

## Clay Minerals of the Bottom Sediments on the Northwestern Continental Shelf in the East China Sea

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**Abstract:** The clay minerals of thirty-four bottom sediments collected from the northwestern continental shelf of the East China Sea have been determined by X-ray diffraction analysis. The clay mineral distribution is mainly controlled by the sediment source and the dominant circulation pattern. The predominant clay mineral in our study area is illite comprising more-than 70% of whole clay fraction. The highest concentration of illite (>72%) is found in the southeastern offshore parts beyond the reach of terrigenous input from the Cheju Island. It means that these illites are largely transported by the Kuroshio Current from the South China Sea. Smectite is highly concentrated in the northwest middle part and in the outer-shelf mud patch. It seems to be due to the high supply of smectite transported from China where the fine-grained sediments are discharged from the modern and ancient Huanghe River. The relatively high abundance of kaolinite is likely derived from the Changjiang River via Taiwan Warm Current. In contrast, the large amounts of chlorite and high chlorite/kaolinite ratios occur in the northwestern area, reflecting the transportation by the Huanghai Sea Coastal Current from the southern Yellow Sea.

**Keywords:** clay minerals, sediment source, continental shelf, East China Sea

### Introduction

The East China Sea extends from the north shore of the Changjiang River estuary mouth to the Cheju Island, and through the Ryukyu Island to the southern tip of Taiwan to Fijian. It has an average water depth of about 65 m and NE-SW trending of submarine contour lines which become deep south-southeastward to the northern area of the Okinawa Trough (Butenko et al., 1985). During the Holocene period, the Changjiang and Huanghe Rivers brought huge amounts of sediments to the East China Sea. In fact, the Changjiang River discharges about  $500 \times 10^6$  tons of sediments annually, and the annual budget of the Huanghe River is about  $1,100 \times 10^6$  tons (Milliman and Meade, 1983). Thus, the continental shelf of the East China Sea is mainly dominated by the sediments from these two rivers (Yang and Milliman, 1983).

Clay mineral studies in various marine depositional systems have amply elucidated the provenance, dispersal patterns, transport agents and depositional sites of fine-grained particles (Naidu and Mowatt, 1983; Hume and Nelson, 1986; Park and Khim, 1992), and understanding the distribution and origin of clay mineral assemblages has proved to be a useful tool to interpret the net pathways of fine-grained sediment transport (Piper and Slatt, 1977; Karlin, 1980; Naidu et al., 1982). There have been some previous works on the clay mineralogy of sediments in the East China Sea and adjacent seas (Aoki et al., 1983; Yang and Milliman, 1983; Park and Han, 1985; Aoki and Oinuma, 1988; Park and Khim, 1992; Lee and Chough, 1989). Due to the limited access, however, the East China Sea has not been investigated easily by the Korean scientists. The purpose of the present study is to describe the clay mineral composition of the surface sediments from the northwestern East China Sea, and to discuss possible source of these clay minerals.

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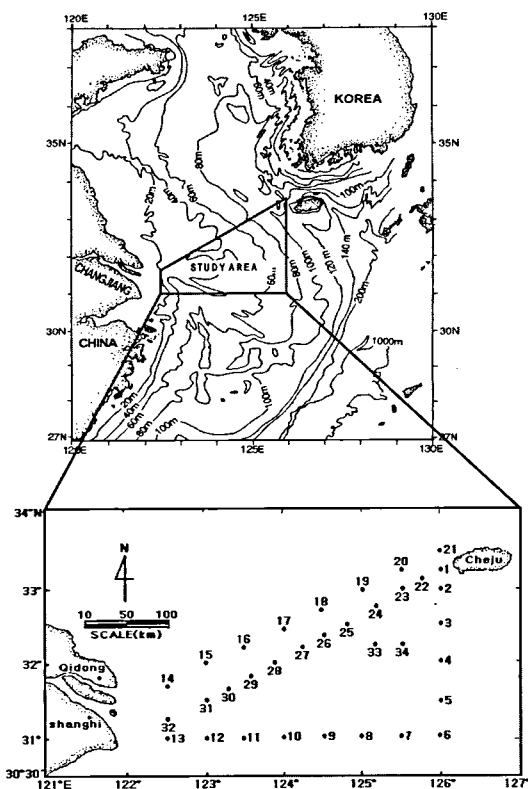


Fig. 1. Map showing the study area, bathymetry, and sites of the surface sediment samples. Depth contours are in meters.

### Sample collection and methods

Thirty-four surficial sediment samples were obtained using grab sampler in the northwestern continental shelf of East China Sea during the period of August 1997 and June 1998 onboard the R/V Ara-1 of Cheju National University (Fig. 1).

Grain-size analyses of the surficial sediments were performed by standard procedure (Krumbein and Pettijohn, 1938). Organic carbon and nitrogen of the sediments were measured using CHN Analyzer following the method of Byers et al. (1978). The less-than  $2\ \mu\text{m}$  clay fraction was smeared on glass slides and air dried (Gibbs, 1965). Glycolation was affected by vapor-phase exposure for 48 hours. All clay specimens were scanned from  $2^\circ$  to  $35^\circ 2\theta$  at  $2^\circ 2\theta/\text{min}$  on a Shimadzu XD-1 X-ray diffractometer using Ni-filtered  $\text{CuK}\alpha$  radiation. A slow scan ( $0.25^\circ 2\theta/\text{min}$ ) between  $24^\circ$  and  $26^\circ 2\theta$  was used to

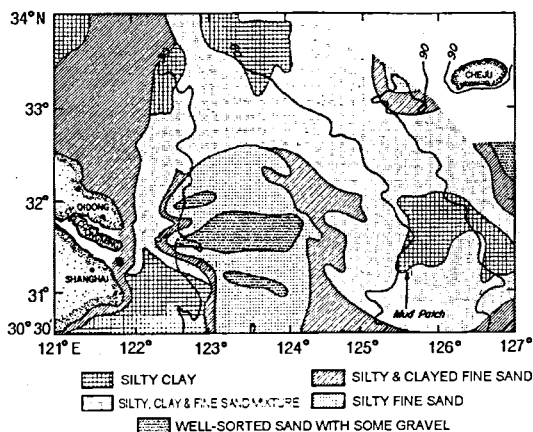


Fig. 2. Distribution of seafloor sediment granulometry in the East China Sea (after Butenko et al., 1985).

differentiate kaolinite from chlorite. Clay minerals were identified from their basal X-ray diffraction peaks according to the criteria outlined by Brindley and Brown (1980). The semi-quantitative determination of relative amounts of major clay minerals was made by measuring the peak area and multiplied by factors of 1, 4, and 2 for smectite, illite and chlorite + kaolinite, respectively (Biscaye, 1965).

## Results and discussions

### Sediment properties

The sediment types near the Changjiang River estuary and on the adjacent continental shelf of the East China Sea are mainly composed of relict sand, silt, mud, silty clay and sand-silt-clay mixture sediments (Fig. 2; Butenko et al., 1985).

Modern terrestrial fine-grained sediments consisting of silty clay, silt and mud are distributed at the mouth of the Changjiang River estuary, extend southward along the coast, and also occur in the southwest offshore of the Cheju Island (Fig. 2). The inner-shelf mud deposits are composed of 8.9% of sand, 59.1% of silt, and 32.1% of clay with  $6.9\phi$  of mean grain size. This sedimentary deposit has more silt than clay fraction, and is coarser than that of the outer-shelf mud deposit to the southwest of Cheju Island (Table 1). The outer-shelf mud deposit

**Table 1.** Sediment type, textural parameter, physical property and C/N ratios

Sedi- mentary region	Station No.	Sediment composition			Sedi- ment type	Textural parameters				Physical property		Organic matter		
		Sand (%)	Silt (%)	Clay (%)		Mean ( $\phi$ )	Sorting ( $\phi$ )	Skew- ness ( $\phi$ )	Kurtosis ( $\phi$ )	Water content (%drywt)	Bulk density (g/cm <sup>3</sup> )	Organic- C (%)	Organic- N (%)	C/N Ratio (atomic)
Inner- Shelf Mud Deposit	CJ97012	3.9	63.3	32.8	Z	7.1	2.39	0.40	0.59	34.2	0.78	0.659	0.061	9.4
	CJ97013	2.2	60.9	36.9	M	7.3	2.27	0.29	0.61	42.6	1.05	0.733	0.075	10.8
	CJ98032	20.6	53.0	26.5	sZ	6.3	2.55	0.52	0.72	42.8	0.75	0.542	0.043	10.2
	Mean	8.9	59.1	32.1		6.9	2.40	0.40	0.64	40.0	0.86	0.645	0.060	10.1
Outer- Shelf Mud Deposit	CJ97004	1.2	34.9	64.0	M	8.7	1.83	-0.21	0.77	59.5	0.59	0.843	0.091	9.7
	CJ97005	0.5	35.3	64.3	M	8.7	1.80	-0.31	0.82	58.2	0.60	0.791	0.095	7.5
	CJ97007	15.9	33.5	50.7	sM	7.5	2.77	-0.31	0.83	51.8	1.45	0.636	0.072	8.8
	Mean	5.8	34.5	59.6		8.3	2.13	-0.28	0.81	56.5	0.88	0.757	0.086	8.7

Note: sM: sandy mud, sZ: sandy silt, M: mud, Z: silt, C: carbon, N: nitrogen

are composed of 5.8% of sand, 34.5% of silt, and 59.6% of clay with 8.3 $\phi$  of mean grain size. This sedimentary deposit is also characterized by abundant clay content, high water content value and high organic matter content (Table 1). Organic carbon and nitrogen values in the outer-shelf mud deposit are 0.76% and 0.09% respectively, which are higher than those in the inner-shelf mud deposit.

At a station near the Changjiang River mouth, the organic carbon and nitrogen contents in the inner-shelf mud area are 0.65% and 0.06% respectively. A close relationship could also be found between grain size of sediment and organic matter. The organic materials in the sediments generally show that higher contents are closely related to the fine-grained sediments. The C/N ratio is commonly used to characterize the origin and type of organic matter (Stein, 1990). The C/N ratios in the study area vary between 7.5 and 10.8 (Table 1), and are higher in the inner-shelf mud deposit (about 10.1) rather than that of the outer-shelf mud deposit (8.7). The C/N ratio exceeding 10 in the inner-shelf mud indicates that large amounts of the organic matter have been supplied from the Changjiang River because C/N ratio of the marine organic matter is normally less than 10 (Stein, 1990).

#### Clay mineral distribution and provenances

In all samples analyzed, illite, chlorite, kaolinite and smectite were identified in varying amounts

(Table 2). The averages of individual clay minerals are 71% for illite, 13% for chlorite, 11% for kaolinite and 5% for smectite.

The illite is the most abundant clay mineral in the northwestern East China Sea, ranging from 69 to 74% of the total clay minerals (Table 2). More than 72% high concentration of illite occurs in the southeastern offshore region beyond the reach of significant terrigenous input, which is opposite to the distribution trend of kaolinite and chlorite in the study area (Fig. 3). Less concentration of illite is found around the Changjiang River estuary and in the west coast of the Cheju Island. The tongue shape of high concentration illite zone on the southeast offshore area tends to decrease gradually toward the southern Yellow Sea from the East China Sea. It is likely interpreted that they are largely transported by Kuroshio Current from the South China Sea via Taiwan Warm Current or Yellow Sea Warm Current. Aoki and Oinuma (1988) also reported that illite is the most dominant constituent (74 to 50%) in the East China Sea and its neighboring oceans, and that the major current affecting sediment dispersal in the East China Sea include the northward flowing Kuroshio Current and Taiwan Warm Current.

Chlorite is the next widespread clay mineral in the study area (Table 2). The highest concentration of chlorite (>14%) and high ratios of chlorite/kaolinite (>1.4) are found in the northwest middle

**Table 2.** Grain-size and clay-mineral data of the surface sediment in the northwestern East China Sea

Station No.	Size analysis (%)				Sediment type	Clay minerals (%)				Chl/Kao
	Granule	Sand	Silt	Clay		Smectite	Illite	Kaolinite	Chlorite	
1	0.3	65.6	9.3	25.3	(g)mS	6	70	12	12	1.0
2	-	91.8	3.4	4.8	S	6	71	11	12	1.1
3	-	27.6	21.4	51.0	sC	4	73	10	13	1.3
4	-	1.2	34.9	63.6	M	6	73	9	12	1.3
5	-	0.5	35.3	64.3	M	6	72	10	12	1.2
6	-	45.7	28.2	26.2	sM	5	72	11	12	1.1
7	-	15.9	33.5	50.7	sM	5	71	12	12	1.0
8	-	48.7	30.0	21.3	sM	4	71	13	12	0.9
9	-	71.1	18.5	10.4	mS	5	72	12	11	0.9
10	-	75.4	12.3	12.3	mS	5	74	10	11	1.1
11	-	68.1	21.0	10.9	mS	3	74	11	12	1.1
12	-	3.9	63.3	32.8	Z	4	72	12	12	1.0
13	-	2.2	60.9	36.9	M	5	70	13	12	0.9
14	-	4.4	64.2	31.4	Z	5	70	12	13	1.1
15	-	83.3	11.6	5.1	zS	7	70	10	13	1.3
16	-	65.2	8.3	26.5	mS	7	71	8	14	1.7
17	-	50.2	30.2	19.6	mS	6	71	9	14	1.5
18	-	68.0	13.3	18.7	mS	5	72	11	12	1.1
19	-	66.3	21.4	12.3	mS	5	72	11	12	1.1
20	-	71.8	21.2	7.1	zS	4	69	13	14	1.1
21	-	68.2	18.3	13.5	mS	6	70	12	12	1.0
22	0.1	52.5	13.6	33.9	(g)mS	6	70	12	12	1.0
23	-	45.9	17.8	36.4	sC	5	71	11	13	1.2
24	-	53.8	14.7	31.5	cS	5	73	10	12	1.2
25	-	42.2	19.7	38.1	sC	5	73	10	12	1.2
26	-	55.6	18.8	25.6	mS	5	73	10	12	1.2
27	-	71.1	17.4	11.5	mS	6	70	10	14	1.4
28	-	71.1	15.0	13.8	mS	6	70	10	14	1.4
29	0.2	71.2	18.6	10.0	(g)mS	7	71	10	12	1.2
30	-	66.8	16.1	17.1	mS	6	71	11	12	1.1
31	-	67.4	15.1	17.5	mS	6	71	11	12	1.1
32	-	20.6	53.0	26.5	sZ	4	70	13	13	1.0
33	-	41.5	28.5	30.3	sM	5	73	10	12	1.2
34	-	32.1	21.6	46.4	sC	5	73	10	12	1.2
Range	0.1~0.3	0.5~91.8	3.4~64.2	4.8~64.3		37	69~74	8~13	11~14	0.9~1.7
Average	0.2	49.6	24.4	26.0		5	71	11	13	1.2

Note. (g)mS: slightly gravelly muddy sand, S: sand, mS: muddy sand, sM: sandy clay, zS: silty sand, Z: silt sC: sandy clay M: mud

part of the sea rather than around the Changjiang River estuary (Fig. 4 and 5). Such occurrence seems to be due to the transportation by Huanghai Sea Coastal Current from the southern Yellow Sea. Yang and Milliman (1983) also suggested that the sediments of Huanghe River origin are characterized by fine-grained size with abundant chlorite. The ratios of chlorite/kaolinite in our study area range from 1.2 to 2.1. However, the sediments of Changjiang

River origin characterized by abundant kaolinite show the ratio of chlorite/kaolinite at about 0.7, one-half of Huanghe River sediment.

Figure 6 shows the kaolinite distribution in the northwestern East China Sea, varying from 9 to 13%. The high concentration areas are near the Changjiang River estuary, southern central offshore region and near the west coast off the Cheju Island. More-than 12% concentration of kaolinite occurs

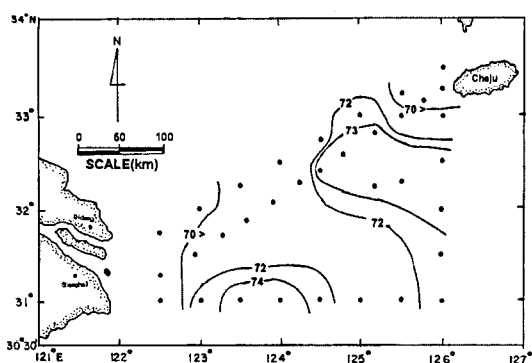


Fig. 3. Distribution pattern of illite in surface sediments of the East China Sea.

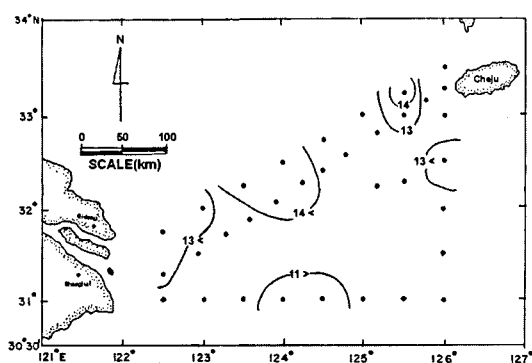


Fig. 4. Distribution pattern of chlorite in surface sediments of the East China Sea.

around the Changjiang River estuary and decreases gradually northeastward, resulting from the supplies of fine-grained kaolinite-rich particles from the Changjiang River. Thus, the Changjiang River deposits have been known to be abundant in kaolinite (av. 14.3%), compared with the Huanghe River sediments (av. 8.4%; Xu, 1983). According to Yang and Milliman (1983), the drainage area of the Changjiang River is located in warm and humid climate zone, where chemical weathering is intensive, and mostly acidic soils are enriched in Al content and kaolinite. The high concentration of kaolinite on the southern central offshore region may be supplied from the South China Sea by Taiwan Warm Current (Aoki and Oinuma, 1988). According to Beardsley et al. (1983), during the June to September, Taiwan Warm Current flows northeastward along the eastern coast of China following the pre-

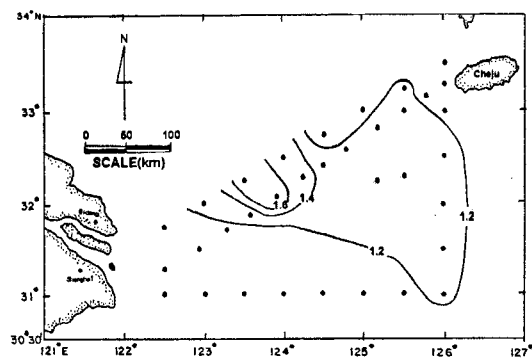


Fig. 5. Distribution pattern of the chlorite/kaolinite ratios in the East China Sea.

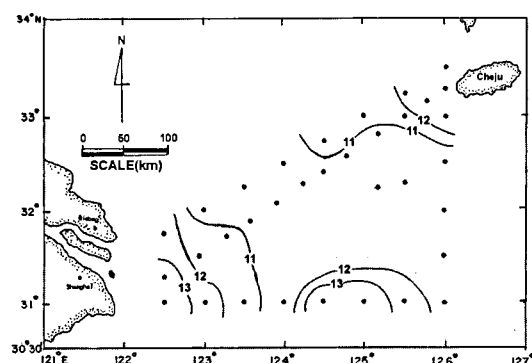


Fig. 6. Distribution pattern of kaolinite in surface sediments of the East China Sea.

dominant southerly wind direction, thus the suspended sediments are dispersed eastward or northeastward from the South China Sea. The high concentration of kaolinite in the west coast off Cheju Island seems to be the transportation from the southern Yellow Sea and the South Sea of Korea. The nearshore of the South Sea of Korea has a characteristic mineralogy of more than 20% of kaolinite content in a wide range influenced by lithofacies from the adjacent land (Park and Han, 1985).

Smectite contents range from 3 to 7% in the study area (Table 2). The low contents of smectite was also reported by Aoki et al. (1983), being less than 9% in the sediments of the East China Sea. The highest concentration of smectite (>6%) is found in the northwest middle part of the sea, the west coast off Cheju Island and the outer-shelf mud

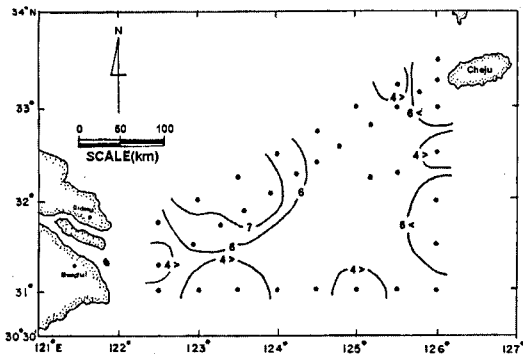


Fig. 7. Distribution pattern of smectite in surface sediments of the East China Sea.

deposit (Fig. 7). The concentration of smectite decreases progressively southeastward from the southern Yellow Sea, suggesting the influence of the high input of smectite via river runoff from the Chinese mainland that received the modern and ancient flow of the Huanghe River. This interpretation is also supported by the clay mineral comparison between the Huanghe and Changjiang River sediments. The Huanghe River sediment is characterized by the highest average concentration of smectite (23.2%) with the lowest kaolinite (8.4%; Xu, 1983), whereas the Changjiang River sediment are distinguished by high amount of kaolinite (14.3%) and minor amounts of smectite (5.9%; Xu, 1983).

Zhao et al. (1990) reported that suspended sediments from the Huanghe River are dispersed southward, passing around the Shandong Peninsula, and being deposited in the middle of the Yellow Sea. The remaining suspended sediments continue to be transported farther southward by Jiangsu Coastal Current and finally deposited in the outer shelf of the East China Sea (Fig. 8). The content of smectite in the outer-shelf mud deposits to the southwest of the Cheju Island is higher than those in the adjacent areas (st. 4 and 5; Fig. 7). This clearly indicates that the resuspended terrigenous materials have been delivered from the Yellow Sea by Jiangsu Coastal Current into this area. Milliman et al. (1985) also suggested that one potential mechanism for southward transport of Huanghe River-derived

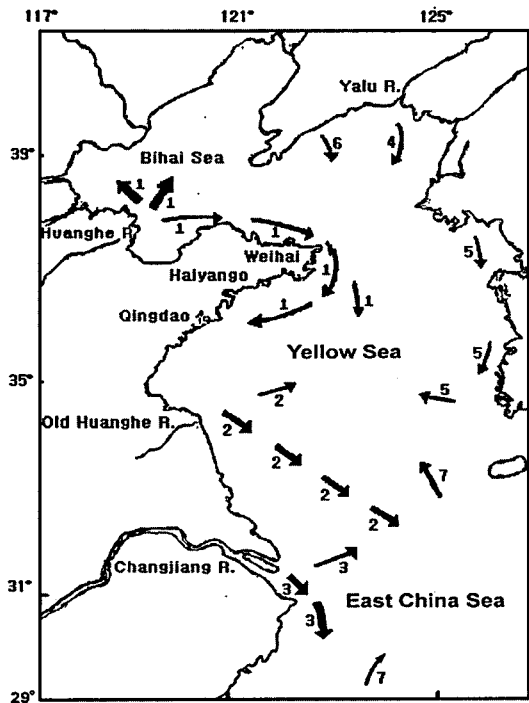


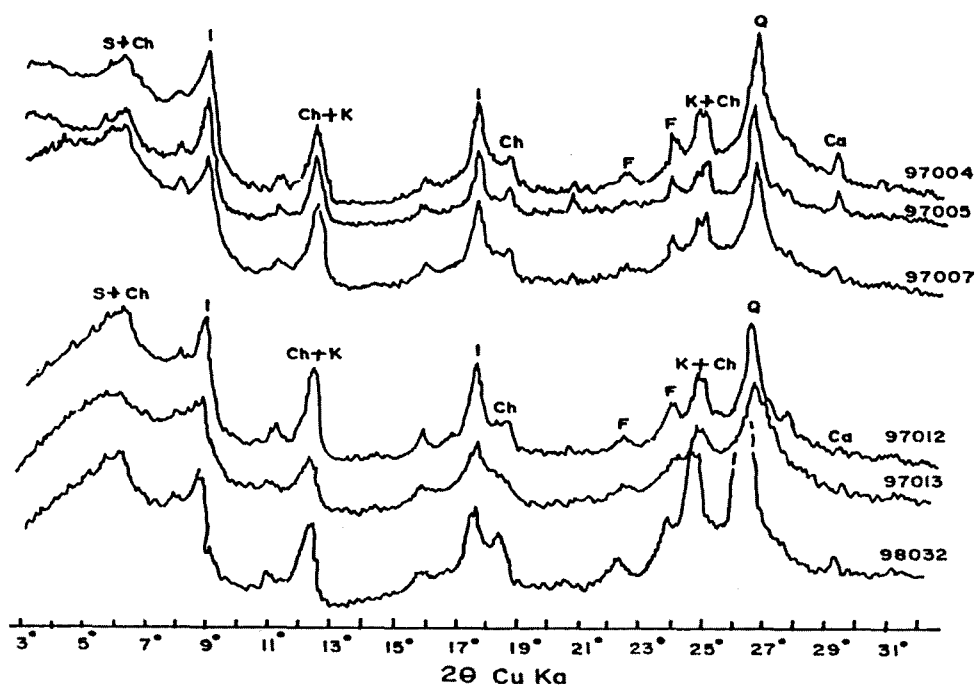
Fig. 8. The transportation trend of the Yellow Sea clays or sediments (Zhao et al., 1990). 1. Materials from the Huanghe River; 2. Materials from the old Huanghe River in north Jiangsu; 3. Materials from the Changjiang River; 4. Materials from the Yalu River; 5. Materials from the east coast of the Yellow Sea; 6. Materials from the north coast of the Yellow Sea; 7. Materials from the open sea.

mud from the western Yellow Sea is seasonal resuspension during winter storms and advection toward the southeast outer-shelf mud deposits by the dominant circulation patterns, such as Jiangsu Coastal Current (Fig. 8). The mud deposits region to the southwest of Cheju Island is a place where northward flowing Taiwan Warm Current and southward flowing Jiangsu Coastal Currents meet, forming a cyclonic gyre that may have deposited muddy sediment at the center (Beardsley et al., 1985). The higher concentration of smectite (>5%) in the west coast of the Cheju Island may be attributed to the relatively large supply of smectite from the volcanics of the Cheju Island.

The relative abundance of clay minerals in the study area and adjacent regions are summarized in Table 3, which may help to understand and distin-

**Table 3.** Relative clay mineral abundances of sediment in the East China Sea, and adjacent sea and rivers (unit: %)

Region	Smectite	Kaolinite	Chlorite	Illite	Chlorite/Kaolinite	Reference
Inner-shelf mud deposit	4	13	12	71	1.0	Present study
Outer-shelf mud deposit	6	10	12	72	1.2	
Huanghe River	23.2	8.4	9.2	59.0	1.1	Xu (1983)
Ancient Huanghe River	24.0	8.9	8.1	59.0	0.9	Xu (1983)
Changjiang River	5.9	14.3	13.1	68.4	0.9	Xu (1983)
East China Sea	3.0	7.0	28.0	62.0	4.0	Aoki et al. (1983)
Central Yellow Sea	13.0	10.0	12.0	67.0	1.2	Khim (1988)
Keum River	0.1	17.0	19.3	63.7	1.1	Choi (1981)
Yeongsan River	0.1	19.2	16.8	63.9	0.9	Kim (1980)

**Fig. 9.** X-ray diffraction record of surface sediment on the East China Sea continental shelf. Note: S = smectite, Ch = chlorite, I = illite, K = kaolinite, S + Ch = smectite + chlorite, K + Ch = kaolinite + chlorite, F = feldspar, Q = quartz, Ca = calcite.

guish the sources of the sediment in the study area. The ancient and modern Huanghe River in China supplies relatively high concentration of smectite, with less amounts of kaolinite and chlorite, while the Changjiang River clay suites are distinguished by high concentration of kaolinite and minor amount of smectite. In case of the Keum and Yeongsan Rivers in Korea, smectite is nearly absent, but kaolinite and chlorite are dominant (Table 3). It implies that smectite in the East China Sea might not be provided from the Korean Peninsula. Instead, it seems to be mainly controlled by the ancient and

modern Huanghe River system.

The clay minerals of the outer-shelf mud deposit are characterized by abundant smectite (6%) and lack of kaolinite (10%), contrasted to those of the inner-shelf mud deposit (smectite: 4%, kaolinite: 13%, and a ratio of chlorite/kaolinite of 1.2). These values are similar to that of the Huanghe River and the central Yellow Sea sediments (Table 3). Yang and Milliman (1983) suggested that the sediments of Huanghe River origin are characterized by abundant smectite and chlorite, and a existence of calcite concretion. In addition, the ratio of chlorite/kaolinite

ranges from 1.2 to 2.1. However, the sediment of Changjiang origin is characterized by abundant kaolinite and absence of calcite concretion, and a ratio of chlorite/kaolinite by 0.7.

Figure 9 showed X-ray diffraction pattern of untreated clay fraction from the inner-shelf mud near the mouth of the Changjiang River and the outer-shelf mud deposit to the southwest of the Cheju Island. The X-ray diffractogram of the outer-shelf mud deposit shows relatively sharp and strong peaks as well as quite apparent calcite at 3.04Å of sample 97004 and 97005. In contrast, the X-ray diffraction of the inner-shelf mud shows relatively low and wide peaks, and no distinct calcite peaks at the station 97012 and 97013 (Fig. 9). The average content of kaolinite (12.5%) in the inner-shelf mud deposit is a little higher than the outer-shelf mud deposit (10.4%), and which is similar to the sediment in the Changjiang River (Table 3). Therefore, we suggest that the clay minerals in the northwestern continental shelf of East China Sea are the mixtures of clays derived predominantly from the modern and ancient Huanghe River, the Changjiang River, and to a small extent from the South China Sea.

## Conclusions

The sediments in the inner-shelf area of the East China Sea are characterized by abundant silt and high amount of kaolinite, and absence of calcite. However, the outer-shelf mud deposit is characterized by rich clay with higher contents of water and organic carbon, and an existence of calcite concretion. The relative content of illite ranges from 69 to 74% of the total clay minerals, and it seems to be transported by Kuroshio Current. Smectite is highly concentrated in the northwest central area of the sea and on the west coast of the Cheju Island. It seems to be mainly controlled by the Huanghe River system, and the result of supplies of smectite altered volcanic materials from Cheju Island. More than 12% of high concentration of kaolinite occurs in the

Changjiang River estuary and off the west coast of the Cheju Island. It can be supplied by the Changjiang River and influenced by lithofacies on adjacent land of the South Sea of Korea. The highest concentration of chlorite (>14%) is found in the northwest central part of the sea. It seems to be carried by Huanghai Sea Coastal Current from the Yellow Sea. The clay minerals in the northwestern East China Sea are characterized by relatively higher smectite and a high ratio of chlorite/kaolinite, which is similar to the sediment of the central Yellow Sea.

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