

Changes in environment and provenance within the Changjiang (Yangtze River) Delta during Pliocene to Pleistocene transition



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ABSTRACT

This study investigates heavy mineral assemblages in sediments of the modern Changjiang Delta (Yangtze River) and the Quaternary/Pliocene (Q/N) boundary strata within the delta area with the aim of deciphering changes in sediment provenance and the depositional environment during the Pliocene to Pleistocene transition. Major heavy mineral assemblages in the modern Changjiang sediments originate from their provenance rocks, while the distribution of unstable (amphibole), ultrastable (zircon, tourmaline, and rutile), and altered minerals (limonite, leucoxene) are closely related to the climate and the hydrodynamic environment within the catchment. The ratio of a certain unstable mineral (amphibole) to an altered mineral (limonite) is considered to be a sensitive indicator of the chemical weathering intensity that is largely affected by climate conditions. Heavy mineral data of core sediments reveal distinctly different environments and provenances between the upper (Quaternary) and lower (Pliocene) sedimentary strata in the Changjiang Delta. The Pliocene sediments are characterized by abnormally high contents of limonite (42%), leucoxene (8%) and zircon (15%), but an extremely low amphibole content (5%). Enrichment of zircon is indicated from the mid-lower Changjiang catchment, where granitoid rocks are widely distributed. The strong enrichment of altered minerals and the considerable great loss of unstable minerals suggest that Pliocene weathering was much stronger than it is nowadays. In contrast, high contents of amphibole, garnet, and pyroxene, and the low stable mineral content in the Quaternary strata indicate that sediment provenances may have extended to the upper Changjiang catchment, which underwent weak chemical weathering. These results imply that Changjiang as a large river system might have drained the East China's continental margin prior to the Q/N transition. The distinct heavy mineral assemblages found at the Q/N boundary in the Changjiang Delta are indicative of a drastic environment with provenance changes in response to intensive neotectonics. It is thus inferred that sensitive heavy mineral indices can be used to indicate the Q/N boundary in the Changjiang Delta, where reliable geochronological proxies have rarely reached an agreement.

1. Introduction

With the intensive uplift of the Tibetan Plateau and drastic climate change during the transition from the Pliocene to Pleistocene, the East Asian continent and marginal seas witnessed significant landscape evolution and terrigenous material cycling (Raymo and Ruddiman, 1992; Peizhen et al., 2001; Clark et al., 2004; Clift, 2006; Shen et al., 2017). In addition, the interaction between neotectonics and the monsoon climate during the late Cenozoic enabled large river systems

to transport huge Tibetan Plateau-derived terrigenous sediments, which were buried in continental margins (France-Lanord and Derry, 1997; Owen et al., 2005; Yang et al., 2006a; Clift et al., 2008; Van Hoang et al., 2009; Liu et al., 2017, 2018). Therefore, the evolutionary histories of tectonics, landscape, and paleoclimate are well preserved in the late Cenozoic fluvial sediments that accumulated in large deltas and marginal seas. These major depocenters and sinks of fluvial sediments in continental margins are thus ideal archives that can be analyzed to reconstruct paleoenvironmental evolution and land-sea interactions on

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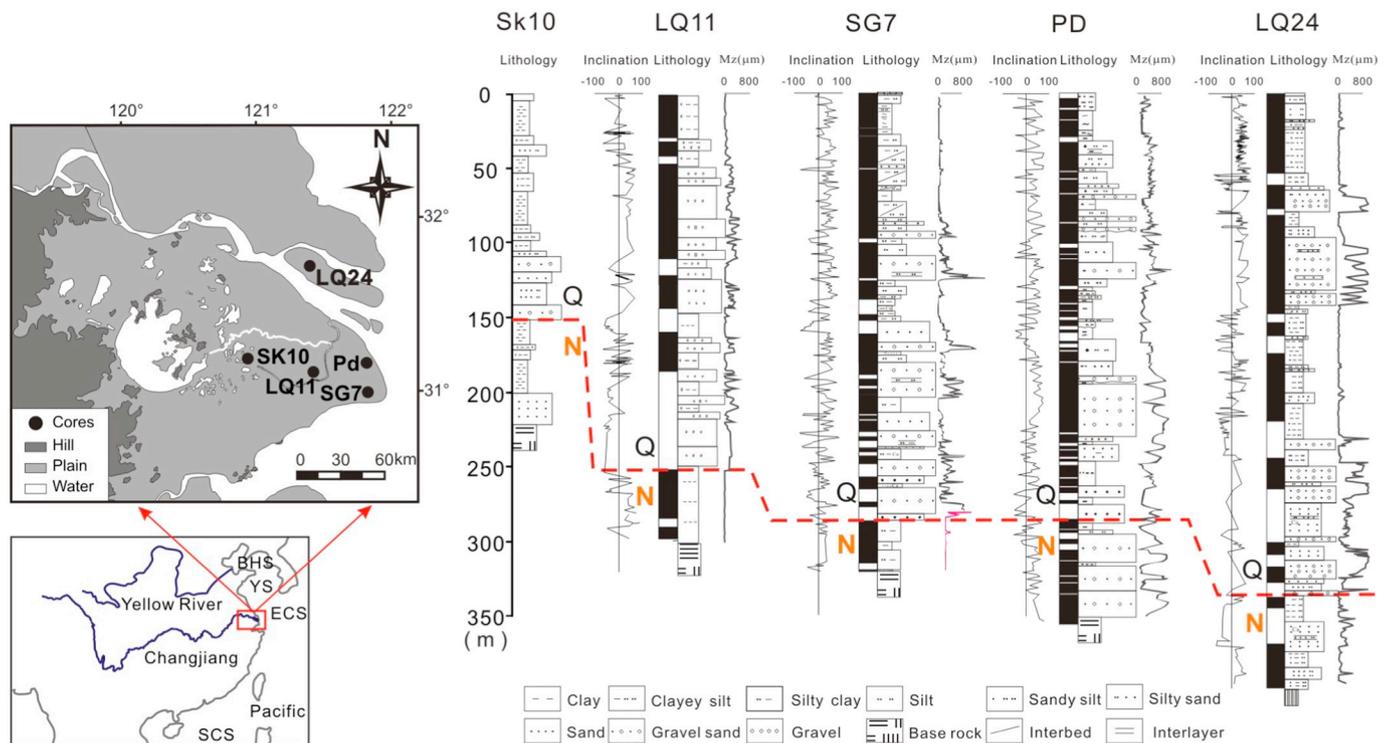


Fig. 1. The Plio-Pleistocene magneto-stratigraphy of the Changjiang Delta (Lithology and paleo-magnetic dating of cores SK10, Pd, SG7, LQ11, and LQ24 were respectively from Chen et al., 2007; Chen et al., 2009; Yue et al., 2016; Liu et al., 2018; and Yue et al., 2018; the red dotted line represents the inferred Q/N boundary based on paleo-magnetic dating result; Mz = median size, N = Neocene, Q = Quaternary, BHS = Bohai Sea, YS = Yellow Sea, ECS = East China Sea, SCS = South China Sea). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

multiple temporal scales (Clift et al., 2002; Giosan et al., 2006; Bishop, 2007; Yang et al., 2008; Bianchi and Allison, 2009; Gao et al., 2016; Gugliotta et al., 2017).

The Changjiang (Yangtze River) is the longest river in Asia; it originates in the eastern Tibetan Plateau and drains through central China before entering the East China Sea. The large Changjiang Delta is one of major depocenters in eastern China during the late Cenozoic, and has been continuously fed by riverine sediments since the Pliocene, with thicknesses of ~300 m siliciclastic sediment overlain above bedrocks (Fig. 1; Chen et al., 1997; Gu et al., 2014). Over the last two decades, the sediment provenance, paleoenvironment and river system evolution during the late Cenozoic have been widely studied by sedimentary stratigraphy, geochemistry, heavy-mineral analysis, and single mineral geochemistry (e.g. monazite, zircon, feldspar, muscovite and magnetite), with the major target areas including the First Bend and Three Gorges in the upper reaches, Jiangnan Basin and delta area in the mid-lower reaches (Kirby et al., 2002; Fan et al., 2004; Yang et al., 2006b; Van Hoang et al., 2010; Richardson et al., 2010; Zheng et al., 2013; Zhang et al., 2014; Tada et al., 2016; Wei et al., 2016; Yue et al., 2018). Such researches have greatly improved our understanding Changjiang River system evolution in the late Cenozoic; however, it has yet been clarified how the delta region responded to the river evolution. Previous studies on the provenance and environmental changes in the Changjiang Delta focus on the late Quaternary, and long and continuous sedimentary records have rarely been investigated. In particular, there is a lack of integrated investigations on the relationship between sediment transfer process in the catchment and paleoenvironmental changes since the Pliocene.

However, the reconstruction of Plio-Pleistocene chronostratigraphy is a challenging problem when using the long sedimentary records in the Changjiang Delta, and it is almost impossible to apply biostratigraphy because the lack of index fossils. Although paleomagnetism was commonly applied (Rea et al., 1998; Ding et al., 1999; Wang et al., 2005a, 2005b; Liu et al., 2014; Duan et al., 2016), ubiquitous sand

layers prevent direct and reliable comparisons with the Standard Geomagnetic Polarity Schedule, especially with respect to the boundary of Pleistocene and Pliocene (Q/N). The magnetostratigraphy of the Chinese Loess Plateau has been well studied (Nat et al., 1990; Spassov et al., 2001; Liu et al., 2015; Zhang et al., 2016), but the sedimentary environments of the delta and coastal regions are different from the successive sedimentation of the Loess Plateau. As they are complex and very dynamic, this has resulted not only in great sedimentary facies diversity, but also ubiquitous sedimentary hiatus in the strata (Fig. 1). In addition, the well-developed coarse sediment layers are not suitable for paleomagnetic dating. Therefore, the Q/N stratigraphic boundary in the Changjiang Delta has yet to be reliably reconstructed due to these factors.

However, climate change occurring during the transition from the Neogene to Quaternary is likely to have made an imprint on fluvial sediments (Shackleton and Opdyke, 1977; An et al., 2001; Ravelo et al., 2004; Clift and Blusztajn, 2005), which thus provides an alternative solution to the establishment of Q/N boundary. In addition to being used to indicate sediment provenance and depositional environments, detrital minerals have been successfully applied in paleoclimatic reconstruction (Foucault and Stanley, 1989; Dill, 1995; Schirmeister et al., 2002; Sinha et al., 2006; Peng et al., 2016; Song et al., 2018). Strong chemical weathering in a warm and wet climate generally results in relatively high mineralogical maturity, which is shown as a high content of stable minerals (such as quartz, ilmenite, zircon, tourmaline, and rutile); but also by a significant decrease in unstable minerals content (such as pyroxene, amphibole, and biotite) (Morton, 1982; Hessler and Lowe, 2017; Garzanti, 2017). In contrast, weak chemical weathering in a low temperature environment (such as glacial period) results in the relative enrichment of unstable minerals. In this respect, therefore, heavy-mineral analysis can be used as a potential indicator of both sediment provenance and environment change.

This study reports an investigation of heavy mineral assemblages in sediments from the modern Changjiang River system and from the late

Cenozoic strata in the delta area. The main aim of this study is to investigate sediment provenances and paleoenvironmental changes, verify the evidence showing that these changes were triggered by neotectonics, and to further explore the possibility of using heavy minerals to indicate the Q/N stratigraphic boundary in the dynamic coastal region.

2. Regional setting and the late Cenozoic stratigraphy

2.1. Geological and geographical backgrounds of the Changjiang River

The Changjiang River is one of the largest rivers in the world; and has a length of over 6300 km and a drainage basin of $1.8 \times 10^6 \text{ km}^2$ (Chen et al., 2001). It originates from the eastern Tibetan Plateau, and flows eastward across the YunGui Plateau, Sichuan Basin, Three Gorges, and Jiangnan Basin before finally entering the East China Sea (Fig. 1). According to a long-term observation at hydrological gauge stations from 1951 to 2016, it annually discharges $900 \times 10^9 \text{ m}^3$ fresh water and 390×10^6 tons sediments into the delta and East China Sea (Changjiang Water Resources Commission, 2016).

Geologically, the Changjiang catchment mainly comprises five tectonic units: including the Qamdo Block, the Songpan-Garze terrane, the Qinling-Dabie orogenic belts, the Cathaysia Block and the Yangtze Craton. The basement of Changjiang drainage covers a variety of rocks. The upper and middle basins are characterized by widely distributed Paleozoic sedimentary rocks, including carbonate rock and Jurassic red sandstone, Archaean-Cenozoic metamorphic rocks, and Mesozoic-Cenozoic igneous rocks (Changjiang Water Resources Commission, 1999). The globally recognized large E'meishan basalt, which is uniquely distributed in the upper Changjiang valley (where it occupies an area of approximately $2.5 \times 10^5 \text{ km}^2$) (Chung and Jahn, 1995; Song et al., 2009), was formed in the late Permian; and the middle-lower Changjiang basin contains different rock types but is dominated by Mesozoic intermediate-acidic igneous rocks, Pre-Paleozoic metamorphic rocks, Paleozoic sedimentary rocks, and Quaternary unconsolidated sediment (Changjiang Water Resources Commission, 1999). Furthermore, Mesozoic granite covers the middle-lower Changjiang mainstream, and medium-low grade metamorphic rocks are widely distributed in Qinling-Dabie orogenic belts.

The Changjiang River is usually classified into three reaches according to these geomorphological characteristics. The upper reaches span from the header water to the Three Gorges near Yichang, covering a length of > 4000 km with a drainage basin area of $1.0 \times 10^6 \text{ km}^2$, and the relief in the upper Changjiang is mainly elevated plateau and high mountains. The middle reaches are from Yichang to Hukou (Fig. 2), and the relief is mainly low with meandering platforms. The lower reaches extend from Hukou to the river mouth in Shanghai; this reach spans a length of approximately 1000 km and has a drainage area of about $0.1 \times 10^6 \text{ km}^2$.

The Changjiang drainage is mainly located within the sub-tropical climate zone, but the climatic regime differs widely between the upper and middle-lower reaches (Fig. 2). Affected by both latitude and elevation, the mean annual temperature in the Changjiang basin is higher in the south and east. In the middle-lower reaches, it is generally higher than 16°C except for the upper reaches of the Hanjiang River (ca. 14°C), which are situated at a much higher latitude than the other drainage basins (Fig. 2). The upper reaches have the mean average temperature mostly below 12°C , and the source region is below freezing point (Fig. 2). Strongly affected by the Asian monsoon, most precipitation in the Changjiang catchment occurs in spring and summer (April to October) and it increases in an eastern direction. The Tibetan Plateau is located in semi-humid and semi-arid zone, and the annual precipitation is only about 300 mm. The Yun-Gui Plateau, Sichuan Basin and the Three Gorges have a subtropical humid climate with the annual precipitation ranging from 1000 mm to 1400 mm, and the middle-lower valley is controlled by East Asian monsoon and its mean

annual precipitation reaches 1600 mm (Changjiang Water Resources Commission, 1999).

2.2. Sedimentary stratigraphy of the Changjiang Delta since the Pliocene

The Changjiang Delta and its coastal sedimentary system have been built up by huge amounts of sediments derived from its large catchment, and unconsolidated sediments have thicknesses between 200 and 400 m (Fig. 1; Chen et al., 1997; Gu et al., 2014). The Pliocene-Quaternary strata are thicker towards the east and range from 30 m to 400 m (Fig. 1; Chen and Stanley, 1995), and the magnetostratigraphic boundary of Matuyama/Gauss (M/G) in the delta is generally located at depths of 160–300 m (Fig. 1; Qiu and Li, 2007).

The previously-recognized Pliocene strata (20–100 m thick) were deposited unconformably on Mesozoic and Cenozoic intermediate-acidic igneous and metamorphic bedrocks. The Pliocene sediments are featured by indurated silty clay embedded in one to two fining upward sequences. Each sequence consists of yellow or grey sand of 5–10 m thick at the bottom and grey silty clay of 5–10 m thick at the top. Poorly-sorted gravels with maximum diameters of up to ca. 5 cm occur widely within the coarse sediments and are indicative of alluvial fans or a meandering river environment (Wang et al., 2005a; Qiu and Li, 2007). In the fine sedimentary facies, calcareous and iron-manganese nodules are widely observed. In addition, a block of grey calcareous sands occur at the bottom strata. No microfossils are observed in sediments of the Pliocene.

The Quaternary sediments in the delta region consist of several upward-grading sedimentary sequences and are generally of thicknesses between 50 m and 300 m. All sequences have similar characteristics, with gravelly sand at the bottom and clayey silt at the top, which, together with the sedimentary structures, implies that they are fluvial and alluvial facies (Chen et al., 1997; Wang et al., 2005a; Fig. 1). Microfossils (including foraminifera) can be frequently observed in the mid-late Quaternary sediments, which indicates that several marine transgressions occurred in eastern China (Chen et al., 1997; Li et al., 2001; Yang et al., 2006a; Yue et al., 2018). The Holocene sediments consist of dark to grey clayey silts or yellowish sandy silts; they are representative of tidal and deltaic facies.

3. Materials and methods

3.1. Sample sources

A total of 27 surface sediment samples were collected on the floodplains of major tributaries, the mainstream of the Changjiang during the dry season (November 2012 and May 2018), and in the upper Jinshajiang River (downstream to the estuary) (Fig. 2). Of the 27 samples, 15 were obtained from main tributaries of the Changjiang and 12 from the mainstream (Fig. 2). For each sample, approximately 500 g of fine-grained sediment was taken (using a clean spoon) from the subsurface of the floodplain below a depth of 20 cm.

A borehole measuring 301 m (LQ11) was drilled in the Changjiang Delta within Shanghai ($31^\circ2.5'N$, $121^\circ23.9'E$, Fig. 2). The borehole was split, logged, and sampled successively in the laboratory. A grain size analysis of a total of 279 subsamples obtained using a sampling interval of ca. 1.0 m was made using the Beckman Coulter Laser Diffraction Particle Size Analyzer (LS-13320). To establish magnetostratigraphy of core LQ11, 539 fine subsamples, taken at sampling interval of 0.2–1.5 m, were analyzed using a magnetometer (2G-755R) at the Institute of Earth Environment in the Chinese Academy of Sciences. The detailed paleomagnetic dating results is presented in Liu et al. (2018). In addition, data from three boreholes (SK10, LQ24, and PD) extracted in the Changjiang Delta were also collected to conduct a stratigraphic correlation and comparison (Fig. 1). The heavy mineral data from these three cores are reported in Chen et al. (2007, 2009) and Yue et al. (2018).

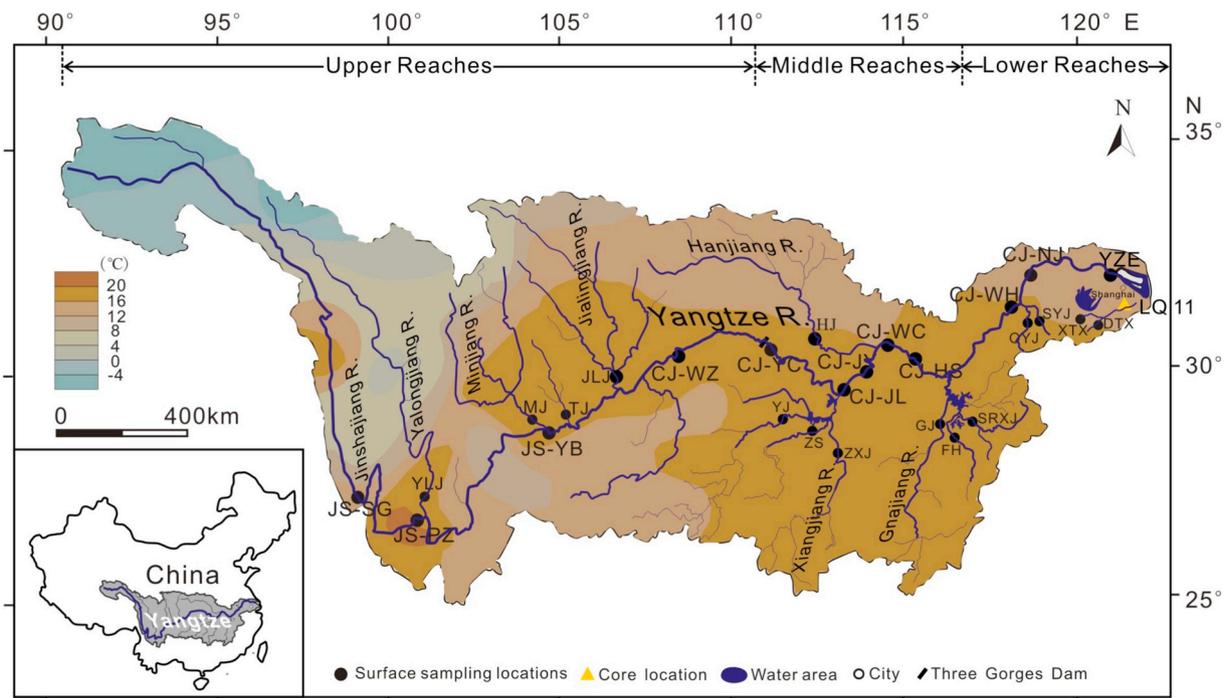


Fig. 2. Surface sampling locations and isotherm in the Changjiang catchment (modified after Changjiang Water Resources Commission, 1999) (The letter “R.” is the abbreviation of “River”; The capital letters indicate: JS-SG = Jinshajiang near Shigu Town, YLJ = Yalongjiang, JS-PZ = Jinshajiang near Panzhihua City, JS-YB = Jinshajiang near Yibin City, MJ = Minjiang, TJ = Tuojiang, JLJ = Jialingjiang, CJ-WZ = Changjiang near Wanzhou City, CJ-YC = Changjiang in the Three Gorges; CJ-JL = Changjiang near Jianli County, YJ = Yuanjiang, ZS = Zishui, XJ = Xiangjiang, CJ-WC = Changjiang near Wuhan City, HJ = Hanjiang, CJ-JY = Changjiang near Jiayu County, GJ = Ganjiang, FH = Fuhe, SRXJ = Xinjiang, CJ-HS = Changjiang near Huangshi City; CJ-WH = Changjiang near Wuhu City, QYJ = Qingyijiang; SYJ = Shuiyangjiang, DTX = Dongtiaoxi, XTX = Xitiaoxi; CJ-NJ = Changjiang near the Nanjing City, YZE = Changjiang estuary).

3.2. Heavy mineral analysis

Heavy mineral analysis was conducted on 27 surface samples obtained from the Changjiang River and 24 core subsamples from core LQ11 at a sampling interval of ca. 10 m. Heavy mineral separation was conducted following the standard procedures described by Mange and Maurer (1992). In our study, a wide grain size window strategy was used following the methods of Garzanti et al. (2009) and Peng et al. (2018). In this respect, a sediment fraction of 32–250 μm was obtained through wet sieving, and it was then separated using sodium polytungstate (at a density of 2.90 g/cm^3 at 20 °C) to collect heavy minerals. More than 200 transparent heavy mineral grains were counted to identify heavy minerals using the ribbon counting method (Galehouse, 1971), and the concentration of each heavy mineral is presented as a mass percent in total coarse silt and fine sand fraction (Garzanti and Andò, 2007a, 2007b). In addition, to analyze surficial weathering, recycling, and diagenesis, altered limonite and leucoxene minerals were identified and counted, and their concentrations were counted as the grain percentages of total heavy minerals and calculated separately from transparent heavy minerals. A scanning electron microscope (SEM) equipped with an energy-dispersive spectrometer (EDS) was employed to examine these minerals at the State Key Laboratory of Marine Geology at Tongji University.

4. Results

4.1. Heavy minerals in modern Changjiang River system

The heavy mineral compositions of Changjiang surface sediments are shown in Table 1. The heavy mineral concentrations (HMC) vary from 0.3% to 12%, which shows that unstable and stable minerals contents vary between the upper and mid-lower Changjiang reaches. Sediments from the upper Changjiang mainstream and tributaries have

HMCs of ca. 0.7–12%, with generally similar heavy mineral assemblages of unstable amphibole (46% on average, range of 25–66%), epidote (mean 28%) and clinopyroxene (19% on average). The altered mineral limonite accounts for only 12% (5–22%). The other stable minerals, such as ZTR (zircon + tourmaline + rutile) and leucoxene are in minor abundance, and each has a content of < 1% (Fig. 3).

In comparison, the HMCs in middle Changjiang tributaries range from 0.3% to 1.7%, but are mostly < 1%, with heavy mineral assemblages dominated by high stable and altered mineral contents (Fig. 3; Yang et al., 2009; Yue et al., 2018). In particular, the mean ZTR is approximately 31%; and this is the highest value found in any samples. Limonite is also highly concentrated in middle tributary samples at an average of 37% and reaching a maximum of 64% in Xiangjiang sediment. Similar to the middle tributaries, the lower tributaries are also characterized by lower HMCs, but higher ZTR and limonite. In addition, a small amount of leucoxene (mean 2%) is commonly found in these tributaries. There is a considerable decrease in the content of amphibole in the mid-lower tributaries (mostly < 30%) except for in the sample from the Hanjiang River (50%).

The mean HMC in the middle Changjiang mainstream is approximately 5%, and it is dominated by amphibole (44% on average), epidote (38%), and clinopyroxene (12%), while the lower Changjiang mainstream has high contents of the unstable mineral, amphibole (average of 57%). Although clinopyroxene decreases significantly towards the lower mainstream, its content still reaches 6% on average; but this is much lower than in the upper mainstream (Fig. 3). Compared to the mid-lower tributaries, the mid-lower mainstream samples are featured by lower amounts of stable and altered minerals; limonite and ZTR account for 6%, and < 1% respectively, and no leucoxene is found in these mid-lower mainstream samples.

Table 1
Heavy mineral compositions in sediments from the modern Changjiang river system (unit: %).

	Samples	HMC	Amp	Ep	Cpx	ZTR	Grt	other	Lm	Leu
Upper tributaries	MJ	0.9	63	26	3	1	3	3	8	0
	JLJ	0.7	54	31	2	1	8	4	14	0
	TJ	1.7	66	24	2	1	3	5	7	0
Upper mainstream	YLJ	12.4	33	26	39	0	0	2	8	0
	JS-SG	1.4	59	34	2	1	3	1	14	0
	JS-PZ	11.5	32	23	42	0	1	1	5	0
	JS-YB	3.3	25	29	40	1	4	0	17	0
	CJ-WZ	5.0	45	33	14	0	7	2	17	0
Middle tributaries	CJ-YC	8.4	37	28	30	0	2	2	22	0
	HJ	0.9	50	46	1	0	2	0	8	0
	ZXJ	0.3	29	24	0	43	2	2	64	2
	YJ	0.5	23	52	0	22	3	0	50	4
	ZS	1.7	31	28	0	36	2	2	51	1
	SRXJ	0.9	14	58	0	21	6	1	33	3
	GJ	0.4	17	37	0	43	1	2	41	2
Middle mainstream	FH	0.5	21	50	0	23	3	3	36	2
	CJ-JL	2.9	42	36	18	1	2	1	15	0
	CJ-WC	5.8	55	34	7	0	3	1	10	0
	CJ-JY	5.4	43	37	15	0	4	1	17	0
Lower tributaries	CJ-HS	5.1	38	46	7	0	6	2	12	0
	DTX	0.5	29	59	0	5	1	6	26	1
	XTX	1.0	25	67	0	5	2	1	28	1
	SYJ	0.5	21	56	0	17	4	1	32	2
Lower mainstream	QYJ	0.8	28	59	0	11	2	0	22	1
	CJ-WH	5.8	53	33	10	0	2	1	5	0
	CJ-NJ	5.3	62	25	5	2	5	3	9	0
	YZE	4.6	57	36	2	0	2	2	5	0

Note: HMC = heavy mineral concentration, Amp = amphibole, Ep = epidote, ZTR = zircon + tourmaline + rutile, Grt = garnet; Lm = limonite, Cpx = clinopyroxene, Leu = leucoxene.

4.2. Heavy minerals in boreholes from Changjiang Delta

In total, 36 different heavy mineral species are identified in sediments from Core LQ11 (Table 2), and based on the vertical heavy mineral distribution, two distinct Units (I and II) are recognized (Fig. 4), where the boundary between the two units is located at a depth of ca. 200 m. The transparent heavy mineral assemblages in the core sediments are generally dominated by amphiboles and epidote, and other heavy minerals (zircon, tourmaline, garnet, and apatite) are present in minor amounts (Fig. 4).

4.2.1. Unit I (301.0 m to 186.0 m)

The HMC is very low at the bottom of core, (mostly < 1%), and the heavy mineral assemblage is featured by high stable mineral concentrations and low unstable mineral concentrations (Table 2, Fig. 4). In this section, amphibole only accounts for an average of 5% (2–10%), while the contents of limonite and leucoxene are 27–79% and 1–23%, respectively. In addition, the stable minerals, ZTR, are prevalent and the ZTR content is the highest (18% on average and ranging from 7 to 38%) of all borehole samples. Furthermore, authigenic minerals (such as pyrolusite) are found sporadically throughout in this unit.

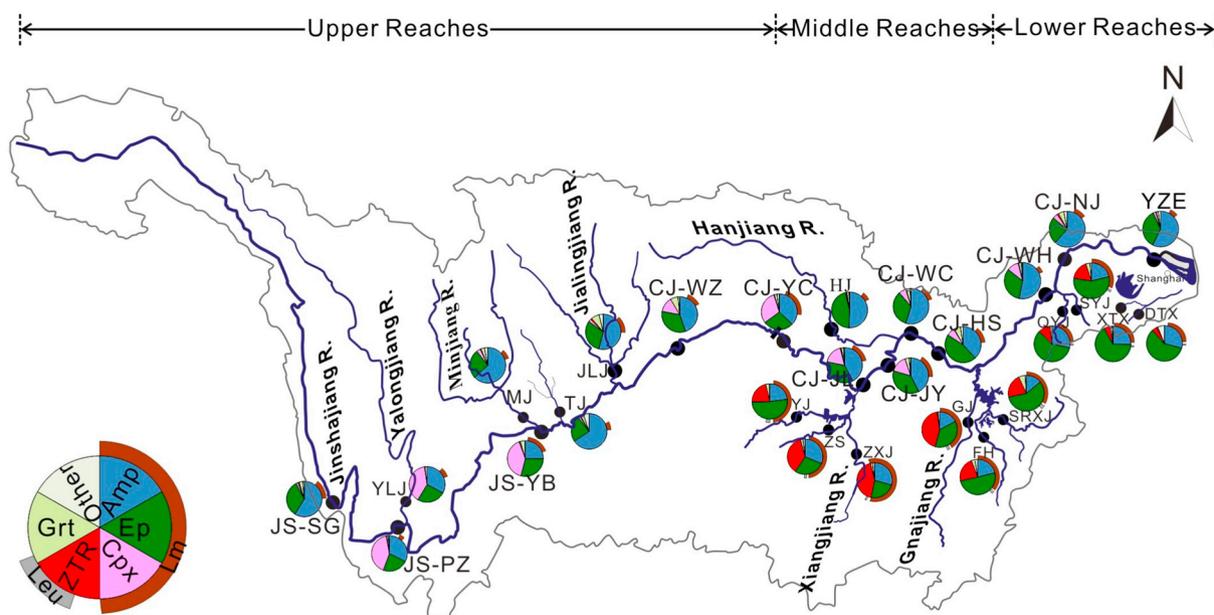


Fig. 3. Heavy mineral distributions in the surface samples of the Changjiang (Amp = amphibole, Cpx = clinopyroxene, Lm = limonite, Leu = leucoxene, ZTR = zircon + tourmaline + rutile). Please see Fig. 2 for the explanation of capital letters.

Table 2
Compositions of major heavy minerals in Core LQ11 from the Changjiang Delta (unit: %).

Depth (m)	HMC	Amp	Ep	Cpx	Zrn	Tur	Rt	Grt	Lm	Leu
3.2	1.3	68	27	1	0	0	0	1	8	0
12.6	2.0	67	29	1	0	0	0	1	7	0
20.9	1.3	64	31	2	0	1	0	1	6	0
29.9	0.8	66	31	2	0	0	0	1	3	0
32.7	1.6	62	31	3	0	0	0	1	5	0
37.6	2.0	64	33	1	0	0	0	0	7	0
44.7	1.3	58	34	2	0	1	0	2	3	0
54.1	0.6	60	34	2	0	0	0	2	5	0
60.4	4.1	65	28	1	1	0	0	3	3	0
72.4	1.5	61	34	1	0	0	0	1	5	0
84.2	8.7	62	30	2	0	1	0	2	5	0
92.5	8.3	54	42	1	0	0	0	2	23	0
108.2	1.3	50	46	0	0	0	0	3	9	0
131.7	2.1	63	36	0	0	0	0	0	6	0
143.8	5.3	58	40	1	0	0	0	0	6	0
162.0	2.4	29	64	1	0	1	0	3	11	0
172.8	0.7	55	42	0	0	0	0	1	23	1
186.0	0.5	2	75	0	17	3	2	0	79	1
198.0	1.1	4	81	0	11	1	0	0	34	3
204.4	0.9	3	81	0	11	1	0	2	30	5
216.7	0.6	8	71	0	16	2	0	2	30	23
238.3	0.5	2	59	0	36	0	1	0	52	12
265.5	0.6	10	70	0	11	2	0	3	27	13
297.0	0.4	6	83	0	3	3	1	3	41	2

Note: HMC = heavy mineral concentration, Amp = amphibole, Ep = epidote, Cpx = clinopyroxene, Zrn = zircon, Tur = tourmaline, Rt = rutile, Grt = garnet; Lm = limonite, Leu = leucoxene.

4.2.2. Unit II (186.0 m to 0.0 m)

Compared to Unit I, there is a dramatic change in the heavy mineral distribution in the mid-upper strata (Unit II; Table 2, Fig. 4). In this section, there is a marked increase in the unstable mineral content but a significant reduction in the stable minerals content. The heavy mineral assemblage is dominated by amphibole, and its content is much higher than other minerals (ranging from 28% to 68%). In addition, there is a remarkable decrease in the stable and altered mineral contents, and the

average contents of limonite and ZTR are 8% (3–23%) and < 1% (0–2%), respectively. Other altered and authigenic mineral, such as psilomelane and pyrolusite, are rarely found. The dramatic increase in unstable heavy minerals corresponds to the relatively high HMC (mean 3%), which is approximately three times higher than in Unit I.

5. Discussion

5.1. Controls on heavy mineral assemblages in modern Changjiang River system

The heavy mineral distribution of siliciclastic depends on their original source rocks, and other factors (such as chemical weathering, hydraulic sorting, and diagenesis) have an influence on the heavy mineral assemblages (Morton, 1985; Nechaev and Isphording, 1993; Garzanti and Andò, 2007a, 2007b; Yang et al., 2009; Jafarzadeh et al., 2014).

5.1.1. Influence of parent rocks

In the modern sediments from the upper Changjiang, a relatively high content of clinopyroxene (19% on average; Fig. 2) indicates they are influenced by basic-ultrabasic rocks in the upper valley where the large E'meishan basalt province (ca. 2.5×10^5 km² in area) is developed (Chung and Jahn, 1995; Song et al., 2009), and the ferromagnesian rocks contain a plenty of clinopyroxene minerals (Poldervaart and Hess, 1951; Song et al., 2009). Previous studies have shown that the upper Changjiang tributaries carry litho-feldspatho-quartzose sand that has a very rich amphibole-dominated suite (including epidote and clinopyroxene) (Yang et al., 2009; Vezzoli et al., 2016). Although clinopyroxene minerals are unstable naturally, they are still widely observed in the lower Changjiang mainstream, where they are more abundant than in the mid-lower Changjiang tributaries (Fig. 3). It can be inferred that clinopyroxene is mainly supplied from the upper Changjiang valley where the basic-ultrabasic rocks are widely distributed. This phenomenon has also been reported in other regions; for example, along the hyper-arid Atlantic coast of southern Africa, where weathering is negligible, most of the sand is derived long-distance from

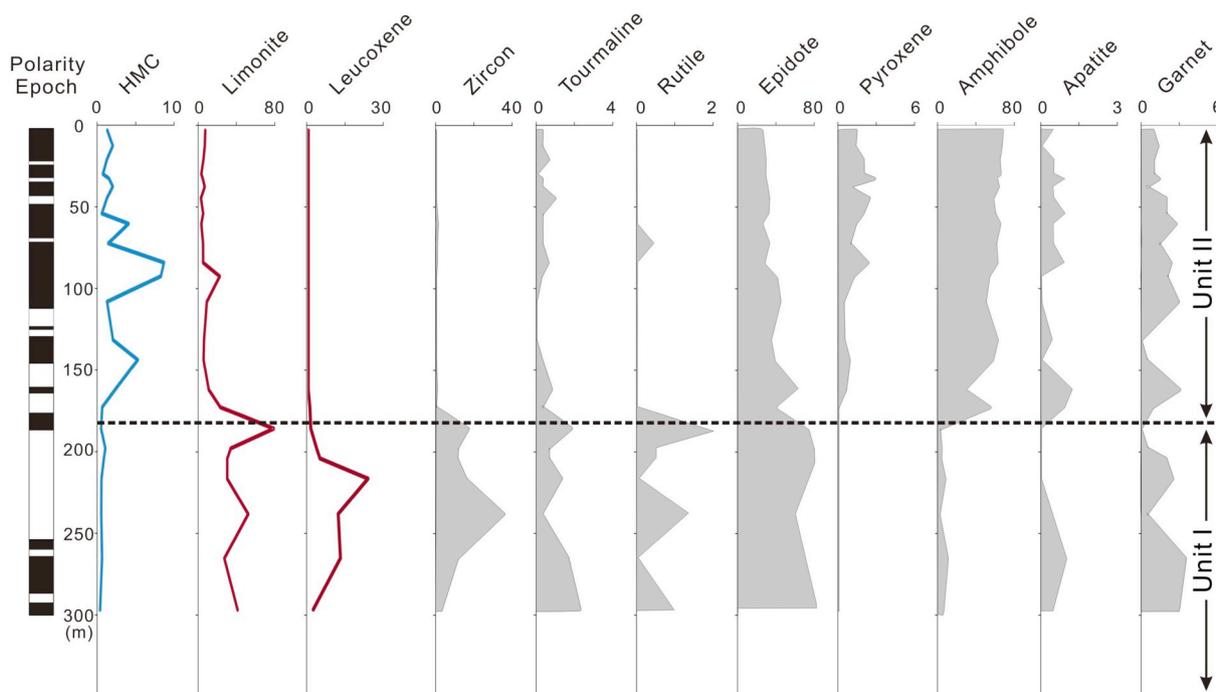


Fig. 4. Distribution of heavy minerals in borehole LQ11 (content unit: %; the dash line represents the depositional unit boundary with sharp changes in heavy mineral assemblages).

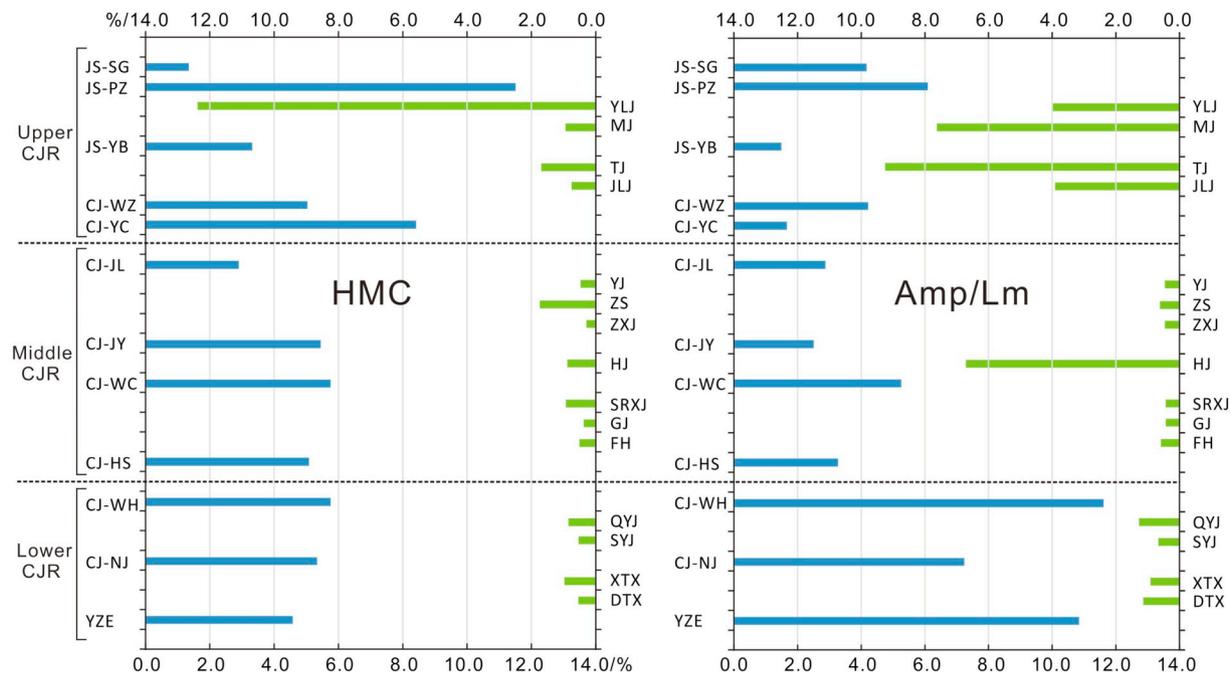


Fig. 5. Amp/Lm ratio (amphibole to limonite) and heavy mineral concentrations in different reaches of the Changjiang River system (CJR = Changjiang Reaches, the blue rectangles represent samples of the Changjiang mainstream, the green rectangles of the tributaries). Please see Fig. 2 for the explanation of capital letters. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the Orange River mouth, and basaltic rock fragments and pyroxene do not decrease relative to quartz, even after being transported for thousands of kilometers through high-energy fluvial, shallow-marine, and eolian environments (Garzanti et al., 2014, 2015; Garzanti, 2017).

In the mid-lower Changjiang tributaries, sediments samples are relatively rich in zircon and tourmaline, which implies the parent rocks are different from those in the upper Changjiang. There are large block of intermediate-acid igneous rocks distributed in the middle basin (Changjiang Water Resources Commission, 1999). The heavy mineral assemblages are evidence that igneous rocks can yield plenty of accessory minerals, such as zircon and tourmaline (Nechaev and Ispording, 1993). These results also show that the heavy minerals in different reaches of the Changjiang still mirror their provenance rocks in sub-catchments, even after undergoing complicated source-to-sink processes, such as mechanical breakdown, hydraulic sorting, and sediment recycling (Morton, 1985; Garzanti, 2017).

5.1.2. Specific heavy minerals indicating basin weathering intensity

Although the heavy mineral characteristics of detrital sediments are predominantly determined by the parent rocks, certain kinds of minerals are predominantly determined by chemical weathering (Morton and Hallsworth, 1999; Svendsen and Hartley, 2002; Garzanti, 2017). Heavy mineral assemblages sourced from the similar parent rocks differ widely with respect to the distinct weathering resistance of each mineral. In addition, stable and altered minerals are usually well preserved in hot climates, while there is a significant decrease in unstable minerals because they have undergone intense chemical weathering and dissolution (Morton, 1982; Morton and Hallsworth, 1999; Garzanti, 2017). As representative unstable minerals, olivine, pyroxene, amphibole, biotite, are sensitive to climate change; therefore, they decrease in high temperature environment with strong weathering. In contrast, olivine and pyroxene are strongly enriched in ultrabasic and basic rocks, and biotite is sensitive to hydrodynamic sorting (Garzanti et al., 2008; Jafarzadeh et al., 2014), which suggests that they are not suitable indicators for reflecting weathering intensity and climate conditions.

However, amphibole can be used as a potential indicator. Previous studies have shown that amphibole content accounts for ca. 20% of

riverine sediments of subtropical areas (SE Asia), which is much lower than amounts in the temperate area of the Changjiang and Huanghe (Yellow River) (ca. 40%) (Garzanti and Andò, 2007a, 2007b; Yang et al., 2009; Sevastjanova et al., 2012; Nie et al., 2015). Stable minerals such as ZTR are completely different from unstable minerals and are resistant to high temperature and strong weathering conditions, leading to high ZTR values (Hubert, 1962; Dill, 1995). For example, ZTR can reach up to 50% in riverine samples from tropical regions (Sevastjanova et al., 2012).

In our study, there are differences in the compositions of unstable and altered minerals that are susceptible to chemical weathering between the upper (above the Three Gorges) and mid-lower Changjiang tributaries (Poyang Lake, Dongting Lake, Hanjiang River, Qingyi River, Shuiyang River, and Tiaoxi River). The upper reaches contain a greater amount of unstable minerals, in which the contents of amphibole and pyroxene are about twice higher than those in the mid-lower tributaries (Fig. 3; Table 1). However, the contents of altered minerals (limonite, leucoxene) in the upper reaches are very low in all samples (Table 1). The average content of limonite is approximately one third of that in the mid-lower Changjiang tributaries. In addition, leucoxene is scarce in the upper Changjiang, but it is commonly found in the mid-lower tributaries (Fig. 3; Table 1). The discrepancy of stable and unstable minerals in different Changjiang sections is likely to be related to the climate and weathering conditions; this has been also suggested by Yang et al. (2009) and Vezzoli et al. (2016).

As stated above, the mean annual temperature and precipitation of the mid-lower Changjiang basin are higher than in the upper valley because of the impact of the monsoon climate (Fig. 2). Consequently, the weathering intensity in the mid-lower reaches is stronger overall than in the upper valley (Shao and Yang, 2012; Wang and Yang, 2013; He et al., 2015), which may cause the relative depletion of unstable minerals (e.g. amphibole) in the mid-lower tributaries (Fig. 3). Conversely, the altered mineral content (such as limonite and leucoxene) increases with respect to the weathering of certain Fe, Ti enriched minerals (such as biotite, ilmenite, amphibole, and epidote), the content of which is much higher in the middle-lower Changjiang tributaries than in the upper Changjiang. In addition, the average heavy mineral

concentration in upper Changjiang sediments is 5%, which is much higher than in the mid-lower tributaries; this also implies stronger weathering corrosion and chemical dissolution of unstable heavy minerals in the mid-lower catchments (Fig. 5).

Based on the heavy mineral characteristics of modern Changjiang sediment, we suggest that a certain unstable mineral (amphibole: Amp) and an altered mineral (limonite: Lm) can be used as respective sensitive minerals to reflect weathering conditions. It is considered that the ratio of amphibole to limonite (Amp/Lm) may reflect the weathering intensity registered in sediments (Fig. 5). However, it is also understood that amphibole is platy and that hydrodynamic conditions and suspension sorting may influence its content in the riverine environment; therefore the Amp/Lm ratio may provide large uncertainties. However, all surface samples were collected from the floodplain in similar depositional environments, and it is thus considered that the sorting effect may be somewhat reduced.

Our analysis indicates that Amp/Lm is much higher in the upper Changjiang (all > 1.5) than in the mid-lower tributaries (mostly < 1.3) (Fig. 5). The sample from Hanjiang River is an exception with Amp/Lm is > 6 (Fig. 5); this is considered to be mainly related to river's sediment origin, which is the upper drainage area (that has a large loess content and a relatively low mean annual temperature) (Fig. 2).

5.2. Sediment provenance evolution in the Changjiang Delta during late Cenozoic

The sediment provenance of the Changjiang Delta in the late Cenozoic has been studied using multiple methods including stratigraphy, mineralogical and geochemical analyses, and single mineral chronological analysis (e.g. zircon and monazite ages) (Chen and Stanley, 1995; Wang et al., 2005a, 2005b; Yang et al., 2006b; Chen et al., 2009; Gu et al., 2014; Yue et al., 2018). Most of these studies have suggested that the pre-Quaternary sediments were mostly derived from the proximal region, and the Tibetan Plateau source is only detected in the Pleistocene strata.

The bottom (below ca. 200 m) sediments of core LQ11 have distinct heavy mineral assemblages that are dominated by the iron oxide mineral of limonite. Moreover, the prevalent distribution of detrital zircon is not only observed in core LQ11 but also in the Pliocene sediments of the whole delta area (Table 2, Fig. 4; Qiu and Li, 2007; Chen et al., 2009; Jia et al., 2010; Yue et al., 2018), which is suggestive of the widespread granitoid rocks in the mid-lower valley (Figs. 3, 4). Previous studies have reported a large number of angular euhedral zircons occurring in Pliocene sediments in the delta area (Jia et al., 2010; Wang, 2016). Furthermore, the zircon U–Pb age spectrum of these Pliocene sediments shows that they are mainly of a Cretaceous age, which suggests the proximal sources in the middle-lower reaches (Jia et al., 2010; Wang, 2016). In short, the heavy-mineral features, zircon typology, and chronology all show that the proximal basins, which are enriched with Mesozoic magmatic rocks, were the predominant provenances of pre-Quaternary sediments in the Changjiang Delta.

Table 3

Comparison of typical heavy mineral assemblages and index values in the Plio-Pleistocene sediments from different cores of the Changjiang Delta (unit: %). The figures in brackets denote standard deviations; the data of cores SK10, Pd and LQ24 are from Chen et al. (2007, 2009) and Yue et al. (2018).

Cores	Units	Sample numbers	Amphibole	Limonite	Leucoxene	ZTR	Amp/Lm
SK10	II	236	8.3 (5.7)	7.0 (4.8)	0.3 (0.5)	3.5 (1.5)	1.2 (4.4)
	I	75	2.4 (4.5)	19.1 (23.1)	2.1 (3.5)	17.8 (15.8)	0.1 (1.7)
LQ11	II	17	59.0 (9.2)	8.0 (6.0)	0.0 (0.2)	0.7 (0.6)	10.5 (5.5)
	I	7	5.1 (3.2)	42.2 (18.4)	8.4 (8.1)	17.8 (9.8)	0.2 (0.1)
Pd	II	89	27.1 (12.6)	0.9 (2.1)	0.1 (0.1)	0.2 (0.4)	30.1 (33.1)
	I	28	2.9 (5.7)	6.3 (19.2)	4.7 (4.6)	1.3 (2.4)	0.5 (1.5)
LQ24	II	53	36.9 (17.2)	9.5 (11.4)	0.2 (0.7)	1.3 (1.4)	3.9 (5.7)
	I	29	1.2 (1.2)	17.6 (20.4)	13.1 (17.2)	6.1 (5.4)	0.1 (0.1)

Note: Unit I indicates the Pliocene strata; Unit II denotes the Quaternary strata.

After the Pliocene until the late stage of the early Pleistocene (core LQ11 ca. 140 m), the sharp increase in clinopyroxene suggests that more sediments were derived from basic igneous sources. The amphibole and epidote assemblages also show great similarities to those of modern Changjiang (Figs. 3, 4). Based on the geologic background, we infer that the upper Changjiang basin (where large basic-ultrabasic rocks prevail) (Chung and Jahn, 1995; Song et al., 2009) supplies a large amount of clinopyroxene to the coastal area. In addition, the lower catchment of Nanjing and Zhenjiang contains basaltic outcrops and layers of basaltic lava intercalated within Neogene deposits (Zhang et al., 2003), which may also supply unstable minerals to the estuarine and delta areas. Furthermore, the lower stream may contribute a certain amount of unstable minerals to the delta plain deposits (not merely the upper stream).

Single mineral analysis suggests that after the Pliocene, the upper Changjiang catchment supplied a large volume of siliciclastic sediments to the lower mainstream and delta area. For example, magnetite grains characterized by high contents of Ti, Mg, and V, which are derived from the upper reaches (mostly the E'meishan basalt province), only occur steadily only in Quaternary sediments (Yue et al., 2016). In addition, rounded zircon grains that have short elongation are prevalent in the Quaternary stratum, which also implies long-distance sediment transport (Wang, 2016; Yue et al., 2018). The detrital zircon age spectrum of Quaternary sediments exhibits multi-peaks with respect to the higher abundance of Neoproterozoic and Paleoproterozoic zircons, which further verifies the existence of a large provenance area that reaches as far as the eastern Tibetan Plateau (Weislogel et al., 2006; Jia et al., 2010; Yang et al., 2012; He et al., 2013; Wang, 2016).

The above integrated analyses show that after the early Pleistocene, the Changjiang delta area received a large amount of sediment from the upper basins as a response to accelerated tectonic subsidence in the East China and tectonic uplift in the source region of Tibet (Wang, 1990; Li and Fang, 1999; Zhang et al., 2003; Yi et al., 2014; Tada et al., 2016). The abrupt increase in unstable minerals (e.g. pyroxene, amphibole, and apatite) and the decrease in stable (e.g. ZTR) and altered minerals (limonite and leucoxene) indicate that the upper stream materials flowed down to the Changjiang deltaic area at the Pliocene-Quaternary transition, which implies that a grand river evolution event occurred and that the Changjiang River might have drained the East China's continental margin no later than 2.5 Ma.

5.3. Environmental change in the Changjiang Delta and heavy mineral indicators for the Q/N stratigraphic boundary in delta

The heavy mineral assemblages in modern Changjiang suggest that the ratio of amphibole to limonite (Amp/Lm) could indicate climate and weathering conditions. To verify the universal applicability of this ratio, four cores (SK10, LQ11, LQ24, and Pd) were collected from the Changjiang Delta to conduct comparisons (Table 3; Fig. 6).

As shown in Fig. 6, two distinct units are recognized in all cores from Changjiang Delta. In the lower parts of these cores, stable minerals

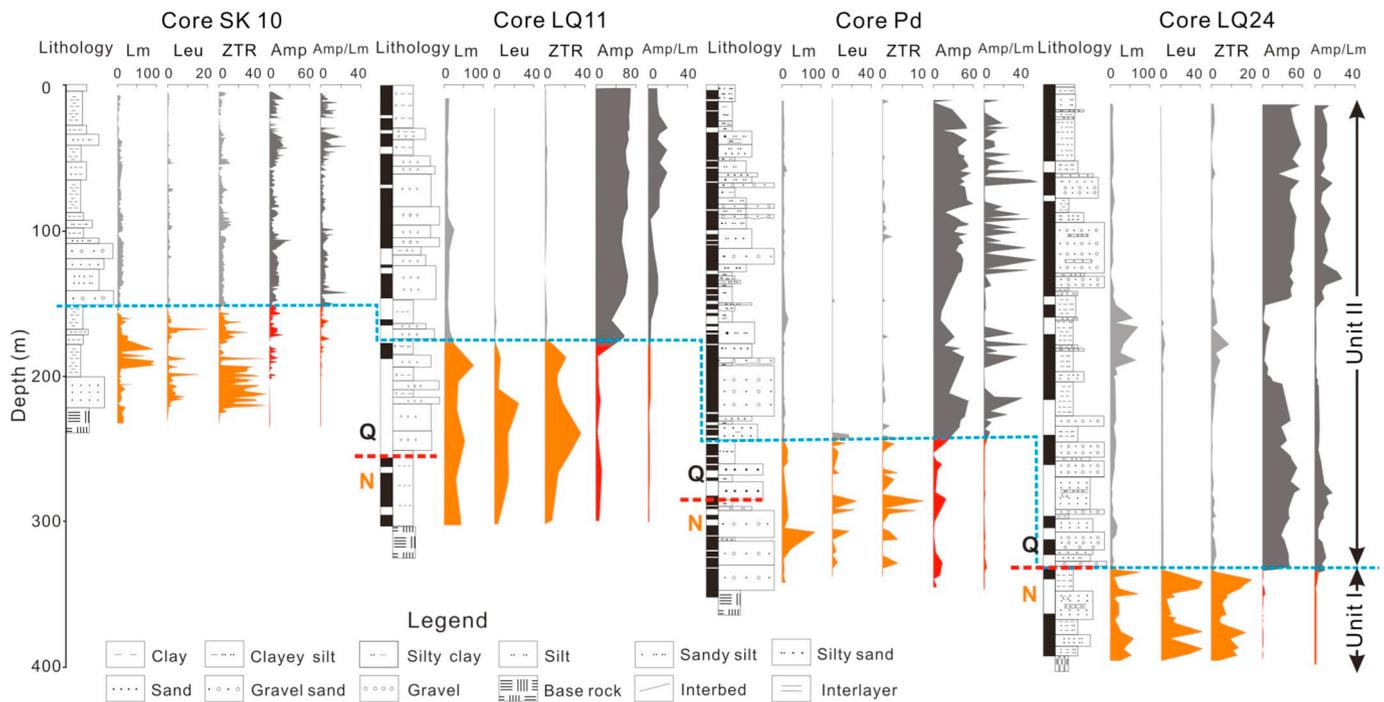


Fig. 6. Changes of typical heavy mineral assemblages across the Q/N stratigraphic boundary of the Changjiang Delta (The blue dash line represents the Q/N boundary based on heavy mineral result; the red dash line represents the Q/N boundary constrained by paleomagnetic dating result). Amp = amphibole, Lm = limonite, Leu = leucoxene, ZTR = zircon + tourmaline + rutile; the data of cores SK10, Pd and LQ24 are respectively from Chen et al. (2007), Chen et al. (2009) and Yue et al. (2018). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

are dominant while unstable minerals are in minor amount. The ratio of the Amp/Lm is the lowest and amphibole is extremely low (5%), but the ZTR and leucoxene have the highest contents. However, heavy minerals display significantly different features in the mid-upper strata of these cores.

Based on the previous information presented about climate sensitive minerals, we infer that the heavy mineral composition differences between the lower and upper strata occur because of the changes in sedimentary environments during different geological times. The sediment source of the bottom strata (mostly Pliocene) is predominantly from the proximal mid-lower tributaries. However, compared to the surface samples, the content of amphibole in these strata (5%, Tables 2, 3) is much lower than that of the present-day mid-lower tributaries (25%, Table 1). This implies that the chemical weathering of Pliocene sediment was stronger than that of the present day. Previous researches documented that warmer and wetter climate conditions were prevalent in East China during the Pliocene (Han et al., 1997; Chen et al., 2001; Yang et al., 2006a; Zhang et al., 2013). As previously stated, stable minerals such as ZTR would be enriched in this relatively hot climate regime because they are resistant to chemical weathering (e.g. Garzanti et al., 2009; Morton et al., 2012), whereas there would be a reduction in unstable minerals (such as amphibole) because of weathering corrosion, and chemical decomposition (Morton, 1982; Andò et al., 2012; Hessler and Lowe, 2017). Abnormally high contents of altered minerals, such as leucoxene, and limonite also suggest strong weathering conditions (Fig. 7).

Strong chemical weathering in the Changjiang Delta during the Pliocene has been well documented by previous studies (Yang et al., 2006a), where low contents of Fe, Mn, Al, Mg, and Na have been determined in Pliocene sediments (Gu et al., 2014). In addition, an extraordinary low HMC (mostly < 0.5%) is observed in Pliocene sediments (Fig. 4), which indicates heavy mineral dissolution under strong weathering conditions.

Above the Q/N boundary, there is a sharp increase in amphibole and an obvious decrease in limonite, ZTR, and leucoxene in the mid-

upper strata, which is in complete contrast to the bottom strata (Tables 2, 3; Fig. 6). Compared with the surface samples, we infer that this mineralogical feature reflects relatively weak to moderate chemical weathering. In the Quaternary, the accelerated Himalayan orogeny led to the continuous uplift of the Tibetan Plateau and degradation of the monsoon climate (Wang, 1990; Li and Fang, 1999; Clark et al., 2004; Lunt et al., 2010). The intensive uplift of the Tibetan Plateau not only changed the landform of East Asia, but it also changed the route of water, heat, and atmospheric circulation (Wang, 1990; Li and Fang, 1999; Lunt et al., 2010). Consequently, Quaternary weathering weakened due to the low temperature and weak precipitation, and thus unstable minerals could be well preserved (Ravelo et al., 2004; Wang et al., 2008; Wan et al., 2010). The striking increase in HMC (3% on average) in the middle-lower strata (Fig. 4) implies an obvious increase in heavy minerals and weak chemical weathering. In addition, regional subsidence accelerated on eastern China's plains in the early Quaternary (Chen and Stanley, 1995; Yi et al., 2014; Liu et al., 2018), and huge changes in elevation made the eastern Tibetan Plateau the main sediment source. The sedimentation rate in the Quaternary was higher than during the Pliocene (Wang et al., 2005a; Wang et al., 2007), and the fast sediment transfer in the catchment and quick burial in the delta depositor provided insufficient time for unstable minerals to be diagenetically altered. In our study, the rare occurrence of pyrite and greigite in Quaternary sediments also suggests weak diagenesis.

In summary, certain special minerals (amphibole, limonite, leucoxene) and the mineral index (Amp/Lm) are used to show that the sedimentary environments in the bottom strata were remarkably different from those in the middle-upper layer (Fig. 6). In view of the distinct climate conditions between the Pliocene and Pleistocene, the heavy mineral assemblages can be further applied as reliable indicators for recognizing the Q/N boundary. Based on the paleomagnetic dating analysis, however, the Q/N boundary in Core LQ11 and Core Pd is located at ca. 250 m and 288 m respectively, which is somewhat different from the identification using this heavy mineral analysis (Fig. 6). However, as there are large uncertainties in magnetostratigraphy and

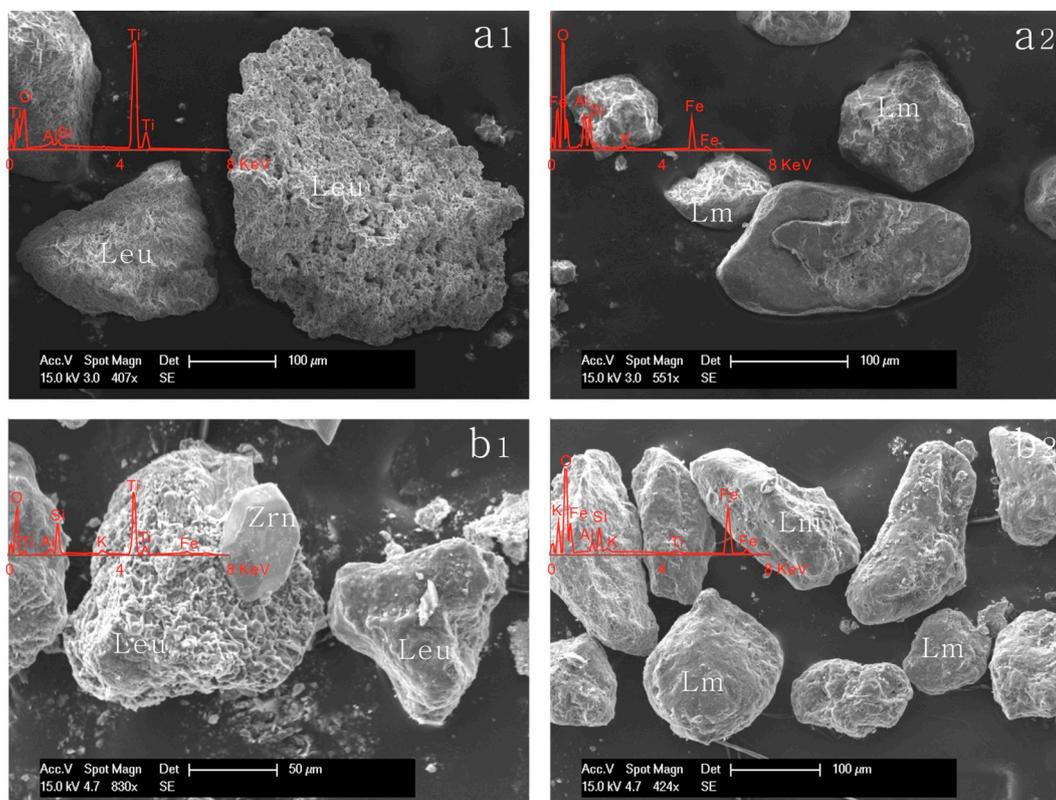


Fig. 7. BES images and EDS spectra (red lines) of leucoxene and limonite in the Pliocene sediments of Changjiang Delta; (a1) Leucoxene image of Core LQ11 at 198 m depth; (a2) Limonite image of Core LQ11 at 265 m depth; (b1) Image of leucoxene with zircon in Core LQ24 at 355 m depth; (b2) Limonite image of Core LQ24 at 378 m depth; Leu = leucoxene, Lm = limonite, Zrn = zircon. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

sedimentary stratigraphy in the Changjiang Delta, determining the Q/N boundary using certain mineral assemblages may be more reliable, because they are sensitive to climate and weathering conditions.

The borehole stratigraphic analysis (Fig. 6) indicates the sedimentary sequences near the Q/N boundary are similar in the Changjiang delta area, showing that fine sediments (grey silty clay) underlie coarse grains (yellow-grey sand or gravel). Gravel is poorly sorted in these coarse sediment layers, which indicates an alluvial fan or braided fluvial facies, and the fine-grained silty clay indicates lacustrine facies (Wang et al., 2005a; Gu et al., 2014). The sedimentary stratigraphic analysis suggests that there has been an almost continuous sedimentation process in the Changjiang Delta since the Pliocene, which has occurred with respect to gradual subsidence (Wang et al., 2005a, 2005b; Duan et al., 2016). Nevertheless, the sedimentary hiatus in the strata cannot be ignored because of the dynamic depositional environments in the estuarine and delta areas. Recent research shows that the magnetostratigraphy can be well established in the strata of fine sediments deposited after the middle Pleistocene (Duan et al., 2016); however determining reliable dates from coarse-grained sand is always a big challenge, and unfortunately, coarse sediments are ubiquitous within the Plio-Pleistocene strata of the delta. Strong chemical weathering in the Pliocene would also have caused the dissolution of magnetic minerals, which further restrain the accuracy of paleomagnetic dating. The Pliocene strata in the Core LQ11 the Pliocene strata is a lacustrine facies with silty clay dominating, while the Core Pd features by an alluvial fan or braided fluvial facies dominated by fine sand.

Integrating the heavy mineral data of our study with the sedimentary stratigraphy, we reexamine the paleomagnetic dating results and speculate the Q/N boundary may locate in the sedimentary facies where the upper grey sandy fluvial sediments are overlain the lower blue-grey indurated lacustrine, overbanking, or tidal mud (Fig. 6). However, it is

considered that an absolute dating analysis is required to establish a more accurate chronostratigraphy.

6. Conclusions

Our study illustrates that heavy mineral characteristics vary significantly within the different reaches of the modern Changjiang River system. In the upper reaches, the content of unstable minerals (such as amphibole and pyroxene) reach up to ca.50% and 20% respectively, while sediments from the mid-lower tributaries are enriched with stable minerals (such as zircon, tourmaline) and the altered minerals of limonite and leucoxene. The heavy mineral assemblages of different Changjiang reaches are mainly controlled by their parent rock types, and the ratios of amphibole to limonite yield values that differ greatly between the upper and mid-lower Changjiang tributaries and are potentially indicative of different environment conditions.

Abrupt changes in heavy mineral distribution patterns between the lower and middle-upper strata are universal in the Changjiang Delta. The bottom stratum (Pliocene) has an abnormally high altered minerals (limonite and leucoxene) content, stable minerals (ZTR) content, but a low of amphibole and amphibole/limonite ratio, which indicates that the sediments were mainly from the middle and that chemical weathering was strong at that time. In the middle-upper stratum (Quaternary), the contents of amphibole, garnet, and pyroxene increased, while there is an obvious reduction in ZTR and altered minerals, which suggests that the sediment provenances extend to the upper Changjiang valley where basic rocks widely outcrop. The unstable mineral assemblages also imply weak chemical weathering during the Quaternary. These results indicate that the complete connection between the Yangtze River to the East China's continental margin occurred no later than the Quaternary. The sediment

provenances are related to complex tectonic and environment change in the Changjiang Delta and the far-reaching effect of the eastern Tibetan uplift. It is considered that combined with sedimentary stratigraphy and paleomagnetic dating, the heavy mineral indices of Amp/Lm and ZTR can be used to identify the Q/N boundary in the Changjiang Delta and surrounding areas.

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