Quaternary clay mineralogy in the northern South China Sea (ODP Site 1146)

—— Implications for oceanic current transport and East Asian monsoon evolution

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Abstract Measurement of clay mineralogy at ODP Site 1146 in the northern South China Sea (SCS) indicates that illite, chlorite, and kaolinite contents increased during glacials and smectite content increased during interglacials. The smectite/(illite+chlorite) ratio and the smectite abundance were determined as mineralogical indicators for the East Asian monsoon evolution. At a 10 ka timescale, prevailing southeasterly surface oceanic currents during interglacials transported more smectite from the south and east areas to the north, showing a strengthened summer monsoon circulation, whereas dominated counter-clockwise surface currents during glacials carried more illite and chlorite from Taiwan as well as from the Yangtze River via the Luzon Strait to the northern SCS, indicating a strongly intensified winter monsoon. Based on a 100 ka timescale, a linear correlation between the smectite/(illite+chlorite) ratio and the sedimentation rate reflects that the winter monsoon has prevailed in the northern SCS in the intervals 2000—1200 ka and 400—0 ka and the summer monsoon did the same in the interval 1200—400 ka. The evolution of the summer monsoon provides an almost linear response to the summer insolation of Northern Hemisphere, implying an astronomical forcing of the East Asian monsoon evolution.

Keywords: clay minerals, sedimentation rate, East Asian monsoon, Quaternary, South China Sea.

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The East Asian monsoon system is a thermodynamic atmospheric circulation induced by the different potential heating between the "Western Pacific Warm Pool" (WPWP) and the Asian continent. The circulation patterns dominate seasonal patterns of winds, precipitation, runoff, and land vegetation over eastern Asia[1, 2]. The winter monsoon is characterized by continental cooling, development of high pressure over northern Asia, and northeast winds across the South China Sea (SCS). In contrast, the summer monsoon circulation is characteristic of continental heating, development of low pressure over Tibet, southwesterly winds across the SCS, and high precipitation over South China[3]. The SCS, the largest marginal sea in the western Pacific, therefore, becomes ideal to record the formation and evolution history of the East Asian monsoon[2, 4−7]. Previous studies based on Late Pleistocene and Holocene records have unraveled that interglacials are characterized by the strengthened summer monsoon circulation and weak winter monsoon winds[2,7] and the strongly intensified winter monsoon and weakened summer monsoon were typical of glacials[2,5]. These studies were mainly based on evidences of micropaleontology, oxygen isotope stratigraphy, or geochemistry. A few studies, however, were performed on clay records in the SCS for the East Asian monsoon evolution[8].

Clay minerals in marine sediments can be implied to track oceanic current variations[9−11], and their vertical distributions have been interpreted widely in terms of contemporaneous paleoclimatic changes prevailing in continental source areas[12−14]. But previous studies on clay minerals in the SCS were dominated by surface sediments and their provenance[15−20]. In this paper, we select an Ocean Drilling Program (ODP) Site 1146 (19°27.40′N, 116°16.37′E, water depth 2092 m), which was collected in the northern SCS in 1999[21]. A total of 508 samples were taken from the upper 191 mcd (meters composite depth) for clay mineral measurement. Based on a preliminary study on variations of clay mineral assemblages[22], potential source and oceanic current transport of individual clay mineral species and their significance on evolution of the East Asian monsoon are evaluated.

1 Methods and age model

The clay minerals were measured by X-ray diffraction (XRD) on oriented mounts of clay-sized particles (<2 μm) [22]. All samples (about 1 cm^3 each) were disaggregated in distilled water and treated with 0.2 N HCl to remove carbonate. The suspensions were kept until pH is checked to avoid overly long exposure to acid. The decarbonated suspensions were washed successively with distilled water to remove excess ions and to help the deflocculation of clays. The particles less than 2 μm were separated following the Stoke's law[23] and were concentrated using a centrifuge. The resulting pastes were spread into calibrated recess into glass slides. Two slides for each sample were made and air-dried to get ready for the XRD analysis. The analysis was conducted using a Philips PW 1710 diffractometer with CuK_a radiation and Ni filter, under a voltage of 40 kV and an intensity of 25 mA. One slide was measured directly as air-dried sample. Then, the slide was measured again after ethylene-glycol solvation for 12 h, as glycolated sample. The other slide was heated at 490°C for 2 h and was measured as heated sample. A few randomly selected samples were investigated under an electronic microscope for crystal shapes of clay minerals. The clay particles were suggested mainly to be continentally clastic and their diagenesis after deposition may be ignorable. Preparation and measurement of the clay mineral analysis were performed at Laboratoire de Sédimentologie et Géodynamique, Université des Sciences et Technologies de Lille, France.

Identification of clay minerals was made mainly according to a comprehensive comparison of three multiple X-ray diffratogrammes obtained under different measurement conditions (fig. 1).

Comparing with the air-dried curve, the heated curve shows that the 14 Å peak decreases visibly and the 10 Å peak increases obviously and a plateau appears between 10 Å and 14 Å peaks. These changes imply the existence of potential illite/smectite random mixed-layers. Furthermore, the 7 Å peak at the heated curve decreases clearly, indicating an existence of kaolinite. Similarly, comparing to the air-dried curve, the glycolated curve shows that a new peak appears at 17 Å and the 14 Å peak decreases obviously, implying the occurrence of smectite. But no any peak or plateau appears between 10 Å and 14 Å at the glycolated curve, which may suggest that reflections of illite/smectite random mixed-layers shift to the interval 14—17 Å. Semi-quantitative calculations of each peak's parameters were carried out on the glycolated curve by using MacDiff software[1)]. 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 illite/smectite random mixed-layers at 17 Å, illite (001) at 10 Å, and kaolinite (001) and chlorite (002) at 7 Å[23]. Kaolinite and chlorite were separated by relative proportions given by a ratio of the 3.57 Å and 3.54 Å peak areas.

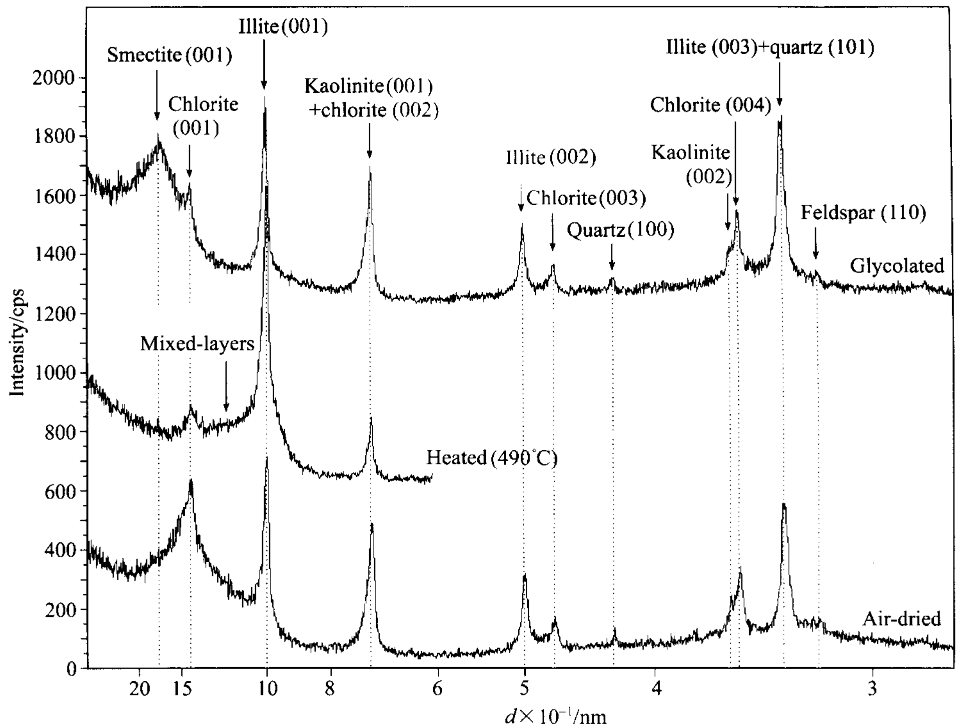

Fig. 1. Multiple X-ray diffratogrammes of a typical sample (1146B-12H-4 110—112 cm, at 114.9 mcd) from ODP Site 1146, showing identification and interpretation of clay-sized fractions. Mixed-layer clay minerals mainly consist of illite/smectite random mixed-layers.

1) Petschick, R., Macdiff 4.2.2, Available: http://Servermac.geologie. un-frankfurt. De/Rainer. html, 2000.

Additionally, some mineralogic characteristics of illite and smectite were determined on the glycolated curve. Illite chemistry index refers to a ratio of the 5 Å and 10 Å peak areas. Ratios lower than 0.5 represent Fe-Mg-rich illites, which are characteristics for physical erosion; ratios higher than 0.5 are found in Al-rich illites, which are formed by strong hydrolysis[24]. Illite crystallinity was obtained from half height width of the 10 Å peak. Lower values represent the higher crystallinity, which is the characteristics for weak hydrolysis in continental sources and dry and cold climate conditions[12,25]. This index also serves as a potential tracer of source regions and transport paths[9]. Smectite abundance was assessed from the ratio 17 and 10 Å peak heights to indicate relative significance of smectite and illite sources [12].

The age model of ODP Site 1146 has been established mainly by using the oxygen isotope stratigraphy[26]. The planktonic foraminifer *G. ruber* (white) was measured on a Finnigan MAT 252 mass spectrometer at Brown University for d^{18}O stratigraphy of the interval 0—185 mcd to determine an age-depth relation of 0—1820 ka with a temporal resolution of 2 ka. For the interval 185—191 mcd, an age of 2050 ka at 2070 mcd was chosen according to the micropaleontology[26]. The oxygen isotope stages of this site were obtained by correlating to the benthic d^{18}O stratigraphy of ODP Site 677[27] (fig. 2).

2 Results

The clay-sized fraction (< 2 μm) minerals over the past 2 Ma (oxygen isotope stages 73-1) at ODP Site 1146 in the northern SCS are mainly composed of four clay mineral species and less important amounts of quartz and feldspar (fig. 1). Among relative clay mineral proportions, smectite (21%—59%) and illite (22%—43%) consist of the major components. These minerals are associated with chlorite (10%—30%) and kaolinite (2%—18%) (fig. 2). The clay mineral percentages indicate a strong glacial-interglacial cyclicity[22]. Generally, illite and chlorite contents have the same pattern of cyclic changes, with higher values during glacials. Their contents decreased by about 6% during the interval 1200—400 ka comparing with that in other periods. On the contrary, smectite content presents a negative variation trend and its values increased by over 15% in the interval 1200—400 ka. The kaolinite content is relatively stable at an average of 12% and indicates higher values during glacials for most of glacial-interglacial cycles.

The illite chemistry index shows a short range of 0.30—0.52 with a general trend of less than 0.50, representing the Fe-Mg-rich illites (fig. 3). The illite crystallinity varies from 0.17º to 0.24º $\Delta 2q$, averaging 0.20º $\Delta 2q$, which is lower than those of loess samples (0.22º—0.33º $\Delta 2q$) on the Chinese Loess Plateau[28], implying a physically weathered environment. The illite chemistry index and crystallinity indicate a complicated fluctuation with no clear glacial-interglacial variation (fig. 3), implying various sources or different climate conditions. Instead, the smectite abundance, ranging from 0.2 to 0.7 with an average of 0.35, indicates a strong correlation to oxygen isotope

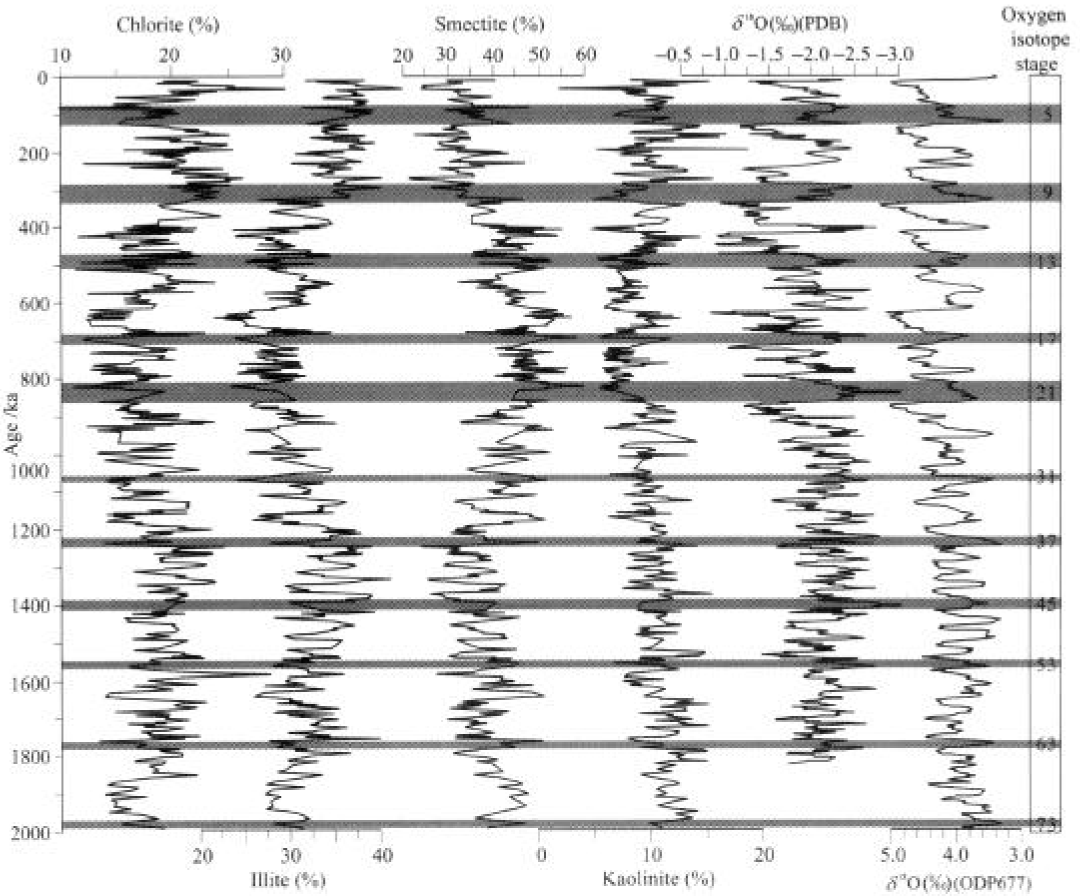

Fig. 2. Clay mineral assemblages and planktonic foraminifer oxygen isotope stratigraphy over the past 2 Ma at ODP Site 1146. Oxygen isotope data of ODP Site 1146 refer to ref. [26] and were performed through 3-point Gauss smoothing; oxygen isotope data of ODP Site 677 see ref. [27].

records with higher values during interglacials, especially during the interval 1200—400 ka (fig. 3). The linear sedimentation rate varied from 4 to 46 cm/ka with an average of 15 cm/ka (fig. 3). The values increased during interglacials from the oxygen isotope stages 37 to 11 (approximate 1200—400 ka) with the exception of stages 17 and 21. But the variation trend was opposed to other periods (specially the interval 400-0 ka) with increased values during glacials.

3 Discussion

3.1 Source analysis and oceanic current transport

The paleoclimatic interpretation of clay minerals in marine sediments requires a detailed knowledge of potential source areas and means of transport processes of individual clay mineral[29,30]. Sources of clay minerals at Site 1146 may be determined by distributions of surface clay minerals and major clay components of surrounding fluvial suspensions[22]. In the areas ranging from the Yangtze River estuary, the Taiwan Strait, to the shallow northern slope with water depth

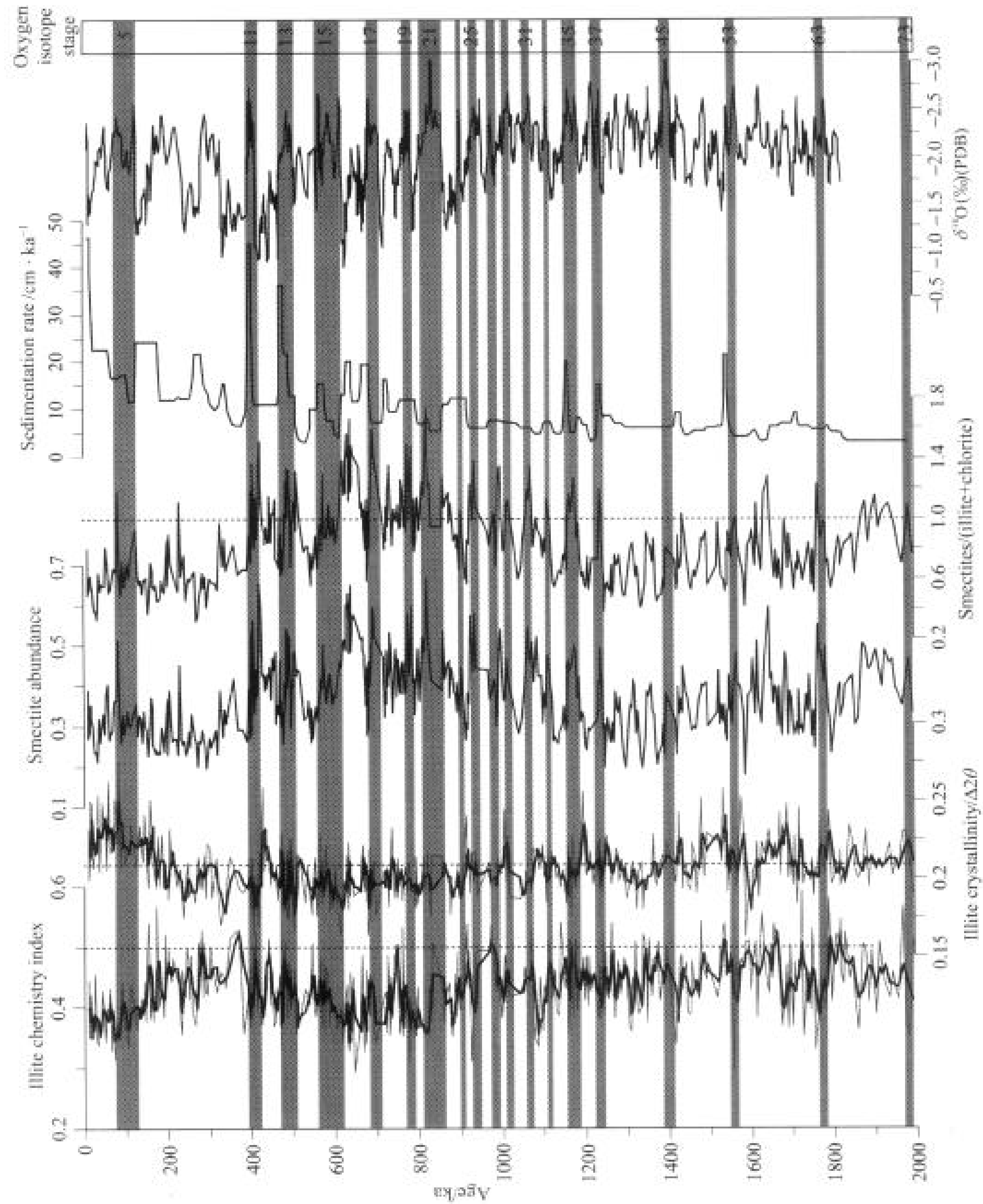

Fig. 3. Illite chemistry index, illite crystallinity, smectite abundance, smectite/(illite+chlorite) ratio, sedimentation rate, and oxygen isotope stratigraphy of ODP Site 1146. Illite chemistry index refers to a ratio of 5 and 10 Å peak areas; illite crystallinity to a half height width of the 10 Å peak; smectite abundance to a ratio of 17 and 10 Å peak heights; all measurements were performed on the glycolated curve. The curves of illite chemistry index and crystallinity (fine gray) were trendlined by 3-point smoothing (coarse dark).

less than 1000 m in the SCS, the illite content varies from 65%[31, 32], 73% to 84%[33]. The chlorite content is generally at 25% and the smectite content is lower than 5%. But the kaolinite content within the Lingdingyang Bay, the Pearl River estuary, is at over 50% and decreases rapidly towards the slope[34]. The illite content nearby the Mekong River estuary is also at about 45% and decreases to 30% in deep-sea areas[15]. The volcanic arc island areas in the eastern and southern parts of the SCS contain a high percentage of smectite, with an average value of over 40%[35]. For example, the smectite content in the Sanda continental slope reaches as high as 50% and the kaolinite content reaches about 20% [15]. The clay mineral contents in the SCS central areas with water depth more than 1000 m vary with an average value of 55% for illite, 21% for chlorite, 12% for kaolinite, and 12% for smectite[15].

The major control on variations of clay mineral assemblages in surface sediments is provenance[15, 18, 20]. Two main sources, which have markedly different geologic characteristics surrounding SCS, are contributing sediments. The northern source is mainly the Asian continent and Taiwan, and the southern source is islands or volcanic arcs. Three large rivers on the Asian continent, Yangtze, Mekong, and Red, discharge 768×10^6 suspended sediments annually into oceans[36]. Their clay mineral fractions consist mainly of illite and chlorite[31, 37]. About 30% sediment discharge from the Yangtze River was observed to escape southward by the nearshore current along the China coast[38]. Major rivers from Taiwan (i.e. Choshui, Tsengwen, Kaoping, Peinan, Hsuikuluan, and Hualien) contribute 185×10^6 sediments annually to oceans[36] with the major clay mineral of illite[39]. The Pearl River discharges 69×10^6 sediments annually[36], but with the major clay fraction of kaolinite[34]. The illite crystallinity at Site 1146 ranges from 0.17° to 0.24° $\Delta 2q$, which are lower than those of loess samples (0.22°—0.33° $\Delta 2q$) and paleosol samples (0.22—0.42° $\Delta 2q$) on the Chinese Loess Plateau[28], implying that the illite in the northern SCS did not mainly come from loess by eolian transport, even during the prevailing winter monsoon periods. The illite chemistry index (0.30—0.52) at Site 1146 is far less than that of the Sanda shelf surface sediments (0.8—1.0)[11].

The clay mineral percentages at Site 1146 indicate a strong glacial-interglacial cyclicity (fig. 2). During interglacial periods, the coastline is approximately similar to the present one and southwesterly surface oceanic currents in summer prevail in most areas of the SCS[2,40] (fig. 4). The currents might transport abundant smectite from islands or volcanic arcs in the south and east to the north. Because the decreased illite and chlorite contents did not follow the changes of smectite (fig. 2), illite and chlorite provided by the Red and Mekong rivers were considered as minor important contribution to Site 1146. During glacial periods, the coastline shifted approximately to the present 100-m isobathic position, and the Borneo Strait and the Gulf of Thailand in the south and the Taiwan Strait in the northeast were closed (fig. 4). The prevailing counter-clockwise surface circulations in winter[40] forced by the winter monsoon transported more illite and chlorite from Taiwan and the Yangtze River via the Luzon Strait to the northern

SCS. In glacials, suspended sediments from the Red and Mekong rivers could not reach to the north because of the opposed current direction. The Paleo-Yangtze River estuary shifted southeastward about 3500 km, but its suspended sediments southward transported could be held relatively by the Okinawa Trough, which starts offshore northeast Taiwan. Therefore, we suggest Taiwan provide very important contents of illite and chlorite for Site 1146. Recent study on geochemical analysis of the past 1.05 Ma for the provenance of ODP Site 1144, which is situated to the northeast of Site 1146, also considered Taiwan as the main source area[41]. In summary, illite and chlorite at Site 1146 came mainly from Taiwan and the Yangtze River, kaolinite mainly from the Pearl River, and smectite mainly from islands or volcanic arcs in the south and east (fig. 4).

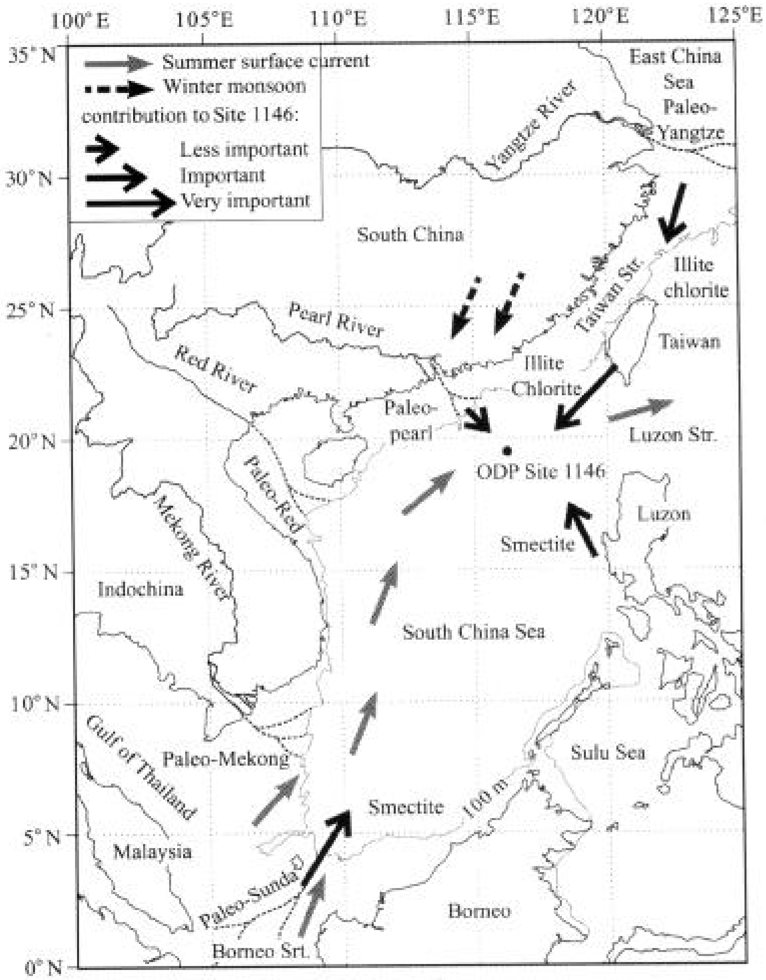

Fig. 4. Source analysis and oceanic current transport of clay minerals at ODP Site 1146. The coastline is approximately the present one during interglacials and shifts to the approximate present 100-m isobath position (point line). The paleo-rivers (dash line) refer to potential rivers developed on the exposed shelf during glacials. Data of surface oceanic currents in summer and winter monsoon see ref. [2].

3.2 Mineralogical indicators of the East Asian monsoon evolution

The illite chemistry index and crystallinity at Site 1146 indicate a physically weathered environment, but their complicated fluctuations do not present clear glacial-interglacial cyclicity (fig. 3). This phenomenon could be produced by various climate conditions and multi-period weathered profiles, which were distributed in the broad Yangtze River drainage basin with an area of 1.94 × 10^6 km^2. Kaolinite provided by the Pearl River primarily originated from red-soil weathered profiles in South China[42] and was formed by weathered profiles of volcanic rocks of various periods. Therefore, the kaolinite compiles various climate characteristics of different geological times. Smectite could be formed preferentially from volcanic rocks, regardless of typical climate conditions, if there is sufficient water to allow hydrolytic processes[12]. For instance, in regions around Iceland and near islands of west Antarctica, high content of smectite is as much as 40%[43]. Consequently, the clay mineral assemblages at Site 1146 directly reflect variations of provenance and oceanic currents, instead of contemporary climates. But the surface currents in the SCS are forced mainly by the East Asian monsoon circulations[2, 7, 40]. Thus, the clay mineral assemblages at Site 1146 actually are implied for the prevailing monsoon variations in the SCS. During interglacials, the prevailing southwesterly surface oceanic currents transport more smectite from the south and east areas to the north; whereas during glacials, the prevailing counter-clockwise surface oceanic currents carried much more illite and chlorite from Taiwan, and sources of the Yangtze River via the Luzon Strait to the position of Site 1146. Therefore, higher contents of illite and chlorite represent the dominating winter monsoon and higher content of smectite represents the dominating summer monsoon.

Generally, multiple sources and mutual dilution of individual clay minerals make it difficult to assign changes of paleoclimate or oceanic currents by a single mineral factor, but by their ratio of several components[30]. In this paper, the mectites/(illite+chlorite) ratio and the smectite abundance are adopted as mineralogical indicators for the East Asian monsoon evolution. The two indicators have a similar variation versus age (fig. 3). At a 10 ka timescale, the relatively high ratio and high abundance occur in interglacials, implying a strengthened summer monsoon circulation and weak winter monsoon winds. In contrast, the relatively low ratio and low abundance happened in glacials, inferring a strongly intensified winter monsoon and weakened summer monsoon. These results could be correlated to previous studies on the Late Pleistocene[2,5,7]. The fluctuations of sedimentation rate can be correlated to the glacial-interglacial cyclicity (fig. 3). In the interval 1200—400 ka, the sedimentation rate and the smectite content increased synchronously during interglacials, indicating that the increased smectite (the relative higher smectite/(illite+chlorite) ratio) became the major component of sediments. The increased smectite was induced by the intensified summer monsoon. In the intervals 2000—1200 ka and 400—0 ka, however, the sedimentation rate increased during glacials, suggesting that the increased illite and chlorite (the relatively low smectite/(illite+chlorite) ratio) could be the major components of sediments. The process represents the prevailing winter monsoon. Based on a 100 ka timescale, therefore, the winter monsoon has prevailed in the northern South China Sea in the intervals 2000—1200 ka and 400—0 ka, so has the summer monsoon in the interval 1200—400 ka.

Recent studies found a linear correlation between the East Asian summer monsoon evolution in the SCS and the summer insolation of Northern Hemisphere[2,44]. The stronger Northern Hemisphere summer insolation can lead to a strengthened land-sea beating contrast, which enhances the summer monsoon precipitation and runoff, in turn causing wind variations between land and sea. We used the average summer insolation of 65 N[45] as a target curve to compare with the mineralogical indicator of the East Asian monsoon evolution for the interval 1200—400 ka, when the summer monsoon prevailed. An almost linear correlation that the insolation maxima correspond to the indicator maxima (the highest smectite/(illite+chlorite) ratios, representing the prevailing summer monsoon is displayed (fig. 5). The comparison indicates that the summer monsoon forced the surface oceanic currents to transport more smectite to the north, implying an astronomical forcing of the East Asian monsoon evolution.

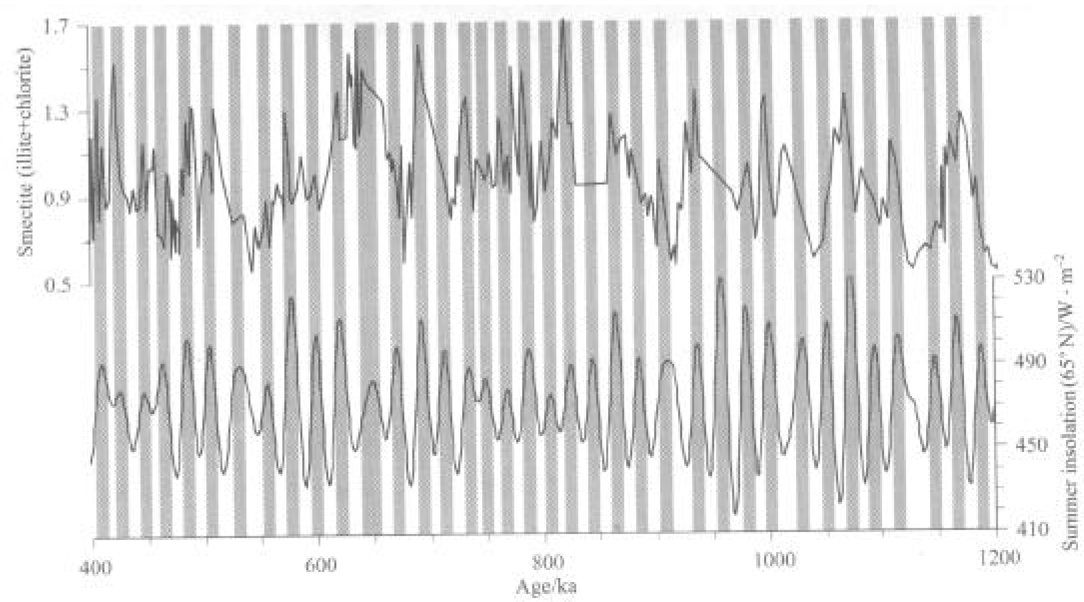

Fig. 5. Comparison between the smectite/(illite+chlorite) ratio at ODP Site 1146 and the summer insolation of Northern Hemisphere during the interval 1200—400 ka. The summer insolation refers to an average insolation during June and July at 65 N following the Laskar (1990) solution[45], using Analyserie software[46].

4 Conclusions

The Quaternary clay mineral assemblages at ODP Site 1146 in the northern SCS consist mainly of smectite (21%—59%) and illite (22%—43%), with associated chlorite (10%—30%) and kaolinite (2%—18%). They indicate a strong glacial-interglacial cyclicity that illite, chlorite, and kaolinite contents increased during glacials and smectite content increased during interglacials.

During both interglacials and glacials, the clay minerals have relatively stable provenances and transport means. The prevailing southeasterly surface oceanic currents during interglacials transport more smectite from the south and east areas to the north; whereas the dominating counter-clockwise surface currents during glacials carried more illite and chlorite from Taiwan and the Yangtze River via the Luzon Strait to the northern SCS; kaolinite is provided mainly by the Pearl River. The surface currents in the SCS are forced mainly by the East Asian monsoon circulations. Therefore, the smectite/(illite + chlorite) ratio and the smectite abundance were determined as mineralogical indicators for the East Asian monsoon evolution. At a 10 ka timescale, the strengthened summer monsoon circulation prevailed during interglacials, whereas a strongly intensified winter monsoon prevailed during glacials. Based on a 100 ka timescale, a linear correlation between the smectite/(illite+chlorite) ratio and the sedimentation rate reflects that the winter monsoon has prevailed in the northern SCS in the intervals 2000—1200 ka and 400—0 ka so has the summer monsoon in the interval 1200—400 ka. The evolution of the summer monsoon provides an almost linear response to the summer insolation of Northern Hemisphere, implying an astronomical forcing of the East Asian monsoon evolution.

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