Chinese Science Bulletin

Clay minerals in surface sediments of the Pearl River drainage basin and their contribution to the South China Sea

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Clay minerals have played a significant role in the study of the East Asian monsoon evolution in the South China Sea by being able to track oceanic current variations and to reveal contemporaneous paleoclimatic changes prevailing in continental source areas. As one of the most important rivers inputting terrigenous matters to the northern South China Sea, the Pearl River was not previously paid attention to from the viewpoint of clay mineralogy. This paper presents a detailed study on clay minerals in surface sediments collected from the Pearl River drainage basin (including all three main channels, various branches, and the Lingdingyang in the estuary) by using the X-ray diffraction (XRD) method. The results indicate that the clay mineral assemblage consists dominantly of kaolinite (35%—65%), lesser abundance of chlorite (20%—35%) and illite (12%—42%), and very scare smectite occurrences (generally <5%). Their respective distribution does not present any obvious difference throughout the Pearl River drainage basin. However, downstream the Pearl River to the northern South China Sea, the clay mineral assemblage varies significantly: kaolinite decreases gradually, smectite and illite increase gradually. Additionally, illite chemistry index steps down and illite crystallinity steps up. These variations indicate the contribution of major kaolinite, lesser illite and chlorite, and very scarce smectite to the northern South China Sea from the Pearl River drainage basin. The maximum contribution of clay minerals from the Pearl River is 72% to the northern margin and only 15% to the northern slope of the South China Sea. In both glacials and interglacials, kaolinite indicates that the ability of mechanical erosion occurred in the Pearl River drainage basin.

clay minerals, surface sediments, provenance, East Asian monsoon, Pearl River drainage basin

The East Asian monsoon is a significant component of the global climate system[1]. Following the successful study on deep-sea sedimentary records of the South Asian monsoon evolution in the Arabian Sea[2], the South China Sea is becoming an international oceanic location for the study on the East Asian monsoon evolution[3]. Specially, the Ocean Drilling Program (ODP) Leg 184[4], which was carried out successfully in the South China Sea in 1999, has impelled the study on the East Asian monsoon evolution not only in micropaleontology and oxygen isotope stratigraphy in the past[5], but also in sedimentology and sedimentary geochemistry[6,7].

Received September 27, 2006; accepted January 23, 2007
doi: 10.1007/s11434-007-0161-9
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Supported by the National Natural Science Foundation of China (Grant Nos. 40331002, 40621063 and 40506014), the Fok Ying Tung Education Foundation (Grant No. 101018), the Doctoral Program of Higher Education (Grant No. 20060247032), the Excellent Young Teachers Program and the Program for New Century Excellent Talents in University of the Ministry of Education of China (Grant No. NCET-04-0372), and the Laboratory of Marginal Sea Geology Grant of the Chinese Academy of Sciences (Grant No. BYH03A08)

Among them, clay minerals can be used to track oceanic current variations and to reveal contemporaneous paleoclimatic changes prevailing in continental source areas[8,9], and therefore have played a significant role in the study of the East Asian monsoon evolution in the South China Sea[7,10−13].

The significance of clay mineral assemblages in the study of the East Asian monsoon evolution requires a detailed knowledge of distribution of clay minerals and investigation of their provenance. Hence, clay mineralogical characteristics of modern sediments in major rivers surrounding the South China Sea and surface sediments on the sea bottom are the keys to study variations in the clay mineral assemblages occurring in the geological past. Provenances of terrigenous matter in the South China Sea are complicated because they involve two markedly different geological regions: the Asian continent and Taiwan in the north and the volcanic arcs in the south and east. A few clay mineral provinces in the South China Sea divided by the surface distribution suggest that illite and chlorite derive mainly from the Asian continent and Taiwan to the north, whereas smectite and kaolinite come mainly from the Sunda shelf and the Indonesian arcs to the south[14]. The less investigated clay mineral assemblage in major rivers surrounding the South China Sea, however, has resulted in important contradictions in the implication of clay minerals in the study on the East Asian monsoon evolution. For example, on basis of the study on ODP Site 1144 in the northern South China Sea, Tamburini et al.[15] and Boulay et al.[16] considered that clay minerals at this site derived mainly from the Pearl River, and therefore deduced that the Pearl River provided most of the terrigenous load to this site. Wehausen and Brumsack[6] also obtained a similar result on basis of an elemental geochemical study at nearby ODP Site 1145. However, in view of high kaolinite contents (>50%) in sediments of the Lingdingyan in the Pearl River estuary[17], Liu et al.[7,12] concluded that clay minerals at ODP Site 1146, very close to Sites 1144 and 1145, derive mainly from Taiwan and the Yangtze River. This conclusion was confirmed by a geochemical tracer study on sediments in the northern South China Sea[18]. The Pearl River transports 69×10^6 tons of suspended sediments annually, accounting for about 14% of total terrigenous discharge to the South China Sea[19−21]. Such a discharge must have played a significant role in sediment components in the South China Sea, especially on the northern slope. But clay mineral investigations in surface sediments were previously carried out only for a few locations in the Pearl River drainage basin[22,23], and the clay mineral assemblages in the whole drainage basin are still poorly known. This study is an investigation of clay mineralogy for 38 surface argillaceous sediments collected along the Pearl River drainage basin in March 2004 and 12 bottom surface sediments collected in the Lingdingyang of the estuary and in the northern margin of the South China Sea in July 1999 and July 2000, respectively. We present the results to reveal clay mineral assemblage characters and to evaluate sources of clay minerals in the northern South China Sea.

1 Materials and methods

Surface sediments, in the Pearl River drainage basin, were collected along three main channels: the East River, the North River, and the West River (Figure 1, Table 1). Knowing that the river water level falls greatly from December to March, and that muddy channel deposits are a close proxy for the components of suspended particles in river water, we sampled argillaceous sediments directly from shallow water river beds or from river beds with water depth < 20 cm. Surface samples of the Lingdingyang in the Pearl River estuary and the northern South China Sea were obtained from the Pearl River Estuary Pollution Research Cruise (PREPP, 1999—2000) operated by the Hong Kong University of Science and Technology and the Second Institute of Oceanography of State Oceanic Administration. The sampling is representative of the whole Pearl River estuary (Figure 1, Table 1). In addition, Holocene clay mineral data at ODP Site 1144 (20°3.18'N, 117°25.14'E, water depth 2037 m)[16], Site 1145 (19°35.04'N, 117°37.86'E, water depth 3157 m)[23], and Site 1146 (19°27.40'N, 116°16.37'E, water depth 2092 m)[12] are included in the discussion of the provenance of clay minerals deposited on the northern slope of the South China Sea.

Clay minerals were identified by X-ray diffraction (XRD) on oriented mounts of clay-sized particles (< 2 μm)[24]. Firstly, all samples were reacted with 0.5% HCl to remove carbonate. Secondly, the samples were washed successively with distilled water. Thirdly, the decarbonated suspensions were reacted with 10% H_2O_2 to remove organic matter. Fourthly, the samples were washed again successively with distilled water to re-

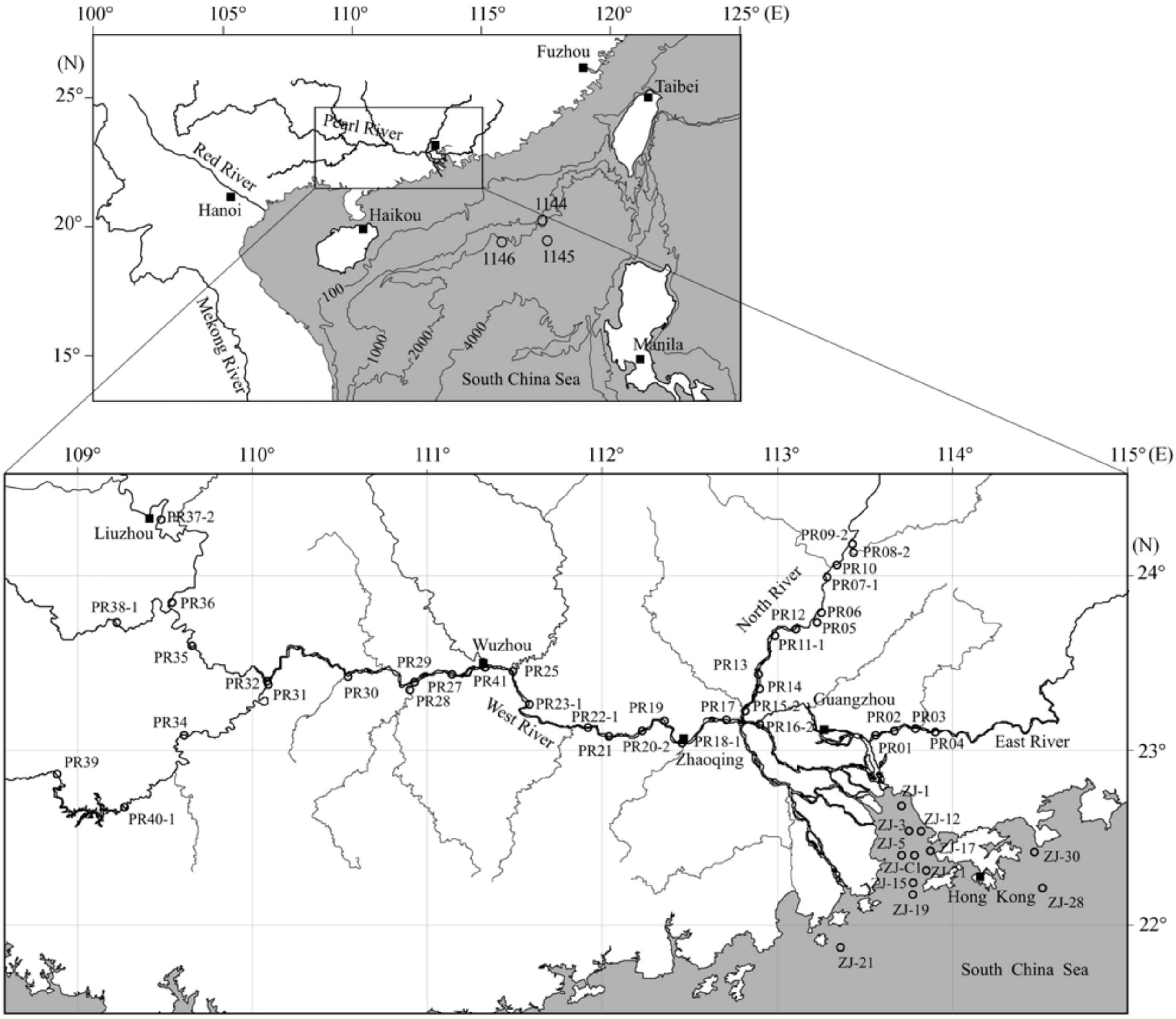

Figure 1 Locations of surface sediment samples in the Pearl River drainage basin. See Table 1 for detailed geographic positions. Locations of ODP Sites 1144, 1145, and 1146 on the northern slope of the South China Sea are also displayed.

move excess ions and to help the deflocculation of clays. The particles less than 2 μm were separated following the Stoke's law[24] and were concentrated using a centrifuge. The resulting paste spread into calibrated recess onto glass slides. The analysis was conducted using a Philips PW 1710 diffractometer with CuKα radiation and Ni filter, under a voltage of 40 kV and an intensity of 25 mA. Three XRD runs were performed, following air-drying, ethylene-glycol solvation for 12 h, and heating at 490℃ for 2 h. A few randomly selected samples were investigated under an electronic microscope for clay crystal shapes. The clay particles were found to be mainly continental clastics and their post deposition diagenesis is considered negligible. Previous studies on clay minerals from ODP Sites 1144, 1145, and 1146 collected from the northern slope of the South China Sea reached the same conclusion[12,16,23]. Following a similar procedure as for the three ODP sites, preparation and analyses of the samples were performed at the Laboratoire de Sédimentologie et Géodynamique, Université des Sciences et Technologies de Lille, France.

Identification and interpretation of clay minerals were made according to a comprehensive comparison of three multiple X-ray diffratograms obtained under different measurement conditions (Figure 2)[7,12]. Semi-quantitative calculations of each peak's parameters were carried out on the glycolated curve by using MacDiff software[25]. The relative contents of each clay mineral species were estimated mainly according to area of (001) series of basal reflections, i.e. smectite (001) including

Table 1 Geographic locations and clay mineral assemblages of surface sediments in the Pearl River drainage basin

No.	Sample	Location	Latitude (N)	Longitude (E)	Chlorite (%)	Illite (%)	Smectite (%)	Kaolinite (%)	Illite chemistry index	Illite crystallinity (°Δ2θ)
1	PR01	Nangang	23°05′10.7″	113°32′59.4″	26	20	1	53	0.54	0.34
2	PR02	Xintang	23°06′52.4″	113°39′30.0″	32	13	1	54	0.77	0.33
3	PR03	Tangxia	23°07′39.7″	113°46′57.0″	35	16	0	49	0.67	0.26
4	PR04	Shipai	23°06′35.8″	113°53′45.5″	28	12	1	58	0.79	0.27
5	PR05	Jiangkou	23°43′58.2″	113°12′59.5″	27	6	1	67	0.80	0.24
6	PR06	Shengping	23°47′19.1″	113°14′30.4″	17	31	2	51	0.48	0.33
7	PR07-1	Lianjiangkou	24°03′27.6″	113°20′02.1″	23	33	2	42	0.51	0.31
8	PR08-2	Yingde	24°07′52.9″	113°25′33.8″	21	30	3	47	0.55	0.25
9	PR09-2	Yingdeqiao	24°10′49.0″	113°25′20.4″	15	33	1	51	0.60	0.27
10	PR10	Lixi	23°59′31.4″	113°16′32.7″	24	33	0	43	0.65	0.27
11	PR11-1	Zhouxin	23°41′44.2″	113°05′57.9″	20	29	2	49	0.63	0.27
12	PR12	Qingyuan	23°39′33.2″	112°58′29.4″	25	41	2	32	0.46	0.31
13	PR13	Datang	23°26′16.8″	112°53′07.0″	26	25	3	47	0.62	0.34
14	PR14	Lubao	23°21′03.8″	112°53′21.2″	25	31	4	40	0.45	0.24
15	PR15-2	Sanshuinan	23°09′04.3″	112°53′27.7″	23	26	3	48	0.58	0.32
16	PR16-2	Sanshuibei	23°13′40.5″	112°48′30.0″	27	12	0	61	0.65	0.32
17	PR17	Yong’an	23°10′33.4″	112°41′51.5″	22	29	4	45	0.72	0.26
18	PR18-1	Zhaoqing	23°02′41.9″	112°26′42.1″	43	24	2	32	0.70	0.28
19	PR19	Sunwei	23°10′20.3″	112°20′46.7″	32	17	2	49	0.69	0.28
20	PR20-2	Yuecheng	23°06′58.3″	112°13′16.9″	34	17	1	48	0.65	0.38
21	PR21	Jiushi	23°04′48.0″	112°01′41.5″	36	19	1	44	0.65	0.26
22	PR22-1	Deqing	23°07′51.4″	111°54′29.6″	28	19	2	51	0.59	0.35
23	PR23-1	Changgang	23°15′54.5″	111°34′39.2″	37	28	4	30	0.69	0.30
24	PR25	Fengkai	23°27′20.9″	111°28′46.8″	25	27	1	46	0.57	0.34
25	PR27	Chishui	23°25′59.6″	111°08′05.1″	36	17	3	44	0.61	0.25
26	PR28	Tenxian	23°23′28.9″	110°54′58.5″	21	36	3	40	0.51	0.26
27	PR29	Tenxiannan	23°20′48.9″	110°53′36.4″	18	13	2	67	0.52	0.42
28	PR30	Wulin	23°25′24.1″	110°32′11.3″	13	34	5	48	0.68	0.38
29	PR31	Shizui	23°27′44.9″	110°09′50.6″	32	33	4	31	0.70	0.34
30	PR32	Guiping	23°22′35.7″	110°04′59.3″	24	33	2	41	0.63	0.29
31	PR34	Guigang	23°05′15.2″	109°35′57.0″	17	40	5	37	0.55	0.26
32	PR35	Wuxuan	23°36′04.4″	109°38′42.2″	24	34	3	39	0.61	0.25
33	PR36	Jinji	23°50′46.1″	109°31′56.8″	15	42	4	39	0.50	0.25
34	PR37-2	Liuzhou	24°18′59.9″	109°28′06.3″	24	18	7	51	0.64	0.31
35	PR38-1	Leibin	23°43′59.2″	109°13′07.4″	23	35	11	31	0.45	0.31
36	PR39	Liujing	22°52′03.1″	108°52′41.9″	21	34	3	42	0.65	0.33
37	PR40-1	Hengxian	22°40′51.0″	109°15′36.1″	23	25	3	50	0.56	0.27
38	PR41	Wuzhou	23°29′43.8″	111°19′0.9″	22	41	2	35	0.77	0.36
39	ZJ-1	Lingdingyang	22°40′37.2″	113°41′34.8″	29	26	17	28	0.44	0.16
40	ZJ-3	Lingdingyang	22°32′16.8″	113°44′16.8″	30	28	3	39	0.49	0.18
41	ZJ-5	Lingdingyang	22°24′57.6″	113°41′56.4″	29	29	1	40	0.54	0.20
42	ZJ-11	Lingdingyang	22°26′16.8″	113°53′38.4″	28	21	14	37	0.47	0.18
43	ZJ-12	Lingdingyang	22°32′16.8″	113°48′0.0″	28	23	3	46	0.52	0.20
44	ZJ-15	Lingdingyang	22°16′04.8″	113°46′04.8″	30	26	3	41	0.56	0.19
45	ZJ-17	Lingdingyang	22°19′58.8″	113°51′0.0″	29	23	5	43	0.53	0.20
46	ZJ-19	Lingdingyang	22°12′03.6″	113°46′01.2″	27	26	4	43	0.52	0.20
47	ZJ-C1	Lingdingyang	22°25′01.2″	113°46′04.8″	23	28	6	43	0.48	0.20
48	ZJ-21	South China Sea	21°52′3.0″	113°20′02.4″	25	27	8	40	0.51	0.19
49	ZJ-28	South China Sea	22°12′0.0″	114°30′03.6″	26	26	19	29	0.43	0.16
50	ZJ-30	South China Sea	22°26′02.4″	113°28′01.2″	23	24	22	31	0.43	0.16

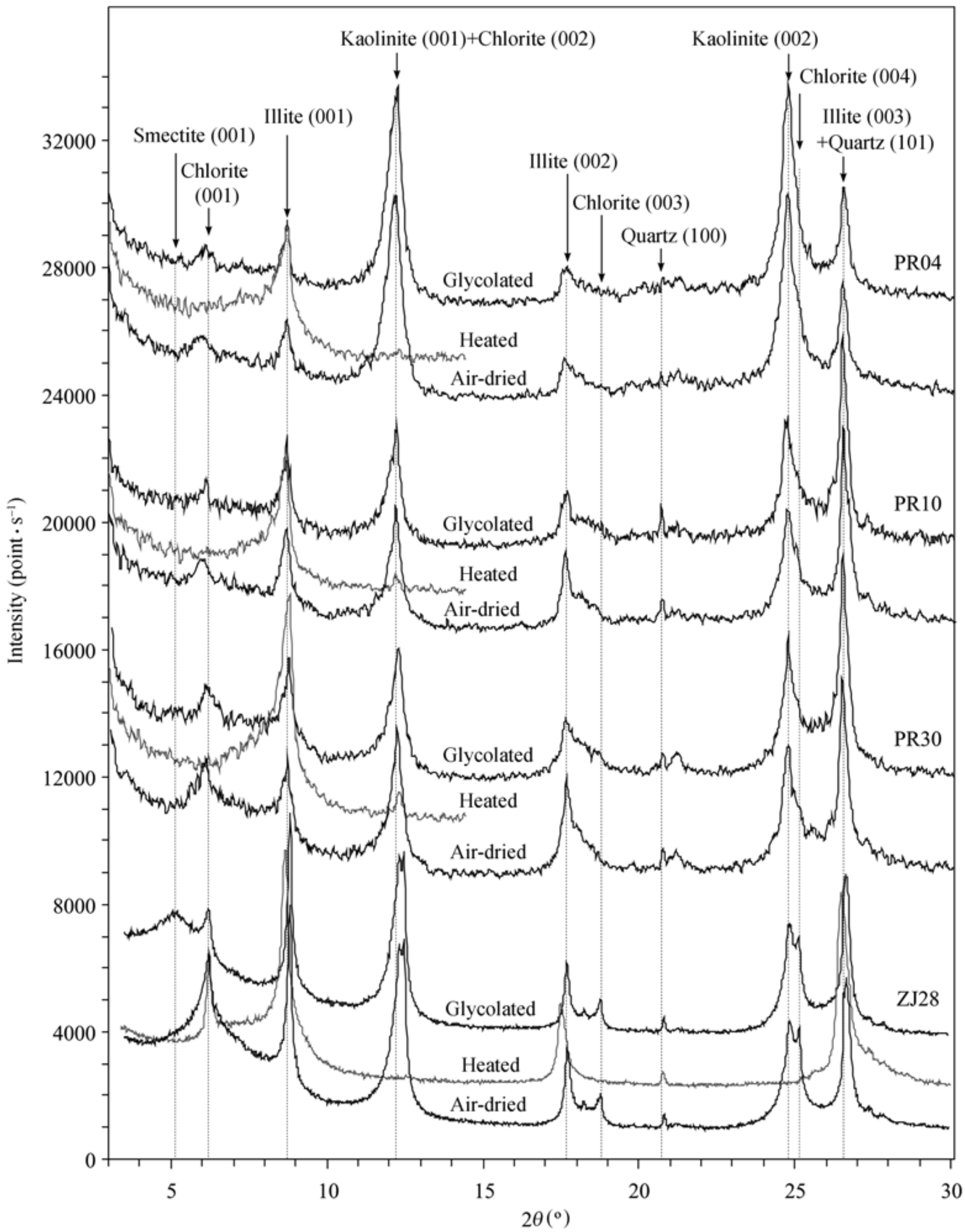

Figure 2 Multiple X-ray diffratograms of typical samples from the Pearl River drainage basin. Samples PR04 from the East River, PR10 from the North River, and PR30 from the West River contain very scarce smectite. Sample ZJ-28 from the northern continental margin of the South China Sea contains relatively more smectite. See Figure 1 and Table 1 for their detailed locations and clay mineral assemblages.

illite/smectite random mixed-layers at 1.7 nm, illite (001) at 1 nm, and kaolinite (001) and chlorite (002) at 0.7 nm[24]. Kaolinite and chlorite were dicriminated according to the relative proportions given by a ratio of the 0.357 nm and 0.354 nm peak areas. Peaks of vermiculite (001) on both glycolated and air-dried conditions are at 1.44 nm. Low content in vermiculite in our samples prevented any semi-quantitative calculation. Additionally, some mineralogical characters of illite were determined on the glycolated curve. Illite chemistry index refers to a ratio of the 0.5 and 1 nm peak areas. Ratios below 0.5 represent Fe-Mg-rich illites (biotites, micas), which are characteristic of physical erosion; ratios above 0.5 are found in Al-rich illites (muscovites), which are released following strong hydrolysis[8,26,27]. Illite crystallinity was obtained from half height width of the 1 nm peak. Lower values represent the higher crystallinity, characteristic of weak hydrolysis in continental sources and arid and cold climate conditions[28−30]. This index is also used as a potential tracer of source regions and transport paths[31].

2 Results

For the Pearl River sediments including all the three main channels: the East River, the North River, and the West River, the clay mineral assemblages consist mainly of kaolinite, chlorite, and illite (Table 1, Nos. 1—38). Kaolinite (35%—65%) is the most dominant clay mineral with an average of 46%. Chlorite (20%—35%) and illite (12%—42%) are in lesser abundance with average contents of 25% and 26%, respectively. Smectite is very scarce (<5%) and is <2% for most of the samples with two exceptions (~10%) in the western West River (Nos. 37 and 38). Illite chemistry index of all Pearl River sediments is usually comprised between 0.45 and 0.80 with most of values >0.50, indicating a strong hydrolysis. Illite crystallinity varies between 0.24° and 0.42° Δ2θ.

For the Lingdingyang sediments in the estuary, similar to the Pearl River sediments, the clay mineral assemblages consist mainly of kaolinite, chlorite, and illite (Table 1, Nos. 39—47). Their average contents are 40%, 28% and 26%, respectively. Smectite slightly increases with an average of 6%. Only few samples can reach 17%. The illite chemistry index varies between 0.44 and 0.56, and the illite crystallinity is between 0.16° and 0.20° Δ2θ.

The samples from the northern margin of the South China Sea (Table 1, Nos. 48—50) were compared to the clay mineral assemblage of the Pearl River and Lingdingyang sediments. In these samples, kaolinite decreases markedly with an average of 33%; smectite clearly increases with an average of 16%, with some assemblages reaching about 22%. On the other hand, chlorite and illite show similar contents and their average abundances are 25% and 26%, respectively. The illite chemistry index varies between 0.43 and 0.51, and the illite crystallinity is between 0.16° and 0.19° Δ2θ.

3 Discussion

3.1 Distribution of clay minerals of surface sediments in the Pearl River drainage basin

The distribution of clay minerals of surface sediments through main channels and various branches of the Pearl River to the Lingdingyang in the estuary, is similar basin-wide and no obvious difference is observed (Table 2, Figure 3). Kaolinite is the most abundant clay mineral in the East River and North River with an average of 54%, whereas it is less abundant (generally 40%—44%) in the West River. Accordingly, smectite is very rare in the East River and North River and the average content is generally less than the semi-quantitative calculation error (2%). No significant differences exist for chlorite and illite contents in the whole Pearl River drainage basin (Figure 3).

However, through the Lingdingyang on the northern margin of the South China Sea, kaolinite decreases on average from 40% to 33%, and smectite increases from 6% to 16% (Figure 3). Through the northern margin to the three ODP sites on the northern slope of the South China Sea, kaolinite decreases continuously to 7% and, inversely, smectite increases to 35%. At the same time, illite also increases to about 40% (Table 2). The distri-

Table 2 Average clay mineral assemblages of various sections in the Pearl River drainage basin and the northern South China Sea[a)]

Region	Sample number	Chlorite (%)	Illite (%)	Smectite (%)	Kaolinite (%)	Illite chemistry index	Illite crystallinity (°Δ2θ)
East River	4	30	15	1	54	0.69	0.30
Northern North River	6	21	28	1	50	0.60	0.28
Southern North River	6	24	27	3	46	0.56	0.30
Eastern West River	7	33	22	2	43	0.67	0.30
Middle West River	8	24	29	3	44	0.63	0.33
Western West River	4	22	32	6	40	0.55	0.28
Southwestern West River	3	20	33	4	43	0.59	0.28
Lingdingyang	9	28	26	6	40	0.51	0.19
Northern margin	3	25	26	16	33	0.46	0.17
Holocene at ODP Site 1144	5	22	43	24	11		
Holocene at ODP Site 1145	13	21	31	41	7		
Holocene at ODP Site 1146	2	21	35	35	9	0.40	0.22

a) Clay mineral data of ODP Site 1144 are from ref. [16], those of Site 1145 from ref. [23], and those of Site 1146 from ref. [12].

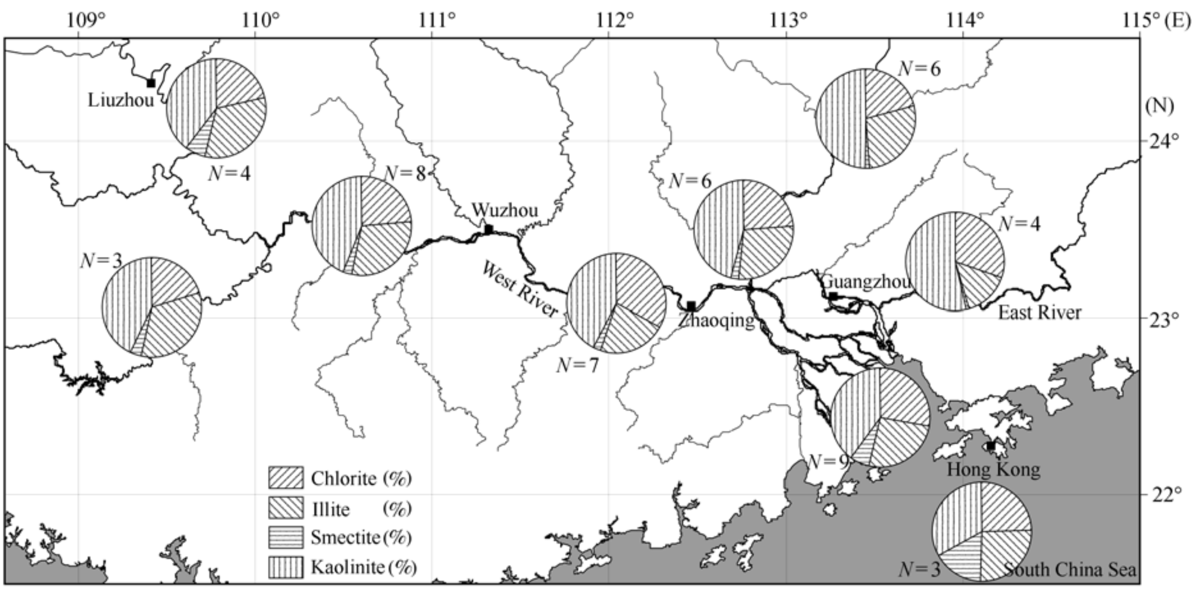

Figure 3 Distribution of average clay mineral assemblages of various sections in the Pearl River drainage basin and the northern South China Sea.

bution of clay minerals in the northern South China Sea presents a significant regional difference. Globally, clay mineral segregation exists in the marine environment[32]. For instance, kaolinite particles are relatively large and smectite particles, have usually a smaller diameter. This observation explains why kaolinite may decrease and smectite may increase gradually form river beds to ocean bottom[32]. However, for clay particles <2 μm in size, and because of the deflocculaton of clays, the physical segregation during sedimentation of clay minerals is not remarkable and cannot explain the 35% difference in kaolinite and smectite contents from the Pearl River to the northern South China Sea. Therefore, we believe that it is the provenance that controls variations in clay mineral assemblages from the Pearl River to the northern South China Sea.

3.2 Contribution of the Pearl River to clay minerals in the northern South China Sea

From previous sections, it can be seen that clay mineral assemblages of the Pearl River drainage basin are obviously different from the northern South China Sea ones (Figure 4). The Pearl River bed sediments are characterized by abundant kaolinite, while the northern slope sediments nearby the ODP sites in the South China Sea are smectite-rich, and the Lingdingyang ones in the estuary and the northern margin of the South China Sea show moderate kaolinite and smectite contents. These variations in clay mineral assemblages indicate that the Pearl River discharges abundant kaolinite and almost no smectite to the northern South China Sea.

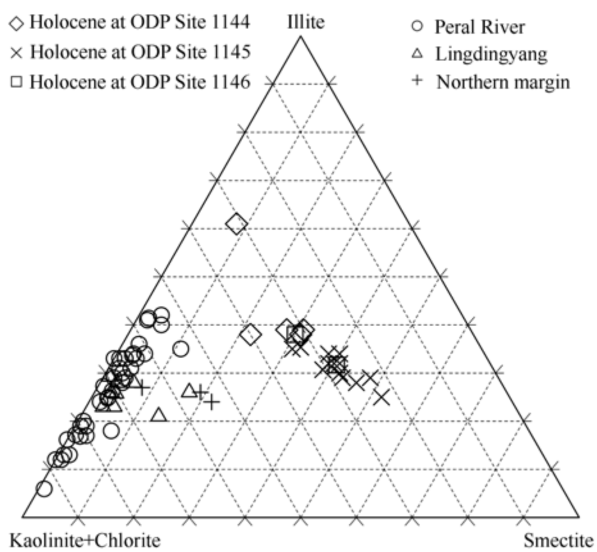

Figure 4 Comparison of clay mineral assemblages between surface sediments in the Pearl River drainage basin and Holocene sediments at ODP Sites 1144, 1145, and 1146 in the northern South China Sea. Clay mineral data of ODP Site 1144 are from ref. [16], those of Site 1145 from ref. [23], and those of Site 1146 from ref. [12].

The average illite content increases by about 10% from the Pearl River to the northern South China Sea (Table 2). At the same time, illite chemistry index decreases gradually from Al-rich illites derived from strong hydrolysis process to Fe-Mg-rich illites representing physical erosion process. In parallel, illite crystallinity also trends to increase from average 0.30° Δ2θ in the Pearl River drainage basin to average 0.20° Δ2θ in the northern South China Sea, indicating gradual drying and cooling climate conditions. A linear correlation of illite chemistry with illite crystallinity indexes exists for surface sediments in the Pearl River drainage basin (Figure 5). A higher illite chemistry index corresponds to a lower illite crystallinity (a higher value), indicating stronger hydrolysis, and vice versa, and further confirming the response of illite chemistry index and illite crystallinity in the hydrolysis and climate conditions during weathering. The inconsistent illite crystallinity pattern, which is also used as a potential tracer of source regions and transport paths[31], suggests that illites in the Pearl River drainage basin and in northern South China Sea have corresponding different provenances. The values of illite chemistry index and illite crystallinity in the Lingdingyang are mid-way in the Pearl River and the northern South China Sea spectrum (Figure 5), indicating transitional illite parameters. Therefore, providing illite contents and its mineralogical characters it is clear that the illite mineral found in the northern South China Sea, is not provided by the Pearl River drainage basin.

The chlorite content remains in the range of 20%—30% from the Pearl River through the northern South China Sea (Table 2) and no obvious regional difference is observed. Therefore, the source of chlorite in the northern South China Sea cannot be determined on basis of the clay mineral assemblage itself. However, previous studies on ODP Sites 1144, 1145, and 1146 in the northern South China Sea revealed similar patterns of variations in illite and chlorite contents, both representing similar climate conditions with strong physical weathering[12,16,23]. Moreover, the study on clay mineral assemblages at ODP Site 1146 suggested that both illite and chlorite were not provided by the Pearl River, but by Taiwan and the Yangtze River[7,12]. Hence, we deduce that similar to illite, the chlorite in the northern South China Sea is, as well, not provided by the Pearl River drainage basin.

In comparison with major contributions of illite and chlorite to the South China Sea from Taiwan and the Yangtze River[33,34], the Pearl River mainly provides kaolinite. Such a result is in agreement with previous studies on surface sediments in the Lingdingyang and in the lower reach of the Pearl River[17,23]. The kaolinite content decreases gradually from the Pearl River, the Lingdingyang, the northern margin, to the northern slope of the South China Sea, with average contents of 46%, 40%, 33%, and 7%, respectively. The variations indicate the gradually decreased contribution of clay minerals to the South China Sea from the estuary to the slope (Figure 5). Assuming that kaolinite in the northern South China Sea is provided completely from the Pearl River drainage basin, the maximum contribution of clay minerals from the Pearl River is 72% to the northern margin and only 15% to the northern slope of the South China

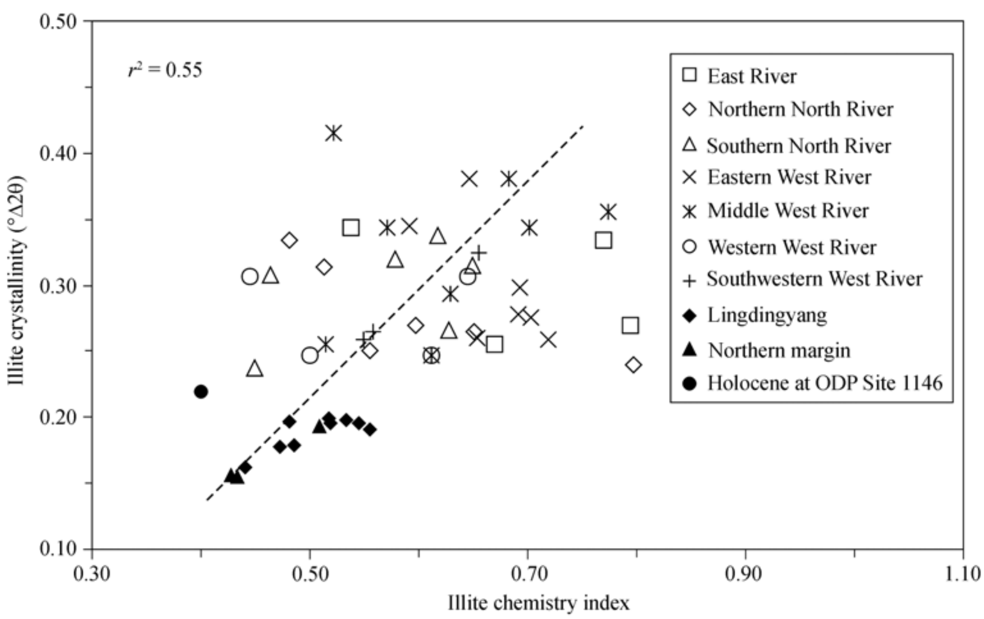

Figure 5 Correlations of illite chemistry index with illite crystallinity of surface sediments in various sections of the Pearl River drainage basin and in northern South China Sea. See Table 1 for illite mineralogical data of the Pearl River drainage basin and Table 2 for illite mineralogical data of ODP Site 1146.

Sea (Figure 5). As such, the maximum contribution of both illite and chlorite from the Pearl River to the northern slope of the South China Sea would be only 4% (estimated on basis of 15% contribution rate), whereas most of illite and chlorite in the northern slope of the South China Sea would come mainly from Taiwan and the Yangtze River[7,12]. We do not interpret this hypothesis as the result of selective clay minerals input to the South China Sea from the Pearl River. Instead, we believe that the Pearl River is not the major provenance of clay-size minerals to the northern slope of the South China Sea as far as the nearby locations of the ODP sites are concerned.

Clay minerals at the surface of the Earth originated mainly from weathering of parent rocks, including physical weathering leading to rock fragmentation and chemical weathering with subtraction of ions[28]. Hydrolysis is by far the most efficient chemical weathering, which consists in the attack of rocks by low ionized water at medium pH conditions. Following an increase of hydrolysis intensity during chemical weathering, subtraction of ions from minerals in parent rocks statistically concerns first the more mobile ions, like Na, K, Ca, Mg, and Sr. This hydrolytic weathering process has been called bisialitization with the formation of 2:1 layer clays (the assemblage of two tetrahedral sheets with one octahedral sheet, e.g., smectite). Transitional elements tend to be expelled later (Mn, Ni, Cu, Co, Fe). This process corresponds to monosialitization with the formation of 1:1 layer clays (the assemblage of one tetrahedral sheet with one octahedral sheet, e.g., kaolinite). Finally, Si tends to be leached completely when compared to Al, which is the less mobile element through the hydrolytic process. The final process of hydrolytic weathering is alitization with the formation of Al hydroxides (e.g., gibbsite)[35]. Warm and humid climate conditions, as well as tectonic stable and low relief morphology, which prevail in the Pearl River drainage basin, have strengthened significantly the intensity of chemical weathering. The well-developed monosialitization and alitization have caused the formation of red-soil profiles in South China[36]. They are composed mainly of kaolinite and Fe-oxides. The bisialitization is generally not developed in South China. Therefore, clay minerals of surface sediments in the Pearl River drainage basin consist mainly of kaolinite with very scarce smectite (Figure 6). However, the long-term East Asian summer monsoon rainfall may directly scour surficial exposed rocks, and can produce primary minerals such as illite and chlorite, which constitute the lesser clay mineral components in surface sediments of the Pearl River basin.

Therefore, kaolinite in the Pearl River drainage basin has been produced by weathering of rocks during dif-

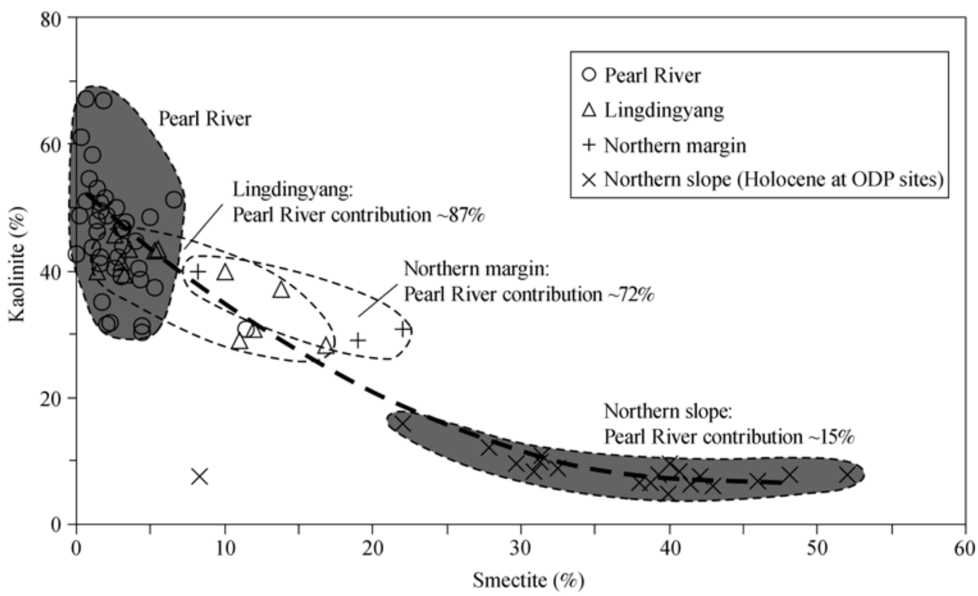

Figure 6 Correlations of kaolinite with smectite contents of surface sediments in the Pearl River drainage basin and the sediments in northern South China Sea. The coarse dash line shows linear gradual decrease of kaolinite content from the Pearl River, the Lingdingyang, and the northern margin to the northern slope of the South China Sea, and vice versa for smectite content. Clay mineral data of Holocene sediments at ODP Sites 1144, 1145, and 1146 on the northern slope of the South China Sea are from refs. [16, 23, 12], respectively. See Table 1 for clay mineral data of the Pearl River, the Lingdingyang, and the northern margin of the South China Sea.

ferent geological periods, and thus cannot reflect contemporaneous paleoclimatic characters. However, because the transport of kaolinite in the Pearl River drainage basin is mainly controlled by river erosion and, considering that transport ability is raised by the East Asian monsoon rainfall, therefore the kaolinite content is the result of the mechanical erosion ability in the Pearl River basin. In both glacial and interglacial periods, the contribution of clay minerals from the Pearl River to the northern South China Sea is mainly kaolinite. During glacial periods, the counter-clockwise surface currents forced by the East Asian winter monsoon, dominantly carried terrigenous matters from Taiwan, as well as from the Yangtze River via the Luzon Strait, to the northern slope of the South China Sea. This circulation pattern provided the illite- and chlorite-dominated clay mineral assemblage[7,12]. Although the monsoon rainfall was relatively weak during glacial periods, the coastline shifted approximately to the present 100-m isobathic position and the paleo-Pearl River estuary was very close to the northern slope of the South China Sea[37]. Consequently, the Pearl River discharged more kaolinite which resulted in consistent variations of kaolinite with illite and chlorite at three ODP sites in the northern South China Sea, and in reversed changes of kaolinite contents relative to smectite[12,16,23]. During interglacial periods, the coastline remained approximately similar to the present one and southwesterly surface currents in summer prevailed in most areas of the South China Sea. The currents might transport abundant smectite from islands or volcanic arcs in the south and east to the north, and make up major sources of smectite in the northern South China Sea. It is known that summer monsoon rainfall might greatly increase the ability of mechanical erosion in the Pearl River drainage basin during interglacial periods. Thus, this factor might increase the kaolinite discharge to the South China Sea from the Pearl River. The contribution of kaolinite material is relatively small when compared to smectite and illite contents provided by other sources. The reason is that the Pearl River estuary is far from the northern slope of the South China Sea[12,16,23], thus indicating that the clay mineral contribution to the northern slope of the South China Sea from the Pearl River is relatively limited even during interglacials when the summer monsoon rainfall is strong.

4 Conclusions

Our results indicate that clay mineral assemblages of surface sediments in the Pearl River drainage basin (including all three main channels, various branches, and the Lingdingyang in the estuary) consist dominantly of kaolinite (35%—65%), less abundant chlorite (20%—35%) and illite (12%—42%), and very scare smectite (generally <5%). Among them, kaolinite is most abundant clay mineral in the East River and North River with an average of 54%; smectite is relatively abundant in the Lingdingyang with an average of 6% (a few samples reach 17%). Their distribution does not show any obvious basin-wide differences. From the Pearl River to the northern margin and the northern slope of the South China Sea, clay mineral assemblages vary significantly: kaolinite decreases gradually to 7%, smectite increases continuously to 35%, and illite also increases gradually to about 40%. Additionally, illite chemistry index steps down and illite crystallinity steps up. This shows that the major sources of illite shift from dominating Pearl River origin strong hydrolysis to Taiwan and the Yangtze River which in turn, are dominated by the physical erosion process. Therefore, the clay mineral contribution to the northern South China Sea from the Pearl River is mainly kaolinite, lesser illite and chlorite, and very scarce smectite. The maximum contribution of clay minerals from the Pearl River is 72% to the northern margin and only 15% to the northern slope of the South China Sea. In both glacial and interglacial periods, the transportation of kaolinite in the Pearl River drainage basin is mainly controlled by river erosion and transport ability augmented by the East Asian monsoon rainfall, representing the mechanical erosion ability in the Pearl River drainage basin.

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