1989).

The most striking feature of the clay mineral suite is the presence of relatively high content of illite and chlorite in almost all specimens. Its been noted that chlorite is always a significant component in areas of illite-high, both in near-shore and offshore environments. Kaolinite and smectite are also important clay mineral species in the sediments but their presence is considerably variable. Kaolinite has a point source and is derived almost entirely from several major rivers containing kaolinite-altered materials while smectite occurs as alteration product of volcanic rocks and is commonly dispersed seaward by active marine currents. The clay mineral 10 Å halloysite is commonly of subordinate amount but nearly present in all samples.

Among the non-clay minerals identified, quartz is predominant in all specimens, along with feldspar and calcite/aragonite. Minor amount of other detrital minerals are present like pyrite, gypsum, hornblende and zeolite (probably clinoptilolite)

It is plausible that all the phyllosilicates and non-clay minerals in the sediments of South Yatsushiro Kai were current transported (density current or bottom-water flow and shallow water currents) and derived from the major rivers draining the area of west central Kyushu.

Identification of detrital minerals and their provenance are discussed below.

CLAY MINERAL COMPONENT

Illite

The diffraction pattern of illite is characterized mainly by intense peak in the region of 10Å and a relatively weaker peak at 5Å. These peaks remain unaltered upon ethylene glycol solvation and upon treatment with 6N HCl at boiling condition (Figure 5). Well defined X-ray reflection is attributed to the moderately high crystallinity of illite mineral in almost all samples.

In electron micrograph, illite appears thin and platy (Figure 4B, 4E) and sometimes shows a crude hexagonal habit. It usually occurs on the surface of fine sediment materials (sandstone and slate grains) and frequently associated with mineral chlorite.

Table 2. List of clay minerals present in the bottom sediments of South Yatsushiro Kai

Sample No.	Clay Mineral Component				
	Smectite	Chlorite	Illite	Kaolinite	Halloysite
St-2	*	**	**	*	**
St-3	*	**	***	*	*
St-7	*	**	**	**	*
St-8	*	**	**	*	**
St-9	*	**	***	**	...
St-10	*	**	**	*	**
St-11	**	**	**	*	*
St-12	*	**	**	**	**
St-13	*	**	***	*	*
St-14	*	**	**	**	**
St-16	*	**	**	**	*
St-17	*	**	**	**	*
St-18	**	***	**	*	*
St-19	*	*	**	**	*
St-20	*	**	**	**	**
St-21	**	**	***	*	*
St-22	**	**	***	*	**
St-23	*	**	***	**	...
St-24	*	*	**	***	**
St-25	*	*	**	**	*
St-27	*	**	***	*	*
St-28	*	**	**	*	*
St-30	*	**	***	*	*
St-31	*	***	***	*	...
St-32	*	**	***	**	...
St-33	*	**	**	**	*
St-34	**	**	**	*	*
St-36	*	**	**	**	*
St-37	*	**	**	*	*
St-38	*	***	**	*	*
St-39	*	**	**	***	...
St-40	*	*	**	**	**
St-41	*	**	***	*	*
St-43	*	***	**	*	*
St-44	*	***	**	*	...
St-45	*	***	**	*	*
St-46	**	**	**	*	*
St-49	**	**	**	*	**
*** abundant ** common * rare ... absent					

Table 2 (cont'd)

Sample No.	Clay Mineral Component				
	Smectite	Chlorite	Illite	Kaolinite	Halloysite
St-50	*	**	**	**	*
St-51	*	**	**	**	...
St-52	*	**	**	*	**
St-54	*	**	***	**	*
St-55	*	*	**	***	*
St-56	**	**	**	*	*
St-57	**	***	**	*	*
St-58	*	**	**	*	*
St-59	*	**	**	**	*
St-60	*	**	**	*	*
St-61	*	**	**	**	*
St-62	*	**	***	**	*
St-63	**	**	***	*	*
St-64	**	**	**	*	*
St-65	**	***	***	*	*
St-66	*	**	**	**	*
St-67	**	**	**	**	...
St-68	*	**	**	*	*
St-69	*	***	**	*	...
St-71	*	**	**	**	*
St-72	*	**	**	**	...
St-74	*	**	**	*	**

*** abundant ** common * rare ... absent

Table 3. List of non-clay minerals present in the bottom sediments of South Yatsushiro Kai

Sample No.	Non-clay Mineral Component							
	Quartz	Feldspar	Calcite	Aragonite	Clinoptilolite	Gypsum	Hornblende	Pyrite
St-2	••	•••	•••	•	...	*	•	...
St-3	•••	••	•	*	...	*	••	*
St-7	••	•••	•••	•	...	...	...	...
St-8	•••	••	••	*	*	...	*	...
St-9	•••	••	•	...	...	*	*	...
St-10	••	••	••	*	*	...	•	•
St-11	••	••	•••	••	...	...	...	*
St-12	••	••	••	•	*	*	•	*
St-13	••	••	••	*	*	*	*	...
St-14	•••	••	••	...	*	...	...	*
St-16	••	•••	••	••	...	...	...	...
St-17	••	•••	••	...	...	...	*	*
St-18	••	••	••	...	*	*	*	•
St-19	••	•	••	...	•	*	*	...
St-20	•••	••	••	••	...	...	*	*
St-21	•••	••	••	*	*	...	•	•
St-22	••	••	••	...	...	...	*	...
St-23	••	•••	••	*	•	•	...	...
St-24	••	••	••	...	...	*	*	•
St-25	•	•	••	•	*	*	...	*
St-27	••	••	•••	*	*	...	•	*
St-28	••	••	•••	...	*	*	•	...
St-30	••	••	•••	••	...	...	*	*
St-31	••	••	•••	••	...	*	*	•
St-32	•••	••	•••	••	...	...	*	...
St-33	•••	••	••	...	*	...	•	...
St-34	•••	•••	•	...	*	...	*	•
St-36	••	•	•••	••	...	...	*	•
St-37	••	••	•••	••	...	*	*	...
St-38	•••	••	•••	••	*	...	•	*
St-39	••	•	••	*	*	*	•	*
St-40	••	••	•	...	•	...	...	...
St-41	••	••	•••	••	*	*	*	...
St-43	•••	••	•••	*	...	...	*	...
St-44	••	•••	••	•	•	*	...	...
St-45	••	••	••	...	*	*	*	•
St-46	•••	•	•	...	...	*	*	...
St-49	••	••	•••	*	*	•	•	...

••• Abundant •• Common • Rare * Traceable ... Absent

Table 3 (cont'd)

Sample No.	Non-clay Mineral Component							
	Quartz	Feldspar	Calcite	Aragonite	Clinoptilolite	Gypsum	Hornblende	Pyrite
St-50	••	•	••	••	...	...	...	*
St-51	••	•	••	••	...	...	...	*
St-52	••	••	••	...	*	...	*	...
St-54	•••	•••	•	...	*	*	...	...
St-55	••	••	••	•	*	*	*	•
St-56	••	••	••	...	*	...	...	*
St-57	••	••	•	...	*	*	•	•
St-58	•••	•••	•	...	...	...	*	...
St-59	••	••	••	*	•	...	...	...
St-60	•••	••	••	*	*	*	...	*
St-61	•••	••	••	*	...	*	•	*
St-62	•••	••	•	...	•	•	•	...
St-63	••	••	•	...	•	•	•	*
St-64	•••	•	•	...	*	*	*	*
St-65	•••	••	••	...	...	...	...	*
St-66	••	••	•	*	...	...	*	...
St-67	•••	•	•	...	...	*	...	•
St-68	••	••	••	••	*	...	*	*
St-69	••	•	•	...	*	*	*	*
St-71	•••	••	•	*	*	•	•	*
St-72	•••	••	•	...	...	...	...	...
St-74	•••	••	•	*	...	*	•	*

••• Abundant •• Common • Rare * Traceable ... Absent

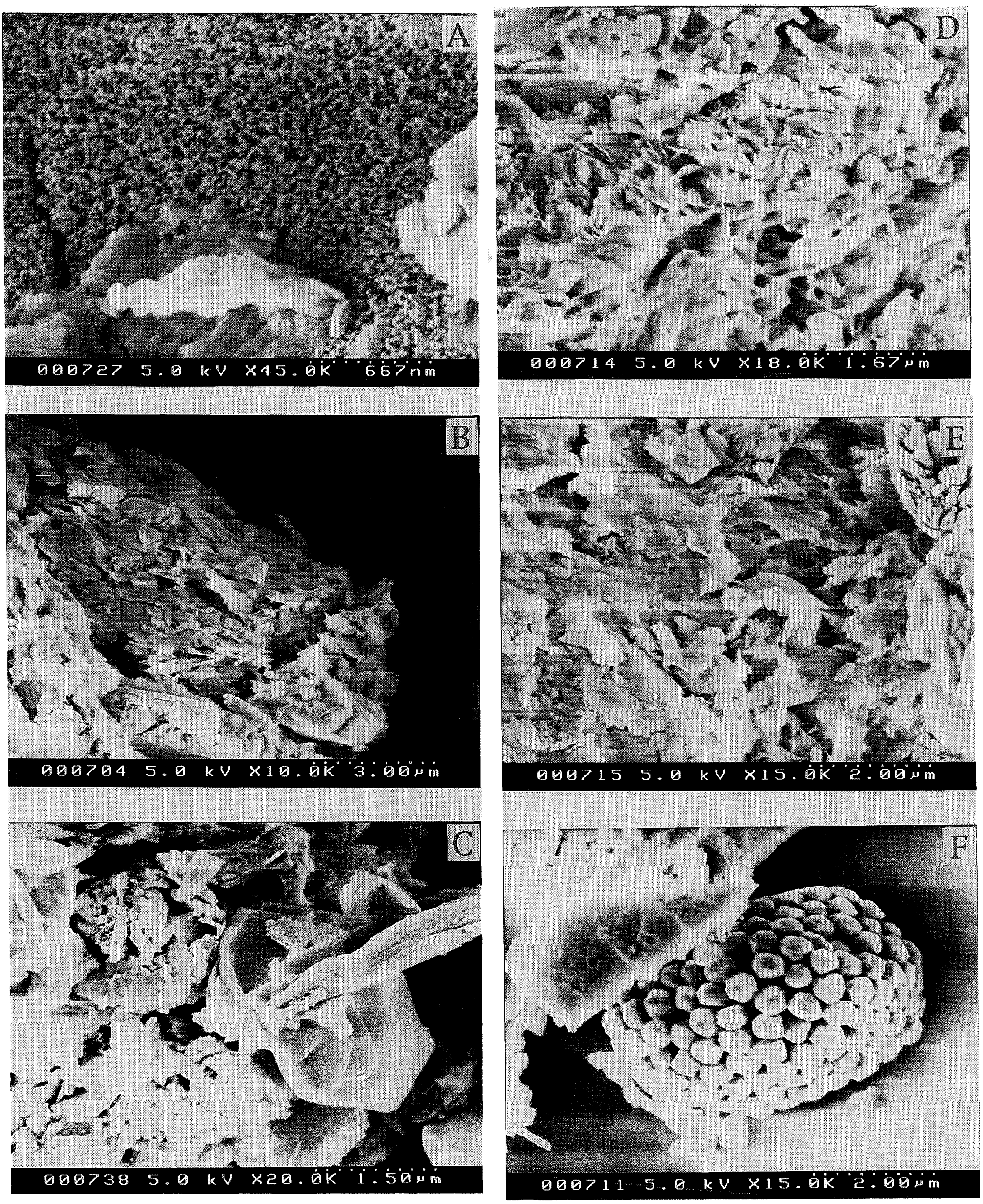

Figure 4. Scanning electron micrographs of selected bulk sediment samples showing smectite - coated surface of volcanic material (A); thin and platy illite minerals associated with minor amount of smectite (B and E); well-formed six sided flakes of kaolinite (C); irregular fluffy masses of smectite (D); and framboidal pyrite (F).

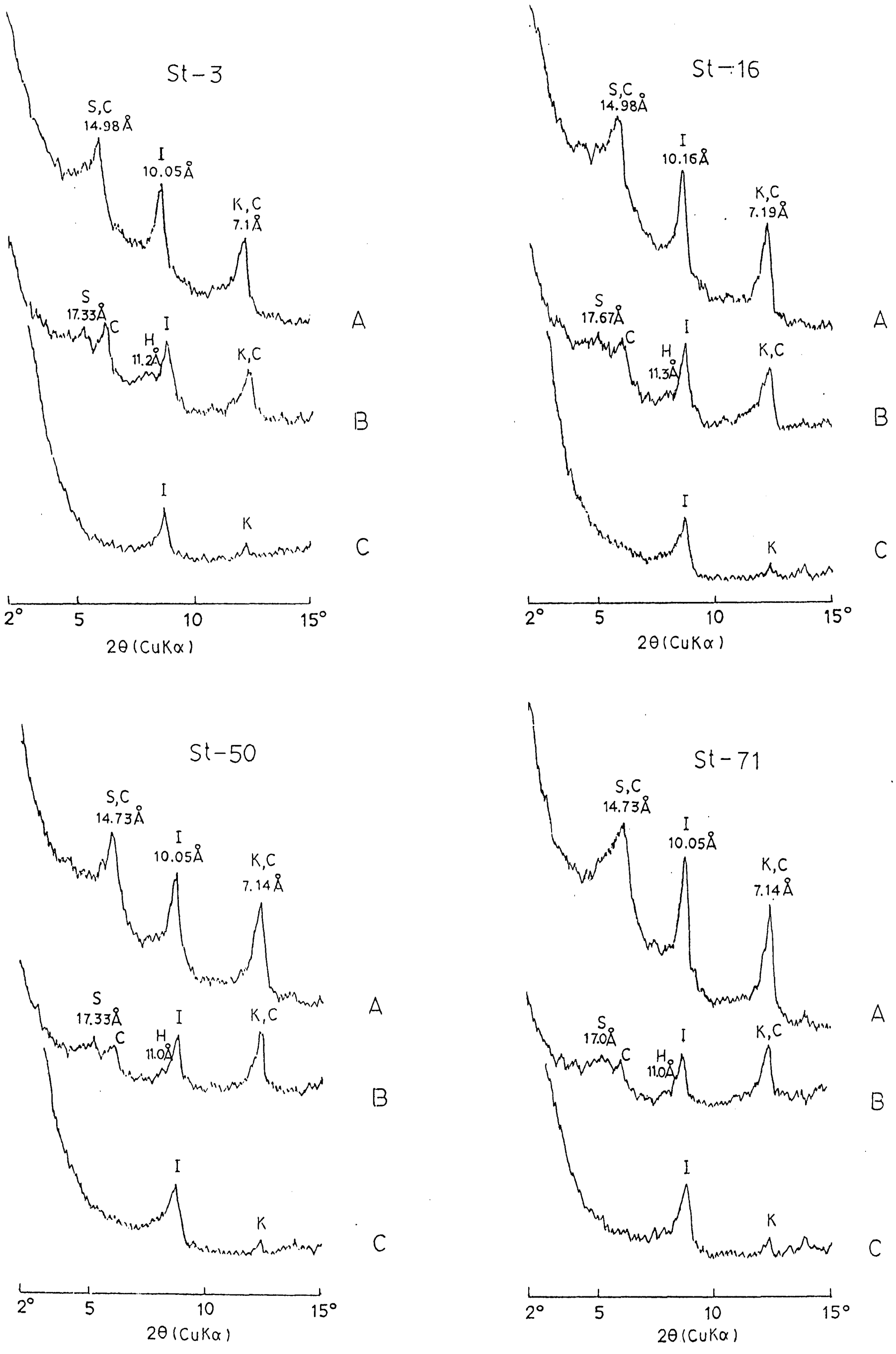

Figure 5. X-ray diffraction patterns of representative clay ($<2\mu m$) samples under various treatments.

A. oriented sample (untreated) ; B. ethylene glycol-treated ; C. HCl-treated (I=illite, C=chlorite, K=kaolinite, S=smectite, H=10Å halloysite)

The high illite content reflects the abundance of Mesozoic sedimentary formations and Paleozoic metamorphic rocks in much of the coastal area fringing the east and west of South Yatsushiro Kai. These rocks might have undergone considerable alteration and weathering to permit accumulation of argillaceous materials in soil sediments which is the ideal condition for illite to become more prevalent.

Chlorite

X-ray identification of chlorite in presence of smectite in the untreated samples is accomplished by the addition of ethylene glycol. This treatment expands smectite to 17-18 Å (Figure 5) whereas chlorite remains unaffected at 14 Å. Differentiation between chlorite and kaolinite with 7 Å reflection was accomplished by subjecting the sample to boiling HCl. This test dissolved the Mg-chlorite mineral and the remaining peak at 7 Å is assigned to kaolinite (Figure 5).

The abundant presence of chlorite not only reflects conditions of mild to no chemical weathering but source rock with a relatively high chlorite content --- low grade metamorphic rock. Generally, chlorite is largely inherited as primary mineral found in slates and igneous rocks near Yatsushiro Kai or it may occur as alteration products from minerals such as hornblende, biotite and other ferro-magnesian minerals (Barnhisel, 1977). In addition, the average shale which has approximately 10% chlorite (Weaver, 1989) may also contribute to its presence.

Kaolinite

As previously mentioned, the 7 Å reflections in the untreated and glycolated samples belong to both kaolinite and chlorite minerals, however, upon HCl treatment only kaolinite reflection remains at 7 Å peak.

Analysis of electron micrographs shows that, in general, poorly crystallized kaolinite showing less distinct six-sided flakes is present in most specimens. The edges of the flakes are somewhat ragged and irregular and the hexagonal outline is only crudely shown. But in few samples, well crystallized kaolinite (Figure 4C) shows well-formed six-sided flakes, frequently with a prominent elongation in one direction.

The concentration of kaolinite mineral in the near-eastern-coast samples is believed to be caused by runoff of coastal rivers which is highly suggestive of the presence of kaolinitic source material, most likely from volcanic terrain.

Smectite

The ethylene glycol saturated samples which have X-ray spacings between 16.9 to 17.1 Å are considered to be due to smectite (Figure 5), however, for untreated samples, the basal reflection peaks appear in the region of 14-16 Å.

In electron micrographs, smectite shows irregular fluffy masses of extremely small particles (Figure 4D). But in some cases the larger masses appear to be stackings of flaked-shape unit without regular outlines. Smectite-coated surface of sand grains (Figure 4A) exhibiting characteristic cellular morphology is noticeable in several specimens.

Commonly, smectite is found to be limited to places where there are neighboring areas of basic volcanic activity. In South Yatsushiro Kai, the most probable source of smectite in the bottom sediments is the prevalence of altered pyroclastic materials and lava flows of hornblende andesite surrounding the bay and in part, the considerable deposition of smectite-rich Cenozoic sedimentary rocks near the coastal area.

Smectite in most cases, preferentially stays in suspension and can be transported a long distance by marine currents. This is supported by the mineral's extremely slow settling velocity which range from .0023 to .088 cm/min in quiescent sea water having low (0.9ppt) to moderate (32.5ppt) salinity (Whitehouse *et al.*, 1960).

The less-abundant character of smectite in the near-coast sediments of South Yatsushiro Kai may be attributed to the active agitation action of waves which causes the fine grained material like smectite to be removed (Oinuma and Kobayashi, 1966).

Halloysite

In the untreated samples, 10 Å halloysite mineral is identified from the 10 Å peak (overlapping illite peak) and shifted to 11 Å upon treatment with etylene glycol (Figure 5). The halloysite peak is characterized by weak, broad reflection owing to its curved disorderly particles.

Some loose aggregates of halloysite spheroids are observed in scanning electron micrographs which are possibly formed from volcanic glass and feldspar. Recent studies from many parts of the world indicate that this highly disordered form of kaolinite has been identified mostly in weathered volcanic and other igneous deposits (Dixon, 1977) and is degraded in materials derived from sedimentary rocks.

NON-CLAY MINERAL COMPONENT

Quartz

Quartz in bulk (raw) samples is easily identified because it yields a characteristic X-ray pattern with an intense, well defined peaks at 3.34 and 4.26 Å (Figure 6). Nearly all of the quartz in the total clay fraction ($<2\mu m$) of sediments is concentrated in the coarse clay fraction (0.2 to 2μm) (Kunze and Oakes, 1957). Surface morphology of quartz is basically recognized by the occurrence of conchoidal breakage pattern and the presence of flat cleavage plates.

Sources of quartz in the area may include sandstone (average sandstone contains 67% quartz) (Clarke, 1924), silica-rich igneous rocks and volcanic ash.

Feldspar

In many samples, intense feldspar reflections occur at at 3.23, 3.33 and 3.79 Å (Figure 6). Like quartz, this mineral is also a significant constituent of the bottom sediments ; a major portion of it was derived from the source sedimentary rocks which are believed to be of igneous origin and accumulated in sedimentary environments as weathering residue of the igneous and metamorphic rocks (Rankama and Sahama, 1950).

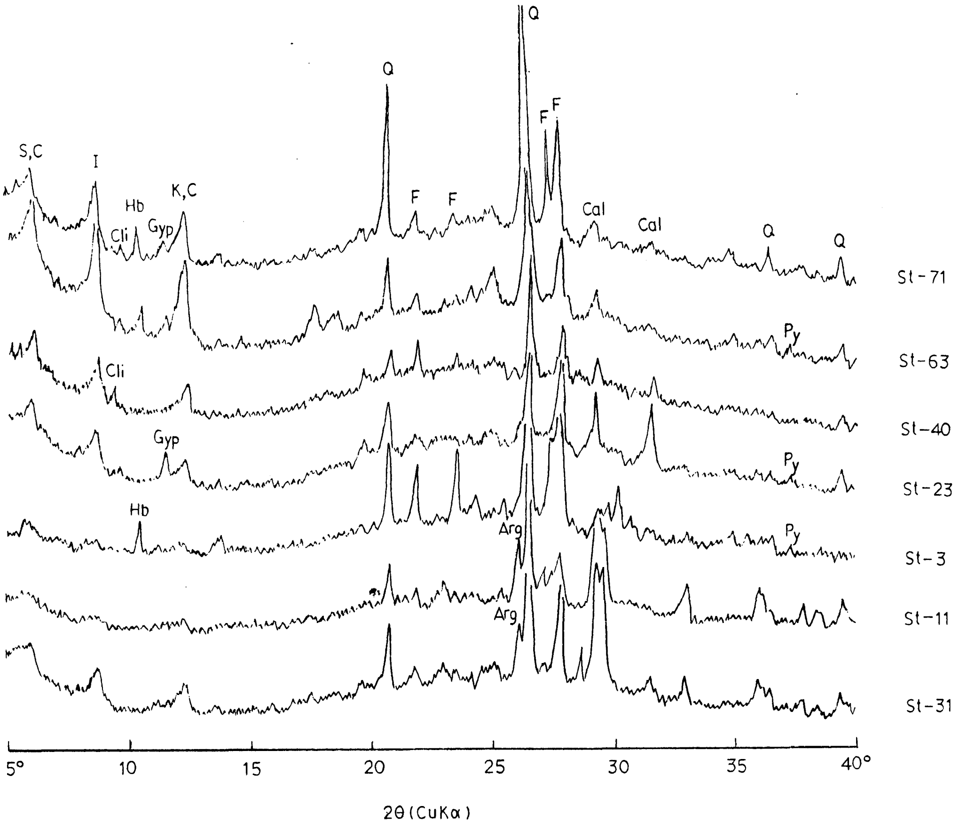

Figure 6. X-ray diffraction patterns of bulk sediment samples
(Q=quartz, F=feldspar, Cal=calcite, Arg=aragonite, Py=pyrite, Gyp=gypsum, Hb=hornblende, Cli=clinoptilolite, S=smectite, C=chlorite, K=kaolinite, I=illite)

Calcite/Aragonite

Calcium carbonate minerals (calcite and aragonite) are present in the samples as particles or coatings over or between other particles. Calcite is identified from the strong 3.03 Å peak while aragonite is very evident at 3.40 Å reflection (Figure 6). Coarse aggregates of coral material and other biogenic debris are frequently associated with fine fractions of calcite; aragonite needles, which are typical in fine carbonate mud, are considerably abundant.

Sources of calcite may occur either from the original soil parent materials or as the result of pedogenic processes and its widespread presence in the bottom sediments of South Yatsushiro Kai indicates that the area has been a favorable condition for its precipitation (Carranza *et al.*, 1994; 1996).

Gypsum

Characteristic strong peak at 7.62Å is attributed to gypsum (Figure 6). Scarse distribution of this mineral gives clue to limited point source. As with the deposition of carbonate minerals, $CaSO_4$ is transported by the soil solution and gypsum precipitates when the solubility of the mineral is exceeded.

Pyrite, Hornblende, Clinoptololite

Pyrite mineral in bulk samples is detected at 2.41Å peak (Figure 6), although other Fe-bearing minerals may also be present. Microscopic framboidal pyrite (Figure 4F) is common in several samples which exhibits spheroidal cluster of pyrite grains resembling raspberry seeds. To many mineralogists, framboid was considered to be the result of colloidal processes but is now linked with the presence of organic materials.

Among the ferromagnesians, only hornblende mineral is observed in the XRD pattern and it shows a distinct well defined peak at 8.5A region. This type of amphibole is widely distributed in igneous rocks surrounding the bay such as hornblende-andesite, diorite and granite porphyry. In most cases, this mineral occurs in the clay fractions of soil that are later transported by rivers and deposited in marine environment.

Fine grained zeolite (probably clinoptilolite) is also detected in limited samples and gives a characteristic x-ray reflection at 9.02Å. SEM shows some clinoptilolite minerals in a generally long, slender crystal clusters that are commonly arranged in divergent bunches. Ordinarily, this type of zeolite is secondary mineral occurring chiefly in igneous and pyroclastic rocks, especially in volcanic rocks.

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