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Provenance discrimination of siliciclastic sediments in the middle Okinawa Trough since 30 ka: Constraints from rare earth element compositions

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Abstract:

The late Quaternary paleoceanography and paleoenvironment in the Okinawa Trough, East China Sea, have been well reconstructed over the last decade, while in contrast the provenance of terrigenous sediments that have accumulated there remains enigmatic. In this study, rare earth elements (REE) were used to investigate provenance changes in sediments from Core DGKS9604, taken from the middle Okinawa Trough. Discrimination plots based on REE fractionation parameters suggest that the cored sediments have variable provenances over the last 30 ka, with the lower part (ca. 31–8.2 ka) ultimately originating mostly from the Changjiang (Yangtze River) and the upper part (7.1–0 ka) primarily from Taiwan. During the Last Glacial Maximum and early deglacial period, sea level was low and the main stream of the Kuroshio Current was deflected to the east of the Ryukyu Islands. As a result the Changjiang-derived sediments might have dominated sedimentation of the middle Okinawa Trough. However, since about 7 ka the main stream of the Kuroshio Current strengthened in the area of the trough, as sea level approximated the modern position. This caused near-bottom transport of fine-grained sediments from the continental margin to the trough to become weak and instead, Taiwan-derived terrigenous sediments dominated in the middle trough. The changing provenances of terrigenous sediments into the middle Okinawa Trough are closely related to the evolution of oceanic circulation and sea level in the East China Sea. Two tephra layers in the core have distinct REE compositions and correlate well with two volcanic eruptions at 7.6 and 25.8 ka in southern Japan.

Keywords: sediment provenance; the Okinawa Trough; Kuroshio Current; rare earth element

1. Introduction

The Okinawa Trough is located in the southeast of the East China Sea and is regarded as an incipient intra-continental basin formed behind the Ryukyu arc-trench system (Clift et al., 2003)(Fig. 1). The continuous sedimentation during the late Quaternary in the Okinawa Trough has been regulated by changing terrigenous sediment supply, sea level, oceanic circulation and the intensity of the East Asian monsoon. As a result the sediment distribution, transport and dispersal patterns in the Trough and adjoining shelf are closely related to these complex controlling factors. In view of this, the Okinawa Trough is an ideal natural laboratory in the Western Pacific marginal seas for the studies of late Quaternary land-sea interaction and paleoenvironmental changes.

Over the last two decades, many scientists have attempted to identify the origin of sediment in the middle Okinawa Trough using mineralogical (Chen et al., 1982; Dou et al., 2010), oceanographic and paleoceanographic (Jian et al., 1998, 2000; Ujiié and Ujiié, 1999; Liu et al., 1999, 2000; Liu, 2005), environmental magnetic (Liu et al., 2007), as well as geochemical methods (Zhao and Yan, 1992; Meng et al., 2007). These studies suggested that the terrigenous sediment sources of the Okinawa Trough predominantly include direct supply from major rivers in East Asia, particularly the Changjiang (Yangtze River) and Huanghe (Yellow River) (Katayama and Watanabe, 2003), and lateral transport from the East China Sea shelf through the bottom layers (Honda et al., 2000; Iseki et al., 2003; Oguri et al., 2003). In particular, during the Last Glacial Maximum (LGM) when the sea level was about 120 m lower than in the present day, Changjiang-derived sediments must have directly emptied into the Okinawa Trough (Milliman et al., 1989; Saito et al., 1998; Yoo et al., 2002; Liu et al., 2007; Dou et al., 2010). In addition, the episodic volcanic eruptions in the west Japan volcanic zone have produced widespread fallout tephra layers in the northern Okinawa Trough (Machida, 1999; Miyairi et al., 2004). Other factors such as submarine hydrothermal activity (Zhai et al., 2001), erosion of tectonically active Taiwan Island (Liu et al., 2008), aeolian transport (Tsunogai et al., 1985), seafloor earthquakes (Huh et al., 2004), as well as intrusion of the Kuroshio Current (Jian et al., 1998; Lee et al., 2004) also contribute to the transport of siliciclastic sediments into and within the trough.

Nevertheless, the ultimate origin of sediment in the Okinawa Trough still remains unresolved at present despite many research efforts (Hu et al., 2001; Katayama and Watanabe, 2003). Sedimentation in the middle Okinawa Trough is primarily controlled by Changjiang diluted waters, the Asian winter monsoon winds that originate from the northwest of China, as well as the Kuroshio Current, which is derived from the northern equatorial current. Whether the Changjiang (and/or Huanghe) - derived particulate materials can directly reach the Okinawa Trough in the present day or during the LGM remains controversial (Li and Zhang, 1995; Oguri et al., 2003). It has been documented that the terrigenous sediments may have been laterally transported from the East China Sea shelf to the trough after sea level reached its highstand during the Holocene (Iseki et al., 2003; Liu et al., 2007). However, the Kuroshio Current strengthened and its main stream returned to the Okinawa Trough at about 7 ka (Jian et al., 2000), which might have significantly reduced the sediment transport at the bottom from the continental shelf of the East China Sea to the trough because of a “water barrier” effect (Guo et al., 2001). Whether and how far the terrigenous sediments from Taiwan Island can be transported northward into the middle and northern Okinawa Trough is another challenging question waiting for more lines of evidences (Hsu et al., 2004).

The reliable provenance tracers of terrigenous sediments from different end-members and high-resolution sampling analysis are urgently needed if the sediment transport patterns in and around the Okinawa Trough are to be understood in detail. Geochemical approaches have been proven to be powerful in identifying sediment provenances in the East Asian marginal seas (Clift et al., 2002, 2006; Yang et al., 2004, 2008; Choi et al., 2007; Yan et al., 2007). Among the various methods, rare earth elements (REEs) have been well accepted as reliable provenance tracers because they behave conservatively during sediment formation being largely water-immobile (Taylor and McLennan, 1985). The main research objectives of this paper are to 1) characterize the REE compositions of sediments from Core DGKS9604 taken from the middle Okinawa Trough; 2) identify the provenance changes of siliciclastic sediments since 30 ka; and 3) discuss the competing roles of sea level change, monsoon variability and Kuroshio Current strength and location in regulating the terrigenous sediment inputs into the middle trough.

2. Samples and method

A piston core (DGKS9604) was taken from the middle Okinawa Trough (28°16.64′N, 127°01.43′E) at a water depth of 766 m during the joint Chinese-French DONGHAI Cruise in 1996 (Fig. 1). The core is 1076 cm long and located on the western continental slope of the middle Okinawa Trough. Different from the adjacent core DGKS9603 (28°08.869′N, 127°16.238′E), which was taken during the same cruise (Fig.1), no visual ash layers and hiatuses are present in Core DGKS9604 (Yu et al., 2008, 2009). A high-resolution age model of the core was established on the basis of the oxygen isotopic compositions of *Globigerinoides sacculifer* and accelerator mass spectrometry (AMS) radiocarbon dating of planktonic foraminifera (Liu et al., 2001; Yu et al., 2009). The depositional age at the bottom of the core is estimated to be 37.0 Cal ka, and the bulk sedimentation rate of the core averages about 29 cm/k.y., which is higher than that of Core DGKS9603 (Liu et al., 1999, 2000). Sediments from Core DGKS9604 are primarily composed of clayey silt, with a mean grain size of 6.7±0.4 Φ (Yu et al., 2008; Dou et al., 2010; Fig. 2). The paleoceanography and clay mineralogy of this core have been recently reported elsewhere (Yu et al., 2008, 2009; Dou et al., 2010).

A total of 106 subsamples were collected from Core DGKS9604 at 4 cm intervals for the uppermost 100 cm and at 8 cm intervals between 100 and 743 cm. To separate the residual fractions from the bulk samples, about 0.2 g bulk sediment samples were leached with 20 mL 1 N HCl (hydrochloric acid) for 24 hours at 50ºC. In this study we followed the 1 N HCl–leaching method by Choi et al. (2007). Recent work by Song and Choi (2009) also used this method to leach the river sediments for separating different phases of bulk REE concentrations. The residues of the leached samples were rinsed using deionized water, and then heated to dryness at 50ºC. All the residual samples were combusted in the muffle furnace for two hours at 600 ºC before the acid digestion. About 50 mg powdered samples were digested with 4 ml HNO_3 and 1 ml $HClO_4$ for 24 hours in a tightly closed Teflon vessel on a hot plate at less than 150 °C, heated to dryness, and then digested with a mixture of 4 ml HF and 1 ml $HClO_4$. Afterwards, the solution was evaporated to dryness, and extracted with 10 ml 1% HNO_3. The digestion method for measuring REE concentrations in river sediments was previously reported by Yang et al. (2002). Concentrations of REEs and other elements in the residual fractions were determined by ICP-MS (PQ3, Thermo Elemental) and ICP-AES (IRIS Advantage) in the State Key Laboratory of Marine

Geology at Tongji University. The precision and accuracy were monitored by national geostandards GSR-5, GSR-6, and GSR-9 provided by National Research Center for Geoanalysis. For REE analysis, the differences between the determined and certified values of these geostandards were less than 5%. The leaching efficiency of 1 N HCl was checked by measuring the concentrations of major elements in the leached and residual fractions of GSR-5, GSR-6, and GSR-9. The recoveries of the measured total concentrations were estimated to above 90%.

A total of 188 subsamples were selected for calcium carbonate analysis using an element analyzer (Carlo-Erba model EA1110, Italy). All the samples were pretreated with 1 N HCl to remove calcium carbonate to measure the total organic carbon (TOC) contents of the residual fractions, and then, measure the total carbon (TC) contents of bulk samples. The contents of calcium carbonate were estimated by the TC and TOC contents: $CaCO_3$ (%) = (TC–TOC)×100/12. For monitoring the analytic error, the pure organic compounds including Crystine, Sulphanilamide and Methionine were used as standards, which yielded a precision of about 0.3%.

3. Results and discussions

3.1 REE compositions of the Core DGKS9604 sediments

The average compositions of rare earth elements in the Core DGKS9604 sediments and the reference materials are given in Table 1, and the raw data see the background dataset. Depth profiles of total REE concentrations, REE fractionation parameters, $CaCO_3$ contents and mean grain size are shown in Figure 2. The anomalies of cerium (δCe) and europium (δEu) as two important REE parameters, were calculated by comparing the measured concentrations of Ce and Eu with their neighboring elements: $\delta Ce = Ce_N/\sqrt{[(La_N) \cdot (Pr_N)]}$; $\delta Eu = Eu_N/\sqrt{[(Sm_N) \cdot (Nd_N)]}$, where N represents the normalization of chondrite.

The REE compositions, including total REE concentrations, $(La/Yb)_{UCC}$, $(La/Sm)_{UCC}$, $(Gd/Yb)_{UCC}$, δCe and δEu, exhibit regular variations in the core with a remarkable and abrupt change occurring in the mid-Holocene (8.2–7.1 ka) (Fig. 2). The upper and younger sediments (Unit 1, 0–142 cm and deposited since 7.1 ka) are significantly enriched in REE concentrations and characterized by lower values of $(La/Yb)_{UCC}$, $(La/Sm)_{UCC}$, δCe and δEu compared to the underlying and older sediments (Unit 2, 158–743 cm, deposited during 8.2–30.3 ka). However, $(Gd/Yb)_{UCC}$ ratios, are relatively higher in Unit 1 than in Unit 2. In contrast to the REE compositions, the oxygen isotopic values ($\delta^{18}O$) of foraminifera, $CaCO_3$ contents and mean grain size of the sediments show large variations at the boundary between oxygen isotope stages (OIS) 1 and 2 at about 10 ka (Fig. 2), yet this change does not affect the source of the siliciclastic component.

The upper continental crust (UCC, Taylor and McLennan, 1985)-normalized REE patterns of Core DGKS9604 sediments show significant fractionations of the middle REE (specially Gd and Eu), as shown by higher values of $(Gd/Yb)_{UCC}$ relative to $(La/Yb)_{UCC}$ and $(La/Sm)_{UCC}$ (Table 1; Fig.2). In addition, different UCC-normalized REE patterns occur between Unit 1 and Unit 2, and it is noteworthy that Gd (gadolinium) anomalies apparently occur in the UCC-normalized REE patterns of the core sediments (Figs. 3, 4). Gadolinium anomaly in river sediments was once suggested to be of anthropogenic origin (Bau and Dulski, 1996). However, the Gd anomalies occurring in the modern fluvial sediments of the Changjiang and Huanghe do not reflect anthropogenic origin but indicate natural middle REE fractionation probably caused by specific minerals (Yang et al., 2002).

Overall, Unit 1 shows relative enrichments in total REE and remarkable fractionations of middle and heavy REEs, with obvious convex shapes in the REE plots, whereas Unit 2 is characterized by relatively flat REE patterns, without obvious Ce and Eu anomalies (Fig. 3).

Two layers with much lower than normal values of $(La/Yb)_{UCC}$, $(La/Sm)_{UCC}$ and $(Gd/Yb)_{UCC}$ occur at core depths of 150 cm and 575 cm respectively (Fig. 2). The AMS ^{14}C ages of these two layers are about 7.6 ka and 25.8 ka respectively, which correspond well with two volcanic eruption events documented in southwestern Japan (Kitagawa et al., 1995; Arakawa et al., 1998).

3.2 Controlling factors of REE compositions in the core sediments

Many factors, including bed rock composition in the provenance area, sediment grain size, mineralogy, intensity of chemical weathering, diagenesis and anthropogenic activity are all responsible for REE compositions in sediments. Among these competing processes, sediment provenance is regarded as the most important control over REE compositions (Taylor and McLennan, 1985; Condie, 1991; Yang et al., 2002; Song and Choi, 2009), at least in the case of the present study. It has been well documented that REE are generally enriched in clay and silt fractions, but depleted in sand fractions, because of dilution by quartz and carbonate minerals (McLennan, 1989; Vital et al., 1999). The Core DGKS9604 sediments are primarily composed of clayey silt with an average grain size ranging from 7.2 *Φ* to 6.3 *Φ* (Yu et al., 2008). Poor correlations are observed between mean grain size, REE concentrations and fractionation parameters (Fig. 2), suggesting that the sediment grain size is not an important factor for controlling the REE concentrations in the sediments we analyzed from the middle Okinawa Trough.

In this study, 1 N HCl was used to leach the bulk sediment samples and the residual fractions were separated for measuring REE concentrations. Therefore, we infer that a major part of the mobile fraction, including carbonate, apatite, and Fe-Mn oxides were removed from the bulk core sediments (Yang et al., 2002; 2006; Choi et al., 2007; Song and Choi, 2009), and thus that the measured REE compositions overall represent the contributions of the siliciclastic fraction to the core sediments. Chemical weathering thus exerts a weak influence on the REE compositions in the residual fractions.

Heavy minerals such as zircon, monazite, garnet, allanite and sphene, despite their low abundances in sediments, may account for a considerable fraction of the bulk REE concentrations because of the high REE concentrations in these minerals (Gromet and Silver, 1983; Tayor and McLennan, 1985; McLennan, 1989; Hannigan and Sholkovitz, 2001). However, recent study suggested that major rock-forming and heavy minerals in total contribute less than 20% of the total REE concentrations in the modern Changjiang riverine sediments (Yang et al., 2002). The mean sediment grain size of Core DGKS9604 ranges from 7.2 Φ to 6.3 Φ, smaller than that of the Changjiang sediment (6.3±0.4 Φ), which suggests that heavy minerals are probably not the primary control on REE compositions in the core sediments. Consequently, variations of REE concentrations, as well as fractionation patterns in the siliciclastic sediments, should reflect the bulk mineralogy and so clearly indicate changes of sediment provenance.

3.3 Provenance discrimination of the Core DGKS9604 sediments

Because of the lower contents of biogenic silica from radiolarians and diatoms and authigenic components (Fe–Mn oxides) in the west slope of the Okinawa Trough (Liu, 2005), the residues of 1N HCl leached samples studied in this paper are primarily composed of detrital silicate minerals, which mainly originated from erosion of terrigenous and volcanic sources. The potential provenances of siliciclastic sediments in the middle Okinawa Trough include terrigenous sources supplied via fluvial and eolian inputs, volcanic and hydrothermal activities, and those carried by the oceanic currents such as the Kuroshio Current from the southern Okinawa Trough. It has long been recognized that the terrigenous particulate matters into the middle and north Okinawa Trough are mainly derived from the shelf of the East China Sea where the sediments predominantly originate from the two largest rivers in China, i.e. Changjiang and Huanghe Rivers (Qin et al., 1987; Iseki et al., 2003; Katayama and Watanabe, 2003; Liu et al., 2007; Dou et al., 2010). Further south however Taiwan dominates because it is one of the greatest producers of terrigenous sediment to the ocean known globally (Milliman and Syvitski, 1992).

Compared to the surface seafloor sediments in the East China Sea, Core DGKS9604 sediments have higher REE concentrations, ratios of $(La/Yb)_{UCC}$ and $(La/Sm)_{UCC}$, and lower of $(Gd/Yb)_{UCC}$ (Table 1). Nevertheless, a detailed comparison of REE composition between the sediments from the continental shelf of the East China Sea and from the Core DGKS9604 can not be made because of inadequate data from the East China Sea and different sample pre-treatment methods. Significant differences in REE concentrations and fractionation patterns between Unit 1 and Unit 2 suggest that these depositional units may have different sediment provenances. Unit 2 sediments have REE compositions that are more similar with the modern Changjiang, rather than Huanghe sediments, in terms of their REE concentrations and fractionation parameters (Table 1, Fig 4a). In contrast, Unit 1 sediments have much higher total REE concentrations and different REE fractionation patterns compared to Unit 2 (Table 1; Figs. 2–4). In this study, a discrimination plot of $(La/Sm)_{UCC}$ vs. $(Gd/Yb)_{UCC}$ was used to identify the provenance of sediment in Core DGKS9604 (Fig. 5). The figure clearly demonstrates that the Unit 2 sediments plot together with the

Changjiang sediments, whereas the Unit 1 sediments plot in another group that is close in character to the sediments from southwestern Taiwan (Chen et al., 2007). Therefore, we infer that the older sediments (Unit 2, 158–743 cm, deposited at 8.2–30.3 ka) were derived predominantly from the Changjiang and partly from the Huanghe, while the upper core sediments (Unit 1, 0–142 cm, deposited at 0–7.1 ka) were primarily sourced from Taiwan Island.

Volcanic materials derived from the sea floor or transported from the volcanoes of the Japanese islands also exert significant influence on the deposition in the middle Okinawa Trough, and therefore, can be considered as another potential contributor to the terrigenous detritus (Machida, 1999). Several tephra layers occurring in the sediments of the north Okinawa Trough are mainly composed of volcanic glasses and pumices (Xu and Oda, 1999; Ujiié et al., 2001; Sun et al., 2003). Two abnormal layers deposited at 7.6 ka and 25.8 ka have extraordinarily strong HREE enrichment and LREE depletion (Figs. 3, 4b), very similar to the surrounding volcanic materials which came from the Kyushu islands of Southwestern Japan (Figs. 1, 4b) (Arakawa et al., 1998; Hamasaki, 2002), but much different from the other core sediment and the riverine sediments of the Changjiang and Huanghe (Figs. 3, 4a). This clearly suggests that these two layers with abnormal REE compositions in Core DGKS9604 predominantly consist of volcanic materials. Two volcanic events, known as Kikai-Akahoya (K-Ah) and Aira-Tanzawa (AT), with eruption ages at 7324 Cal yr BP (Kitagawa et al., 1995) and 25,120±270 Cal yr BP respectively (Miyairi et al., 2004), are known in southwest Japan (Fig. 1). They were regarded as the origin of two widely distributed tephra layers in the sediments of the middle and north Okinawa Trough, which predominantly consist of volcanic glasses and pumices (Xu and Oda, 1999; Liu et al., 2000; Ujiié et al., 2001; Sun et al., 2003). The two geochemically abnormal layers in Core DGKS9604 are thus estimated to be dominated by K-Ah tephra and AT tephra, especially considering that they have similar ages.

The average REE concentrations of the volcanic glass (Liu and Meng, 2004) and volcanic rocks around the southwestern Japan Island (Shinjo and Kato, 2000) are 93.7 μg/g and 109.6 μg/g respectively, much lower than those found in this core, or in sediments from the Changjiang and Huanghe Rivers (Table 1). Indeed, the total REE concentrations of these two layers are 174.0 μg/g and 129.0 μg/g respectively, much higher than those of the documented volcanic materials, which implies that these two tephra layers are probably mixed with other terrigenous sediments, which have higher REE concentrations. The discrimination plot suggests that the abnormal layer 1 deposited at 7.6 ka was probably K-Ah tephra mixed with fine-grained terrigenous sediments from Taiwan, while abnormal layer 2, which was deposited at 25.8 ka was formed by mixture between the AT tephra and riverine sediments from mainland China (Fig. 5). In addition, no visual volcanic glasses and pumices were observed in the core sediments (Yu et al., 2008, 2009), further suggesting that the volcanic materials from the surrounding islands might not have dominated the late Quaternary sedimentation in the middle Okinawa Trough.

3.4 Transport pattern of detrital sediments into the middle Okinawa Trough during the last 30 ka

The identification of sediment sources in the Okinawa Trough is of great significance for understanding the depositional history and paleoenvironmental changes of the East China Sea during the late Quaternary. REE concentrations and fractionation patterns strongly suggest that the siliciclastic sediments accumulated in the middle Okinawa Trough during the late Quaternary might originate from different provenances. During the period from late last glaciation (30 ka) to the early-middle Holocene (8.0~7.0 ka) the Changjiang was the primary sediment supplier, whereas during the late Holocene of the last 7 ka Taiwan Island could be the dominant sediment supplier. The provenance discrimination results based on REE compositions are basically similar with the observation of clay mineral assemblages (Dou et al., 2010).

The lowest sea level during the Last Glacial Maximum (LGM) was about 120~135 m lower than that of today (Emery et al., 1971; Qin et al., 1987; Henderson, 2002), which implies that the continental shelf of the East China Sea would have been largely exposed and correspondingly, that the river mouths of the paleo-Changjiang and/or paleo-Huanghe must have been positioned significantly closer to present-day's outer shelf (Fig. 6a). Furthermore, the main stream of the Kuroshio Current deflected to the east of the Ryukyu Islands Arc during the LGM (Ujiié et al., 1991; Ahagon et al., 1993; Xiang et al., 2003) (Fig. 6a). As a result, the terrigenous fine-grained particulate materials from the Changjiang and/or Huanghe might have been transported directly into the middle Okinawa Trough and dominated the deposition there, since there would have been no influence from the Kuroshio Current.

Previous studies suggested that the aeolian input from the loess area in western China into the East Asian marginal seas increased during the LGM (Irino and Tada, 2002; Nagashima et al., 2007). It is well known that the Huanghe sediments have very similar REE compositions with the loess because the latter is the main sediment provider of the Huanghe (Yang et al., 2002). The Unit 2 and Huanghe sediments plot in different groups in the discrimination plot (Fig. 5), suggesting that the Huanghe-derived and aeolian materials did not contribute significantly to the siliciclastic deposition in the middle Okinawa Trough during the LGM.

During the deglacial and early Holocene period, the Changjiang river mouth retreated with the postglacial sea-level rising to its present position at about 6~7 ka (Liu et al., 2007). The main stream of the Kuroshio Current re-entered the Okinawa Trough and/or strengthened at about 7.5–6.0 ka, and the modern oceanic circulation in the East China Sea was finalized at that time (Ujiié et al., 1991; Jian et al., 1998, 2000; Xiang et al., 2003). Since then, sedimentation in the Okinawa Trough has been dominated by the competing processes of the Kuroshio Current and the oceanic circulations in the East China Sea (Lee et al., 2004). Sediment trap experiments revealed that a significant amount of suspended terrigenous particles are transported through the bottom layer from the outer shelf of the East China Sea to the Okinawa Trough (Iseki et al., 2003; Katayama and Watanabe, 2003). Near-bottom transport may be a key process for shelf-to-deep sea export of biogenic/lithogenic particles (Iseki et al., 2003).

The REE compositions of the Unit 1 sediments are remarkably different from the Unit 2 and riverine sediments from mainland China, but very similar with those of Taiwan-derived sediments (Table 1; Figs. 2 and 5). This implies that the siliciclastic sediments deposited since 7 ka in the middle Okinawa Trough were primarily sourced from Taiwan, and probably transported northward by the main stream of the Kuroshio Current, after their initial deposition from the Lanyang River delta and fan. In contrast, the sediment transport through the bottom layer from the outer shelf of the East China Sea to the middle Okinawa Trough during the late Holocene was relatively weak as a result of the blocking effect of the Kuroshio and Taiwan Warm Currents, which act as a barrier deflecting other currents from the area (Fig. 6b). Therefore, the suspended or resuspended fine-grained sediments of the Changjiang and/or Huanghe would not have been able to dominate the sedimentation in the middle Okinawa Trough since the high stand of sea level at ca. 6~7 ka.

The basins offshore northeastern Taiwan experience an extremely energetic sediment transport regime due to the passage of the Kuroshio Current and its interaction with the high rugged bathymetry in the southern trough. Annual loading of riverine suspended particulate matter from northern Taiwan (ca. 2×10^{7} ton yr^{-1}) makes the island an important source for sediments to the subduction margin accretionary wedge (Hung et al., 1999; Dadson et al., 2003). The sediments in the Southern Okinawa Trough have been suggested to be primarily derived from the Lanyang River in northern Taiwan and other eastern Taiwanese rivers, transported by the Kuroshio Current (Hsu et al., 2004; Jian et al., 2000; Jeng et al., 2003; Huh et al., 2004; Lee et al., 2004). Due to the lack of end-member data of Taiwan rivers, in the present study we can not make a detailed and direct comparison of REE composition between the core and Taiwan riverine sediments. Nevertheless, the discrimination plot implies that the Taiwan-derived fine-grained sediments might have contributed considerably to the middle Okinawa Trough since the middle Holocene.

It is interesting to note that the REE compositions of Core DGKS9604 sediments do not vary simultaneously with the oxygen isotopes of foraminifera or with bulk $CaCO_3$ compositions, which show abrupt and large variations at ca. 10 ka (Fig. 2). For example, the oxygen isotopes of foraminifera and bulk $CaCO_3$ show heavier and lower values respectively during the LGM than in the Holocene (Fig. 2) (Jian et al., 2000; Liu et al., 2001). The relatively uniform REE compositions of the Unit 2 sediments suggest that from ca. 30 to 7.0 ka the provenances of the terrigenous sediments of the middle Okinawa Trough remained stable, despite large fluctuations of sea level, monsoon activity and depositional environments as well. In comparison, the oxygen isotopes of foraminifera and bulk $CaCO_3$ compositions in the core sediments that reflect the in-situ paleoenvironment and primary productivity are more sensitive to changing sea level, depositional environments and/or monsoon activity during the late Quaternary. The depositional flux of REE to the Core DGKS9604 sediments varied significantly since 30 ka, shown by higher fluxes at 30–22, 17.8–11.8, 6–4, and 2–0 ka and were lower at 22–17.8, 11.8–6.0, 4–2 ka (Fig. 2). The variable depositional fluxes of REE strongly suggests that the changing supply rates of terrigenous sediments into the middle Okinawa Trough during the late Quaternary, were probably related to the weathering intensity and sediment production in the large drainage basins. However, it is noteworthy that other factors control the REE flux at the core location, most notably the distance of the river mouth from the middle Okinawa Trough and transport processes of fine-grained sediments in the continental margin (Meng et al., 2007). In particular, we note that 11.8–6.9 ka is a period of lower REE flux, yet this time is generally recognized as a period of strengthening summer monsoon rains (Wang et al., 2001; Herzschuh, 2006). If stronger summer monsoon rains were driving stronger continental erosion, as has been seen in South Asia (Clift et al., 2008), then the sedimentation should increase not decrease. Probably, major part of these increased terrigenous sediments resulted from stronger continental erosion was trapped in the continental shelf with rapidly rising sea level during the early postglacial period. Nevertheless, the controls of depositional flux of terrigenous sediments in the middle Okinawa Trough is beyond this study and will not be considered in greater detail in this paper.

4. Conclusions

One hundred and six sub-samples of Core DGKS9604, which spans the past 30 k.y. and comprises clayey silt, were collected from the middle Okinawa Trough for sediment provenance study. Based on REE geochemical characteristics of the residual fractions leached by 1N HCl, we conclude that Core DGKS9604 can be divided into an upper Unit 1 (0–142 cm, <7.1 ka) and Unit 2 (158–743 cm, 8.2–30.3 ka). Total REE concentrations and fractionation parameters, including $(La/Yb)_{UCC}$, $(La/Sm)_{UCC}$, $(Gd/Yb)_{UCC}$, Ce and Eu anomalies, are significantly different between Units 1 and 2, with large and abrupt variations occurring at 8.2–7.1 ka. The UCC-normalized REE patterns of the Unit 2 sediments are very similar with those of the riverine sediments from mainland China, especially from the Changjiang. This observation suggests that the fine-grained terrigenous sediments which accumulated in the middle Okinawa Trough from LGM to the middle Holocene might originate predominantly from the Changjiang. During that period, the main stream of the Kuroshio Current was deflected to the east of the Ryukyu Islands and the sea level remained lower than present day, so that the river mouth of the Changjiang lay at the shelf edge. As a result, terrigenous materials from the Changjiang and perhaps the Huanghe may have been more easily transported into the middle Okinawa Trough.

The REE compositions of the Unit 1 sediments are more similar to Taiwan-derived sediments than to Changjiang sediments, suggesting that the terrigenous sediments deposited since 7 ka primarily came via transport from Taiwan in the south. Since the middle Holocene at ca. 7 ka when sea level reached a high stand and the main stream of the Kuroshio Current returned to the Okinawa Trough, a large quantity of fine-grained terrigenous particulate matters sourced from Taiwan could have been transported northwards to the middle trough. In contrast, the Changjiang sediment has been restricted to the inner shelf since that time.

Two geochemically abnormal layers with depositional ages at 7.6 and 25.8 ka are characterized by distinct REE compositions, and are interpreted to be dominated by Japanese volcanic material from the Kikai-Akahoya and Aira-Tanzawa tephras respectively. However, we argue that these volcanic glasses are mixed with fine-grained terrigenous sediments from Taiwan and mainland China respectively.

The provenances of the terrigenous sediments in the middle Okinawa Trough remained stable from the LGM to the middle Holocene, despite large fluctuations of sea level, monsoon activity and depositional environments. Nevertheless, the large variations of depositional fluxes of REE strongly suggest the complex controls of sediments supply rates into the trough during the late Quaternary. In contrast, the erosional effects of the varying monsoon onshore are of secondary importance.

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Figure captions:

Fig. 1

Schematic map showing the locations of Core DGKS9604 and other reference cores. The regional circulation pattern in the East China Sea and the adjacent areas are sourced from Huh and Su (1999) and Yu et al (2008). YSCC=Yellow Sea Coastal Current; ZFCC=Zhejiang-Fujian Coastal Current; CDW=Changjiang Diluted Water; TWC=Taiwan Warm Current; YSWC=Yellow Sea Warm Current.

Fig. 2

Depth profiles of mean grain size (Mz), $\delta^{18}O$ of foraminifera, $CaCO_3$, and REE fractionation parameters of Core DGKS9604 sediments. The age model, $\delta^{18}O$ and grain size data after Yu et al (2008; 2009). OIS denotes oxygen isotope stage. The lightly shadowed area indicates two tephra layers, i.e. Kikai-Akahoya (K-Ah) and Aira-Tanzawa (AT) (Kitagawa et al., 1995; Arakawa et al., 1998).

Fig. 3

UCC–normalized patterns of Core DGKS9604 sediments. The samples of Unit 1 and Unit 2 show similar fractionation patterns, much different from two samples primarily of volcanic origins.

Fig. 4

Comparisons of REE patterns between the sediments of Core DGKS9604, Changjiang and Huanghe (Yang et al., 2002), and volcanic source materials (Arakawa et al., 1998; Hamasaki, 2002; Liu et al., 2004).

Fig. 5

Discrimination plot of $(Gd/Yb)_{UCC}$ vs $(La/Sm)_{UCC}$ for the sediments of Core DGKS9604. Values of modern Changjiang and Huanghe riverine sediments (Yang et al., 2002), pumice and lavas samples of Aira pyroclastic eruption (Arakawa et al., 1998), volcanic rocks of Okinawa Trough (Shinjo and Kato, 2000), and core sediments of southwestern Taiwan (Chen et al., 2007) are also shown for comparison.

Fig. 6

Schematic diagrams showing the influences of sea level change and oceanic circulation patterns on the terrigenous sediment inputs to the Okinawa Trough and adjoining shelf of the East China Sea during the LGM (a) and the mid-late Holocene (0~ca.7 Cal ka BP; b). The direction of the Kuroshio Current at the LGM is after Ujiié and Ujiié (1999)

Provenance discrimination of siliciclastic sediments in the middle Okinawa Trough since 30 ka: Constraints from rare earth element compositions

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Abstract:

The late Quaternary paleoceanography and paleoenvironment in the Okinawa Trough, East China Sea, have been well reconstructed over the last decade, while in contrast the provenance of terrigenous sediments that have accumulated there remains enigmatic. In this study rare earth elements (REE) were used to reveal provenance changes in sediments from Core DGKS9604, taken from the middle Okinawa Trough. Discrimination plots based on REE fractionation parameters suggest that the cored sediments have variable provenances over the last 30 ka, with the lower part (ca. 31–8.2 ka) ultimately originating mostly from the Changjiang (Yangtze River) and the upper part (0–7.1 ka) primarily from Taiwan. During the Last Glacial Maximum and early Holocene sea level was low and the main stream of the Kuroshio Current was deflected to the east of the Ryukyu Islands. As a result sediment from the Changjiang dominated sedimentation of the middle Okinawa Trough. However, since about 7 ka the main stream of the Kuroshio Current strengthened in the area of the trough, as sea level reached the modern position. This caused near-bottom transport of fine-grained sediments from the continental margin to the trough to become weak and instead, Taiwan-derived terrigenous sediments dominated in the middle trough. The changing provenances of terrigenous sediments into the middle Okinawa Trough are closely related to the evolution of oceanic circulations and sea level in the East China Sea. Two tephra layers in the core have distinct REE compositions and correlate well with two volcanic eruptions at 7.6 and 25.8 ka in southern Japan.

Keywords: Sediment provenance; the Okinawa Trough; Kuroshio Current; Rare earth element

1. Introduction

The Okinawa Trough is located in the southeast of the East China Sea and is regarded as an incipient intra-continental basin formed behind the Ryukyu arc-trench system (Clift et al., 2003)(Fig. 1). The continuous sedimentation during the late Quaternary in the Okinawa Trough has been regulated by changing terrigenous sediment supply, sea level, oceanic circulation and the intensity of the East Asian monsoon. As a result the sediment source to sink patterns in the Okinawa Trough and adjoining shelf are closely related to these complex controlling factors. In view of this, the Okinawa Trough is an idea natural laboratory in the Western Pacific marginal seas for the studies of later Quaternary land-sea interaction and paleoenvironmental changes.

Over the last two decades, many scientists have attempted to identify the origin of sediment in the middle Okinawa Trough using mineralogical (Chen et al., 1982), oceanographic and paleoceanographic (Jian et al., 1998, 2000; Ujiié and Ujiié, 1999; Liu et al., 1999, 2000), as well as geochemical methods (Zhao and Yan, 1992; Guo et al., 2001; Meng et al., 2001). These studies suggested that the terrigenous sediment sources of the Okinawa Trough predominantly include direct supply from major rivers in East Asia, particularly the Changjiang (Yangtze River) and Huanghe (Yellow River) (Katayama and Watanabe, 2003), and lateral transport from the East China Sea shelf through the bottom layers (Honda et al., 2000; Iseki et al, 2003; Oguri et al., 2003). In particular, during the Last Glacial Maximum (LGM) when the sea level was about 135 m lower than present-day, Changjiang-derived sediments must have directly emptied into the Okinawa Trough (Milliman et al., 1989; Saito et al., 1998; Yoo et al., 2002; Liu et al., 2007). In addition, the episodic volcanic eruptions in the west Japan volcanic zone have produced widespread fallout tephra layers in the northern Okinawa Trough (Machida, 1999; Miyairi et al., 2004). Other factors such as submarine hydrothermal activity (Zeng et al., 2000; Zhai et al., 2001, 2007), erosion of tectonically active Taiwan Island (Liu et al., 2008), eolian transport (Tsunogai et al., 1985), seafloor earthquakes (Huh et al., 2004), as well as intrusion of the Kuroshio Current (Jian et al., 1998; Lee et al., 2004) also contribute to the transport of siliciclastic sediments into and within the trough.

Nevertheless, the origin of sediment in the Okinawa Trough still remains unresolved at present despite many research efforts (Hu et al., 2001; Katayama and Watanabe, 2003). Sedimentation in the middle Okinawa Trough is primarily controlled by Changjiang diluted waters, the Asian winter monsoon winds that originate from northwest of China, as well as the Kuroshio Current, which is derived from the northern equatorial current. Whether the Changjiang (and/or Huanghe) derived particulate materials can directly reach the Okinawa Trough in the present day or during the LGM remains controversial (Li and Zhang, 1995; Xia and Liu, 2001). It has been documented that the terrigenous sediments may have been laterally transported from the East China Sea shelf to the trough after sea level reached its highstand during the mid Holocene (Iseki et al., 2003; Liu et al., 2007). However, the Kuroshio Current strengthened and its main stream returned to the Okinawa Trough at about 7 ka (Jian et al., 2000), which significantly reduced the sediment transport at the bottom from the continental shelf of the East China Sea to the trough because of a “water barrier” effect (Guo et al., 2001).

Whether and how far the terrigenous sediments from Taiwan Island can be transported northward into the middle and north Okinawa Trough is another challenging question waiting for more lines of evidences (Hsu et al., 2004). The reliable provenance tracers of terrigenous sediments from different end-members and high-resolution sampling analysis are urgently needed if the sediment transport patterns in and around the Okinawa Trough are to be understood in detail.

Geochemical approaches have been proven to be powerful in identifying sediment provenances in the East Asian marginal seas (Clift et al., 2002, 2006; Yang et al., 2004, 2008; Yan et al., 2007). Among the various methods, rare earth elements (REEs) have been well accepted as reliable provenance tracers because they behave conservatively during sediment formation being largely water-immobile (Taylor and McLennan, 1985; Rollinson, 1993). The main research objectives of this paper are to 1) characterize the REE compositions of sediments from Core DGKS9604 taken from the middle Okinawa Trough; 2) identify the provenance changes of siliciclastic sediments since 30 ka; and 3) discuss the competing roles of sea level change, monsoon variability and Kuroshio Current strength and location in regulating the terrigenous sediment inputs into the middle trough.

2. Samples and methods

A piston core (DGKS9604) was taken from the middle Okinawa Trough (28°16.64′N, 127°01.43′E) at a water depth of 766 m during the joint Chinese-French DONGHAI Cruise in 1996 (Fig. 1). The core is 1076 cm long and located on the western continental slope of the middle Okinawa Trough. In contrast to the adjacent core DGKS9603 (28°08.869′N, 127°16.238′E), which was taken during the same cruise (Fig.1), no visual ash layers and hiatuses are present in Core DGKS9604 (Yu, 2006). A high-resolution age model of the core was established on the basis of the oxygen isotopic compositions of *Globigerinoides sacculifer* and accelerator mass spectrometry (AMS) radiocarbon dating of planktonic foraminifera (Liu et al., 2001;

Yu et al., 2008). The depositional age at the bottom of the core is estimated to be 37.01 Cal ka, and the bulk sedimentation rate of the core averages about 29 cm/k.y., which is higher than that of Core DGKS9603 (Liu et al, 1999, 2000). Sediments from Core DGKS9604 are primarily composed of clayey silt, with a mean grain size of 6.7±0.4 Φ (Yu, et al, 2008; Fig. 2).

A total of 106 subsamples were collected from Core DGKS9604 at 4 cm intervals for the uppermost 100 cm and at 8 cm intervals between 100 and 743 cm. To separate the residual fractions from the bulk samples, 0.2 g bulk sediment samples were leached with 20 ml 1 N HCl for 24 hours at 50ºC. The residues of the leached samples were then completely digested with concentrated $HF–HNO_3–HClO_4$ in an airtight Teflon container. Concentrations of REEs and other elements in the residual fractions were determined by ICP-MS (PQ3, Thermo Elemental) and ICP-AES (IRIS Advantage) in the State Key Laboratory of Marine Geology at Tongji University. The precision and accuracy were monitored by geostandard GSR-5, GSR-6, and GSR-9. Differences between the determined and certified values were less than 5%, and the leaching efficiency checked by GSR-5, GSR-6, and GSR-9 was above 90%.

3. Results and discussions

3.1 REE compositions of the Core DGKS9604 sediments

The average rare earth element compositions of the Core DGKS9604 sediments and the reference materials are given in Table 1. Depth profiles of total REE concentrations, REE fractionation parameters, $CaCO_3$ contents and mean grain size are shown in Figure 2. The REE compositions, including total REE concentrations, $(La/Yb)_{ucc}$, $(La/Sm)_{ucc}$, $(Gd/Yb)_{ucc}$, δCe (Eu anomalies) and δEu (Ce anomalies), exhibit regular variations in the core with a remarkable and abrupt change occurring in the mid-Holocene (8.2–7.1 ka) (Fig. 2). The younger sediments (Unit 1, 0–142 cm and deposited since 7.1 ka) are significantly enriched in REE concentrations and characterized by lower values of $(La/Yb)_{ucc}$, $(La/Sm)_{ucc}$, δCe and δEu compared to the underlying sediments (Unit 2, 158–743 cm, deposited during 8.2–30.3 ka). However, $(Gd/Yb)_{ucc}$ ratios, are relatively higher in Unit 1 than in Unit 2. In contrast to the REE compositions, the oxygen isotopic values ($\delta^{18}O$) of foraminifera, $CaCO_3$ contents and mean grain size of the sediments show large variations at the boundary between oxygen isotope stages (OIS) 1 and 2 at about 10 ka (Fig. 2), yet this change does not affect the source of the siliciclastic component.

The upper continental crust (UCC)-normalized (Taylor and McLennan, 1985) REE patterns of Core DGKS9604 sediments show relative enrichments in total REE and light REEs (LREE) compared to UCC (Table 1), but also exhibit different forms between the two depositional units. Unit 1 shows remarkable fractionations of middle and heavy REEs with obvious convex shapes in the REE plots, whereas Unit 2 is characterized by relatively flat REE patterns, with no obvious Ce and Eu anomalies (Fig. 3).

Two layers with much lower than normal values of $(La/Yb)_{ucc}$, $(La/Sm)_{ucc}$ and $(Gd/Yb)_{ucc}$ occur at core depths of 150 cm and 575 cm respectively (Fig. 2). The AMS ^{14}C ages of these two layers are about 7.6 ka and 25.8 ka respectively, which corresponds well with volcanic eruption events documented in southwestern Japan (Kitagawa et al., 1995; Arakawa et al., 1998).

3.2 Controlling factors of REE compositions in the core sediments

Many factors, including the bed rock composition in the source area, sediment grain size, mineralogy, intensity of chemical weathering, diagenesis and anthropogenic activity are responsible for REE compositions in sediments. Among these competing processes, sediment provenance is regarded as the most important control over REE compositions (Taylor and McLennan, 1985; Condie, 1991; Yang et al., 2002), at least in the case of the present study. It has been well documented that REE are generally enriched in clay and silt fractions, but depleted in sand fractions, because of dilution by quartz and carbonate minerals (McLennan, 1989; Vital and Stattegger, 1999). The Core DGKS9604 sediments are primarily composed of clayey silt with an average grain size ranging from 7.2 Φ to 6.3 Φ (Yu et al., 2006). Poor correlations are observed between mean grain size, REE concentrations and fractionation parameters (Fig. 2), suggesting that the sediment grain size is not an important factor for controlling the REE concentrations in the sediments we analyzed from the middle Okinawa Trough.

In this study, 1 N HCl was used to leach the bulk sediment samples and the residual fractions were analyzed for REE concentrations. Therefore, we infer that a major part of the mobile fraction, including carbonate, apatite, Fe-Mn oxides and organic-bound phases were removed from the bulk core sediments (Yang et al., 2002; 2006), and thus that the measured REE compositions overall represent the contributions of the siliciclastic fraction to the sediments. Chemical weathering thus exerts a weak influence on the REE compositions in the residual fractions.

Heavy minerals such as zircon, monazite, garnet, allanite and sphene, despite their low abundances in sediments, may account for a considerable fraction of the bulk REE concentrations because of the high REE concentrations in these minerals (Gromet and Silver, 1983; Tayor and McLennan, 1985; McLennan, 1989; Hannigan and Sholkovitz, 2001). However, recent study suggested that major rock-forming and heavy minerals in total contribute less than 20% of the total REE concentrations in the modern Changjiang riverine sediments (Yang et al., 2002). The mean sediment grain size of Core DGKS9604 ranges from 7.2 Φ to 6.3 Φ, smaller than that of the Changjiang sediment (6.3±0.4 Φ), which suggests that heavy minerals are probably not the primary control on REE compositions in the core sediments. Consequently, variations of REE concentrations, as well as fractionation patterns in the siliciclastic sediments, should reflect the bulk mineralogy and so clearly indicate changes of sediment provenance.

3.3 Provenance discrimination of the Core DGKS9604 sediments

Because of the lower contents of biogenic (opals, radiolarians and diatoms) and authigenic components (Fe–Mn oxides) in the Okinawa Trough (Liu, 2005), the residues of 1N HCl leached samples studied in this paper are primarily composed of detrital silicate minerals, which mainly originated from erosion of terrigenous and volcanic sources. The potential provenances of siliciclastic sediments in the middle Okinawa Trough include terrigenous sources supplied via fluvial and eolian inputs, volcanic and hydrothermal activities, and those carried by the oceanic currents such as the Kuroshio Current from the southern Okinawa Trough. It has long been recognized that the terrigenous flux into the middle and north Okinawa Trough is mainly derived from the shelf of the East China Sea where the sediments predominantly originate from the two largest rivers in China, i.e. Changjiang and Huanghe Rivers (Qin et al., 1987; Iseki et al., 2003; Katayama and Watanabe, 2003; Liu et al., 2007). Further south however Taiwan dominates because it is one of the greatest producers of sediment to the ocean known globally (Milliman and Syvitski, 1992).

Compared to the surface seafloor sediments in the East China Sea, Core DGKS9604 sediments have higher REE concentrations, ratios of $(La/Yb)_{ucc}$ and

$(La/Sm)_{ucc}$, and lower of $(Gd/Yb)_{ucc}$ (Table 1). Nevertheless, a detailed comparison of REE composition between the sediments from the continental shelf of the East China Sea and from the Core DGKS9604 can not be made because of inadequate data from the East China Sea and different sample pre-treatment methods. Significant differences in REE concentrations and fractionation patterns between Unit 1 and Unit 2 suggest that both depositional units may have different sediment provenances. Unit 2 sediments have REE compositions that are more similar with the modern Changjiang, rather than Huanghe sediments, in terms of their REE concentrations and fractionation parameters (Table 1, Fig 4a). In contrast, Unit 1 sediments have much higher total REE concentrations and different REE fractionation patterns compared to Unit 2 (Table 1; Figs. 2–4). In this study, a discrimination plot of $(La/Sm)_{ucc}$ vs. $(Gd/Yb)_{ucc}$ was used to identify the provenance of sediment in Core DGKS9604 (Fig. 5). The figure clearly demonstrates that the Unit 2 sediments plot together with Changjiang sediments, whereas the Unit 1 sediments plot in another group that is close in character to the sediments from southwestern Taiwan (Chen et al., 2007). Therefore, we infer that the older sediments (Unit 2, 158–743 cm, deposited at 8.2–30.3 ka) were derived predominantly from the Changjiang and partly from the Huanghe, while the upper core sediments (Unit 1, 0–142 cm, deposited at 0–7.1 ka) were primarily sourced from Taiwan Island.

Volcanic materials derived from the sea floor or transported from the volcanoes of the Japanese islands also exert significant influence on the deposition in the middle Okinawa Trough, and therefore, can be considered as another contributor to the terrigenous detritus (Machida, 1999). Several tephra layers occurring in the sediments of the north Okinawa Trough are mainly composed of volcanic glasses and pumices (Xu et al., 1999; Ujiie et al., 2001; Sun et al., 2003). Two abnormal layers deposited at

7.6 ka and 25.8 ka have extraordinarily strong HREE enrichment and LREE depletion (Figs. 3, 4), very similar to the surrounding volcanic materials which came from the Kyushu islands of Southwestern Japan (Fig. 1) (Arakawa et al., 1998; Hamasaki, 2002), but much different from the riverine sediments of the Changjiang and Huanghe. This clearly suggests that these two abnormal layers in the Core DGKS9604 predominantly consist of volcanic materials. Two volcanic events, known as Kikai-Akahoya (K-Ah) and Aira-Tanzawa (AT), with eruption ages at 7324 Cal yr BP (Kitagawa et al., 1995) and 25,120±270 Cal yr BP respectively (Miyairi et al., 2004), are known from southwest Japan (Fig. 1). They were regarded as the origin of two widely-distributed tephra layers in the sediments of the middle and north Okinawa Trough, which predominantly consist of volcanic glasses and pumices (Xu et al., 1999; Liu et al., 2000; Ujiie et al., 2001; Sun et al., 2003). The two geochemically abnormal layers in the Core DGKS9604 are thus estimated to be dominated by K-Ah tephra and AT tephra, especially considering that they have similar ages.

The average REE concentrations of the volcanic glass (Liu et al., 2004) and volcanic rocks around the southwestern Japan Island (Shinjo and Kato, 2000) are 93.7 ug/g and 109.6 ug/g respectively, much lower than those found in this core, or in sediments from the Changjiang and Huanghe Rivers (Table 1). Indeed, the total REE concentrations of these two layers are 174.0 ug/g and 129.0 ug/g respectively, much higher than those of the documented volcanic materials, which implies that these two tephra layers are probably mixed with other terrigenous sediments, which have higher REE concentrations. The discrimination plot suggests that the abnormal layer 1 deposited at 7.6 ka was probably mixed by the K-Ah tephra with fine-grained sediments from Taiwan, while abnormal layer 2, which was deposited at 25.8 ka was formed by mixture between the AT tephra and riverine sediments from mainland

China (Fig. 5).

3.4 Transport pattern of detrital sediments into the middle Okinawa Trough during the last 30 ka

The identification of sediment sources in the Okinawa Trough is of great significance for understanding the depositional history and paleoenvironmental changes of the East China Sea during the late Quaternary. REE concentrations and fractionation patterns strongly suggest that the siliciclastic sediments accumulated in the middle Okinawa Trough during the late Quaternary might originate from different provenances. During the period from late last glaciation (30 ka) to the early-middle Holocene (8.0~7.0 ka) the Changjiang was the primary sediment supplier, whereas during the mid-late Holocene of the last 7 ka Taiwan could be the dominant sediment supplier.

The lowest sea level during the Last Glacial Maximum (LGM) was about 135 m lower than that of today (Emery et al., 1971; Qin et al., 1987; Henderson, 2002), which implies that the continental shelf of the East China Sea would have been largely exposed and correspondingly, that the river mouths of the paleo-Changjiang and/or paleo-Huanghe must have been positioned significantly closer to present-day's outer shelf (Fig. 6a). Furthermore, the main stream of the Kuroshio Current deflected to the east of the Ryukyu Islands Arc during the LGM (Ujiie et al., 1991; Ahagon et al., 1993; Xiang et al., 2003) (Fig. 6a). As a result, the terrigenous fine-grained particulate materials from the Changjiang and/or Huanghe might have been transported directly into the middle Okinawa Trough and dominated the deposition there, since there would have been no influence from the Kuroshio Current.

Previous studies suggested that the eolian input from the loess area in western

China into the East Asian marginal seas increased during the LGM (Irino and Tada, 2002; Nagashima et al., 2007). It is well known that the Huanghe sediments have very similar REE compositions with the loess because the latter is the main sediment provenance of the Huanghe (Yang et al., 2002). The Unit 2 and Huanghe sediments plot in different groups in the discrimination plot (Fig. 5), suggesting that the Huanghe-derived and eolian materials did not contribute significantly to the siliciclastic deposition in the middle Okinawa Trough during the LGM.

During the deglacial period and early Holocene, the Changjiang river mouth retreated with the postglacial sea-level rising to its present position at about 7 ka (Liu et al., 2007). The main stream of the Kuroshio Current re-entered the Okinawa Trough and/or strengthened at about 7.5–6.0 ka (Ujiie et al., 1991; Jian et al., 1998, 2000; Xiang et al., 2003). Since then, sedimentation in the Okinawa Trough has been dominated by the competing processes of the Kuroshio Current and the oceanic circulations in the East China Sea (Yang et al., 1992; Lee et al., 2004). Sediment trap experiments revealed that a significant amount of suspended terrigenous particles are transported through the bottom layer from the outer shelf of the East China Sea to the Okinawa Trough (Iseki et al., 2003; Katayama and Watanabe, 2003; Pang and Wang, 2004). Near-bottom transport may be a key process for shelf-to-deep sea export of biogenic/lithogenic particles (Iseki et al., 2003).

The REE compositions of the Unit 1 sediments are remarkably different from the Unit 2 and riverine sediments from mainland China, but very similar with those of Taiwan-derived sediments (Table 1; Figs. 2 and 5). This implies that the siliciclastic sediments deposited since 7 ka in the middle Okinawa Trough were primarily sourced from Taiwan, and probably transported northward by the main stream of the Kuroshio Current, after their initial deposition from the Lanyang River delta and fan. In contrast, the sediment transport through the bottom layer from the outer shelf of the East China Sea to the middle Okinawa Trough during the mid-late Holocene was relatively weak as a result of the blocking effect of the Kuroshio and Taiwan Warm Currents, which act as a barrier deflecting other currents from the area (Fig. 6b). Therefore, the suspended or resuspended fine-grained sediments of the Changjiang and/or Huanghe would not have been able to dominate the sedimentation in the middle Okinawa Trough since the high stand of sea level at ca. 7 ka.

The basins offshore northeastern Taiwan experience an extremely energetic sediment transport regime due to the passage of the Kuroshio Current and its interaction with the high rugged bathymetry in the southern trough. Annual loading of riverine suspended particulate matter from northern Taiwan (ca. 2×10^7 ton yr^{-1}) makes the island an important source for sediments to the subduction margin accretionary wedge (Hung et al., 1999; Dadson et al., 2003). The sediments in the Southern Okinawa Trough has been suggested to be primarily derived from the Lanyang River in northern Taiwan and other eastern Taiwanese rivers, transported by the Kuroshio Current (Hsu et al., 2004; Jian et al., 2000; Jeng et al., 2003; Huh et al., 2004; Lee et al., 2004).

It is interesting to note that the REE compositions of Core DGKS9604 sediments do not vary simultaneously with the oxygen isotopes of foraminifera or with bulk $CaCO_3$ compositions, which show abrupt and large variations at ca. 10 ka (Figs. 2). For example, the oxygen isotope of foraminifera and bulk $CaCO_3$ show heavier and lower values respectively during the LGM than in the Holocene (Fig. 2) (Jian et al., 2000; Liu et al., 2001). The relatively uniform REE compositions of the Unit 2 sediments suggest that from ca. 30 to 7.0 ka the provenances of the terrigenous sediments of the middle Okinawa Trough remained stable, despite large fluctuations of sea level, monsoon activity and depositional environments as well. In comparison, the oxygen isotope of foraminifera and bulk $CaCO_3$ compositions in the core sediments that reflect the in-situ paleoenvironment and primary productivity are more sensitive to changing sea level, depositional environments and/or monsoon activity during the late Quaternary. The depositional flux of REE to the Core DGKS9604 sediments varied significantly since 30 ka, shown by higher fluxes at 30–22, 17.8–11.8, 6–4, and 2–0 ka and were lower at 22–17.8, 11.8–6.0, 4–2 ka (Fig. 2). The variable depositional fluxes of REE strongly suggests that the changing supply rates of terrigenous sediments into the middle Okinawa Trough during the late Quaternary, were probably related to the weathering intensity and sediment production in the large drainage basins. However, it is also clear that other factors control the REE flux at the core location, most notably the distance of the river mouth from the middle Okinawa Trough and transport processes of fine-grained sediments in the continental margin (Guo et al., 2001; Xiong and Liu, 2004; Meng, 2007). In particular, we note that 11.8–6.9 ka is a period of lower REE flux, yet this time is generally recognized as a period of strengthening summer monsoon rains (Wang et al., 2001; Herzschuh, 2006). If stronger summer monsoon rains were driving stronger continental erosion, as has been seen in South Asia (Clift et al., 2008), then the sedimentation should increase not decrease. Probably, major part of these increased terrigenous sediments resulted from stronger continental erosion was trapped in the continental shelf with rapidly rising sea level during the early postglacial period. Nevertheless, the controls of depositional flux of terrigenous sediments in the middle Okinawa Trough is beyond this study and will not considered in greater detail in this paper.

4. Conclusions

One hundred and six sub-samples of Core DGKS9604, which spans the past 30 k.y. and comprises clayey silt, were collected from the middle Okinawa Trough for sediment provenance study. Based on REE geochemical characteristics of the residual fractions leached by 1N HCl, we conclude that Core DGKS9604 can be divided into an upper Unit 1 (0–142 cm, <7.1 ka) and Unit 2 (158–743 cm, 8.2–30.3 ka). Total REE concentrations and fractionation parameters, including $(La/Yb)_{ucc}$, $(La/Sm)_{ucc}$, $(Gd/Yb)_{ucc}$, Ce and Eu anomalies, are significantly different between Units 1 and 2, with large and abrupt variations occurring at 8.2–7.1 ka. The UCC-normalized REE patterns of the Unit 2 sediments are very similar compared to those of the riverine sediments from mainland China, especially from the Changjiang. This observation suggests that the terrigenous fine-grained sediments which accumulated in the middle Okinawa Trough from LGM to the middle Holocene might originate predominantly from the Changjiang. During that period, the main stream of the Kuroshio Current was deflected to the east of the Ryukyu Islands and the sea level remained lower than present day, so that the river mouth of the Changjiang lay at the shelf edge. As a result terrigenous materials from the Changjiang and perhaps the Huanghe may have been more easily transported into the middle Okinawa Trough.

The REE compositions of the Unit 1 sediments are more similar to Taiwan-derived sediments than to Changjiang sediments, suggesting that the terrigenous sediments deposited since 7 ka primarily came via transport from Taiwan in the south. Since the middle Holocene when sea level reached a high stand and the main stream of the Kuroshio Current returned to the Okinawa Trough, a large quantity of fine-grained terrigenous particulate matters sourced from Taiwan could have been transported northwards to the middle trough. In contrast, the Changjiang sediment has been restricted to the inner shelf since that time.

Two geochemically abnormal layers with depositional ages at 7.6 and 25.8 ka are characterized by distinct REE compositions, and are interpreted to be dominated by Japanese volcanic material from the Kikai-Akahoya and Aira-Tanzawa tephras respectively. However, we argue that these volcanic glasses are mixed with fine-grained terrigenous sediments from mainland China and Taiwan.

The provenances of the terrigenous sediments in the middle Okinawa Trough remained stable from the LGM to the middle Holocene, despite large fluctuations of sea level, monsoon activity and depositional environments. Nevertheless, the large variations of depositional fluxes of REE strongly suggest the complex controls of sediments supply rates into the trough during the late Quaternary. The erosional effects of the varying monsoon onshore are in contrast of secondary importance.

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Figure captions:

Fig. 1

Schematic map showing the locations of Core DGKS9604 and other reference cores. The regional circulation pattern in the East China Sea and the adjacent areas are sourced from Huh and Su (1999) and Yu (2006). YSCC=Yellow Sea Coastal Current; ZFCC=Zhejiang-Fujian Coastal Current; CDW=Changjiang Diluted Water; TWC=Taiwan Warm Current; YSWC=Yellow Sea Warm Current.

Fig. 2

Depth profiles of mean grain size (Mz), $\delta^{18}O$ of foraminifera, $CaCO_3$, and REE fractionation parameters of Core DGKS9604 sediments. The age model, $\delta^{18}O$ and grain size data after Yu et al. (2006, 2008). OIS denotes oxygen isotope stage. The lightly shadowed area indicates two tephra layers, i.e. Kikai-Akahoya (K-Ah) and Aira-Tanzawa (AT) (Kitagawa et al., 1995; Arakawa et al., 1998).

Fig. 3

UCC–normalized patterns of Core DGKS9604 sediments. The samples of Unit 1 and Unit 2 show similar fractionation patterns respectively, much different from two samples primarily of volcanic origins.

Fig. 4

Comparisons of REE patterns between the sediments of Core DGKS9604, Changjiang and Huanghe (Yang et al., 2002), and volcanic source materials (Arakawa et al., 1998; Hamasaki, 2002; Liu et al., 2004).

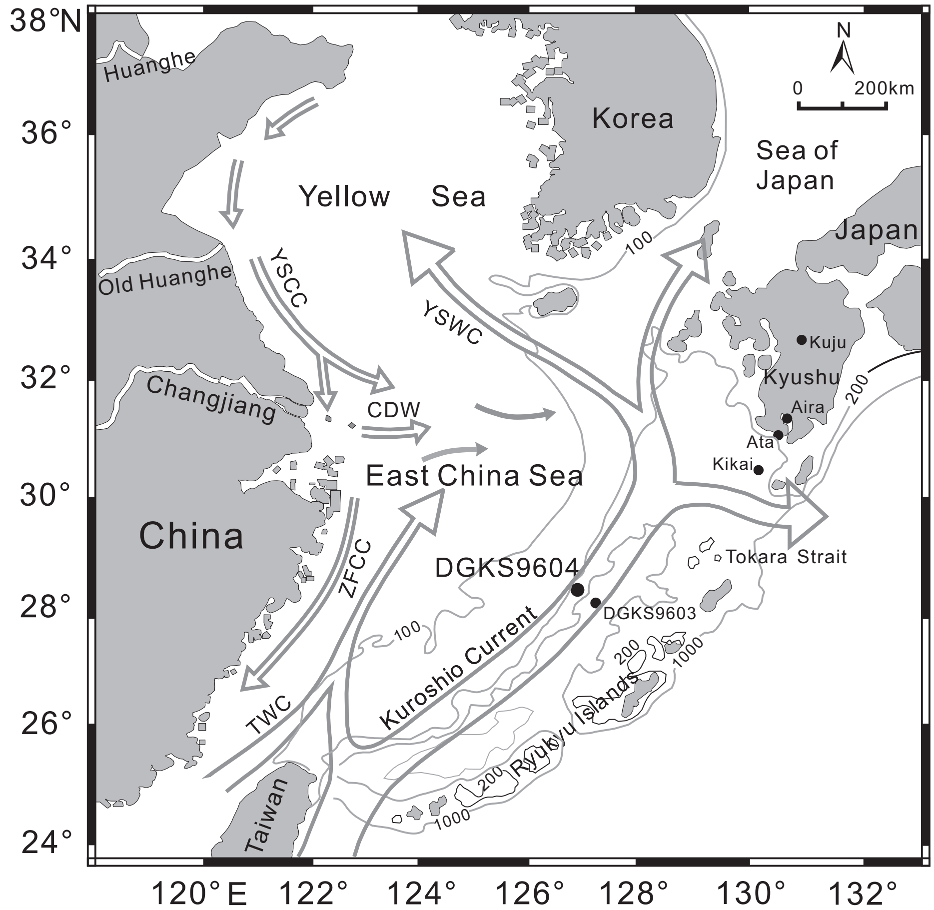
Fig. 5

Discrimination plot of $(Gd/Yb)_{ucc}$ vs $(La/Sm)_{ucc}$ for the sediments of Core DGKS9604. Values of modern Changjiang and Huanghe riverine sediments (Yang et al, 2002), pumice and lavas samples of Aira pyroclastic eruption (Arakawa et al, 1998), volcanic rocks of Okinawa Trough (Shinjo and Kato, 2000), and core sediments of southwestern Taiwan (Chen et al., 2007) are also shown for comparison.

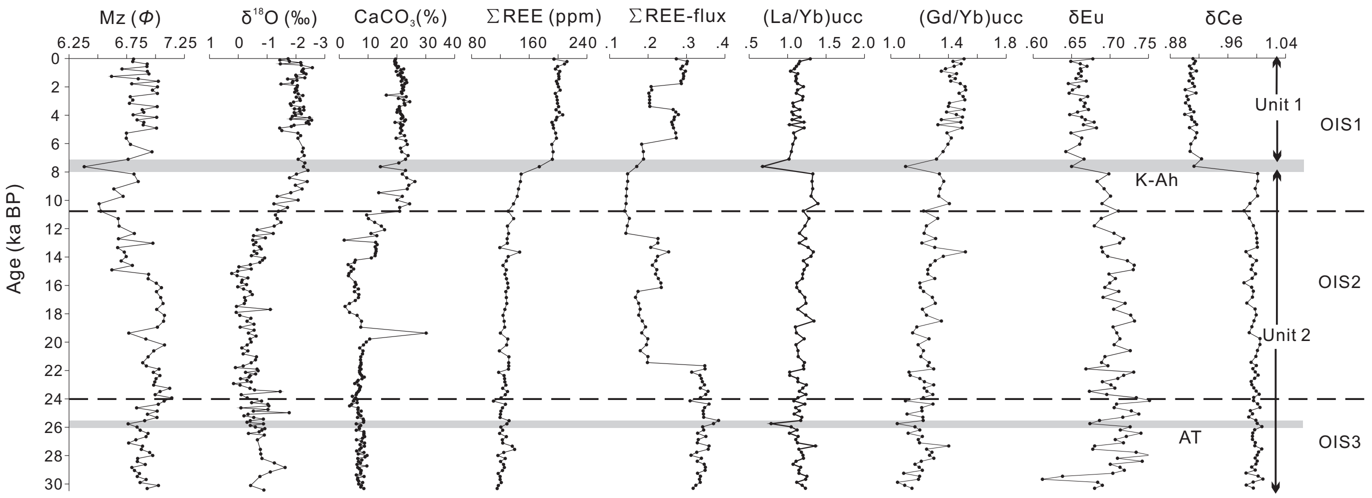
Fig. 6

Schematic diagrams showing the influences of sea level change and oceanic circulation patterns on the terrigenous sediment inputs to the Okinawa Trough and adjoining shelf of the East China Sea during the LGM (a) and the mid-late Holocene (0~ca.7 Cal ka BP; b). The direction of the Kuroshio Current at the LGM is after Ujiié and Ujiié (1999).

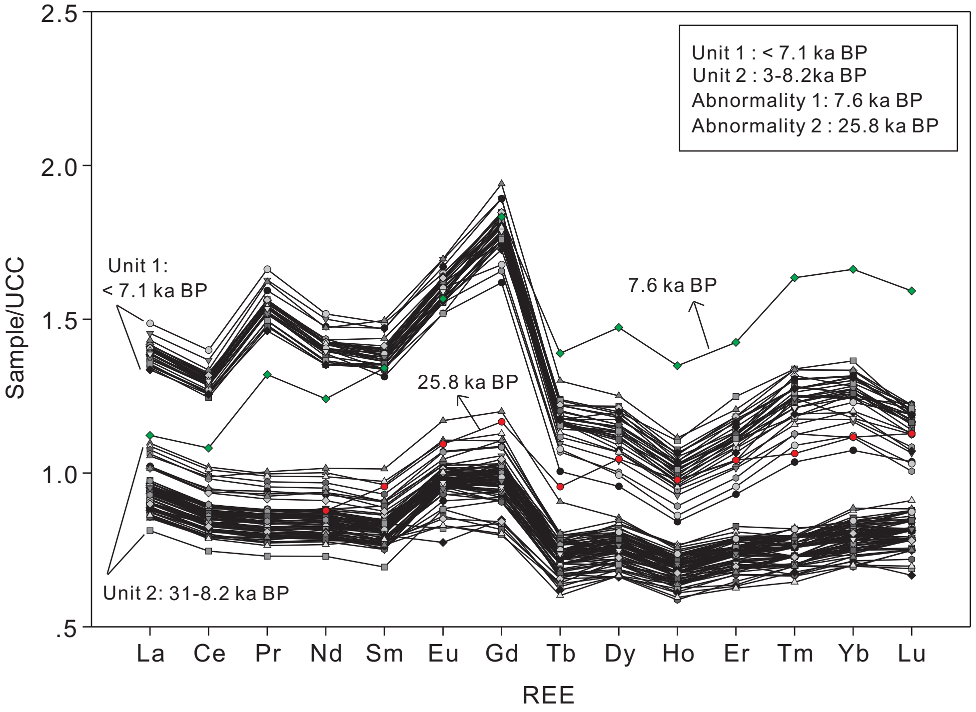
revised Figure 1



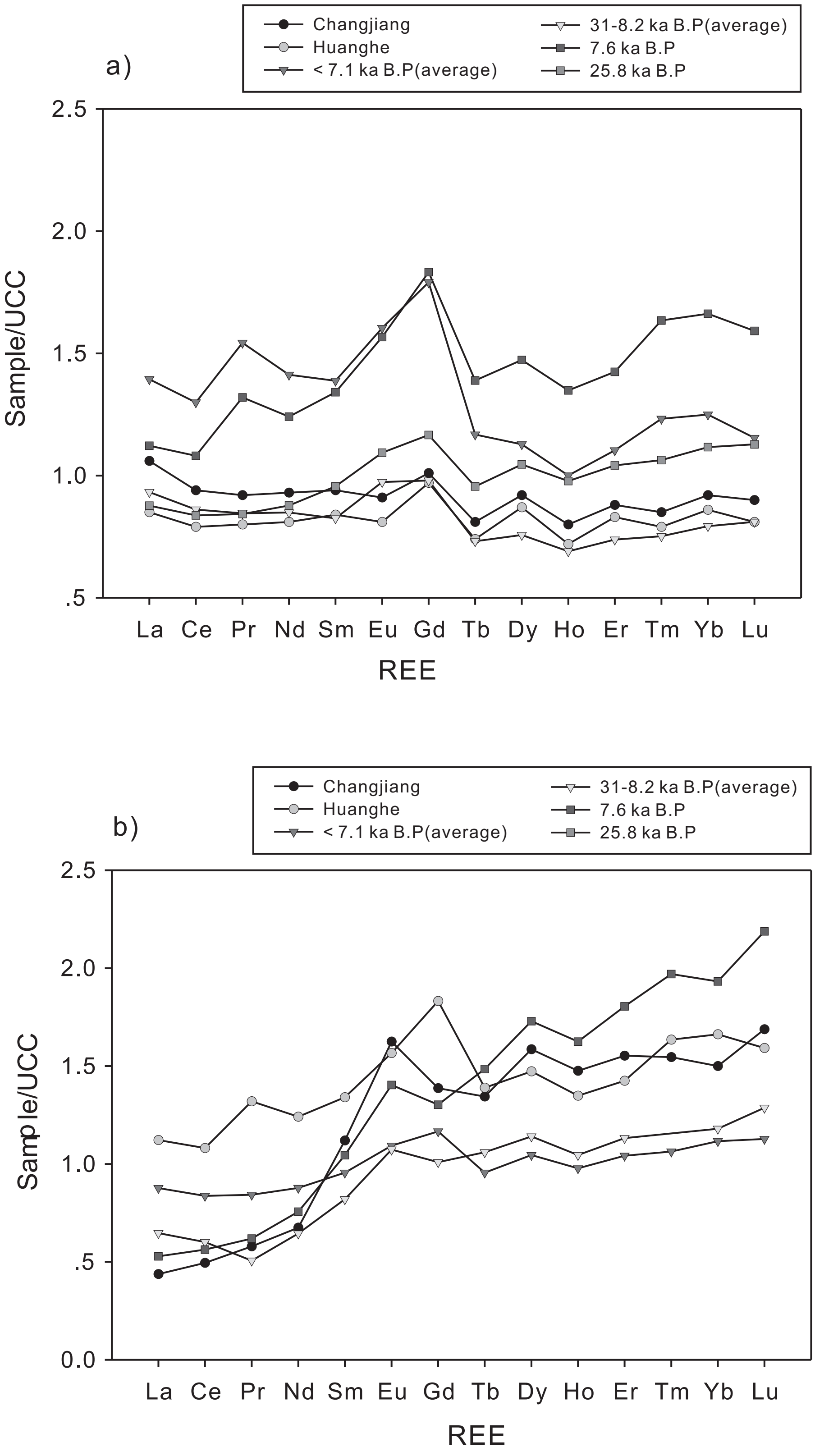
revised Figure 2



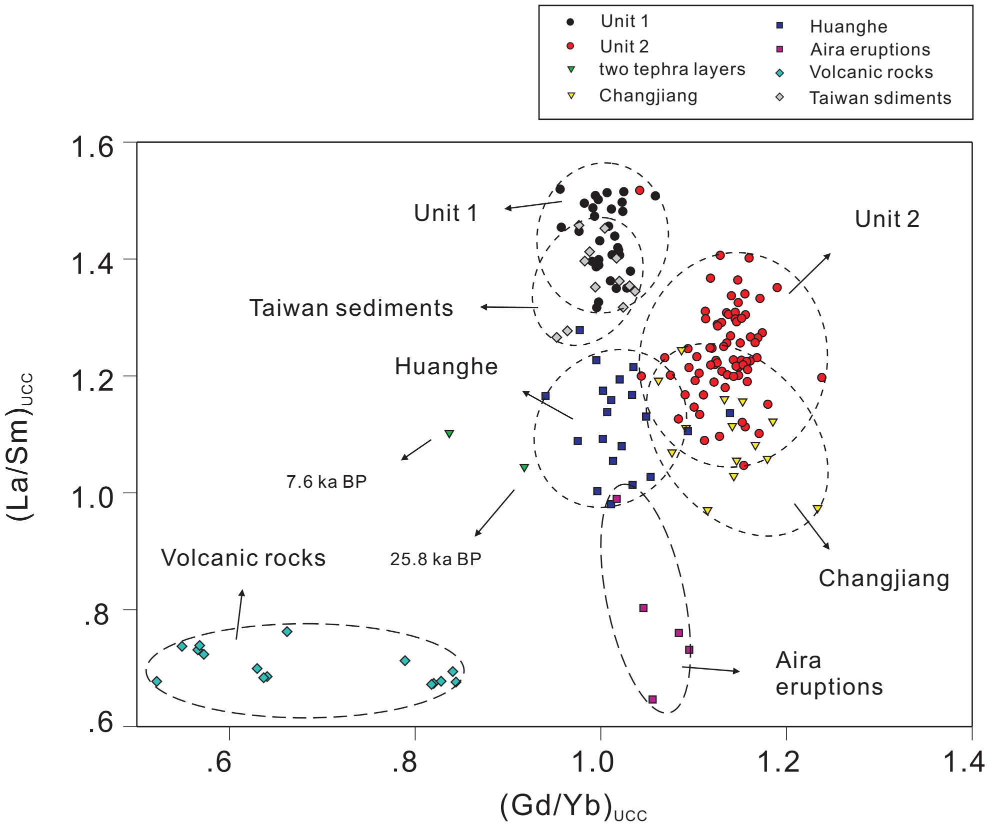
revised Figure 3



revised Figure 4



revised Figure 5



revised Figure 6

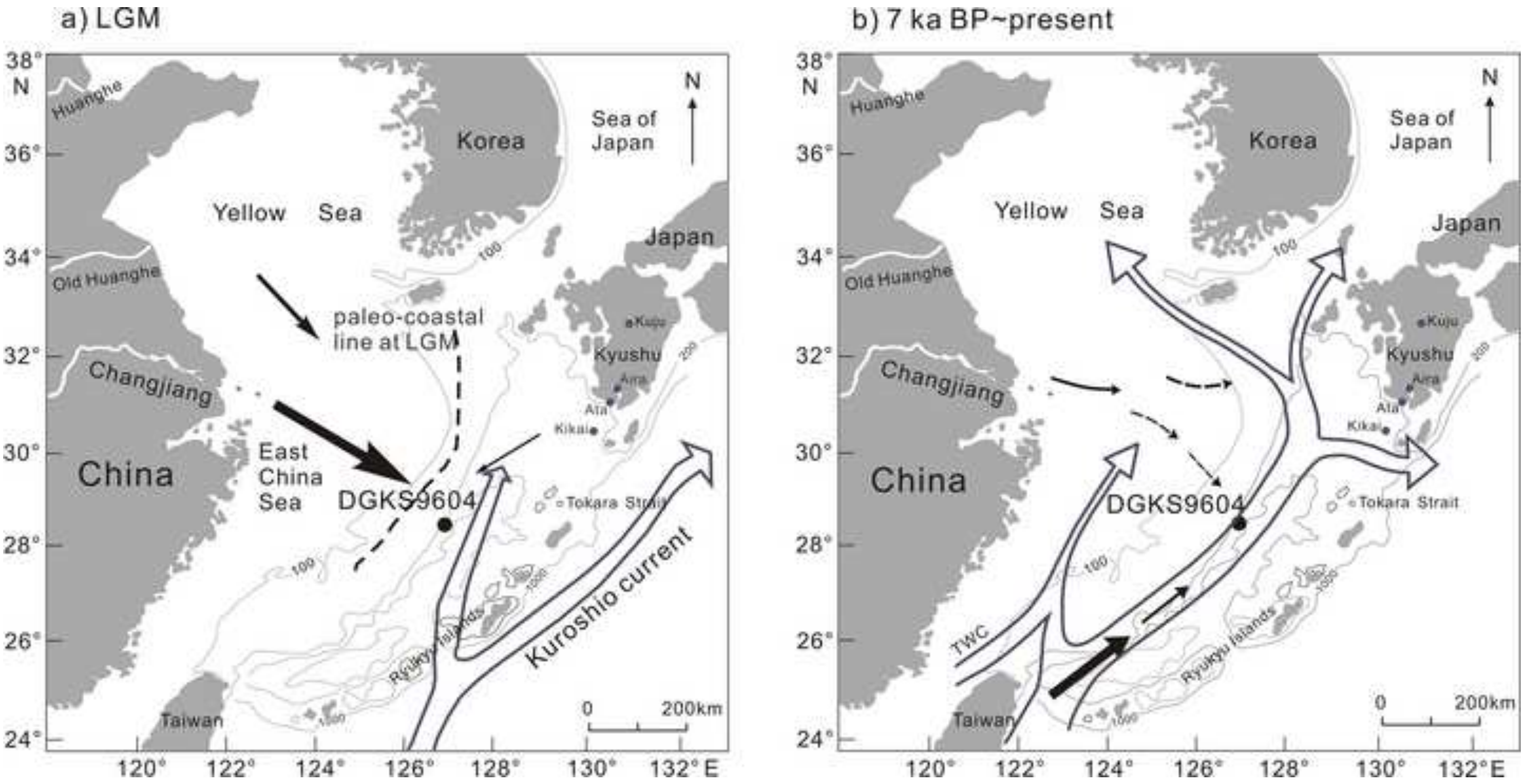


Table 1

Comparisons of rare earth element compositions in Core DGKS9604 sediments with those of upper continental crust (UCC, Taylor and McLennan, 1985), East China Sea sediments (ECS, Zhao et al., 1990), core sediments of southwestern Taiwan (Chen et al., 2007), residual fractions of the Changjiang and Huanghe riverine sediments (Yang et al., 2002), and volcanic rocks of Okinawa Trough (Shinjo and Kato, 2000).

Samples	Core depth (cm)	Age (ka BP)	∑REE (ppm)	δEu	δCe	$(La/Yb)_{UCC}$	$(Gd/Yb)_{UCC}$	$(La/Sm)_{UCC}$
Unit 1	0–142	0–7.1	198.6±4.8	0.66±0.01	0.91±0.01	1.12	1.44	1.01
Unit 2	158–743	8.2–30.3	127.0±9.3	0.71±0.02	1.00±0.01	1.19	1.24	1.14
Abnormality 1	150	7.6	174.0	0.65	0.91	0.68	1.10	0.84
Abnormality 2	575	26.8	129.0	0.67	1.00	0.79	1.04	0.92
Whole core	0–743	0–30.3	148.4±33.5	0.69±0.03	0.97±0.04	1.16	1.29	1.10
UCC	–	–	146.4	0.65	1.03	1.00	1.00	1.00
ECS	Sea floor	modern	120.2	0.60	1.03	0.99	1.56	0.75
Taiwan sediments	0–2340	–	–	0.65	–	1.41	1.33	0.96
Huanghe	Floodplain	modern	119.4	0.60	0.98	0.98	1.12	1.02
Changjiang	Floodplain	modern	140.6	0.61	0.98	1.15	1.10	1.14
Volcanic rocks	–	–	109.6	0.76	0.99	0.37	0.74	0.62

Note: ∑REE denotes total REE concentrations (ppm) ± 1 standard deviation; δEu and δCe are Eu and Ce anomalies respectively, and see the text for the calculation. $(La/Yb)_{UCC}$, $(Gd/Yb)_{UCC}$ and $(La/Sm)_{UCC}$ refer to UCC-normalized REE fractionation parameters. Note that the REE data of the Huanghe and Changjiang refer to the leached residues while those of UCC, ECS, Taiwan sediments and volcanic rocks mean bulk compositions.