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## Clay mineral evolution in the central Okinawa Trough since 28 ka: Implications for sediment provenance and paleoenvironmental change

Yanguang Dou<sup>a</sup>, Shouye Yang<sup>a, b, \*</sup>, Zhenxia Liu<sup>c</sup>, Peter D. Clift<sup>d</sup>, Hua Yu<sup>e</sup>, Serge Berne<sup>f</sup> and Xuefa Shi<sup>c</sup>

<sup>a</sup> State Key Laboratory of Marine Geology, Tongji University, Shanghai 200092, China

<sup>b</sup> Key Laboratory of Yangtze River Water Environment, MOE, Tongji University, Shanghai 200092, China

<sup>c</sup> First Institute of Oceanography, State Oceanic Administration, Qingdao 266071, China

<sup>d</sup> School of Geosciences, University of Aberdeen, Aberdeen AB24 3UE, United Kingdom

<sup>e</sup> Department of Earth Sciences, Rice University, Houston, TX 77005, USA

<sup>f</sup> IFREMER, DRO/GM, P.O. Box 70, 29280 Plouzané, France

\*: Corresponding author : Shouye Yang, Tel.: +86 21 6598 9130; fax: +86 21 6598 6278, email address : [syyang@tongji.edu.cn](mailto:syyang@tongji.edu.cn)

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### Abstract:

The Okinawa Trough is a natural laboratory for the study of later Quaternary land–ocean interaction and paleoenvironmental changes. In this study we reconstruct the evolution of clay mineral assemblages in Core DGKS9604 retrieved from the central Okinawa Trough. Illite dominates the clay mineral compositions, with average contents above 60%. Clay mineral evolution since 28 ka is closely related to changes in sediment provenance and paleoenvironment. Sea level rise and the strength of the Kuroshio Current control the dispersal and deposition of clays on the East China Sea shelf and in the Okinawa Trough, and thus, determine the clay mineral compositions in the core sediments. During the late last glacial period (28.0–14.0 ka), the paleo-Changjiang River mouth was situated at the shelf edge close to the central Okinawa Trough and thus, together with the outer shelf, supplied large volumes of terrigenous sediments directly into the trough. From 14.0 to 8.4 ka influence from the Changjiang decreased while the mid-outer shelf of the East China Sea became the dominant sediment source to the central Okinawa Trough as sea level rose and the Changjiang river mouth migrated west. Strong sediment reworking and erosion at the shelf edge at 15–13 ka significantly increased the lateral transport of fine-grained shelf sediments to the central Okinawa Trough.

Since ca. 8.4 ka clays from Taiwan have dominated the sediment flux to the site, coinciding with the re-entry of the Kuroshio Current into the trough. The increasing influence of the Changjiang-sourced sediments since 1.5 ka was probably related to the weakening of the Kuroshio Current and/or a higher river flux.

**Keywords:** Clay mineral; Sediment; Provenance; East China Sea; Okinawa Trough

## 1. Introduction

The impact that climate change has on continental environments is a topic of interest to those trying to understand the Earth's climate system, and possibly predicting responses to future climate change. In theory marine sediments may store a record of the environmental conditions and allow us to compare these with changes in oceanographic circulation and with global temperatures. However, in order to read this record we need to understand what controls the delivery of clastic sediment to continental margins. Here we present an example from East Asia where sediments from the Okinawa Trough are influenced by strong changes in the Asian monsoon since the Last Glacial Maximum (LGM), as well as by rising sea level and fluctuations of the powerful Kuroshio Current that flows northward along the eastern edge of Asia.

The Okinawa Trough, extending from southwest Kyushu, Japan, to the Lanyang Plain of northeast Taiwan, is a back-arc basin separating the continental shelf of the East China Sea (ECS) from the Ryukyu arc (Sibuet et al., 1998). It is an ideal research area for paleoceanography and continent-ocean interactions because of its continuous sedimentation and because the region is affected by a series of rapid paleoenvironmental changes since the late Quaternary (Liu et al., 1999, 2001; Li et al., 2001, 2005; Lin et al., 2006; Xiang et al., 2007). The supply, transport and deposition of terrigenous materials in the Okinawa Trough are controlled by many factors, including sea-level fluctuation (Lambeck et al., 2002; Liu et al., 2004), variability in the strength and location of the Kuroshio Current (Ujiié et al., 1991, 2003; Ujiié and Ujiié, 1999; Jian et al., 2000), changes in fluvial run-off, many of which are controlled or linked to variability of the summer monsoon, which dominates the regional climate (Wang et al., 1999; Clift et al., 2002, 2008; Boulay et al., 2005).

The strong relation between environmental changes and variations in terrigenous sediment supply can be addressed through clay mineral signals in the marine sediments that bear information on terrigenous sediment provenance. Clay minerals are common components in most marine sediments, especially those deposited on continental margins, where there is a significant terrigenous input (Biscaye, 1965; Griffin et al., 1968). Clay minerals can provide valuable information on sediment provenance, such as relative contributions of river and eolian inputs, but can also constrain sediment transport and depositional processes, as well as providing information on weathering regimes within the continental interior (Biscaye, 1965; Petschick et al., 1996; Thiry, 2000). Recent studies suggest that clay mineral assemblages in west Pacific marginal seas can be closely related to climate fluctuation on orbital timescales, such as those known to affect the evolution of the Asian monsoon (Wehausen et al., 2002; Liu et al., 2003, 2005; Tamburini et al., 2003; Boulay et al., 2005). During the Quaternary and Tertiary the flux of clay minerals into the Pacific marginal seas is mostly controlled by changes in sea level, oceanic circulation, and the relative intensities of physical erosion and chemical weathering in the source areas. However, over shorter time scales, such as in the last glacial-interglacial interval, clay mineral assemblages can predominantly reflect changing influence of different sediment sources, together with partitioning processes during transport (Steinke et al., 2008). These latter are in turn tightly linked to sea level and paleoenvironmental changes (Diekmann et al., 2008). Reconstruction of the evolving clay mineral assemblage in the terrestrial sediment fraction within a continental margin sequence may help to better understand the interactions between detrital sediment supply, paleoceanographic changes and paleoclimatic variability in the source areas.

The clay mineral assemblages in the East China Sea primarily consist of illite, which constitutes average contents of about 60–70 %, while smectite, kaolinite and chlorite are subordinate (Aoki et al., 1974; Xu et al., 1983; Yang et al., 1988; Fan et al., 2001; Yang et al., 2003). Most previous studies on clay mineral assemblages in the ECS focused on the estuarine area and continental shelf, while the Okinawa Trough was less studied. Yu et al. (2008) used clay mineral compositions in Core DGKS9603 from the central trough to define a cooling event at 8.2 ka (Fig. 1). In the southern Okinawa Trough, clay mineral data pointed to increased sediment supply from northwestern Taiwan between 28 and 19.5 ka and from sources of eastern mainland China between 19.5 and 11.2 ka (Diekmann et al., 2008). During the Holocene the southern Okinawa Trough was dominated by the sediments delivered by northeast Taiwanese rivers (Diekmann et al., 2008). The change in provenance at 19.5 ka is interpreted to reflect increased fluvial runoff from the Changjiang (Yangtze River) and strong sediment reworking from the ECS shelf caused by increased humidity (Diekmann et al., 2008).

In this paper, we present a high-resolution reconstruction of clay mineral evolution in Core DGKS9604 taken from the central Okinawa Trough. The main objectives of this study are to quantitatively estimate the relative contributions of fine-grained terrigenous sediments from different provenances, and thus to explore the link between clay mineral assemblages and paleoenvironmental changes in the sea and surrounding continental areas since the Last Glacial Maximum.

## **2. Geological and oceanographic settings**

The Okinawa Trough is 230 km wide at its northern end, and 60–100 km wide in the south, and extends for about 1200 km between Taiwan and the Kyushu Islands

(Fig. 1). The maximum water depth is about 200 m in the north and ~2300 m in the south. The thickness of the sedimentary cover decreases from ~8 km in the north to about 2 km in the south (Sibuet et al., 1987). Sediment accumulation rates in the middle and southern Okinawa Trough were very low during the early Pleistocene as a result of the blocking of the Changjiang and Huanghe (Yellow River) river sediments by the uplift of the Taiwan–Sinzi folded belt, but became very high (up to 4 km/m.y) since 0.5 Ma (Sibuet et al., 1987). Previous studies have demonstrated that the terrigenous sediments in the central Okinawa Trough predominantly originate from the Changjiang and Huanghe Rivers (Qin et al., 1987; Iseki et al., 2003; Katayama and Watanabe, 2003; Liu et al., 2007). In addition, episodic volcanic eruptions from the western Japanese volcanic zone (Machida, 1999; Miyairi et al., 2004), submarine hydrothermal activity (Zeng et al., 2000; Zhai et al., 2001), erosion from Taiwan (Liu et al., 2008), eolian transport (Tsunogai et al., 1985), seafloor earthquakes (Huh et al., 2004), as well as the intrusion of the Kuroshio Current (Jian et al., 2000; Lee et al., 2004), also contribute to the transport of siliciclastic sediments into and within the trough. Thus interpreting the sedimentary record from the trough is potentially a complicated process.

The most striking oceanographic feature in the Okinawa Trough is the Kuroshio Current which is the major western boundary current of the North Pacific Ocean, and significantly impacts surface water character and climatic conditions in the northwestern Pacific region (Jian et al., 2000; Lee and Chao, 2003; Ujiie et al., 2003). The Kuroshio Current originates in the westward-flowing North Equatorial Current of the central Pacific Ocean and deflects towards the northeast offshore the Philippines. The main axis of the Kuroshio Current enters the Okinawa Trough northeast of Taiwan across the I-Lan Ridge, and flows northeastward along the outer edge of the

continental shelf of the ECS, with minor, seasonal lateral shifts of the flow path (Fig. 1; Andres et al., 2008). The current bifurcates into the Ryukyu Arc through the Tokara Strait and into the Yellow Sea and Japan Sea, respectively (Fig. 1; Andres et al., 2008). On the continental slope, the suspended particulate matter and lithogenic elements increase with water depth, implying strong lateral transport of lithogenic particles (Hung et al., 1999). According to Hsu et al. (1998), the Kuroshio Current may transport large volumes of fluvial sediments from east Taiwanese rivers, in particular the Lanyang River, to the southern Okinawa Trough. However, the main Kuroshio Current was deflected from the Okinawa Trough during the LGM because of the emergence of a land bridge connecting Taiwan and the central of southern Ryukyu Arc (Ujiié et al., 1991, 1999; Ahagon et al., 1993; Ijiri et al., 2005). At that time, the main current shifted to the east at the southern end of the Ryukyu Arc (Ujiié et al., 1991, 1999). However, the intensity and flow path of the Kuroshio Current within the Okinawa Trough during the LGM and Holocene are still controversial, as is the timing of re-entry of the main current into the trough region, which has been suggested to be between 16 ka and 7.3 ka based on a series of different proxy records (Li et al., 1997; Ujiié and Ujiié, 1999; Xu and Oda, 1999; Jian et al., 2000; Li et al., 2001, 2005; Ujiié et al., 2003; Kao et al., 2005, 2008; Xiang et al., 2007).

### 3. Materials and methods

Piston core DGKS9604 was taken from the central Okinawa Trough at water depth of 766 m, during the joint Chinese-French DONGHAI cruise of R/V L'Atalante in 1996 (Fig. 1). The core is 10.76 m long and mainly composed of clayey silt without apparent ash layers. The age model for Core DGKS9604 is based on the oxygen isotope of Globigerinoides sacculifer ( $\delta^{18}\text{O}$ ), and accelerator mass spectrometry (AMS) radiocarbon dating of planktonic foraminifers (Liu et al., 2001; Yu, 2006; Yu et al.,

2008, 2009). The depositional age at the core bottom is estimated at 37.01 ka, with bulk sedimentation rate averaging about 29 cm/k.y. For this study, a total of 88 samples were taken at about 8.4 cm intervals from the upper 630 cm of Core DGKS9604.

The clay minerals analyses were performed by standard X-ray diffraction (XRD) following the procedure described by Holtzapffel (1985). All the samples were first decalcified with 0.2 N HCl. Excess acid was removed by repeated rinse with distilled water and centrifuging. Particles smaller than 2 $\mu$ m were separated by exploiting Stoke's Law and concentrated by centrifuging. Each sample was transferred to two slides by wet smearing. Samples were then air-dried prior to XRD analysis. One slide was first measured directly after air-drying, and then measured again after ethylene-glycol solvation for 48 hours. Another slide was heated at 490 °C for two hours and then analyzed (Liu et al., 2003). The analyses were conducted using a Rigaku D/max-rB X-Ray diffractometer (CuK $\alpha$  radiation, 40 kV voltages; a 100 mA intensity and 2°/min ( $2\theta$ ) speed). The analyses were run from 3° to about 35°  $2\theta$ . Identification of specific clay minerals was made using the basal layer plus the interlayer revealed by the XRD patterns (Brown and Brindley, 1980). Peak areas were calculated after manual baseline correction using MacDiff software version 4.2.2 (Petschick, 2000), following the semi-quantitative method of Biscaye (1965, 1997). The error of this method is estimated to be about 5% of the relative abundance of each clay mineral.

#### 4. Results

Downcore variations of clay mineral assemblages in Core DGKS9604 are shown in Figure 2. The contents of clay fraction in the core sediments vary between 14.0%

and 28.9%, with an average of 24.9%. The assemblages are dominated by illite (63.0–87.3%), while smectite (and mixed-layer minerals) (1.0–13.1%), chlorite (7.1–22.8%) and kaolinite (2.0–8.8%) are less abundant. Based on the downcore variations of clay minerals, the stratigraphy of Core DGKS9604 can be divided into three units: Unit 1 (~8.4–0 ka), Unit 2 (~14.0–8.4 ka) and Unit 3 (~28.0–14.0 ka) (Fig. 2). Smectite contents show large downcore fluctuations, with high values in Unit 2 at 14.0–8.4 ka but lower values at around 14–17 ka and after 8 ka. It is noteworthy that the smectite contents decrease gradually within Unit 2 and during the period of 0–2 ka in Unit 1 and 19–14 ka in Unit 3. Overall, kaolinite and chlorite have similar variations and their contents are stable in Unit 2 and 3, except for a number of abnormal values. Illite is more abundant in Unit 3 than in Units 1 and 2. Illite/smectite ratios, which may be used as proxies for the relative intensities of physical erosion versus chemical weathering, show weak downcore variations, except for higher values at 14–17 ka and 0–2 ka. These changes are obviously driven by low smectite contents at those times. In comparison, chlorite/kaolinite ratios vary from 1.8 to 6.6, exhibiting an ascending trend from bottom to top (Fig. 2).

The contents of clay minerals within the three units of Core DGKS9604 are somewhat different from those in the neighboring core (DGKS9603) from the central Okinawa Trough (Guo et al., 2000; Yu et al., 2008; Table 1). However, clays exhibit the same overall trends. The contents of clay minerals in Unit 1 (~8.4–0 ka) are very close to those defined by Aoki and Oinuma (1974), as well as those in the sediments from ODP Site 1202B (9–0 ka) from the southern Okinawa Trough (Diekmann et al., 2008; Table 1).

## 5. Discussion



### 5.1 Provenance discrimination of the fine-grained terrigenous sediments

Clay minerals in marine sediments are derived from two main sources: one is terrigenous detritus, which contain most of the provenance information; another is authigenic processes which may be caused by alternation of volcanic seafloor basement or hydrothermalism. Provenance discrimination of clay minerals requires a detailed understanding of potential source areas, as well as constraints on transport media and processes (Chamley, 1989; Diekmann et al., 1996; Steinke et al., 2008). The potential provenances of clay minerals in the central Okinawa Trough include terrigenous sources supplied via fluvial and aeolian inputs, as well as volcanic alternation and hydrothermal processes. Intermittent hydrothermalism has primarily taken place at the bottom and east slope of the central Okinawa Trough (Zeng et al., 2000). However, Core DGKS9604 is located on the west slope where the sediments are mainly composed of terrigenous and biogenic materials (Meng et al., 1997; Yu et al., 2006). As a result, the influence of submarine hydrothermal activity on clay mineral compositions in the core is weak and can be neglected for the purposes of this study. While eolian particles from mainland China play an important part in the detrital fluxes to the pelagic Pacific Ocean (Rea et al., 1990; Nakai et al., 1993), the supply of windborne materials to the East Asian marginal seas is overwhelmed by fluvial-derived detritus (Chen, 1978; Diekmann et al., 2008). Furthermore, it is hard to distinguish eolian materials from the mainland-derived fluvial sediments in the central Okinawa Trough, because they are expected to have similar mineralogy. Furthermore, the proximal aeolian particles are usually  $>2 \mu\text{m}$  in size (Jan-Berend Stuut, personal communication), and thus did not contribute to the clay component in the marine sediments.

Previous studies suggest that the terrigenous sediments in the central Okinawa

Trough predominantly originate from the two largest rivers in China, i.e. the Changjiang and Huanghe, and partly from the ECS shelf (Qin et al., 1987; Meng et al., 1997; Iseki et al., 2003; Katayama and Watanabe, 2003; Liu et al., 2007). The clay mineral assemblages in modern Changjiang and Huanghe sediments are overall similar, albeit with a higher content of smectite in the Huanghe (Table 1; Xu, 1983; Yang, 1988; Fan et al., 2001; Yang et al., 2003).

In contrast, the clay mineral compositions on the open shelf of the ECS (Chen, 1973; Aoki and Oinuma, 1974; Li, 1990) are different from those in the Changjiang and Huanghe deltas, probably because of hydrodynamic sorting and mixing processes that the fluvial sediments experience after entry into the sea. For example, the contents of kaolinite decrease from 9.4% to 7.5% while chlorite increases from 13.2% to 23.9% between the Changjiang Estuary and the open shelf (Table 1; Chen, 1973; Fan et al., 2001). Furthermore, the clay mineralogy in the subaqueous delta of the Changjiang is characterized by remarkably low smectite content (Fang et al., 2007), compared to those measured in the modern Changjiang sediments or from the open shelf. There is no doubt that the clay minerals in the subaqueous delta front and prodelta are ultimately sourced from the Changjiang. As a result, we infer that the differences are caused by strong hydrodynamic fractionation of the river-borne clay minerals by wave and tidal currents. In particular, smectite which has the smallest size of any of clay minerals, is prone to winnowing by oceanic currents (Chamley, 1989). Terrigenous sediments from east Taiwan may be another potential source supplying sediment to the Okinawa Trough, because of the strong influence of the Kuroshio Current. Sediments derived from east Taiwanese rivers, especially the Lanyang River, are characterized by high chlorite contents (Chen, 1973; Su et al., 2009). This mineral is predominantly weathered from chlorite-rich greenschists and slates exposed in the

high mountain ranges of east Taiwan (Diekmann et al., 2008; Su et al., 2009).

In order to constrain the provenance of clay minerals sampled from Core DGKS9604, we developed a discrimination plot of illite/smectite vs chlorite/kaolinite ratios and a ternary diagram of smectite – chlorite – (illite+kaolinite)(Figs. 3, 4). The clay mineral assemblages in the core are compared with reference data from potential provenances including the modern Changjiang, the Huanghe (Xu, 1983; Yang, 1988; Fan et al., 2001; Yang et al., 2003), the ECS shelf including the subaqueous delta (Chen, 1973; Aoki and Oinuma, 1974; Li, 1990; Fang et al., 2007), and East Taiwan shelf (Chen, 1973) including the south Okinawa Trough (ODP Site 1202B)(Diekmann et al., 2008).

Our plot clearly suggests that the clay minerals in the lower unit (Unit 3) of Core DGKS9604 are most similar to those of the subaqueous Changjiang Delta, and are close in composition to those sampled from the ECS shelf, because both have low illite/smectite ratios (Fig. 3). This indicates that the clays accumulated in the central Okinawa Trough during the period of 28.0–14.0 ka were primarily supplied by the paleo-Changjiang River. The clay mineral assemblages in the middle unit (Unit 2) largely overlap with those from the ECS shelf and partly with the east Taiwan shelf and southern Okinawa Trough, suggesting that the middle-outer shelf of the ECS could be the main source for clays deposited in the central trough between the last deglacial period and the Early Holocene (14.0–8.4 ka). The upper unit (Unit 1) of the Core DGKS9604 shows high chlorite/kaolinite ratios and lower contents of kaolinite, a characteristic which is comparable with those found at ODP Site 1202B in the southern Trough, as well as from the ECS shelf (Figs. 3, 4). We therefore infer that since ca. 8.4 ka the clays deposited in the central Okinawa Trough have predominantly originated from east Taiwan and partly from the ECS shelf.

## 5.2 Quantitative estimation of different provenance contributions

Based on the source discrimination presented above, the three end members, Changjiang (here the subaqueous delta was used for reference considering hydrodynamic fractionation of clay minerals in the sea), and the ECS shelf, and the east Taiwan shelf, are considered as the main sources of clay minerals to the central Okinawa Trough (Fig. 4). A simple three end-member mixing model was developed to quantify the contributions of each source to the clay mineral assemblages in the core sediments. The following equations were extrapolated by using the contents of chlorite, smectite and illite+ kaolinite in the three end-members and sediments from Core DGKS9604.

$$1.1X_i+14.8Y_i+3Z_i=M_i$$

$$16.4X_i+15.2Y_i+26Z_i=Ch_i$$

$$82.5X_i+70Y_i+71Z_i=KI_i$$

$X_i$ ,  $Y_i$  and  $Z_i$  represent the contributions of clay minerals from the Changjiang-, ECS shelf- and Taiwan respectively in each sample (i) of Core DGKS9604;  $M_i$ ,  $Ch_i$ ,  $KI_i$  represent the smectite contents, chlorite, and kaolinite+illite in every sample of the core respectively.

The contributions from each of the three end-member sources to the fine-grained sedimentation at Core DGKS9604 are displayed graphically in Figure 5. The Changjiang- and ECS shelf-derived clays dominated the central Okinawa Trough during the period from 28.0 to 14.0 ka. The Changjiang contribution increased gradually from about 30% to more than 80% over that time period, while the ECS shelf-derived clays decreased from around 50% to less than 10%. From the last deglaciation to early-mid Holocene (14.0–8.4 ka), the proportion of clays supplied from the ECS shelf increased abruptly, rising to more than 60% and then decreasing

gradually to around 30%. Meanwhile, the proportion of Changjiang-derived clays decreased from more than 60% to less than 30% within a short time (15–13 ka). At the same time, the clays sourced from east Taiwan have gradually increased from less than 20% at 12 ka to more than 50% at 8 ka. This result is consistent with clay mineral assemblages at ODP Site 1202. The recent study demonstrated that the Holocene sediments in the southern Okinawa Trough were dominated by sediments originated from rivers in northeast Taiwan (Diekmann et al., 2008). Therefore, since the Early Holocene, eastern Taiwan-derived clays have dominated the central Okinawa Trough, whereas the fluxes of clays from the Changjiang and the ECS shelf have become meager, estimated at around 20% each (Fig. 5). Furthermore, since 1.5 ka, the clay contribution from the Changjiang increased abruptly to 60% and the ECS shelf-derived diminished gradually.

### *5.3 Paleoenvironmental implication of clay mineralogy*

Several factors influenced the depositional regimes in the Okinawa Trough, including sea level, fluvial discharges, flow path of the Kuroshio Current, and monsoon climate (Wei, 2006). The extent of influence of each factor is closely related to global and regional climate changes and marine environments. Here we describe how these competing processes have controlled sedimentation in the central Okinawa Trough since the LGM.

#### *5.3.1 Late last glacial to early deglacial period (28–14 ka)*

During the late last glacial period, the climate was cooler and drier than at present and global sea level was low (Chappell et al., 1996; Siddall et al., 2003; Clark et al., 2009), which led to progradation of the coast line and emergence of an exposed ECS shelf (Saito et al., 1998). This exposure must have significantly influenced sedimentation on the continental slope and deep-sea basin (Steinke et al., 2008).

Furthermore, during that time interval, the paleo-Changjiang River mouth was situated at the shelf edge, close to the central Okinawa Trough (Ujiié et al., 2001), while the main axis of the Kuroshio Current was deflected east of the Ryukyu Islands (Ujiié et al., 1991; Ahagon et al., 1993; Xiang et al., 2003). A bridge connecting the central-southern part of the Ryukyu Arc with Taiwan Island prevented the flow of the Kuroshio Current into the Okinawa Trough (Ujiié et al., 1991; Ujiié and Ujiié, 1999).

As a result of these changes, a large volume of fine-grained terrigenous sediments from the paleo-Changjiang and the mid-outer shelf of the ECS were transported directly into the central Okinawa Trough. The high sedimentation rates in Core DGKS9604 during the LGM are a reflection of rapid sediment supply from the paleo-Changjiang and the exposed open shelf (Fig. 5). Sediment geochemical studies also confirmed the dominance of Changjiang-derived sediments to the sedimentation in the trough at that time (Meng et al., 2007).

Variability of the East Asian monsoon might influence the weathering in the drainage basins and thus, determine the rates and compositions of fluvial fluxes into the marginal seas (Morley and Heusser, 1997; Wang et al., 1999). The monsoon also influenced oceanic circulation and sediment transportation in the ECS. Sediment trap records in the blue water South China Sea indicate that terrigenous material flux reaches a maximum when winter monsoon dominates (Jennerjahn et al., 1992). High sedimentation rates and dominance of the ECS shelf-derived sediments in the central Okinawa Trough during the LGM might reflect the location of the core site close to the river mouth due to the low sea level. A strengthened winter monsoon would have significantly enhanced the lateral cross-shelf transport of fine-grained terrigenous sediments to the continental slope (Fig. 5).

### *5.3.2 Late deglacial to early Holocene period (14–8.4 ka)*

During the last deglaciation the East Asian winter monsoon weakened while the summer monsoon strengthened, accompanied with rising sea level (Chappell et al., 1996; Siddall et al., 2003; Wang et al., 2008). During the early deglacial period at ca. 18–15 ka the contribution from the Changjiang still dominated the clay sedimentation in the central Okinawa Trough, probably because of the slow sea level rise (Fig. 5). Nevertheless, rapidly rising sea level significantly increased the distance between the Changjiang river mouth and the Okinawa Trough, causing the Changjiang contribution to the central trough to decrease abruptly between 15 and 13 ka (Fig. 5). In contrast, the decrease in Changjiang-derived sediments was counterbalanced by a rapid increase of the contribution from the ECS shelf. The abrupt sea level rise at ca. 15–13 ka (Clark et al., 2009) may have resulted in strong sediment reworking and erosion at the shelf edge (Steinke et al., 2008), which may have increased the lateral transport of fine-grained shelf sediments, and correspondingly increased sedimentation rates in the central trough at that time (Fig. 5). With continually rising sea level around 13–8.4 ka, the sources of clayey sediment to the central Okinawa Trough changed from the Changjiang to reworked materials from the ECS shelf. We support this suggestion by Ujiie et al. (2001) that the weakening influence of Changjiang-derived clays was caused by the large-scale landward retreat of the river mouth. Furthermore, the clay contribution from the ECS shelf also decreased at 13–8.4 ka (Fig. 5), suggesting that the continuously rising sea level increased the transport distance from the open shelf to the trough.

### *5.3.3 Mid-late Holocene period (8.4–0 ka)*

Sea level rose to a level of approximately 50 m to 40 m below the present between 11 and 9 ka (Lambeck et al., 2002), while the main axis of the Kuroshio Current re-entered the Okinawa Trough (Ujiie et al., 1991, 1999; Xiang et al., 2007;

Diekmann et al., 2008). The changes of the Kuroshio Current exerted a significant influence on sediment dispersal and depositional processes in the ECS, given that this transported large volumes of Taiwan-sourced sediments into the southern Okinawa Trough (Liu et al., 2003; Diekmann et al., 2008). The proportion of Taiwan-derived sediments in Core DGKS9604 increased from less than 20% in the Early Holocene to more than 50% at present (Fig. 5), supporting the importance of the Kuroshio Current acting as an effective transporter of fine-grained sediment into the trough. Sea level rose to a highstand similar to the present level at ca. 7 ka. Modern oceanic circulation and depositional patterns were established in the ECS at that time (Feng, 1983). The strengthening of the Asian summer monsoon in the Early Holocene may have enhanced continental erosion and run-off, and thereby increased the fluvial flux into the sea (Wang et al., 1999, 2005; Clift et al., 2008). The small contributions of the Changjiang-derived sediments to deposition in the central Okinawa Trough during the early-mid Holocene, however, implies that the presence of a strong Kuroshio Current along the trough may act as a barrier and block the lateral cross-shelf transport of Changjiang-derived clays. In contrast, Taiwan-sourced clays, which are transported by the Kuroshio Current, dominated the deposition in the central trough at that time. However, there was an abrupt increase in the sediment input from the Changjiang at ~1.5 ka, reflected in the increase of illite/smectite ratios and decrease of smectite contents seen at that time (Figs. 3, 5). The abrupt increase of the Changjiang-sourced clays since 1.5 ka was probably related to the weakening of the Kuroshio Current (Jian et al., 2000). Moreover, the strengthening East Asian summer monsoon since 1.5 ka accompanied with high precipitation on land (Wang et al., 2005) might be expected to have increased the flux of the Changjiang sediments into the open shelf. Sedimentological studies also revealed that the fluvial sediments escaping from the



Changjiang Delta and estuarine area to the open shelf over the last 2 ka significantly increased (Saito et al., 1998; Li et al., 2003).

It is interesting to note that the source contribution from the ECS shelf has the same downcore trend as the smectite content (Fig. 5). We interpret this relationship to be linked to the hydrodynamic behavior of smectite in the sea. The crystal size of smectite is the smallest in any of clay minerals, so that it is more sensitive to depositional environmental change than other clay minerals, and can be easily winnowed and transported by oceanic currents. The abrupt changes of smectite content at around 15–13 ka correspond well with a rapid sea level rise (Clark et al., 2009). The transportation and deposition of clays in the ECS shelf are closely related to sea level fluctuation. Therefore, the coherency between the smectite content and the clay contribution from the ECS shelf to the Okinawa Trough, may imply that the content of smectite in marine sediments can be a good proxy for the hydrodynamic sedimentary environment.

## 6. Conclusions

The clay mineralogy of sediments recovered by Core DGKS9604 from the central Okinawa Trough was examined, with an aim to identify the sources of fine-grained terrigenous sediments and to decipher paleoenvironmental changes since 28 ka, prior to the LGM. The clay mineral assemblages in the core predominantly consist of illite, with average contents above 60%. Downcore variability of clay mineral contents allow Core DGKS9604 to be divided into three units, Unit 1 (ca. 8.4–0 ka), Unit 2 (ca. 14.0–8.4 ka) and Unit 3 (ca. 28.0–14.0 ka).

Comparison of clays with possible sources indicates that the clays in the central Okinawa Trough during the period of 28.0–14.0 ka were primarily supplied by the

paleo-Changjiang River, and partly by the ECS shelf, given that the shoreline was then at the modern shelf edge, placing the river mouth close to the core site. During the late deglacial period and early Holocene (14.0–8.4 ka) the middle-outer shelf of the ECS was the main source of clays transported to the central trough. Sediments derived from the ECS shelf increased to above 60%, while the Changjiang-derived clays decreased from more than 60% to less than 30% coinciding at ca. 15–13 ka accompanied with an abrupt sea level rise. This suggests that strong sediment reworking and erosion at the shelf edge during that period increased the lateral transport of fine-grained sediments from the ECS shelf to the central trough. Interestingly, we do not link any of the major changes in the clay mineral evolution to intensification in the Asian summer monsoon during the Early Holocene.

Variability of the flow path of the Kuroshio Current might have exerted a significant influence on sediment dispersal and deposition in the ECS after the Early Holocene when it transported large volumes of Taiwan-sourced sediments to the southern Okinawa Trough. Fine-grained Holocene sediments in the central trough predominantly originated from east Taiwan (> 50%), which may be related to the return and strengthening of the main axis of the Kuroshio Current in the trough since the Early Holocene at ca. 8 ka. Furthermore, an abrupt increase in the proportion of Changjiang-sourced clays since 1.5 ka was probably related to the weakening of the Kuroshio Current and increase of fluvial flux in relation to strengthening summer monsoon.

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Table 1

Comparisons of clay mineral assemblages between Core DGKS9604 and potential provenances

Sediments	n	Smectite (%)	Illite (%)	Kaolinite (%)	Chlorite (%)	Ill/Sm	Chl/Kao	References
9604-Unit 1	23	5.3	69.4	4.9	20.5	14.4	4.4	this study
9604-Unit 2	20	8.9	67.4	5.9	17.8	9.4	3.1	this study
9604-Unit 3	45	5.5	71.5	6.4	16.6	18.1	2.7	this study
9603-Unit 1	6	2.9	71.3	6.5	19.4	24.6	3.0	Guo et al., 2000
9603-Unit 2	7	6.4	63.4	9.0	21.3	9.9	2.4	Guo et al., 2000
9603-Unit 3	25	4.9	67.7	10.0	17.4	13.8	1.7	Guo et al., 2000
Changjiang	8	6	66	16	12	11.0	0.8	Yang et al., 2003
	8	6.6	70.8	9.4	13.2	12.1	1.4	Fan et al., 2001
	87	10	65	14	11	6.5	0.8	Yang, 1988
	–	5.5	68	12.7	13.9	12.4	1.1	Xu, 1983
Huanghe	8	12	62	10	16	5.2	1.6	Yang et al., 2003
	14	15.2	62.5	9.7	12.5	4.3	1.3	Fan et al., 2001
	35	16	62	10	12	3.9	1.2	Yang, 1988
	–	23.2	59	8.5	9.3	2.5	1.1	Xu, 1983
Delta front	35	2	58	16	23	29	1.4	Fang et al., 2007
Prodelta	30	3	63	13	20	21	1.5	Fang et al., 2007
East China Sea shelf	62	11.8	59.7	8.9	19.6	5.1	2.2	Li, 1990
	14	8	60.5	7.5	23.9	7.6	3.2	Chen, 1973
	6	5.8	55	13.7	25.4	10.0	1.9	Aoki and Oinuma, 1974
ODP1202B	38	7	67	6.1	19.9	9.9	3.3	Diekmann et al., 2008
East Taiwan Shelf	14	6.4	61.9	5	26.6	9.7	5.3	Chen, 1973
Okinawa Trough	8	5.3	69	6	19.7	17.5	3.3	Aoki and Oinuma, 1974

Note: n=sample numbers; Ill=illite; Sm=smectite; Kao=kaolinite; Chl=chlorite; – means data

unavailable

Figure captions:

Fig. 1

Location map of the East China Sea with the location of Core DGKS9604 and other reference cores. The regional circulation pattern in the East China Sea and adjacent areas are from Huh and Su (1999) and Yu (2006). YSCC: Yellow Sea Coastal Current, YSWC: Yellow Sea Warm Current, CDW: Changjiang Diluting Water, ZFCC: Zhenjiang and Fujian Coastal Current, TWC: Taiwan Warm Current.

Fig. 2

Temporal variability of clay contents and clay mineralogy in the carbonate-free <2 $\mu$ m size fraction of sediments from Core DGKS9604.

Fig. 3

Plot of clay mineral ratios from Core DGKS9604, compared with sediments from the Changjiang and Huanghe Rivers (Xu, 1983; Yang, 1988; Fan et al., 2001; Yang et al., 2003), Changjiang subaqueous delta (Fang et al., 2007), East China Sea shelf (Chen, 1973; Aoki and Oinuma, 1974; Li, 1990), East Taiwan shelf (Chen, 1973) and Holocene samples from ODP Site 1202 in the southern Okinawa Trough (Diekmann et al., 2008).

Fig. 4

Ternary diagram of clay minerals for sediments from Core DGKS9604 compared with those of the Changjiang and Huanghe Rivers (Xu, 1983; Yang, 1988; Fan et al., 2001; Yang et al., 2003), East China Sea shelf (Chen, 1973; Aoki and Oinuma, 1974; Li, 1990), East Taiwan shelf (Chen, 1973) and Holocene sediments from ODP Site 1202B

in the Southern Okinawa Trough (Diekmann et al., 2008).

Fig. 5

Contributions of the three end-member provenances (Changjiang, East China Sea shelf and eastern Taiwan shelf) to clay mineral compositions of Core DGKS9604. The sea-level fluctuations within the East China Sea (Feng, 1983), sedimentation rates (LSR), oxygen isotopes (Yu et al., 2006, 2009), mean grain size (Mz) (Yu et al., 2006), and East Asian monsoon variability (Wang et al., 1999) are also shown for references.

ACCEPTED MANUSCRIPT



Fig. 1

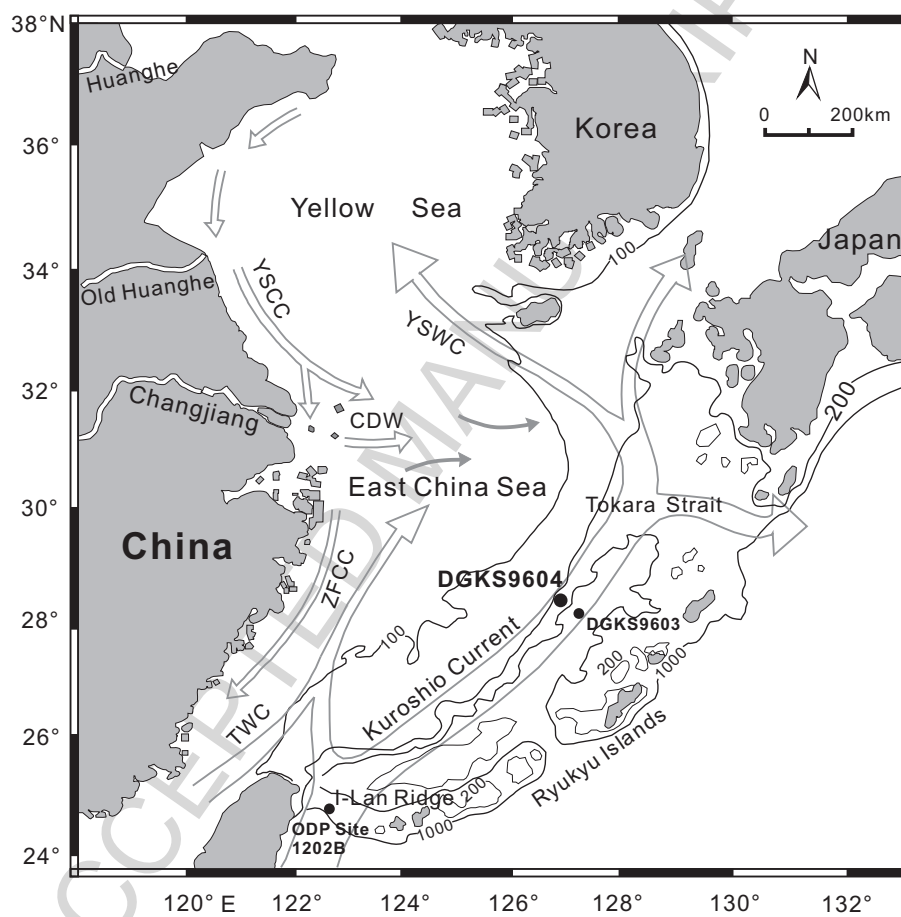


Fig. 2

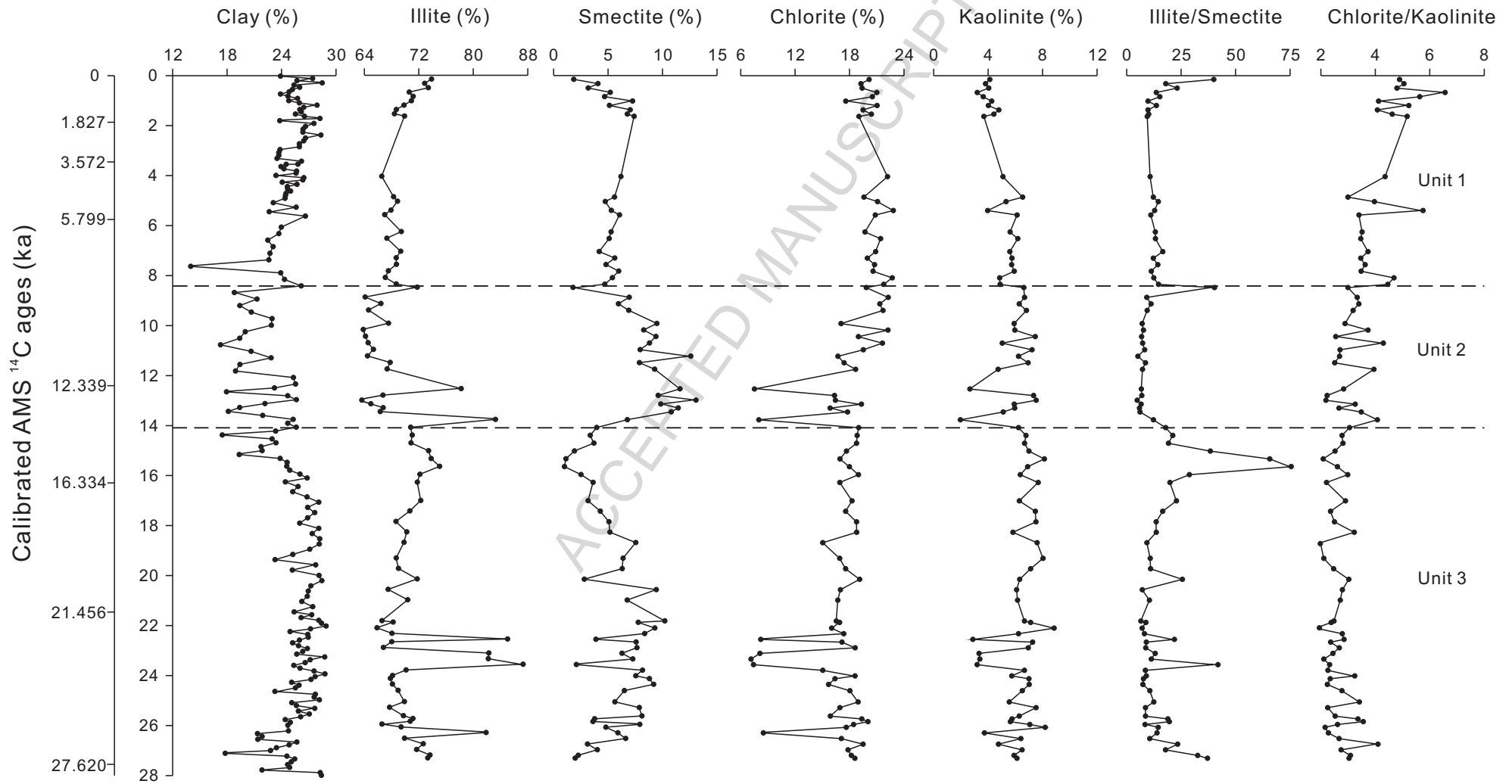


Fig. 3

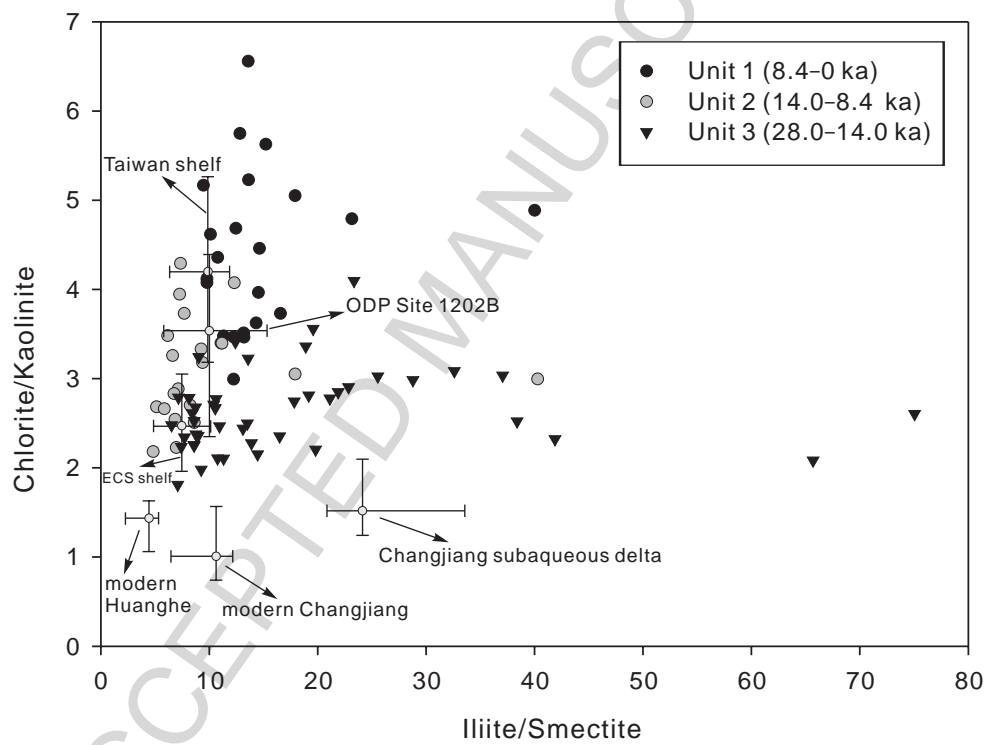


Fig. 4

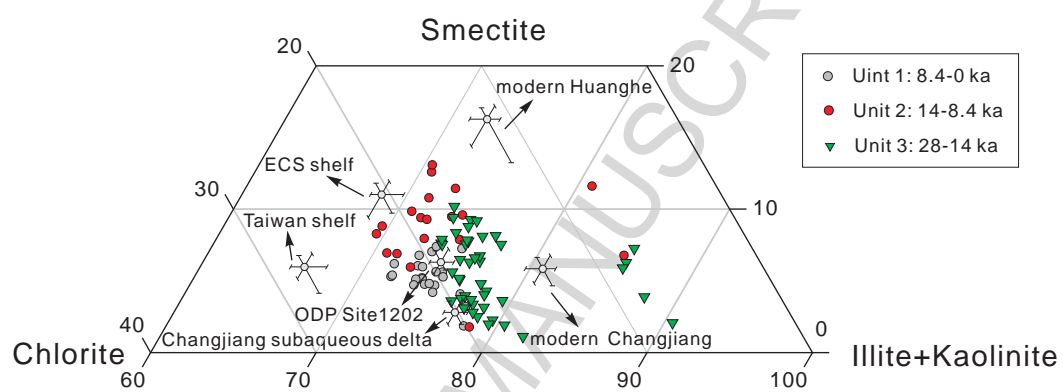


Fig. 5

