Magnetite with anomalously high Cr₂O₃ as a fingerprint to trace upper Yangtze sediments to the sea

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ABSTRACT

This paper examines geochemical properties of detrital magnetite, in order to link sediments in a Plio-Quaternary core taken in the delta area to their sources in the Yangtze River basin. A total of 40 sediment samples were collected from both the main river channel/tributaries and a sediment core from the Yangtze delta. The geochemical compositions of detrital magnetite in these sediments were analyzed by electron microprobe, including FeO, TiO₂, CoO, MgO, Cr₂O₃, MnO, ZnO, Al₂O₃ and V₂O₃. The results revealed that the detrital magnetite grains with anomalously high Cr₂O₃ occurred

exclusively in the upper reaches of the Yangtze (upstream of the Three Gorges Dam), where the E'mei Basalt block is located. This type of magnetite could therefore be considered a unique sediment proxy of the upper river basin to help identify sediment source in the delta area. Our analysis found such magnetite grains with high Cr_2O_3 occurring throughout the core depth above 186.5 m, in contrast to the extremely low Cr_2O_3 below this depth. The boundary between high and low Cr_2O_3 in magnetite grains of the core sediments was dated by paleomagnetism at ca. $\sim 1.2\text{-}1.0$ Ma, signifying that the linkage between the Yangtze River course and the sea was before $\sim 1.2\text{-}1.0$ Ma. This demonstrates that the sediment provenance of the Yangtze delta has experienced a change from local to distal Yangtze River, which took place with the uplift of the Tibetan plateau and coastal subsidence during the Plio-Quaternary.

Key words: Magnetite; E'mei Basalt; Tectono-geomorphology; Yangtze River

1. Introduction

The Yangtze River originates from the Tibetan Plateau at an elevation of above 5000 m. The river is >6300 km long with a drainage basin of 1.80 million km^2 (Chen et al., 2001). It discharged ca. $900 \times 10^9 \text{ m}^3$ of freshwater and $4.5 \times 10^8 \text{ t}$ of sediments each year during the period of 1951-2001 before construction of the Three Gorges Dam ($1.5 \times 10^8 \text{ t}$ per year thereafter) into the East China Sea (Changjiang Water Resources Commission, 2014). The large amount of riverine sediments accumulates on the coast to build the extensive delta-coast system. This process progresses as the result of the coupling between tectonic movement and climate change (Clark et al., 2004). Inevitably, the

sediment fingerprints of the upper Yangtze basin can be identified in the Plio-Quaternary sediment at the river mouth (Yang et al., 2006). Sediment fingerprinting provides us a unique approach to explore not only the timing of the upper Yangtze sediment contribution to the depositional evolution of the delta but also the evolution of the Yangtze fluvial system.

Since the early 20th century, the geomorphological evolution of the Yangtze River has become an academic focus. Much was related to tectono-geomorphological processes in the upper part of the river (Willis et al., 1907; Lee, 1933; Barbour, 1936; Shen, 1965; Yang, 1985; Li et al., 2001; Kong et al., 2012). There are several key arguments on-going among geomorphologists. Firstly, it has been hypothesized that the Yangtze River once flowed into the South China Sea via the Red River before the river was captured at Shigu of the upper Yangtze basin (Fig. 1; Zhao, 1996; Clark et al., 2004), although the timing of the capture is unclear. Secondly, the timing of the opening of the Three Gorges valley (Fig. 1) vs. the river capture has been highly questionable. The Yangtze River would not flow into the East China Sea had the Three Gorges valley not been channelized. This had taken place through uplift of the Tibetan plateau and subsidence of the eastern China coast, the so-called Cenozoic Topographic Reversal (Wang, 2005). Thirdly, the connection of inter-mountainous basins, represented by the Sichuan basin of the upper Yangtze, Jiangnan basin of the middle Yangtze and the coastal basin is ambiguous in terms of the surficial processes accompanying substantial subsidence of eastern China continent (Schneiderman et al., 2003; Gu et al., 2014).

Apart from these debates, more attention has recently been paid to the Yangtze coast with regard to the river connection to the sea by means of a sediment source-to-sink

approach. Previous studies used detrital minerals (such as monazite and zircon) in a chronostratigraphic investigation of sediments of the lower Yangtze basin to trace the sediment sources, especially the diagnostic sediment proxies derived from the upper Yangtze (Yang et al., 2006; Chen et al., 2009; Jia et al., 2010; Zheng et al., 2013). This approach is effective for sediment source identification of the lower Yangtze basin sediments. However, results showed various diagnostic detrital minerals with a wider range of timings of the Yangtze River connection to the sea (Fan et al., 2005; Yang et al., 2006; Huang et al., 2009; Chen et al., 2009; Gu et al., 2014). The key point was the reliability of provenance interpretation from the proxies used, and that these may be influenced by climatic factors and tectonic cycles in the source region (Suttner et al., 1981; Dickinson et al., 2009), hydrodynamic sorting during transport or reformation during deposition (Garzanti et al., 2009; Lawrence et al., 2011). However, the unclear spatial distribution of the provenance proxies in the source regions of Yangtze coastal sediment pool has been a limitation of provenance interpretations. The question then is how to link diagnostic sediments with regional outcrops of the Yangtze basin and improve the reliability of provenance proxies?

Detrital magnetite is often considered as a sediment source tracer due to its stable properties and widespread distribution, easy separation and differential geochemical composition (Pettijohn, 1987; Grigsby, 1990). The present study specifically focused on the linkage of magnetite geochemistry of detrital minerals in the modern river sediment to the large block of E'mei basalt rock that exclusively occurs in the upper Yangtze (upstream of the Three Gorges Dam) (Fig. 1; Bureau of Geology and Mineral Resources of Sichuan Province, 1987; Xu et al., 2001; Xiao et al., 2004). By establishing their

inter-correlation, we applied this linkage to a Plio-Quaternary core sediment from the delta area, which enabled us to define the occurrence and timing of such minerals in well-dated core strata. This interpretation would then provide a better understanding of the connection of the Yangtze River to the sea in association with long-term tectono-geomorphological evolution of the river basin.

2. Materials and Methods

Six sediment samples were collected from the mainstream of the upper, middle and lower Yangtze River (YZ-1, YZ-2, YZ-3 YZ-4, YZ-5 and YZ-6; Fig. 1). One long and continuous sediment core (SG7, 320.9 m long) was taken from the Yangtze delta, penetrating the Holocene, Pleistocene, and Late Pliocene until bedrock (basalt) (Fig. 1). A total of 267 fine sediment samples were taken at an interval of 1.0-1.5 m for paleomagnetic dating at Nanjing Institute of Geology and Mineral Resources, China. Thirty-seven samples, including 6 from the modern river, were taken for geochemical analysis on detrital magnetite grains. To better understand the geochemical differences of detrital magnetite in relation to regional bedrock outcrops, 3 samples were also collected from the upper mainstream and the mid-lower Yangtze tributaries, including the Hanjiang River and Dongting Lake (CJ-7, CJ-11 and CJ-13; Fig. 1; Wang et al., 2007).

Detrital magnetite grains were separated from the bulk sediments using a bar magnet wrapped by vinyon repeatedly. More than 150 magnetite grains were mounted on copper tubes full of resin glue for each sample, polished and then coated with carbon. More than 10 magnetite grains with a size range of 63-125 μm were randomly selected for geochemical determination, i.e. Fe, Ti, Mg, Al, Co, Mn, V, Cr and Zn by electron

microprobe analyzer (JXR-8900M) in the Department of Materials and Optoelectronic Science, the National Sun Yat-sen University, Taiwan. The working time of the microprobe was 30-s for Fe, Mg, Al, Co, Mn, V, Cr and Zn and 60 s for Ti. The content of Fe was synthesized by the percent of Fe₂O₃ and FeO according to Carmichael's method (1967). The test was repeated on three points and on one plane for each grain under a 15 kV accelerating voltage and a 20 nA current. The averaged value of the three points and one plane was regarded as representative of each magnetite grain in this study. Elemental oxide concentrations of magnetite were calculated from the values of above elements according to Bence-Albee correction methods (1968). Coefficient of Variability was calculated for each oxide according to the formula of $CV = \text{standard deviation} / \text{average value}$, which demonstrated compositional variability in the magnetite. The texture of magnetite grains were also imaged for analysis during geochemical determination.

3. Results

3.1 Geochemical characteristics of detrital magnetite in the Yangtze River

The average values of FeO, TiO₂, MgO, Al₂O₃, CoO, MnO, V₂O₃, Cr₂O₃ and ZnO in detrital magnetite of the river surficial sediment are listed in Table 1 in an order from the upper, middle to lower Yangtze reaches. These results showed that magnetite was mainly composed of FeO, (90.62% on average) and TiO₂ (3.75%). Also, it contained some minor element oxides: Cr₂O₃, Al₂O₃ and V₂O₃ (ca. 1%), MnO and MgO (ca. 0.5%) and CoO and ZnO (< 0.2%) (Table 1).

The magnetite of the upper Yangtze mainstream contained high Cr₂O₃, i.e. 3.56% on average (Table 1). This contrasts the low Cr₂O₃ (<1%) occurring in the magnetite from

the middle Yangtze tributaries (Dongting Lake and the Hanjiang River) (Table 1). The MgO and MnO concentrations of magnetite were also higher in the upper Yangtze mainstream than the middle tributaries, but the difference was much smaller than that of Cr₂O₃ (Table 1). Results also revealed that magnetite grains with anomalously high Cr₂O₃ (ca. 40%) account for ca. 10% of total measured grains from the upper Yangtze. No such grains were found in the middle Yangtze tributaries (Fig. 2). A similar situation can be seen for magnetite grains with high MgO (Fig. 2). Meanwhile, the upper Yangtze yielded more magnetite grains with high TiO₂, Al₂O₃, MnO and V₂O₃, when comparing the Dongting Lake and the Hanjiang River of the middle Yangtze reach (Fig. 2). Higher CV values of geochemical elements of magnetite showed various magnetite compositions in the upper Yangtze compared to the middle Yangtze tributaries (Fig. 2). The magnetite grains with anomalously high Cr₂O₃ that occurred in the middle and lower mainstream were primarily transported from the upper reach to the delta-coast (Fig. 2).

Table 1

Average element oxide contents of the magnetite in the Yangtze River sediments (%). The diagnostic magnetite contribution with high Cr₂O₃ is highlighted in bold.

Reach	Samples (grain numbers)	FeO	TiO ₂	CoO	MgO	Cr ₂ O ₃	MnO	ZnO	Al ₂ O ₃	V ₂ O ₃
Upper mainstream	YZ-1(19)	89.86	2.21	0.26	0.97	4.02	0.42	0.07	1.52	0.68
	CJ-7(6)*	85.92	1.72	0.13	1.82	7.48	0.22	0.08	3.39	0.38
	YZ-2(20)	89.46	5.22	0.21	0.53	2.15	0.29	0.06	1.20	0.88
Dongting Lake	CJ-13(6)*	87.30	1.17	0.16	0.15	0.10	0.07	0.08	5.74	0.29
Hanjiang River	CJ-11(5)*	94.68	4.30	0.13	0.26	0.20	0.08	0.12	0.10	0.71
	YZ-3(19)	91.18	5.39	0.25	0.23	0.12	0.64	0.18	1.07	0.94
	YZ-4(18)	90.03	3.18	0.24	0.49	3.90	0.36	0.06	1.01	0.73
Middle mainstream	YZ-5(29)	96.42	1.82	0.19	0.22	0.12	0.14	0.04	0.37	0.69
	YZ-6(10)	91.37	3.40	0.19	0.46	2.95	0.16	0.09	0.57	0.81

* Data refers Wang et al, 2007.

3.2 Chronology, lithology and sedimentary facies of core SG7

The chrono-stratigraphic framework of core SG7 was based on paleomagnetic results, Optically Stimulated Luminescence (OSL) dating and stratigraphic features of the delta area. The details of this have been documented in our previous studies (Wang et al., 2008a; 2008b). The paleomagnetic results showed the boundaries of Gauss/Matuyama (~2.58 Ma) at the core depth of 286.3 m and Matuyama/Brunhes (~0.78 Ma) at 136.9 m, separating the Pliocene and the Early Pleistocene, and the Early Pleistocene and the Middle Pleistocene (Fig. 3). OSL dating showed that the boundary of the Middle and Late Pleistocene (ca.0.12 Ma) occurred at the core depth of 83.55 m (Fig. 3). Upward in the core, a dramatic lithological change occurred at core depth of 24.35 m, marked by the stiff muds of Late Glacial Maximum (Fig. 3). This was regarded as a stratigraphic marker showing the boundary of the Late Pleistocene and Holocene in the delta area (Qiu and Li, 2007; Wang et al., 2008a).

Vermiculated hard muds of Late Pliocene of core SG7 rested on the top of basalt bedrock with a weathering unconformity (Fig. 3). This was followed by several sedimentary cycles consisting of the Early Pleistocene strata, each cycle showing gravelly coarse sands of 10-20 m thick at the bottom and fine sand, silt and clay of 2-15 m thick at the top. Plant fragments were often present. These strata recorded the presence of alluvial fan sediment facies (Fig. 3). The strata of the Middle Pleistocene were usually made up of fine sand and silt with a thickness of 50-60 m, showing fluvial (meandering) deposition (Qiu and Li, 2007). The sedimentary strata of the Late Pleistocene were represented by a thick sediment layer (ca. 60 m) of gravelly sands at the bottom and fine sand and silt at the top. Shell fragments and iron-manganese nodules occurred in the sediments and

foraminifera occurred throughout (Fig. 3). Shells and foraminifera indicated the Late Pleistocene marine transgression, which occurred widely on eastern China coast (Wang et al., 2008b). The Holocene deltaic sediment developed above the core depth of ca. 24 m, was underlain by the stiff mud (Fig. 3).

3.3 Geochemical characteristics of detrital magnetite in SG core

Similar to the river surficial sediment, the magnetite in core SG7 also mainly consisted of FeO (ca. 91.69%) and TiO₂ (4.51%), with other minor (< 4% in total) elements (Cr₂O₃, Al₂O₃, V₂O₃, MnO, CoO and MgO) (Fig. 3). There were three zones (I-III) identified in terms of elemental variations upward the core (Fig. 3). Zone I (320.9-280.0 m) mainly belonged to the Late Pliocene, Zone II (280.0-186.5 m) to the early and middle stage of the Early Pleistocene, and Zone III (186.5-0 m) to the late Early Pleistocene, Middle Pleistocene, Late Pleistocene and Holocene (Fig. 3). The bottom of Zone III (186.5 m) was located at one normal Polarity Event below the boundary of Matuyama/Brunhes that is thought of the Jalamillo Event (1.21-0.99 Ma) compared to the Standard Geomagnetic Polarity Schedule (Fig. 3). Therefore, the age of the bottom of Zone III was deduced ca. 1.2-1.0 Ma.

The magnetite of Zone I was generally characterized by high MgO, Al₂O₃, TiO₂, and MnO (Fig. 3), but Cr₂O₃ was almost absent. Most magnetite grains were inhomogeneous with stripe structures (Fig. 4A). Skeletal crystal magnetite particles were found at a core depth of 279.7 m (Fig. 4A). In Zone II, FeO reached a maximum throughout the core sediments (97.72%) while other element oxides exhibited very low concentrations. Most magnetite grains were homogeneous in texture (Fig. 4B). The magnetite grains with

anomalously high Cr₂O₃ appeared initially and frequently throughout Zone III. The grains with high MgO, Al₂O₃, TiO₂, MnO and ZnO also increased significantly in this zone compared to Zone II. Both homogeneous and inhomogeneous magnetite grains coexisted in this zone (Fig. 4C). The CV values of Cr₂O₃, MgO, Al₂O₃, TiO₂ and MnO showed a dramatic increase in Zone III in contrast to the low values in both Zone I and II (Fig. 3).

4. Discussion

4.1 Magnetite with anomalously high Cr₂O₃: a unique sediment fingerprint of the upper Yangtze basin

Magnetite is a stable mineral that can be derived from various igneous and metamorphic rocks (Xue et al., 1991). However, elemental compositions in magnetite vary with different parent rocks (Basu and Molinaroli, 1989; Grigsby, 1990, 1992). Therefore, magnetite with unique geochemical properties can be used as diagnostic sediment proxies to trace sediment provenance (Hounslow and Morton, 2004; Yang et al., 2009).

The present analysis of elemental oxide compositions in magnetite has presented an obvious difference in the sub-basins of the Yangtze River. The three surficial sediment samples of the upper Yangtze (YZ-1, CJ-7, YZ-2; upstream of the Three Gorges Dam) contained more magnetite grains with high Cr₂O₃, MgO, TiO₂, MnO and Al₂O₃ than those from the tributaries (Dongting Lake basin and the Hanjiang River) of the middle Yangtze basin (Table 1, Fig. 2). These magnetite grains were reasonably linked to the large-scale E'mei Basalt block (ca. 5.0 x10⁵ km² in area), which occurs exclusively in the upper Yangtze above the Three Gorges valley (Fig.1; Hou et al., 1999; Xu et al., 2001;

Xiao et al., 2004; Yan et al., 2006). The E'mei Basalt is rich in magnetite with a high amount of Cr_2O_3 , MgO , TiO_2 , Al_2O_3 , MnO and V_2O_3 compared to other parent rocks in the Yangtze basin (Lin, 1982; Basu and Molinaroli, 1989; Grigsby, 1990, 1992; Lei et al., 2009). In particular, this study has found that the magnetite grains with anomalously high Cr_2O_3 of ca. 40% occurred distinctively in the upper Yangtze (Fig. 2). Previous investigations have revealed that such anomalously high Cr_2O_3 was detected in Ti-Cr magnetite of the E'mei Basalt block (Bureau of Geology and Mineral Resources of Sichuan Province, 1987; Wu, 1998; Fig.1). It can be concluded that these magnetite grains with anomalously high Cr_2O_3 thus came from E'mei Basalt block.

Most magnetite grains derived from the middle Yangtze tributaries were characterized by high FeO and low proportions of other elements oxides (Fig. 2). Magnetite generally originated from felsic igneous or metamorphic rocks that were the major source rocks in the middle and lower Yangtze basin (Fig. 1; Grigsby, 1990; Wang, et al. 2007). Undoubtedly, the magnetite grains with anomalously high Cr_2O_3 found in the middle and lower mainstreams were mainly derived from the upper Yangtze by the river flow (Fig. 2). Therefore, this signal could be used as a unique sediment fingerprint of the upper Yangtze source and to thence trace its temporal distribution in core sediments in the Yangtze delta area (discussion given below).

4.2. Plio-Quaternary sediment source of the Yangtze delta-coast: from local to distal

Magnetite grains characterized by high MgO , Al_2O_3 , TiO_2 , MnO and ZnO in Zone I of the Late Pliocene showed a mafic origin, similar to those of the upper Yangtze reach (Chen et al., 1987; Grigsby, 1992; Fig. 3). However we do not think these grains came

from the upper Yangtze because Cr_2O_3 was virtually undetected in the magnetite. The low CV values of Cr_2O_3 , MgO , Al_2O_3 , TiO_2 , MnO implied a unique origin for the magnetite, unlike the high CV values of those from the upper Yangtze indicating various origins (Fig. 3). Well-preserved skeletal crystal magnetite particles suggested a short transport distance of basaltic origin (Fig. 4A; Neuendorf et al., 2005), which was recognized as the local basalt bedrock at the base of core SG7. Previous regional geological investigations confirmed that the occurrence of basalt bedrock of Tertiary origin is limited in the Yangtze delta (Guo et al., 1997; Wang et al., 2008a). Therefore we inferred that the study area received local sediments during the Late Pliocene when the eastern China coast still remained relatively high as a part of Zhe-Min uplift (also called Fukien-Reinan massif), with weathering processes recorded by the vermiculated hard muds over the bedrock (Fig. 3; Chen and Stanley, 1995; Wang et al., 2005; Gu et al., 2014; Yi et al., 2014).

Magnetite of Zone II was composed almost entirely of FeO with a homogeneous texture (Fig. 3, 4B). The CV values were much lower than those of Zone III, although they were overestimated due to extremely low average values of the elements oxides, implying a unitary magnetite origin (Fig. 3). The magnetite characteristics were similar to those from the Hanjiang River and Dongting Lake of the middle Yangtze basin (Fig. 2, 3), indicating an extended sediment provenance of SG7 from local sources to the middle Yangtze basin as subsidence occurred extensively on the eastern China coast in the early Quaternary (Chen and Stanley, 1995). Another possible sediment source might be the neighboring Zhe-Min uplift where intermediate-acidic igneous rocks dominate and these rocks could provide such a type of magnetite (Jin, 1992; Grigsby, 1992; Liu et al., 2003).

Magnetite with anomalously high Cr_2O_3 occurred uniquely and frequently in Zone

III, accompanied with magnetite with high MgO, Al₂O₃, TiO₂, MnO and V₂O₃. These magnetite grains were similar to those of the upper Yangtze (Fig. 2, 3). Such similarity was also confirmed by the high CV values of these elements and coexistences of homogeneous and inhomogeneous magnetite grains, being of multiple magnetite origins (Fig. 3, 4C). This evidence implies that the sediment provenance of the study area has extended to the upper Yangtze basin, starting from the bottom of Zone III when further subsidence occurred on the eastern China coast when marine transgressions occurred, especially the large-scale marine transgression in the Late Pleistocene (Qiu and Li., 2007; Wang et al., 2008b).

4.3 Timing of the Yangtze linkage to the sea

Tracing sediment source-to-sink from the upper Yangtze basin to coast can help better understand the evolution of the fluvial system during geological history. The use of magnetite as a diagnostic sediment tracer was applicable to the sediment core SG7 in the delta area. The obvious change in magnetite geochemistry in Zone III from the late Early Pleistocene (the Jalamillo Event of ~1.2-1.0 Ma) provided a clear linkage to the nature of parent sediment in the upper Yangtze basin (Fig. 3). The anomalously high Cr₂O₃ magnetite, identified as the unique sediment source of the upper Yangtze basin, initiated in this core at a depth of 186.5 m (bottom of Zone III, ~1.2-1.0 Ma), indicating the timing of the upper Yangtze sediment supply to the Yangtze delta and estuary (Fig. 2, 3). Therefore, the river's complete connection to the sea was established before ~1.2-1.0 Ma (Fig. 3). This timing is also supported by the results from monazite age, magnetic susceptibility, geochemical elements and Sr-Nd isotope in the Yangtze delta and estuary

(Yang et al., 2006, 2007; Wang et al., 2008a; Gu et al., 2014). Besides the Yangtze delta, sediments of Jiangnan basin in the middle Yangtze immediately below the Three Gorges valley also recorded the dramatic change in sediment provenance related to the upper Yangtze at the similar time during the Quaternary (Zhang et al., 2008; Shao et al., 2015).

5. Conclusion

Magnetite with anomalously high Cr_2O_3 can be used as the sediment fingerprint to trace the upper Yangtze sediment provenance in the delta-coast core sediment of Plio-Quaternary age. In light of the magnetite geochemical characteristics of the Yangtze basin, our results showed that the sediment provenance of the delta changed from local sources to the upper Yangtze basin. This process coincided with the dramatic landform change characterized by the uplift of the Tibetan plateau and the subsidence of eastern China coast during the Plio-Quaternary time period. The anomalously high Cr_2O_3 magnetite that first occurs at a core depth of 186.5 m shows that the timing of the Yangtze River thorough connection to the sea was established before ~1.2-1.0 Ma.

Acknowledgements

The Ministry of Science and Technology (formerly the National Science Council) of Taiwan provided funding for the electron microprobe analysis under the grant number NSC 96-2611-M-110-010. This work was also financially supported by the National Science Foundation of China (Grant No. 41301214). We also wish to thank Dr. Wayne Stephenson, the reviewers and the editor (Andrew James Plater) for their valuable comments.

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Figure caption

Fig. 1 Sampling locations and main rock types in the Yangtze drainage basin (modified from Changjiang Water Resources Commission, 1999).

Fig. 2 Element oxide concentrations and coefficients of variability (CV) of the magnetite grains in the Yangtze mainstream and tributaries. The data of CJ-7, CJ-13 and CJ-11

was from Wang et al, 2007. The pink bar marked the samples of tributaries.

Fig. 3 Elemental oxide concentrations in magnetite grains of core SG7 (paleo-magnetic results and OSL dating were from Wang et al., 2008a; 2008b).

Fig. 4 Backscattered electron scanning images of magnetite grains in core SG7. Images of A are representative crystal structure of Zone I; images of B are of Zone II; images of C are of Zone III.