
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

49

50 **1. Introduction**

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

61 Over the last two decades, many scientists have attempted to identify the origin
62 of sediment in the middle Okinawa Trough using mineralogical (Chen et al., 1982;
63 Dou et al., 2010), oceanographic and paleoceanographic (Jian et al., 1998, 2000; Ujiie
64 and Ujiie, 1999; Liu et al., 1999, 2000; Liu, 2005), **environmental magnetic** (Liu et al.,
65 2007), as well as geochemical methods (Zhao and Yan, 1992; Meng et al., 2007).
66 These studies suggested that the terrigenous sediment sources of the Okinawa Trough
67 predominantly include direct supply from major rivers in East Asia, particularly the
68 Changjiang (Yangtze River) and Huanghe (Yellow River) (Katayama and Watanabe,
69 2003), and lateral transport from the East China Sea shelf through the bottom layers
70 (Honda et al., 2000; Iseki et al., 2003; Oguri et al., 2003). In particular, during the
71 Last Glacial Maximum (LGM) when the sea level was about **120 m** lower than **in the**
72 **present day**, Changjiang-derived sediments must have directly emptied into the
73 Okinawa Trough (Milliman et al., 1989; Saito et al., 1998; Yoo et al., 2002; Liu et al.,

74 2007; Dou et al., 2010). In addition, the episodic volcanic eruptions in the west Japan
75 volcanic zone have produced widespread fallout tephra layers in the northern
76 Okinawa Trough (Machida, 1999; Miyairi et al., 2004). Other factors such as
77 submarine hydrothermal activity (Zhai et al., 2001), erosion of tectonically active
78 Taiwan Island (Liu et al., 2008), aeolian transport (Tsunogai et al., 1985), seafloor
79 earthquakes (Huh et al., 2004), as well as intrusion of the Kuroshio Current (Jian et al.,
80 1998; Lee et al., 2004) also contribute to the transport of siliciclastic sediments into
81 and within the trough.

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

99 question waiting for more lines of evidences (Hsu et al., 2004).

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

114

115 **2. Samples and method**

116 A piston core (DGKS9604) was taken from the middle Okinawa Trough
117 (28°16.64'N, 127°01.43'E) at a water depth of 766 m during the joint Chinese-French
118 DONGHAI Cruise in 1996 (Fig. 1). The core is 1076 cm long and located on the
119 western continental slope of the middle Okinawa Trough. Different from the adjacent
120 core DGKS9603 (28°08.869'N, 127°16.238'E), which was taken during the same
121 cruise (Fig.1), no visual ash layers and hiatuses are present in Core DGKS9604 (Yu et
122 al., 2008, 2009). A high-resolution age model of the core was established on the basis
123 of the oxygen isotopic compositions of *Globigerinoides sacculifer* and accelerator

124 mass spectrometry (AMS) radiocarbon dating of planktonic foraminifera (Liu et al.,
125 2001; Yu et al., 2009). The depositional age at the bottom of the core is estimated to
126 be 37.0 Cal ka, and the bulk sedimentation rate of the core averages about 29 cm/k.y.,
127 which is higher than that of Core DGKS9603 (Liu et al., 1999, 2000). Sediments from
128 Core DGKS9604 are primarily composed of clayey silt, with a mean grain size of
129 $6.7\pm 0.4 \Phi$ (Yu et al., 2008; Dou et al., 2010; Fig. 2). The paleoceanography and clay
130 mineralogy of this core have been recently reported elsewhere (Yu et al., 2008, 2009;
131 Dou et al., 2010).

132 A total of 106 subsamples were collected from Core DGKS9604 at 4 cm intervals
133 for the uppermost 100 cm and at 8 cm intervals between 100 and 743 cm. To separate
134 the residual fractions from the bulk samples, about 0.2 g bulk sediment samples were
135 leached with 20 mL 1 N HCl (hydrochloric acid) for 24 hours at 50°C. In this study
136 we followed the 1 N HCl-leaching method by Choi et al. (2007). Recent work by
137 Song and Choi (2009) also used this method to leach the river sediments for
138 separating different phases of bulk REE concentrations. The residues of the leached
139 samples were rinsed using deionized water, and then heated to dryness at 50°C. All the
140 residual samples were combusted in the muffle furnace for two hours at 600 °C before
141 the acid digestion. About 50 mg powdered samples were digested with 4 ml HNO₃
142 and 1 ml HClO₄ for 24 hours in a tightly closed Teflon vessel on a hot plate at less
143 than 150 °C, heated to dryness, and then digested with a mixture of 4 ml HF and 1 ml
144 HClO₄. Afterwards, the solution was evaporated to dryness, and extracted with 10 ml
145 1% HNO₃. The digestion method for measuring REE concentrations in river
146 sediments was previously reported by Yang et al. (2002). Concentrations of REEs and
147 other elements in the residual fractions were determined by ICP-MS (PQ3, Thermo
148 Elemental) and ICP-AES (IRIS Advantage) in the State Key Laboratory of Marine

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

156 A total of 188 subsamples were selected for calcium carbonate analysis using an
157 element analyzer (Carlo-Erba model EA1110, Italy). All the samples were pretreated
158 with 1 N HCl to remove calcium carbonate to measure the total organic carbon (TOC)
159 contents of the residual fractions, and then, measure the total carbon (TC) contents of
160 bulk samples. The contents of calcium carbonate were estimated by the TC and TOC
161 contents: $\text{CaCO}_3 (\%) = (\text{TC} - \text{TOC}) \times 100 / 12$. For monitoring the analytic error, the pure
162 organic compounds including Crystine, Sulphanilamide and Methionine were used as
163 standards, which yielded a precision of about 0.3%.

164 **3. Results and discussions**

165 *3.1 REE compositions of the Core DGKS9604 sediments*

166 The average compositions of rare earth elements in the Core DGKS9604
167 sediments and the reference materials are given in Table 1, and the raw data see the
168 background dataset. Depth profiles of total REE concentrations, REE fractionation
169 parameters, CaCO_3 contents and mean grain size are shown in Figure 2. The
170 anomalies of cerium (δCe) and europium (δEu) as two important REE parameters,
171 were calculated by comparing the measured concentrations of Ce and Eu with their
172 neighboring elements: $\delta\text{Ce} = \text{Ce}_N / \sqrt{[(\text{La}_N) \cdot (\text{Pr}_N)]}$; $\delta\text{Eu} = \text{Eu}_N / \sqrt{[(\text{Sm}_N) \cdot (\text{Nd}_N)]}$, where N
173 represents the normalization of chondrite.

174 The REE compositions, including total REE concentrations, $(\text{La}/\text{Yb})_{\text{UCC}}$,
175 $(\text{La}/\text{Sm})_{\text{UCC}}$, $(\text{Gd}/\text{Yb})_{\text{UCC}}$, δCe and δEu , exhibit regular variations in the core with a
176 remarkable and abrupt change occurring in the mid-Holocene (8.2–7.1 ka) (Fig. 2).
177 The **upper and** younger sediments (Unit 1, 0–142 cm and deposited since 7.1 ka) are
178 significantly enriched in REE concentrations and characterized by lower values of
179 $(\text{La}/\text{Yb})_{\text{UCC}}$, $(\text{La}/\text{Sm})_{\text{UCC}}$, δCe and δEu compared to the underlying **and older**
180 sediments (Unit 2, 158–743 cm, deposited during 8.2–30.3 ka). However, $(\text{Gd}/\text{Yb})_{\text{UCC}}$
181 ratios, are relatively higher in Unit 1 than in Unit 2. In contrast to the REE
182 compositions, the oxygen isotopic values ($\delta^{18}\text{O}$) of foraminifera, CaCO_3 contents and
183 mean grain size of the sediments show large variations at the boundary between
184 oxygen isotope stages (OIS) 1 and 2 at about 10 ka (Fig. 2), yet this change does not
185 affect the source of the siliciclastic component.

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

197 Overall, Unit 1 shows relative enrichments in total REE and remarkable
198 fractionations of middle and heavy REEs, with obvious convex shapes in the REE

199 plots, whereas Unit 2 is characterized by relatively flat REE patterns, without obvious
200 Ce and Eu anomalies (Fig. 3).

201 Two layers with much lower than normal values of $(La/Yb)_{UCC}$, $(La/Sm)_{UCC}$ and
202 $(Gd/Yb)_{UCC}$ occur at core depths of 150 cm and 575 cm respectively (Fig. 2). The
203 AMS ^{14}C ages of these two layers are about 7.6 ka and 25.8 ka respectively, which
204 correspond well with two volcanic eruption events documented in southwestern Japan
205 (Kitagawa et al., 1995; Arakawa et al., 1998).

206 3.2 Controlling factors of REE compositions in the core sediments

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

221 In this study, 1 N HCl was used to leach the bulk sediment samples and the
222 residual fractions were separated for measuring REE concentrations. Therefore, we
223 infer that a major part of the mobile fraction, including carbonate, apatite, and Fe-Mn

224 oxides were removed from the bulk core sediments (Yang et al., 2002; 2006; Choi et
225 al., 2007; Song and Choi, 2009), and thus that the measured REE compositions
226 overall represent the contributions of the siliciclastic fraction to the core sediments.
227 Chemical weathering thus exerts a weak influence on the REE compositions in the
228 residual fractions.

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

242

243 *3.3 Provenance discrimination of the Core DGKS9604 sediments*

244 Because of the lower contents of biogenic silica from radiolarians and diatoms and
245 authigenic components (Fe–Mn oxides) in the west slope of the Okinawa Trough (Liu,
246 2005), the residues of 1N HCl leached samples studied in this paper are primarily
247 composed of detrital silicate minerals, which mainly originated from erosion of
248 terrigenous and volcanic sources. The potential provenances of siliciclastic sediments

249 in the middle Okinawa Trough include terrigenous sources supplied via fluvial and
250 eolian inputs, volcanic and hydrothermal activities, and those carried by the oceanic
251 currents such as the Kuroshio Current from the southern Okinawa Trough. It has long
252 been recognized that the terrigenous **particulate matters** into the middle and north
253 Okinawa Trough **are** mainly derived from the shelf of the East China Sea where the
254 sediments predominantly originate from the two largest rivers in China, i.e.
255 Changjiang and Huanghe Rivers (Qin et al., 1987; Iseki et al., 2003; Katayama and
256 Watanabe, 2003; Liu et al., 2007; Dou et al., 2010). Further south however Taiwan
257 dominates because it is one of the greatest producers of **terrigenous** sediment to the
258 ocean known globally (Milliman and Syvitski, 1992).

259 Compared to the surface seafloor sediments in the East China Sea, Core
260 DGKS9604 sediments have higher REE concentrations, ratios of $(La/Yb)_{UCC}$ and
261 $(La/Sm)_{UCC}$, and lower of $(Gd/Yb)_{UCC}$ (Table 1). Nevertheless, a detailed comparison
262 of REE composition between the sediments from the continental shelf of the East
263 China Sea and from the Core DGKS9604 can not be made because of inadequate data
264 from the East China Sea and different sample pre-treatment methods. Significant
265 differences in REE concentrations and fractionation patterns between Unit 1 and Unit
266 2 suggest that **these** depositional units may have different sediment provenances. Unit
267 2 sediments have REE compositions that are more similar with the modern
268 Changjiang, rather than Huanghe sediments, in terms of their REE concentrations and
269 fractionation parameters (Table 1, Fig 4a). In contrast, Unit 1 sediments have much
270 higher total REE concentrations and different REE fractionation patterns compared to
271 Unit 2 (Table 1; Figs. 2–4). In this study, a discrimination plot of $(La/Sm)_{UCC}$ vs.
272 $(Gd/Yb)_{UCC}$ was used to identify the provenance of sediment in Core DGKS9604 (Fig.
273 5). The figure clearly demonstrates that the Unit 2 sediments plot together with **the**

274 Changjiang sediments, whereas the Unit 1 sediments plot in another group that is
275 close in character to the sediments from southwestern Taiwan (Chen et al., 2007).
276 Therefore, we infer that the older sediments (Unit 2, 158–743 cm, deposited at
277 8.2–30.3 ka) were derived predominantly from the Changjiang and partly from the
278 Huanghe, while the upper core sediments (Unit 1, 0–142 cm, deposited at 0–7.1 ka)
279 were primarily sourced from Taiwan Island.

280 Volcanic materials derived from the sea floor or transported from the volcanoes
281 of the Japanese islands also exert significant influence on the deposition in the middle
282 Okinawa Trough, and therefore, can be considered as another potential contributor to
283 the terrigenous detritus (Machida, 1999). Several tephra layers occurring in the
284 sediments of the north Okinawa Trough are mainly composed of volcanic glasses and
285 pumices (Xu and Oda, 1999; Ujiie et al., 2001; Sun et al., 2003). Two abnormal layers
286 deposited at 7.6 ka and 25.8 ka have extraordinarily strong HREE enrichment and
287 LREE depletion (Figs. 3, 4b), very similar to the surrounding volcanic materials
288 which came from the Kyushu islands of Southwestern Japan (Figs. 1, 4b) (Arakawa et
289 al., 1998; Hamasaki, 2002), but much different from the other core sediment and the
290 riverine sediments of the Changjiang and Huanghe (Figs. 3, 4a). This clearly suggests
291 that these two layers with abnormal REE compositions in Core DGKS9604
292 predominantly consist of volcanic materials. Two volcanic events, known as
293 Kikai-Akahoya (K-Ah) and Aira-Tanzawa (AT), with eruption ages at 7324 Cal yr BP
294 (Kitagawa et al., 1995) and 25,120±270 Cal yr BP respectively (Miyairi et al., 2004),
295 are known in southwest Japan (Fig. 1). They were regarded as the origin of two
296 widely distributed tephra layers in the sediments of the middle and north Okinawa
297 Trough, which predominantly consist of volcanic glasses and pumices (Xu and Oda,
298 1999; Liu et al., 2000; Ujiie et al., 2001; Sun et al., 2003). The two geochemically

299 abnormal layers in Core DGKS9604 are thus estimated to be dominated by K-Ah
300 tephra and AT tephra, especially considering that they have similar ages.

301 The average REE concentrations of the volcanic glass (Liu and Meng, 2004) and
302 volcanic rocks around the southwestern Japan Island (Shinjo and Kato, 2000) are 93.7
303 $\mu\text{g/g}$ and 109.6 $\mu\text{g/g}$ respectively, much lower than those found in this core, or in
304 sediments from the Changjiang and Huanghe Rivers (Table 1). Indeed, the total REE
305 concentrations of these two layers are 174.0 $\mu\text{g/g}$ and 129.0 $\mu\text{g/g}$ respectively, much
306 higher than those of the documented volcanic materials, which implies that these two
307 tephra layers are probably mixed with other terrigenous sediments, which have higher
308 REE concentrations. The discrimination plot suggests that the abnormal layer 1
309 deposited at 7.6 ka was probably K-Ah tephra mixed with fine-grained terrigenous
310 sediments from Taiwan, while abnormal layer 2, which was deposited at 25.8 ka was
311 formed by mixture between the AT tephra and riverine sediments from mainland
312 China (Fig. 5). In addition, no visual volcanic glasses and pumices were observed in
313 the core sediments (Yu et al., 2008, 2009), further suggesting that the volcanic
314 materials from the surrounding islands might not have dominated the late Quaternary
315 sedimentation in the middle Okinawa Trough.

316

317 *3.4 Transport pattern of detrital sediments into the middle Okinawa Trough during the*
318 *last 30 ka*

319 The identification of sediment sources in the Okinawa Trough is of great
320 significance for understanding the depositional history and paleoenvironmental
321 changes of the East China Sea during the late Quaternary. REE concentrations and
322 fractionation patterns strongly suggest that the siliciclastic sediments accumulated in
323 the middle Okinawa Trough during the late Quaternary might originate from different

324 provenances. During the period from late last glaciation (30 ka) to the early-middle
325 Holocene (8.0~7.0 ka) the Changjiang was the primary sediment supplier, whereas
326 during the late Holocene of the last 7 ka Taiwan Island could be the dominant
327 sediment supplier. The provenance discrimination results based on REE compositions
328 are basically similar with the observation of clay mineral assemblages (Dou et al.,
329 2010).

330 The lowest sea level during the Last Glacial Maximum (LGM) was about
331 120~135 m lower than that of today (Emery et al., 1971; Qin et al., 1987; Henderson,
332 2002), which implies that the continental shelf of the East China Sea would have been
333 largely exposed and correspondingly, that the river mouths of the paleo-Changjiang
334 and/or paleo-Huanghe must have been positioned significantly closer to present-day's
335 outer shelf (Fig. 6a). Furthermore, the main stream of the Kuroshio Current deflected
336 to the east of the Ryukyu Islands Arc during the LGM (Ujiié et al., 1991; Ahagon et
337 al., 1993; Xiang et al., 2003) (Fig. 6a). As a result, the terrigenous fine-grained
338 particulate materials from the Changjiang and/or Huanghe might have been
339 transported directly into the middle Okinawa Trough and dominated the deposition
340 there, since there would have been no influence from the Kuroshio Current.

341 Previous studies suggested that the aeolian input from the loess area in western
342 China into the East Asian marginal seas increased during the LGM (Irimo and Tada,
343 2002; Nagashima et al., 2007). It is well known that the Huanghe sediments have very
344 similar REE compositions with the loess because the latter is the main sediment
345 provider of the Huanghe (Yang et al., 2002). The Unit 2 and Huanghe sediments plot
346 in different groups in the discrimination plot (Fig. 5), suggesting that the
347 Huanghe-derived and aeolian materials did not contribute significantly to the
348 siliciclastic deposition in the middle Okinawa Trough during the LGM.

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

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

374 high stand of sea level at ca. 6~7 ka.

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

389 It is interesting to note that the REE compositions of Core DGKS9604 sediments
390 do not vary simultaneously with the oxygen isotopes of foraminifera or with bulk
391 CaCO₃ compositions, which show abrupt and large variations at ca. 10 ka (Fig. 2). For
392 example, the oxygen isotopes of foraminifera and bulk CaCO₃ show heavier and
393 lower values respectively during the LGM than in the Holocene (Fig. 2) (Jian et al.,
394 2000; Liu et al., 2001). The relatively uniform REE compositions of the Unit 2
395 sediments suggest that from ca. 30 to 7.0 ka the provenances of the terrigenous
396 sediments of the middle Okinawa Trough remained stable, despite large fluctuations
397 of sea level, monsoon activity and depositional environments as well. In comparison,
398 the oxygen isotopes of foraminifera and bulk CaCO₃ compositions in the core

399 sediments that reflect the in-situ paleoenvironment and primary productivity are more
400 sensitive to changing sea level, depositional environments and/or monsoon activity
401 during the late Quaternary. The depositional flux of REE to the Core DGKS9604
402 sediments varied significantly since 30 ka, shown by higher fluxes at 30–22,
403 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
404 variable depositional fluxes of REE strongly suggests that the changing supply rates
405 of terrigenous sediments into the middle Okinawa Trough during the late Quaternary,
406 were probably related to the weathering intensity and sediment production in the large
407 drainage basins. However, it is noteworthy that other factors control the REE flux at
408 the core location, most notably the distance of the river mouth from the middle
409 Okinawa Trough and transport processes of fine-grained sediments in the continental
410 margin (Meng et al., 2007). In particular, we note that 11.8–6.9 ka is a period of lower
411 REE flux, yet this time is generally recognized as a period of strengthening summer
412 monsoon rains (Wang et al., 2001; Herzschuh, 2006). If stronger summer monsoon
413 rains were driving stronger continental erosion, as has been seen in South Asia (Clift
414 et al., 2008), then the sedimentation should increase not decrease. Probably, major
415 part of these increased terrigenous sediments resulted from stronger continental
416 erosion was trapped in the continental shelf with rapidly rising sea level during the
417 early postglacial period. Nevertheless, the controls of depositional flux of terrigenous
418 sediments in the middle Okinawa Trough is beyond this study and will not be
419 considered in greater detail in this paper.

420

421 **4. Conclusions**

422

423 One hundred and six sub-samples of Core DGKS9604, which spans the past 30

424 k.y. and comprises clayey silt, were collected from the middle Okinawa Trough for
425 sediment provenance study. Based on REE geochemical characteristics of the residual
426 fractions leached by 1N HCl, we conclude that Core DGKS9604 can be divided into
427 an upper Unit 1 (0–142 cm, <7.1 ka) and Unit 2 (158–743 cm, 8.2–30.3 ka). Total
428 REE concentrations and fractionation parameters, including $(La/Yb)_{UCC}$, $(La/Sm)_{UCC}$,
429 $(Gd/Yb)_{UCC}$, Ce and Eu anomalies, are significantly different between Units 1 and 2,
430 with large and abrupt variations occurring at 8.2–7.1 ka. The UCC-normalized REE
431 patterns of the Unit 2 sediments are very similar **with** those of the riverine sediments
432 from mainland China, especially from the Changjiang. This observation suggests that
433 the **fine-grained terrigenous** sediments which accumulated in the middle Okinawa
434 Trough from LGM to the middle Holocene might originate predominantly from the
435 Changjiang. During that period, the main stream of the Kuroshio Current was
436 deflected to the east of the Ryukyu Islands and the sea level remained lower than
437 present day, so that the river mouth of the Changjiang lay at the shelf edge. As a result,
438 terrigenous materials from the Changjiang and perhaps the Huanghe may have been
439 more easily transported into the middle Okinawa Trough.

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

448 Two geochemically abnormal layers with depositional ages at 7.6 and 25.8 ka are

449 characterized by distinct REE compositions, and are interpreted to be dominated by
450 Japanese volcanic material from the Kikai-Akahoya and Aira-Tanzawa tephras
451 respectively. However, we argue that these volcanic glasses are mixed with
452 fine-grained terrigenous sediments from **Taiwan and mainland China respectively**.

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

459

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468

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704

705 **Figure captions:**

706 Fig. 1

707 Schematic map showing the locations of Core DGKS9604 and other reference cores.

708 The regional circulation pattern in the East China Sea and the adjacent areas are

709 sourced from [Huh and Su \(1999\)](#) and [Yu et al \(2008\)](#). YSCC=Yellow Sea Coastal

710 Current; ZFCC=Zhejiang-Fujian Coastal Current; CDW=Changjiang Diluted Water;

711 TWC=Taiwan Warm Current; YSWC=Yellow Sea Warm Current.

712

713 Fig. 2

714 Depth profiles of mean grain size (Mz), $\delta^{18}\text{O}$ of foraminifera, CaCO_3 , and REE

715 fractionation parameters of Core DGKS9604 sediments. The age model, $\delta^{18}\text{O}$ and

716 grain size data after [Yu et al \(2008; 2009\)](#). OIS denotes oxygen isotope stage. The

717 lightly shadowed area indicates two tephra layers, i.e. Kikai-Akahoya (K-Ah) and

718 Aira-Tanzawa (AT) ([Kitagawa et al., 1995](#); [Arakawa et al., 1998](#)).

719

720 Fig. 3

721 UCC-normalized patterns of Core DGKS9604 sediments. The samples of Unit 1 and

722 Unit 2 show similar fractionation patterns, much different from two samples primarily

723 of volcanic origins.

724

725 Fig. 4

726 Comparisons of REE patterns between the sediments of Core DGKS9604, Changjiang

727 and Huanghe ([Yang et al., 2002](#)), and volcanic source materials ([Arakawa et al., 1998](#);

728 [Hamasaki, 2002](#); [Liu et al., 2004](#)).

729

730 Fig. 5
731 Discrimination plot of $(Gd/Yb)_{UCC}$ vs $(La/Sm)_{UCC}$ for the sediments of Core
732 DGKS9604. Values of modern Changjiang and Huanghe riverine sediments (Yang et
733 al., 2002), pumice and lavas samples of Aira pyroclastic eruption (Arakawa et al.,
734 1998), volcanic rocks of Okinawa Trough (Shinjo and Kato, 2000), and core
735 sediments of southwestern Taiwan (Chen et al., 2007) are also shown for comparison.

736

737 Fig. 6
738 Schematic diagrams showing the influences of sea level change and oceanic
739 circulation patterns on the terrigenous sediment inputs to the Okinawa Trough and
740 adjoining shelf of the East China Sea during the LGM (a) and the mid-late Holocene
741 (0~ca.7 Cal ka BP; b). The direction of the Kuroshio Current at the LGM is after Ujiie
742 and Ujiie (1999)

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

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23

24 **Abstract:**

25 The late Quaternary paleoceanography and paleoenvironment in the Okinawa
26 Trough, East China Sea, have been well reconstructed over the last decade, while in
27 contrast the provenance of terrigenous sediments that have accumulated there remains
28 enigmatic. In this study rare earth elements (REE) were used to reveal provenance
29 changes in sediments from Core DGKS9604, taken from the middle Okinawa Trough.
30 Discrimination plots based on REE fractionation parameters suggest that the cored
31 sediments have variable provenances over the last 30 ka, with the lower part (ca.
32 31–8.2 ka) ultimately originating mostly from the Changjiang (Yangtze River) and the
33 upper part (0–7.1 ka) primarily from Taiwan. During the Last Glacial Maximum and
34 early Holocene sea level was low and the main stream of the Kuroshio Current was
35 deflected to the east of the Ryukyu Islands. As a result sediment from the Changjiang
36 dominated sedimentation of the middle Okinawa Trough. However, since about 7 ka
37 the main stream of the Kuroshio Current strengthened in the area of the trough, as sea
38 level reached the modern position. This caused near-bottom transport of fine-grained
39 sediments from the continental margin to the trough to become weak and instead,
40 Taiwan-derived terrigenous sediments dominated in the middle trough. The changing
41 provenances of terrigenous sediments into the middle Okinawa Trough are closely
42 related to the evolution of oceanic circulations and sea level in the East China Sea.
43 Two tephra layers in the core have distinct REE compositions and correlate well with
44 two volcanic eruptions at 7.6 and 25.8 ka in southern Japan.

45

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

48

49 **1. Introduction**

50

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

61 Over the last two decades, many scientists have attempted to identify the origin
62 of sediment in the middle Okinawa Trough using mineralogical (Chen et al., 1982),
63 oceanographic and paleoceanographic (Jian et al., 1998, 2000; Ujiie and Ujiie, 1999;
64 Liu et al., 1999, 2000), as well as geochemical methods (Zhao and Yan, 1992; Guo et
65 al., 2001; Meng et al., 2001). These studies suggested that the terrigenous sediment
66 sources of the Okinawa Trough predominantly include direct supply from major rivers
67 in East Asia, particularly the Changjiang (Yangtze River) and Huanghe (Yellow River)
68 (Katayama and Watanabe, 2003), and lateral transport from the East China Sea shelf
69 through the bottom layers (Honda et al., 2000; Iseki et al., 2003; Oguri et al., 2003). In
70 particular, during the Last Glacial Maximum (LGM) when the sea level was about
71 135 m lower than present-day, Changjiang-derived sediments must have directly
72 emptied into the Okinawa Trough (Milliman et al., 1989; Saito et al., 1998; Yoo et al.,
73 2002; Liu et al., 2007). In addition, the episodic volcanic eruptions in the west Japan

74 volcanic zone have produced widespread fallout tephra layers in the northern
75 Okinawa Trough (Machida, 1999; Miyairi et al., 2004). Other factors such as
76 submarine hydrothermal activity (Zeng et al., 2000; Zhai et al., 2001, 2007), erosion
77 of tectonically active Taiwan Island (Liu et al., 2008), eolian transport (Tsunogai et al.,
78 1985), seafloor earthquakes (Huh et al., 2004), as well as intrusion of the Kuroshio
79 Current (Jian et al., 1998; Lee et al., 2004) also contribute to the transport of
80 siliciclastic sediments into and within the trough.

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

96 Whether and how far the terrigenous sediments from Taiwan Island can be
97 transported northward into the middle and north Okinawa Trough is another
98 challenging question waiting for more lines of evidences (Hsu et al., 2004). The

99 reliable provenance tracers of terrigenous sediments from different end-members and
100 high-resolution sampling analysis are urgently needed if the sediment transport
101 patterns in and around the Okinawa Trough are to be understood in detail.

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

113

114 **2. Samples and methods**

115 A piston core (DGKS9604) was taken from the middle Okinawa Trough
116 (28°16.64'N, 127°01.43'E) at a water depth of 766 m during the joint Chinese-French
117 DONGHAI Cruise in 1996 (Fig. 1). The core is 1076 cm long and located on the
118 western continental slope of the middle Okinawa Trough. In contrast to the adjacent
119 core DGKS9603 (28°08.869'N, 127°16.238'E), which was taken during the same
120 cruise (Fig.1), no visual ash layers and hiatuses are present in Core DGKS9604 (Yu,
121 2006). A high-resolution age model of the core was established on the basis of the
122 oxygen isotopic compositions of *Globigerinoides sacculifer* and accelerator mass
123 spectrometry (AMS) radiocarbon dating of planktonic foraminifera (Liu et al., 2001;

124 Yu et al., 2008). The depositional age at the bottom of the core is estimated to be
125 37.01 Cal ka, and the bulk sedimentation rate of the core averages about 29 cm/k.y.,
126 which is higher than that of Core DGKS9603 (Liu et al, 1999, 2000). Sediments from
127 Core DGKS9604 are primarily composed of clayey silt, with a mean grain size of
128 $6.7\pm 0.4 \Phi$ (Yu, et al, 2008; Fig. 2).

129 A total of 106 subsamples were collected from Core DGKS9604 at 4 cm intervals
130 for the uppermost 100 cm and at 8 cm intervals between 100 and 743 cm. To separate
131 the residual fractions from the bulk samples, 0.2 g bulk sediment samples were
132 leached with 20 ml 1 N HCl for 24 hours at 50°C. The residues of the leached samples
133 were then completely digested with concentrated HF–HNO₃–HClO₄ in an airtight
134 Teflon container. Concentrations of REEs and other elements in the residual fractions
135 were determined by ICP-MS (PQ3, Thermo Elemental) and ICP-AES (IRIS
136 Advantage) in the State Key Laboratory of Marine Geology at Tongji University. The
137 precision and accuracy were monitored by geostandard GSR-5, GSR-6, and GSR-9.
138 Differences between the determined and certified values were less than 5%, and the
139 leaching efficiency checked by GSR-5, GSR-6, and GSR-9 was above 90%.

140

141 **3. Results and discussions**

142 *3.1 REE compositions of the Core DGKS9604 sediments*

143 The average rare earth element compositions of the Core DGKS9604 sediments
144 and the reference materials are given in Table 1. Depth profiles of total REE
145 concentrations, REE fractionation parameters, CaCO₃ contents and mean grain size
146 are shown in Figure 2. The REE compositions, including total REE concentrations,
147 $(La/Yb)_{ucc}$, $(La/Sm)_{ucc}$, $(Gd/Yb)_{ucc}$, δCe (Eu anomalies) and δEu (Ce anomalies),
148 exhibit regular variations in the core with a remarkable and abrupt change occurring

149 in the mid-Holocene (8.2–7.1 ka) (Fig. 2). The younger sediments (Unit 1, 0–142 cm
150 and deposited since 7.1 ka) are significantly enriched in REE concentrations and
151 characterized by lower values of $(La/Yb)_{ucc}$, $(La/Sm)_{ucc}$, δCe and δEu compared to the
152 underlying sediments (Unit 2, 158–743 cm, deposited during 8.2–30.3 ka). However,
153 $(Gd/Yb)_{ucc}$ ratios, are relatively higher in Unit 1 than in Unit 2. In contrast to the REE
154 compositions, the oxygen isotopic values ($\delta^{18}O$) of foraminifera, $CaCO_3$ contents and
155 mean grain size of the sediments show large variations at the boundary between
156 oxygen isotope stages (OIS) 1 and 2 at about 10 ka (Fig. 2), yet this change does not
157 affect the source of the siliciclastic component.

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

165 Two layers with much lower than normal values of $(La/Yb)_{ucc}$, $(La/Sm)_{ucc}$ and
166 $(Gd/Yb)_{ucc}$ occur at core depths of 150 cm and 575 cm respectively (Fig. 2). The AMS
167 ^{14}C ages of these two layers are about 7.6 ka and 25.8 ka respectively, which
168 corresponds well with volcanic eruption events documented in southwestern Japan
169 (Kitagawa et al., 1995; Arakawa et al., 1998).

170

171 *3.2 Controlling factors of REE compositions in the core sediments*

172 Many factors, including the bed rock composition in the source area, sediment
173 grain size, mineralogy, intensity of chemical weathering, diagenesis and

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

186 In this study, 1 N HCl was used to leach the bulk sediment samples and the
187 residual fractions were analyzed for REE concentrations. Therefore, we infer that a
188 major part of the mobile fraction, including carbonate, apatite, Fe-Mn oxides and
189 organic-bound phases were removed from the bulk core sediments (Yang et al., 2002;
190 2006), and thus that the measured REE compositions overall represent the
191 contributions of the siliciclastic fraction to the sediments. Chemical weathering thus
192 exerts a weak influence on the REE compositions in the residual fractions.

193 Heavy minerals such as zircon, monazite, garnet, allanite and sphene, despite
194 their low abundances in sediments, may account for a considerable fraction of the
195 bulk REE concentrations because of the high REE concentrations in these minerals
196 (Gromet and Silver, 1983; Taylor and McLennan, 1985; McLennan, 1989; Hannigan
197 and Sholkovitz, 2001). However, recent study suggested that major rock-forming and
198 heavy minerals in total contribute less than 20% of the total REE concentrations in the

199 modern Changjiang riverine sediments (Yang et al., 2002). The mean sediment grain
200 size of Core DGKS9604 ranges from 7.2 Φ to 6.3 Φ , smaller than that of the
201 Changjiang sediment (6.3 \pm 0.4 Φ), which suggests that heavy minerals are probably
202 not the primary control on REE compositions in the core sediments. Consequently,
203 variations of REE concentrations, as well as fractionation patterns in the siliciclastic
204 sediments, should reflect the bulk mineralogy and so clearly indicate changes of
205 sediment provenance.

206

207 *3.3 Provenance discrimination of the Core DGKS9604 sediments*

208 Because of the lower contents of biogenic (opals, radiolarians and diatoms) and
209 authigenic components (Fe–Mn oxides) in the Okinawa Trough (Liu, 2005), the
210 residues of 1N HCl leached samples studied in this paper are primarily composed of
211 detrital silicate minerals, which mainly originated from erosion of terrigenous and
212 volcanic sources. The potential provenances of siliciclastic sediments in the middle
213 Okinawa Trough include terrigenous sources supplied via fluvial and eolian inputs,
214 volcanic and hydrothermal activities, and those carried by the oceanic currents such as
215 the Kuroshio Current from the southern Okinawa Trough. It has long been recognized
216 that the terrigenous flux into the middle and north Okinawa Trough is mainly derived
217 from the shelf of the East China Sea where the sediments predominantly originate
218 from the two largest rivers in China, i.e. Changjiang and Huanghe Rivers (Qin et al.,
219 1987; Iseki et al., 2003; Katayama and Watanabe, 2003; Liu et al., 2007). Further
220 south however Taiwan dominates because it is one of the greatest producers of
221 sediment to the ocean known globally (Milliman and Syvitski, 1992).

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

224 (La/Sm)_{ucc}, and lower of (Gd/Yb)_{ucc} (Table 1). Nevertheless, a detailed comparison of
225 REE composition between the sediments from the continental shelf of the East China
226 Sea and from the Core DGKS9604 can not be made because of inadequate data from
227 the East China Sea and different sample pre-treatment methods. Significant
228 differences in REE concentrations and fractionation patterns between Unit 1 and Unit
229 2 suggest that both depositional units may have different sediment provenances. Unit
230 2 sediments have REE compositions that are more similar with the modern
231 Changjiang, rather than Huanghe sediments, in terms of their REE concentrations and
232 fractionation parameters (Table 1, Fig 4a). In contrast, Unit 1 sediments have much
233 higher total REE concentrations and different REE fractionation patterns compared to
234 Unit 2 (Table 1; Figs. 2–4). In this study, a discrimination plot of (La/Sm)_{ucc} vs.
235 (Gd/Yb)_{ucc} was used to identify the provenance of sediment in Core DGKS9604 (Fig.
236 5). The figure clearly demonstrates that the Unit 2 sediments plot together with
237 Changjiang sediments, whereas the Unit 1 sediments plot in another group that is
238 close in character to the sediments from southwestern Taiwan (Chen et al., 2007).
239 Therefore, we infer that the older sediments (Unit 2, 158–743 cm, deposited at
240 8.2–30.3 ka) were derived predominantly from the Changjiang and partly from the
241 Huanghe, while the upper core sediments (Unit 1, 0–142 cm, deposited at 0–7.1 ka)
242 were primarily sourced from Taiwan Island.

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

249 7.6 ka and 25.8 ka have extraordinarily strong HREE enrichment and LREE depletion
250 (Figs. 3, 4), very similar to the surrounding volcanic materials which came from the
251 Kyushu islands of Southwestern Japan (Fig. 1) (Arakawa et al., 1998; Hamasaki,
252 2002), but much different from the riverine sediments of the Changjiang and Huanghe.
253 This clearly suggests that these two abnormal layers in the Core DGKS9604
254 predominantly consist of volcanic materials. Two volcanic events, known as
255 Kikai-Akahoya (K-Ah) and Aira-Tanzawa (AT), with eruption ages at 7324 Cal yr BP
256 (Kitagawa et al., 1995) and $25,120 \pm 270$ Cal yr BP respectively (Miyairi et al., 2004),
257 are known from southwest Japan (Fig. 1). They were regarded as the origin of two
258 widely-distributed tephra layers in the sediments of the middle and north Okinawa
259 Trough, which predominantly consist of volcanic glasses and pumices (Xu et al., 1999;
260 Liu et al., 2000; Ujiie et al., 2001; Sun et al., 2003). The two geochemically abnormal
261 layers in the Core DGKS9604 are thus estimated to be dominated by K-Ah tephra and
262 AT tephra, especially considering that they have similar ages.

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

274 China (Fig. 5).

275

276 *3.4 Transport pattern of detrital sediments into the middle Okinawa Trough during the*
277 *last 30 ka*

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

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

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

299 China into the East Asian marginal seas increased during the LGM (Irimo and Tada,
300 2002; Nagashima et al., 2007). It is well known that the Huanghe sediments have very
301 similar REE compositions with the loess because the latter is the main sediment
302 provenance of the Huanghe (Yang et al., 2002). The Unit 2 and Huanghe sediments
303 plot in different groups in the discrimination plot (Fig. 5), suggesting that the
304 Huanghe-derived and eolian materials did not contribute significantly to the
305 siliciclastic deposition in the middle Okinawa Trough during the LGM.

306 During the deglacial period and early Holocene, the Changjiang river mouth
307 retreated with the postglacial sea-level rising to its present position at about 7 ka (Liu
308 et al., 2007). The main stream of the Kuroshio Current re-entered the Okinawa Trough
309 and/or strengthened at about 7.5–6.0 ka (Ujiie et al., 1991; Jian et al., 1998, 2000;
310 Xiang et al., 2003). Since then, sedimentation in the Okinawa Trough has been
311 dominated by the competing processes of the Kuroshio Current and the oceanic
312 circulations in the East China Sea (Yang et al., 1992; Lee et al., 2004). Sediment trap
313 experiments revealed that a significant amount of suspended terrigenous particles are
314 transported through the bottom layer from the outer shelf of the East China Sea to the
315 Okinawa Trough (Iseki et al., 2003; Katayama and Watanabe, 2003; Pang and Wang,
316 2004). Near-bottom transport may be a key process for shelf-to-deep sea export of
317 biogenic/lithogenic particles (Iseki et al., 2003).

318 The REE compositions of the Unit 1 sediments are remarkably different from the
319 Unit 2 and riverine sediments from mainland China, but very similar with those of
320 Taiwan-derived sediments (Table 1; Figs. 2 and 5). This implies that the siliciclastic
321 sediments deposited since 7 ka in the middle Okinawa Trough were primarily sourced
322 from Taiwan, and probably transported northward by the main stream of the Kuroshio
323 Current, after their initial deposition from the Lanyang River delta and fan. In contrast,

324 the sediment transport through the bottom layer from the outer shelf of the East China
325 Sea to the middle Okinawa Trough during the mid-late Holocene was relatively weak
326 as a result of the blocking effect of the Kuroshio and Taiwan Warm Currents, which
327 act as a barrier deflecting other currents from the area (Fig. 6b). Therefore, the
328 suspended or resuspended fine-grained sediments of the Changjiang and/or Huanghe
329 would not have been able to dominate the sedimentation in the middle Okinawa
330 Trough since the high stand of sea level at ca. 7 ka.

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

341 It is interesting to note that the REE compositions of Core DGKS9604 sediments
342 do not vary simultaneously with the oxygen isotopes of foraminifera or with bulk
343 CaCO₃ compositions, which show abrupt and large variations at ca. 10 ka (Figs. 2).
344 For example, the oxygen isotope of foraminifera and bulk CaCO₃ show heavier and
345 lower values respectively during the LGM than in the Holocene (Fig. 2) (Jian et al.,
346 2000; Liu et al., 2001). The relatively uniform REE compositions of the Unit 2
347 sediments suggest that from ca. 30 to 7.0 ka the provenances of the terrigenous
348 sediments of the middle Okinawa Trough remained stable, despite large fluctuations

349 of sea level, monsoon activity and depositional environments as well. In comparison,
350 the oxygen isotope of foraminifera and bulk CaCO₃ compositions in the core
351 sediments that reflect the in-situ paleoenvironment and primary productivity are more
352 sensitive to changing sea level, depositional environments and/or monsoon activity
353 during the late Quaternary. The depositional flux of REE to the Core DGKS9604
354 sediments varied significantly since 30 ka, shown by higher fluxes at 30–22,
355 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
356 variable depositional fluxes of REE strongly suggests that the changing supply rates
357 of terrigenous sediments into the middle Okinawa Trough during the late Quaternary,
358 were probably related to the weathering intensity and sediment production in the large
359 drainage basins. However, it is also clear that other factors control the REE flux at the
360 core location, most notably the distance of the river mouth from the middle Okinawa
361 Trough and transport processes of fine-grained sediments in the continental margin
362 (Guo et al., 2001; Xiong and Liu, 2004; Meng, 2007). In particular, we note that
363 11.8–6.9 ka is a period of lower REE flux, yet this time is generally recognized as a
364 period of strengthening summer monsoon rains (Wang et al., 2001; Herzschuh, 2006).
365 If stronger summer monsoon rains were driving stronger continental erosion, as has
366 been seen in South Asia (Clift et al., 2008), then the sedimentation should increase not
367 decrease. Probably, major part of these increased terrigenous sediments resulted from
368 stronger continental erosion was trapped in the continental shelf with rapidly rising
369 sea level during the early postglacial period. Nevertheless, the controls of depositional
370 flux of terrigenous sediments in the middle Okinawa Trough is beyond this study and
371 will not considered in greater detail in this paper.

372

373 **4. Conclusions**

374

375 One hundred and six sub-samples of Core DGKS9604, which spans the past 30
376 k.y. and comprises clayey silt, were collected from the middle Okinawa Trough for
377 sediment provenance study. Based on REE geochemical characteristics of the residual
378 fractions leached by 1N HCl, we conclude that Core DGKS9604 can be divided into
379 an upper Unit 1 (0–142 cm, <7.1 ka) and Unit 2 (158–743 cm, 8.2–30.3 ka). Total
380 REE concentrations and fractionation parameters, including $(La/Yb)_{UCC}$, $(La/Sm)_{UCC}$,
381 $(Gd/Yb)_{UCC}$, Ce and Eu anomalies, are significantly different between Units 1 and 2,
382 with large and abrupt variations occurring at 8.2–7.1 ka. The UCC-normalized REE
383 patterns of the Unit 2 sediments are very similar compared to those of the riverine
384 sediments from mainland China, especially from the Changjiang. This observation
385 suggests that the terrigenous fine-grained sediments which accumulated in the middle
386 Okinawa Trough from LGM to the middle Holocene might originate predominantly
387 from the Changjiang. During that period, the main stream of the Kuroshio Current
388 was deflected to the east of the Ryukyu Islands and the sea level remained lower than
389 present day, so that the river mouth of the Changjiang lay at the shelf edge. As a result
390 terrigenous materials from the Changjiang and perhaps the Huanghe may have been
391 more easily transported into the middle Okinawa Trough.

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

399 been restricted to the inner shelf since that time.

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

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

411

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666

667 **Figure captions:**

668 Fig. 1

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

674 Fig. 2

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

680 Fig. 3

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

684 Fig. 4

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

688 Fig. 5

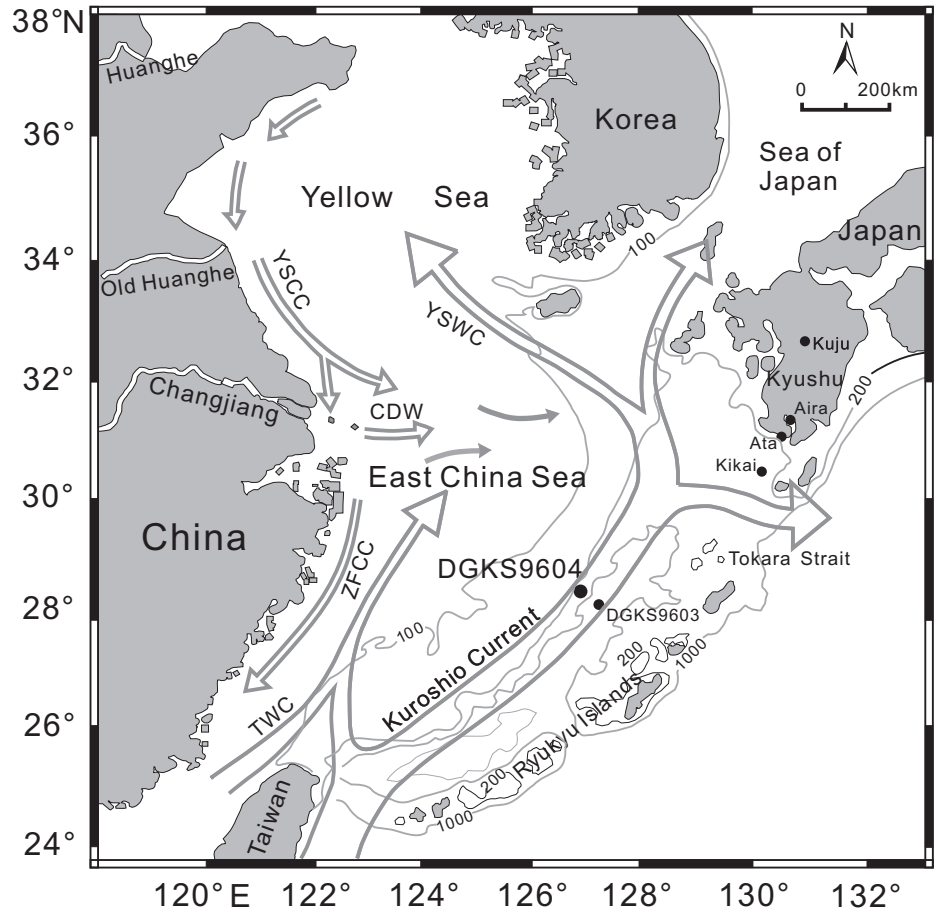
689 Discrimination plot of $(\text{Gd}/\text{Yb})_{\text{ucc}}$ vs $(\text{La}/\text{Sm})_{\text{ucc}}$ for the sediments of Core DGKS9604.
690 Values of modern Changjiang and Huanghe riverine sediments ([Yang et al, 2002](#)),
691 pumice and lavas samples of Aira pyroclastic eruption ([Arakawa et al, 1998](#)), volcanic

692 rocks of Okinawa Trough ([Shinjo and Kato, 2000](#)), and core sediments of
693 southwestern Taiwan ([Chen et al., 2007](#)) are also shown for comparison.

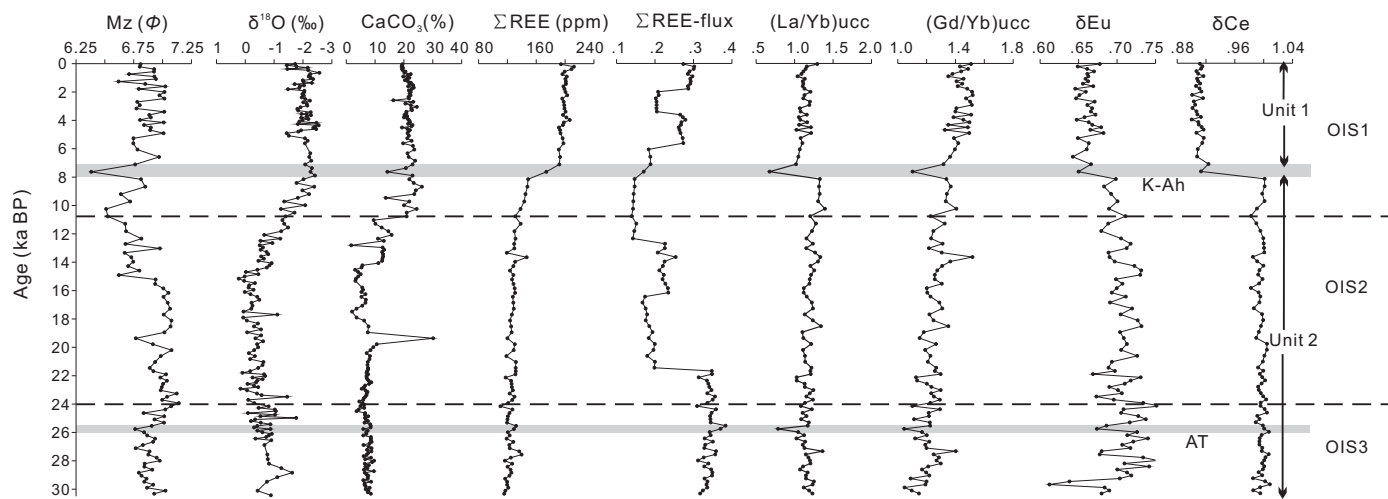
694 Fig. 6

695 Schematic diagrams showing the influences of sea level change and oceanic
696 circulation patterns on the terrigenous sediment inputs to the Okinawa Trough and
697 adjoining shelf of the East China Sea during the LGM (a) and the mid-late Holocene
698 (0~ca.7 Cal ka BP; b). The direction of the Kuroshio Current at the LGM is after [Ujiié](#)
699 [and Ujiié \(1999\)](#).

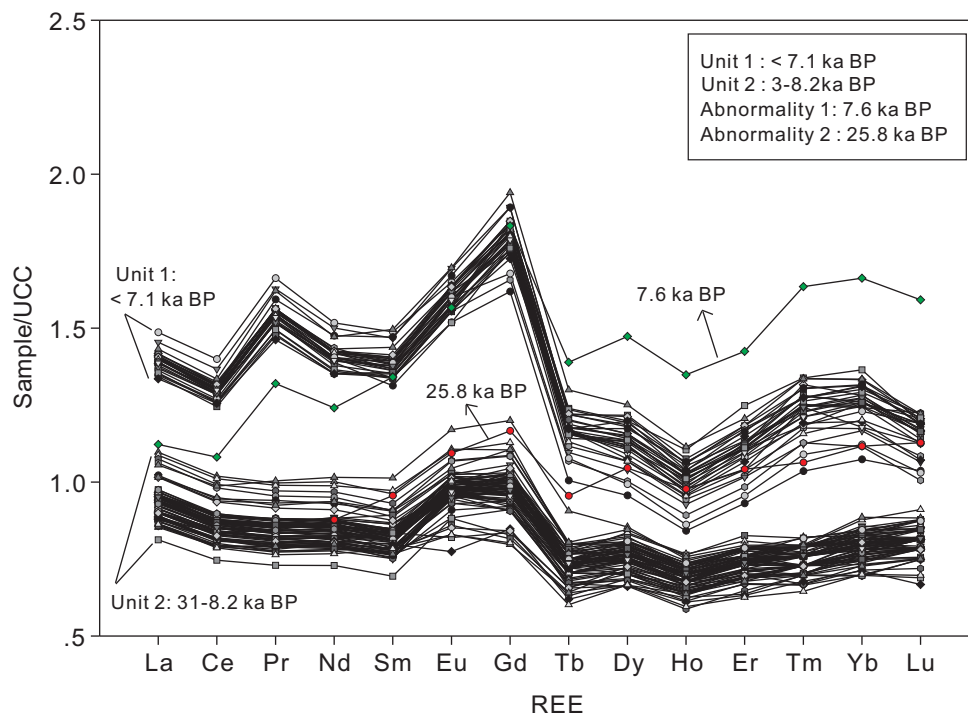
revised Figure 1

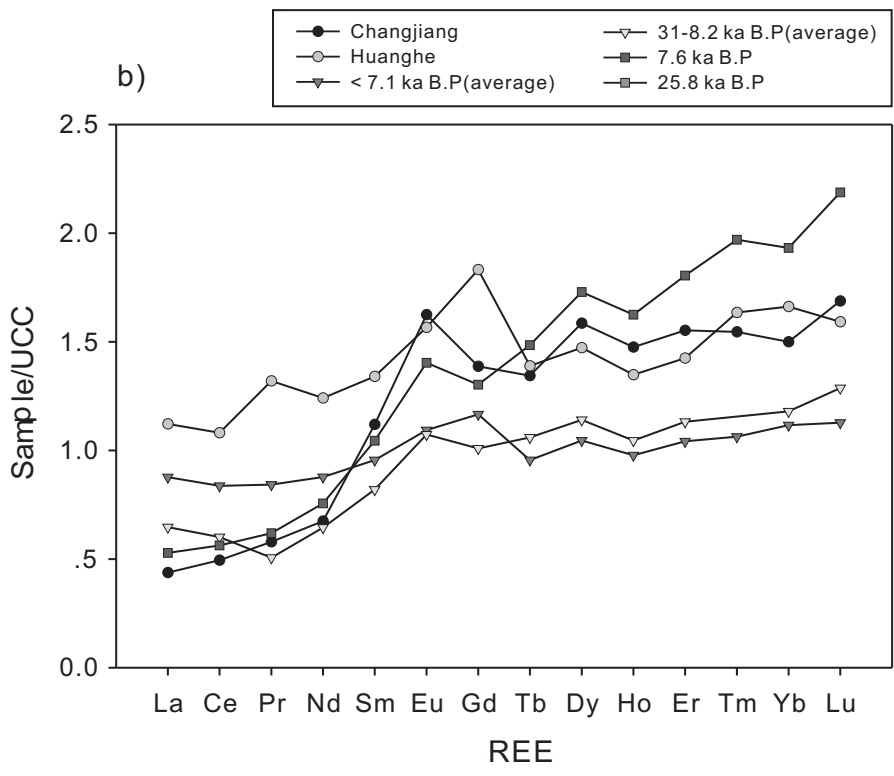
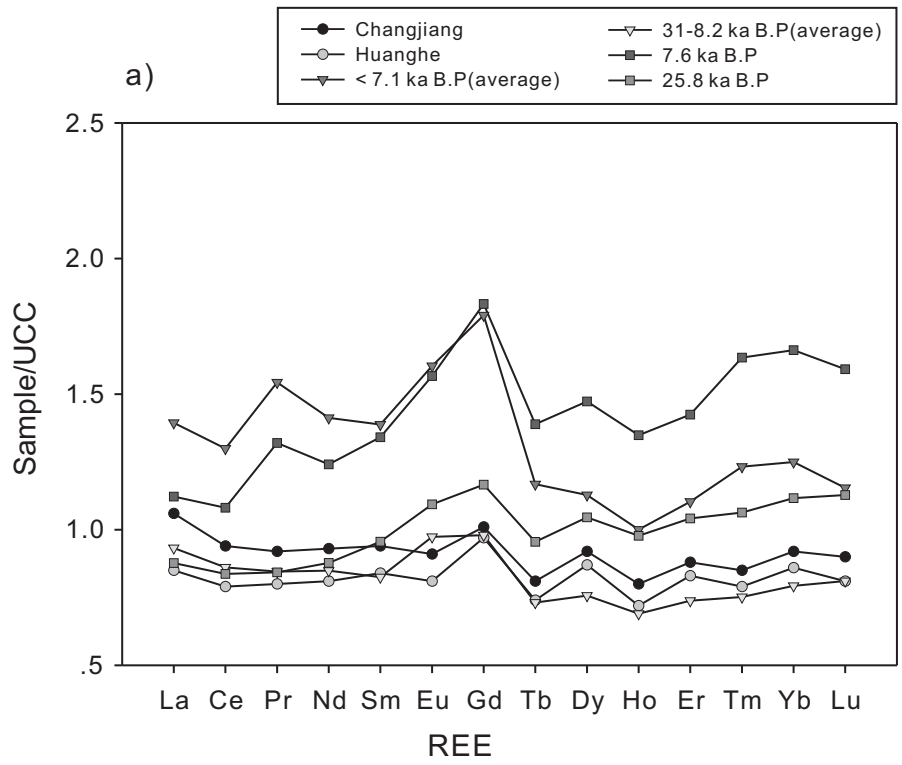


revised Figure 2

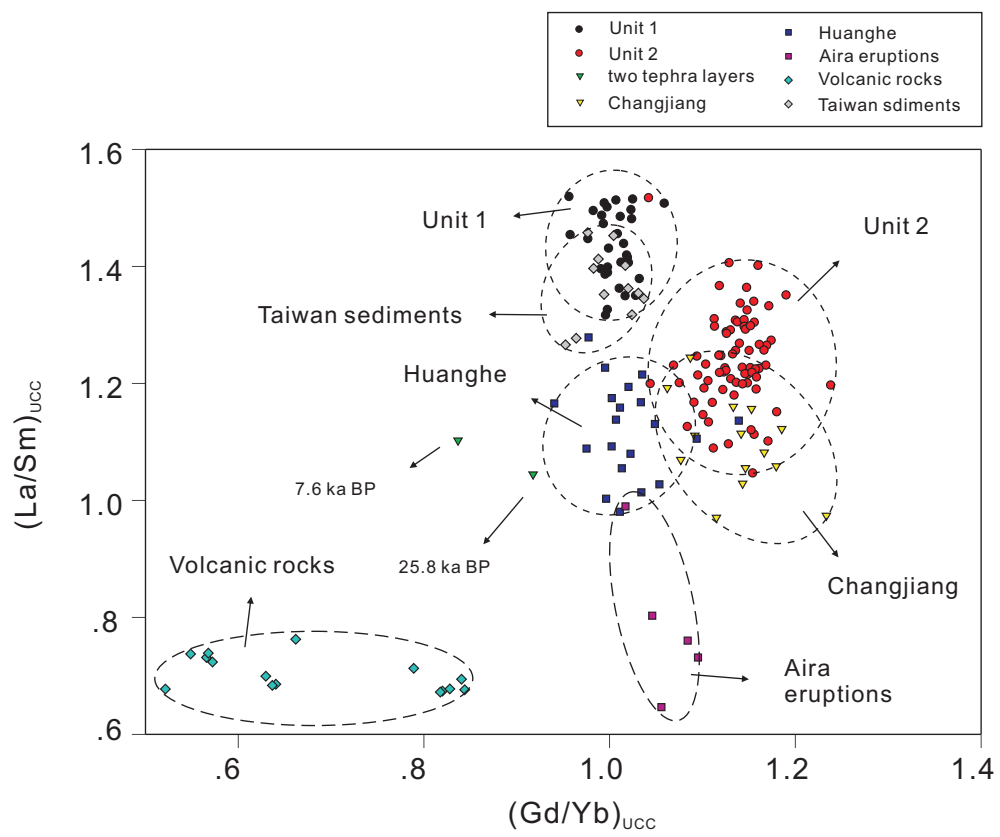


revised Figure 3





revised Figure 5



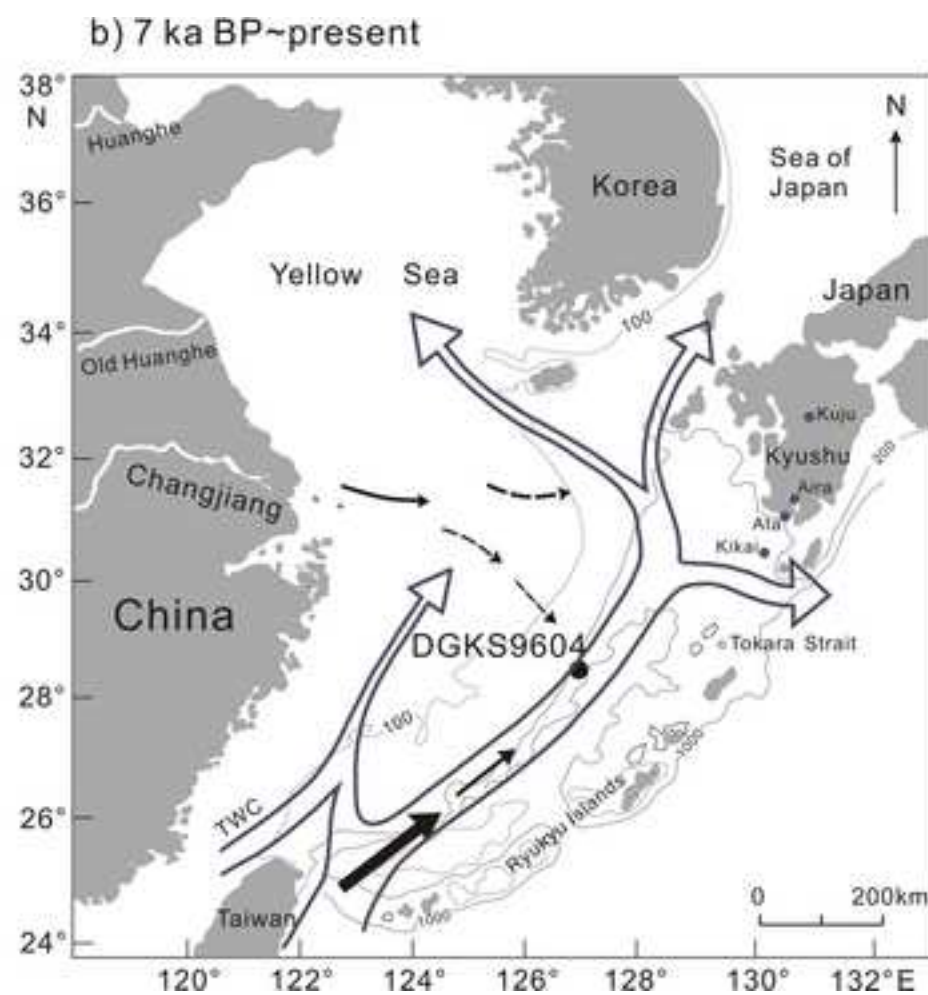
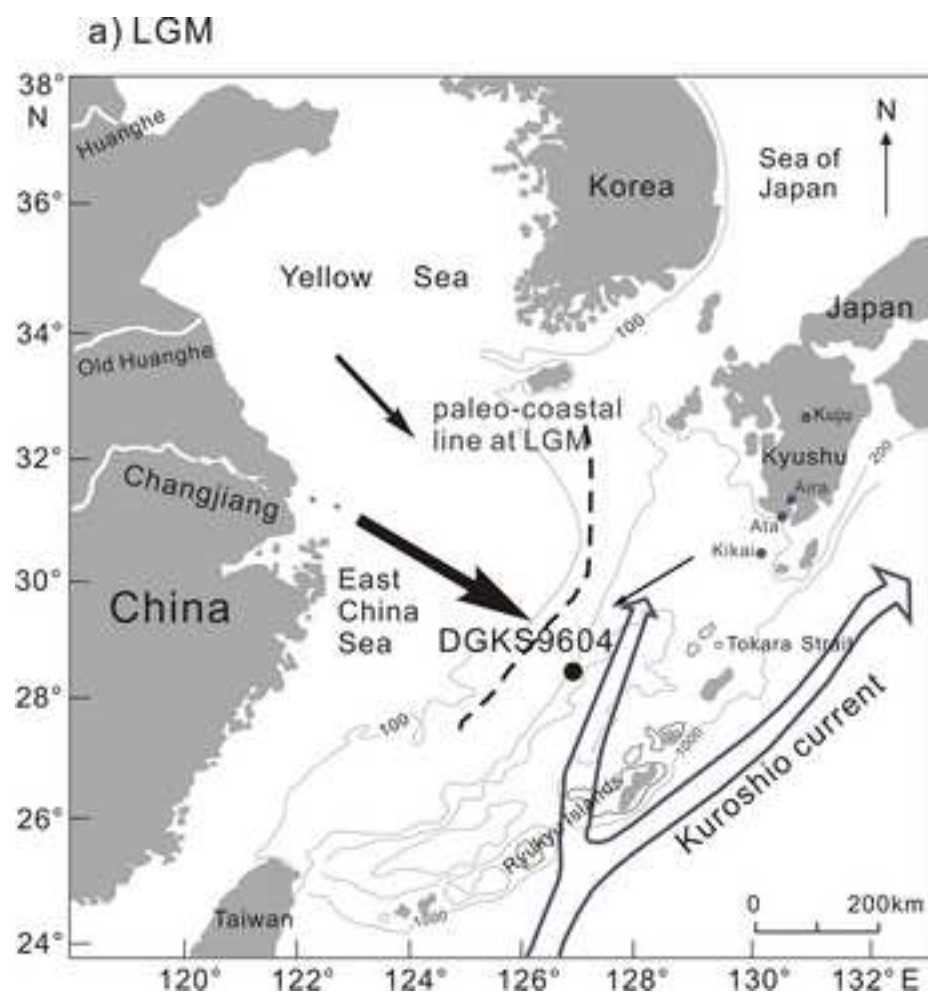


Table 1

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)	Σ REE (ppm)	δ Eu	δ Ce	(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: Σ REE denotes total REE concentrations (ppm) \pm 1 standard deviation; δ Eu and δ Ce 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.

