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Magnetic properties of surface sediments from the Pearl River Estuary and its adjacent waters: Implication for provenance

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ABSTRACT

Environmental magnetism has been widely used as a rapid, cost-effective, and non-destructive method in various fields including sediment source identification. Nineteen surface sediments from ten transects in the Pearl River Estuary (PRE) and its adjacent seas were studied for magnetic measurements. Combined with mineral analysis for representative samples, magnetic implications for provenance are discussed. The results indicate that concentration-dependent magnetic parameters decrease gradually and change sharply between the PRE and its adjacent waters. The magnetic mineral assemblage is basically consistent at all sites, i.e. consisting of magnetite and hematite. However, the grain size of magnetite particles is clearly different probably due to different material sources. Magnetic parameters allow separating the sediments from the PRE and its adjacent seas into three groups, more distinctively than deduced from clay mineral analysis. These new findings indicate that magnetic properties of surface sediments bring complementary information of material source as well as marine geology.

1. Introduction

The study on provenance in the estuary-ocean area is not only an important field of sedimentology, but also directly related with the marine geological problems such as characteristics of seabed, marine evolution, ocean currents, etc. The South China Sea (SCS) is one of the typical largest margin seas. Its northern part receives sediments from various sources including the Pearl River (PR), the Red River, the Taiwan Island, and the Luzon Island (Liu et al., 2008). The contribution of these different sources to the sediments in the SCS has attracted much attention. Many researchers have attempted to identify the sediment provenances of the northern SCS using sedimentological, geochemical, and mineralogical methods (Shao et al., 2000; Zhang et al., 2002; Tang et al., 2009; Cai et al., 2010; Li et al., 2011; Yan et al., 2012; X.F. Zhang et al., 2012). Shao et al. (2009) analyzed Nd isotopic compositions of recent sediments in the northern SCS and found that materials from the PR mainly influence the southwestern area of the Lingdingyang to Dongsha Island. Ge et al. (2010) suggested that kaolinite within the northern SCS sediments mostly came from the PR, while illite and chlorite are sourced from the Changjiang River and the Taiwan Island. Z.F. Liu et al. (2007) analyzed clay minerals of surface sediments from the PR drainage basin and discussed their contribution to the SCS. They found that the clay mineral assemblage of the PR sediments dominantly consists of kaolinite, with less chlorite and illite, and very scarce smectite. They concluded that "the maximum contribution of clay minerals from the PR is 72% in the northern margin and only 15% in the northern slope of the South China Sea". Liu et al. (2010a) confirmed the clay mineral assemblage as an indicator of the PR input and suggested that materials from the PR were predominantly distributed in the area between the PR estuary and northeast of Hainan Island.

Magnetic methods have been widely used for researching paleoenvironmental evolution as well as modern environmental pollution due to its fast, economical and non-destructive application (Thompson and Oldfield, 1986; Evans et al., 1997; Maher, 2007; Su et al., 2015). Magnetic properties of marine sediments often provide reliable information on environmental change (Kumar et al., 2005; Sangode et al., 2007; Yang et al., 2008; Zheng et al., 2010). In particular magnetic concentration parameters such as magnetic susceptibility (MS) have become a kind of basic data for marine sediment research (Evans and

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Heller, 2003; Yim et al., 2004; Ghilardi et al., 2008; Mohamed et al., 2011). Identifying sediment provenances is one important application of magnetic measurements (Yu and Oldfield, 1989; Hatfield and Maher, 2009; Lyons et al., 2012; Larrasoaña et al., 2015). Though interpretation of magnetic features of marine sediments is complex due to various influencing factors such as terrigenous input and diagenesis of magnetic minerals, magnetic characteristics of marine surface sediments can be regarded as an effective means for identification of sediment provenance (Walden et al., 1997; Rotman et al., 2008; Nguyen et al., 2016).

Recently, magnetic data has been used to identify the origin of surface sediments in the SCS (Liu et al., 2010b). Many previous researches indicated that magnetic response to climate differed at different region of the SCS due to difference in magnetic properties of different sources and transportation pathways (Liu et al., 2003; Wang et al., 2009, 2010; Liu et al., 2010c). However, investigation of magnetic property and its provenance implication for estuary-ocean area along the SCS margin has not yet been reported but is clearly important for the magnetic-based paleo-environmental reconstruction and geological evolution of the SCS. The main purpose of our study is to investigate the similarities and differences of magnetic characteristics of surface sediments collected from the Pearl River Estuary (PRE) and its adjacent seas. Combing magnetic characteristics and mineral analysis, we aim to discuss the significance and effectiveness of magnetic methods to trace provenance features in the study area and thus provide a valuable tool for sediment source tracing of estuary-ocean elsewhere.

2. Materials and methods

2.1. Site characteristics

The Pearl River estuary (PRE) and its adjacent sea waters were selected as the study area of the present study. The PRE is in the warm and humid subtropical area of South China. The average annual rainfall and total runoff of the PR drainage area are 1600 to 2300 mm and 345.78 billion m^3, respectively. Though the suspended sand concentration of the PR is relatively low, the PRE annually discharges about 64 million tons of suspended sediments and more than 0.1 billion tons of total sediments into the SCS from its eight outlets. The PR suspended sediments are the main material source of the study area (Mo and Chen, 1986; Lan, 1996; Long, 1997). However, recent studies indicated that the sediment source of the study area differs at different water depths (Zhou et al., 1991). The eastern four outlets named Humen, Jiaomen, Hongqimen and Hengmen pour waters into a trumpet-shaped estuary called Lingdingyang, which is referred to the PRE in the present study (Fig. 1). The unique topographic features of the PRE are formed under the interaction of river runoff, tidal current, geological structure and geographical factors (Song and Ruan, 1986). The runoff from the East River and North River and part of the runoff from the West River discharge into the SCS through the mentioned four gates. Meanwhile, the Pacific Ocean tidal wave enters into the SCS through the Bashi strait and then spreads across each entrance of the PR. Naturally, two deep grooves in the East and West and three shoals in the East, middle and west were formed within the Lingdingyang waters. Due to the effects of differences in sediment source amount and dynamic conditions, a development pattern characterized with rapid expansion of Western shoal, constriction of the west groove, northwest-southeast elongation of the central shoal, little water depth changes and slight constriction of the East groove, and basically stable of the eastern shoal has been formed (Dong, 1986).

According to the results of hydrological survey of the PRE coastal area, the hydrological characteristics of this area is the result of the two main dynamic factors of runoff and tidal current as well as the influence of other factors, such as the outlet flow system and the wind field. The east wind and east or north wind dominates the PRE coastal area during flood and dry seasons, respectively. The maximum wind speed of this area is 4.0 to 9.9 m/s (Mo and Yan, 1986). The reciprocating flow dominates the thalassic area and coastal current and rotational flow interacts at the waters between the bay mouth and the 20 m water depth isobaths. At the Lingdingyang waters, a strong tidal current and a significant influence of runoff affects the eastern and western areas, respectively (Department of Coastal Hydrology, 1986). According to the result s of sediment test of the Lingdingyang closed area, total deposition dominates this area at both flood and dry seasons (Yang, 1986). Mo and Chen (1986) suggested that the main sedimentary characteristics of the PRE was that the particles ranged from relatively coarse, fine, and gradually coarse from estuary, coastal shallow water area, to the outer waters.

2.2. Sampling

Nineteen surface sampling stations with 14 in the estuary and 5 in the shelf were selected for the present study (Fig. 1). According to previous mineral analysis performed by Z.F. Liu et al. (2007, 2008), sediments at sampling sites A9 and A8 mostly come from the Pearl River (52%), and some smaller fractions from Taiwan (29%) and Luzon (19%). The contribution of the PR, Taiwan, and Luzon to the area of sampling sites A7, A6, and A5 are 31%, 23%, and 46%, respectively. The samples from the PRE were collected in autumn 2010 by using grabbers. Sediments from the adjacent sea were collected in summer 2009, using a box corer. The samples are distributed from the shallow estuary with 2 m water depth to the outer shelf with water depth up to 102 m. All samples were freeze dried before performing measurements.

2.3. Mineral and magnetic analysis

Mineral analysis is a widely used method for provenance identification in the study area. In our study, mineral composition analysis was performed for nine representative samples using a BRUKER D8 ADVANCE X-ray diffractometer (XRD), Cu (monochrome).

A suite of mineral magnetic analyses were performed for all samples in order to determine magnetic concentration and magnetic grain size (i.e. magnetic domain state). Magnetic susceptibility (MS) and temperature variation of MS (κ-T curves) were measured at the Guangzhou Institute of Geochemistry, Chinese Academy of Sciences. All other magnetic measurements were performed at the Department of Geosciences, University of Tuebingen.

Low (976 Hz) and high (15,616 Hz) frequency MS (mass-specific χ_{lf} and χ_{hf}, respectively) were measured using a Kappabridge MFK1-FA (AGICO). Frequency dependent MS was calculated from the expression χ_{fd} (%) = $[(\chi_{lf} - \chi_{hf}) / \chi_{lf}] \times 100$. Variation of MS with temperature (κ-T curves) was measured using CS4/CSL high and low temperature units attached to the MFK1-FA for both bulk samples and magnetic extracts of representative samples. Anhysteretic remanent magnetization (ARM), expressed as susceptibility of ARM (χ_{ARM}) in this paper, was imparted with an AF peak field of 100 mT and a DC biasing field of 0.1 mT using a 2G Enterprises 755 cryogenic magnetometer with attached degausser system. Stepwise thermal demagnetization of triaxial isothermal remanent magnetization (IRM) (applied fields 1.0 T, 0.3 T, and 0.1 T) and alternating field (AF) demagnetization were performed using the 2G magnetometer with the degausser. Hysteresis parameters, which are helpful to identify the type and particle size of magnetic minerals (Day et al., 1977; Dunlop 2002; Zang et al., 2010), were measured using a MicroMag 2900 AGM, with a maximum applied field of 0.6 T. Isothermal remanent magnetization (IRM) acquisition curves with twenty-five steps up to 2500 mT were acquired using a MMPM9 pulse magnetizer and a Molspin Minispin magnetometer. Then IRM was measured at backfields of 100 mT and 300 mT, respectively. The IRM at an applied field of 2500 mT was regarded as saturation IRM (SIRM).

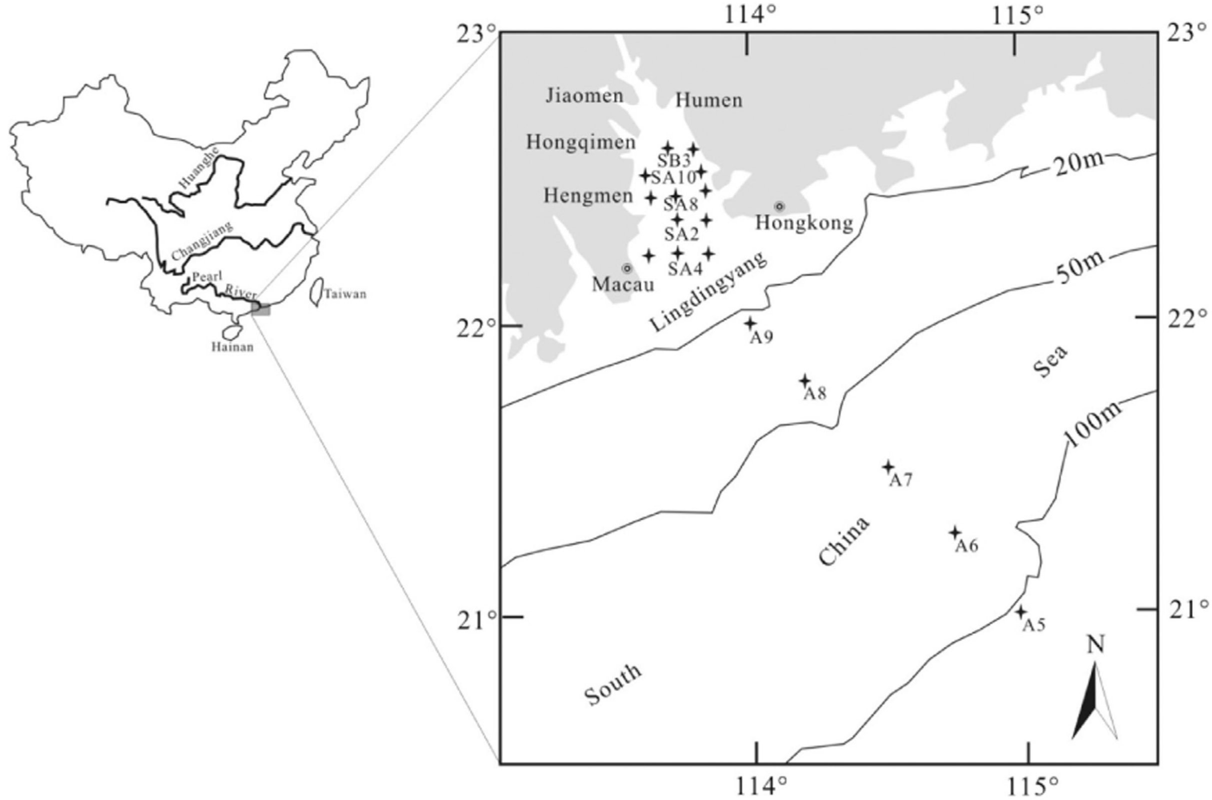

Fig. 1. Study area (including bathymetric lines) and sampling sites. The sampling codes represent the ten cross-sections. A5–9 are termed SEA sites, the others PRE sites.

3. Results

3.1. Magnetic mineralogy

The shape of the κ-T curves is similar for all PRE sediments but differs clearly for the SEA sediments (Fig. 2). There are several interesting features in the κ-T curves of the PRE samples. First, the heating curves of the PRE bulk samples dramatically increase above ~300 °C and decay toward the Curie temperature of magnetite at about 580 °C, with cooling curves running at a much higher level. This indicates the production of new magnetite by the reaction of hematite and organic matter or clay minerals (C.X. Zhang et al., 2012). In the PRE extracts, new formation of magnetite is much less, as one expects because of the lower content of non-ferrimagnetic materials. Second, the κ values already start to increase from room temperature, which is likely an effect of the original presence of small-sized magnetite in a single-domain (SD) or small pseudo-SD (PSD) domain state. Third, the small decrease of κ from about 250 to 300 °C for the PRE bulk samples could be due to greigite. This bump is not present in the κ-T curves of magnetic extracts from the PRE, possibly because of selective separation. The magnetic extracts of SEA samples additionally show a small decrease between about 300 to 400 °C, probably due to some maghemite that transforms into hematite (Liu et al., 2005; Jelenska et al., 2010). An important feature of the SEA samples is the clear κ signal above 580 °C with a Curie temperature around 680 °C, in both bulk samples and magnetic extracts, evidencing the presence of hematite. The occurrence of a hematite signal in κ-T curves suggests a very large proportion of hematite in these samples.

Cumulative log-Gaussian (CLG) analysis of IRM acquisition curves is widely used for magnetic composition analysis (Kruiver et al., 2001; Heslop et al., 2002; Gong et al., 2009; Ouyang et al., 2015). The results of the gradient acquisition plot (GAP) analysis for the PRE and SEA sediments are illustrated in Fig. 3. According to the results the magnetic composition consists of magnetite (component 1; C1) and some hematite (component 2; C2) for both the PRE and SEA sediments (Fig. 3a). The SIRM variations of magnetite and hematite indicate a decreasing magnetite and hematite concentration from the PRE to its adjacent seas (Fig. 3b). The gradually increasing $B_{1/2}$ and relatively stable DP of magnetite suggest the grain size tends to be on average finer. The varying $B_{1/2}$ and DP patterns of hematite indicate a variable grain size distribution of hematite.

3.2. Magnetic granularity

The Day plot (Day et al., 1977; Dunlop, 2002), is regarded as an effective method for magnetic domain identification when the magnetite is the dominating ferrimagnetic mineral. The domain state of the measured samples is illustrated in Fig. 4. From the modified Day plot (Dunlop, 2002), it is indicated that magnetite in the PRE samples are more MD-like than in the SEA samples. Although A8 and A9 are located within the SEA area, some samples from these sites are still more similar to the PRE samples (Fig. 4).

3.3. Clay mineral composition

Mineral analysis has been a widely used method for provenance identification in the study area. The XRD results indicate that the mineral composition of the PRE differs clearly from the SEA sediments (Fig. 5). Calcite as well as relatively high contents of illite and chlorite was found to be dominant within the SEA sediments, while kaolinite was scarcely identified. However, sediments from the PRE show a very different mineral composition. They are characterized by the absence of calcite, but relatively high contents of kaolinite and low contribution of illite and chlorite. These mineral analysis results indicate that the source of the PRE and the SEA sediments may be very different. The semi-quantitative analysis of the XRD results indicates that the concentration of chlorite and the ratio of quartz to feldspar are clearly different between the PRE and the SEA sediments (Fig. 5a). The PRE and SEA sediments can be separated through the triangular plot of clay mineral composition (Fig. 5b).

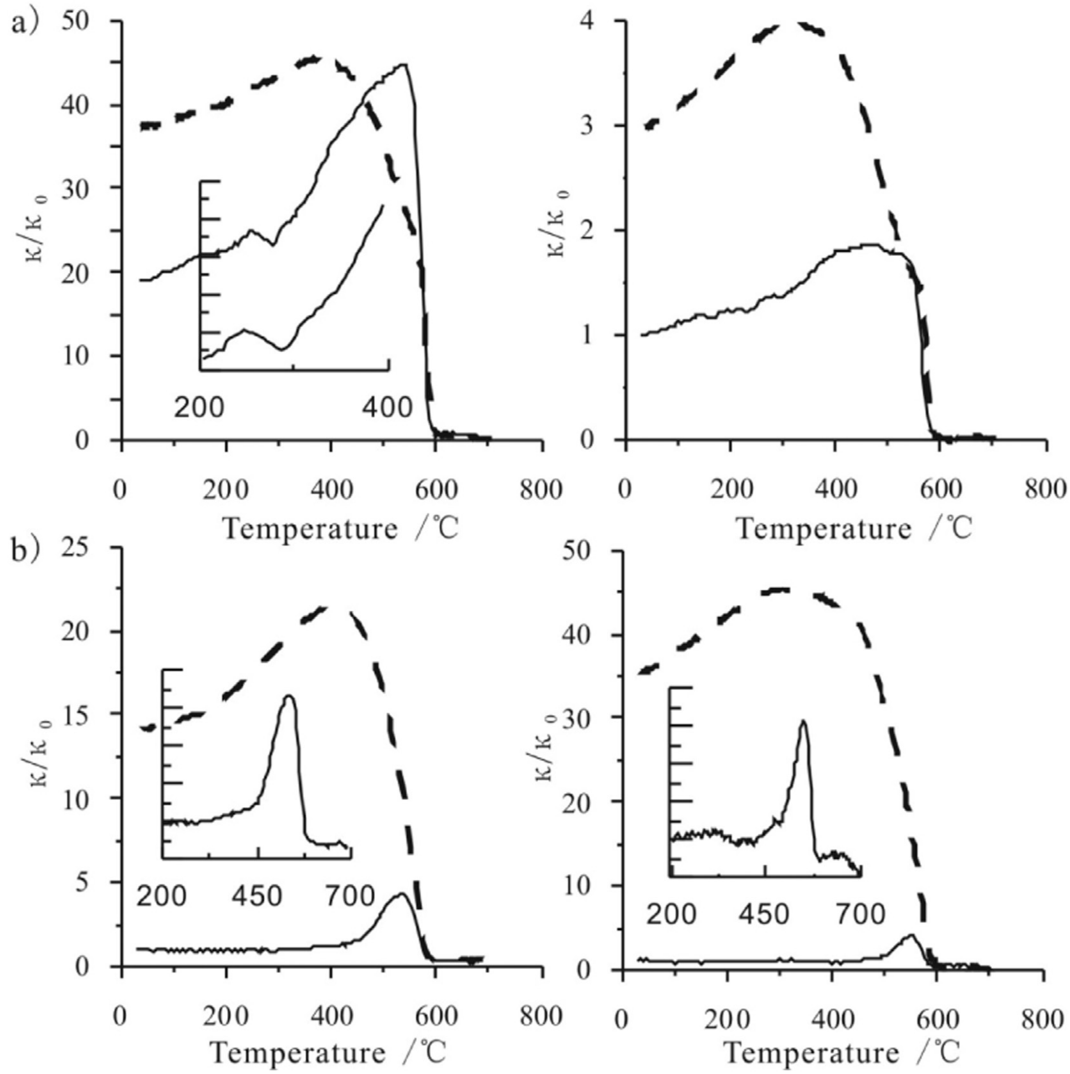

Fig. 2. Typical thermomagnetic (κ-T) curves measured in argon atmosphere for samples from a) the PRE, and b) SEA sediments. The left and right figures are for bulk samples and magnetic extracts, respectively. Solid and dashed lines represent heating and cooling curves, respectively. The small inserts show enlargements of heating curves in specific temperature intervals.

3.4. Variation of magnetic properties from the PRE to its adjacent outer sea

Fig. 6 shows the variation of discriminative magnetic parameters from the PRE toward the adjacent waters. From the PRE to the sea, the concentration parameters χ, χ_{ARM}, and SIRM gradually decrease, and drop sharply between the PRE and the outer sea. Values of χ and SIRM of the PRE surface sediments are five and four times higher than those of surface sediments from waters outside of the PRE, respectively (Fig. 6). The gradual increase of B_c reveals that magnetic minerals get harder from the PRE to the SEA (Fig. 6). Except the sampling site SB3, the relative contribution of magnetite/hematite within sediments reflected by the ratio of $SIRM_{(c1)}/SIRM_{(c2)}$ (from CLG analysis) showed a decrease from the PRE to sampling sites A9 and A8, then followed by a sharp increase with water depth over 50 m (Fig. 6). The frequency dependent magnetic susceptibility $\chi_{fd\%}$ is relatively stable within the sediments with water depth below 50 m, and then gradually increases. The χ_{ARM}/χ and χ_{ARM}/SIRM ratios, reflecting magnetic particle size, also show a significant fining of magnetic particle sizes from the PRE to its adjacent sea. These results are consistent with the results of the sedimentary characteristics survey for the PRE (Mo and Chen, 1986).

4. Discussion

4.1. Controls of magnetic properties

With increasing water depth, magnetic concentration values decrease significantly due to the decreasing sediment deposition. A significant negative correlation between log (χ_{lf}) and water depth is observed (Fig. 7). The reason for this relationship is complex as water depth can reflect many influencing factors such as sedimentary environment, sediment influx, sediments particle size, etc.

A significant positive linear correlation exists between χ_{lf} and SIRM for all samples (Fig. 8a) indicating that the variation of these two parameters is largely controlled by a common type of magnetic minerals. A significant negative correlation exists between χ_{lf} and $B_{1/2}$ of component 1 (magnetite) when all samples are included. This correlation disappears when the sediment samples are separated into two groups i.e., the PRE and SEA samples (Fig. 8b). These results indicate that the difference of magnetic properties between the PRE and SEA sediments is mainly controlled by the concentration of soft magnetic minerals, predominantly magnetite, and a fining of magnetic grain size in this fraction. These results exactly correspond to the sediment characteristics of the PRE and its outer waters (Mo and Chen, 1986).

The relationship between χ_{lf} and χ_{fd} (%) shows a clear difference

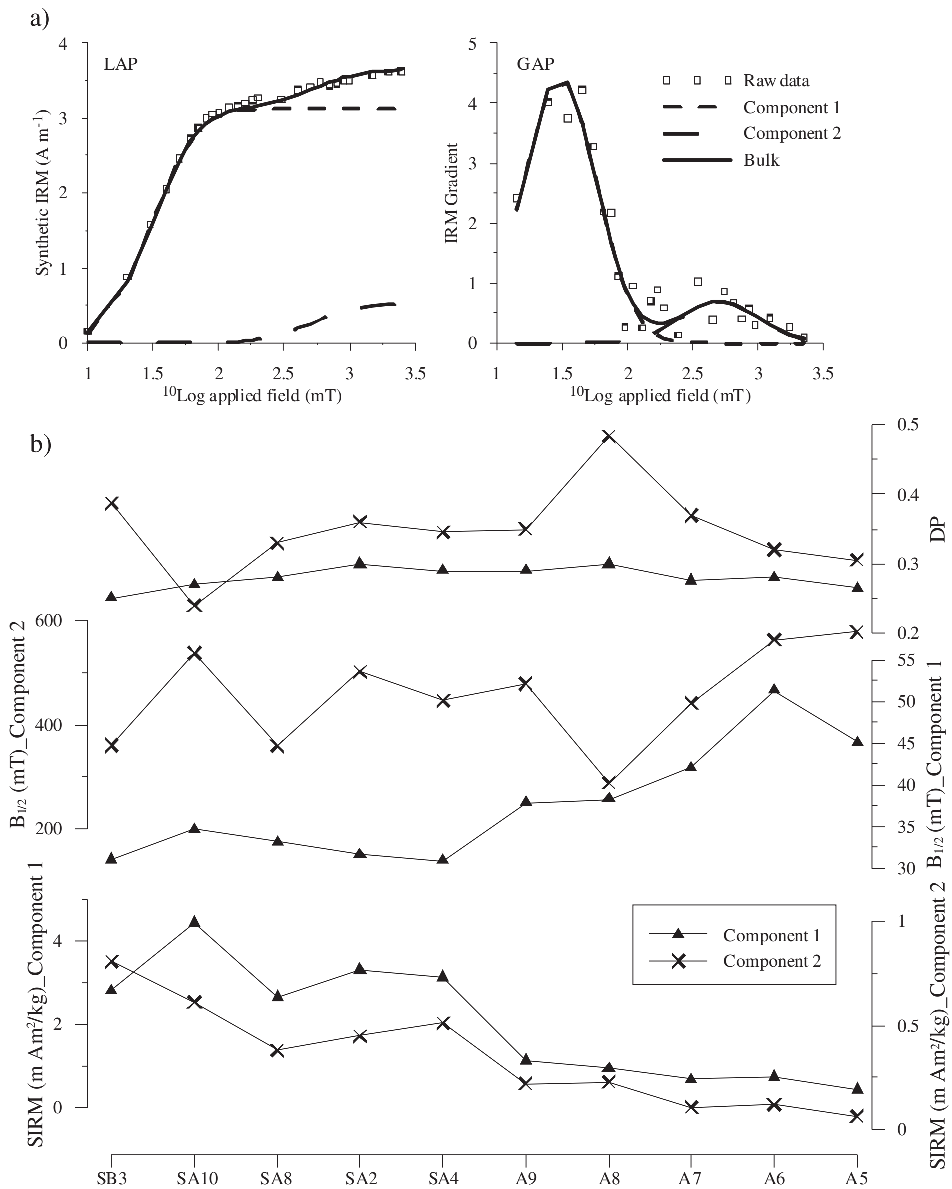

Fig. 3. a) Example of a linear acquisition plot (LAP) and gradient acquisition plot (GAP) of IRM acquisition curves analyzed after Kruiver et al. (2001). b) Variations in fitted IRM parameters from the PRE (SB3-SA4) to the SEA sediments (A9–A5). The possible magnetic mineral composition includes two components (predominantly magnetite and hematite) for all samples.

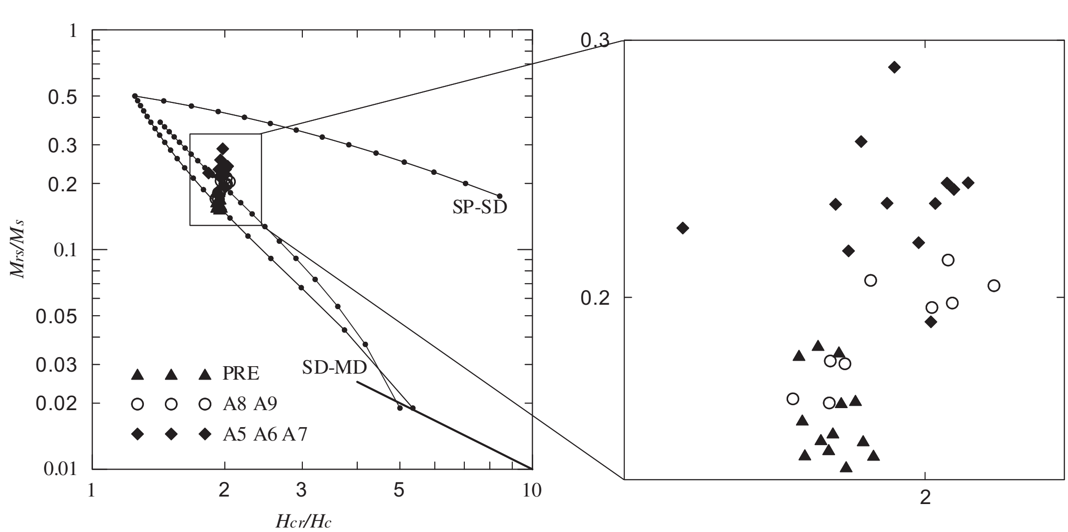

Fig. 4. Magnetic grain size (domain state) determination using the modified Day-plot (Dunlop, 2002).

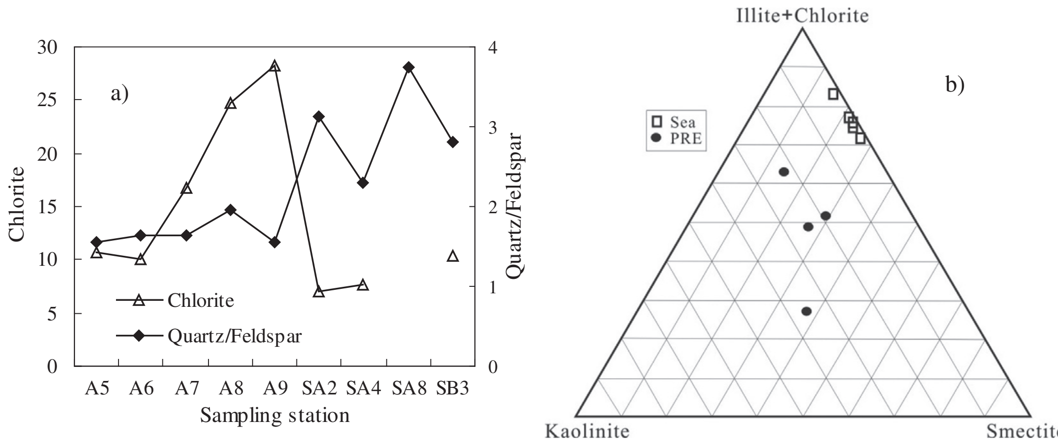

Fig. 5. Results of clay mineral analysis. a) Variation of mineral composition at the studied stations, and b) triangular plot of clay minerals.

between sediments from the PRE and its adjacent waters. The χ_{fd} (%) values are scattered but on average higher in the PRE sediments than that in the SEA sediments. A significant positive linear correlation between χ_{lf} and χ_{fd} (%) exists for the SEA sediments (Fig. 9a). The linear correlation of χ and χ_{fd} in the SEA samples indicates that higher χ values are partly caused by a larger fraction of superparamagnetic (SP) particles. Although the average χ_{fd} (%) value is higher in PRE sediments, no obvious correlation is found for these samples. The two trend lines of the clear positive linear correlation between χ_{lf} and χ_{ARM} for both PRE and SEA samples are not parallel (Fig. 9b). Overall, these results indicate that the size distribution of the magnetic particles in the PRE sediments differs from the one in the SEA sediments, which may imply different sources for the sediments in the PRE and its adjacent seas.

4.2. Source significance of magnetic properties

As discussed above both concentration and particle size of magnetic

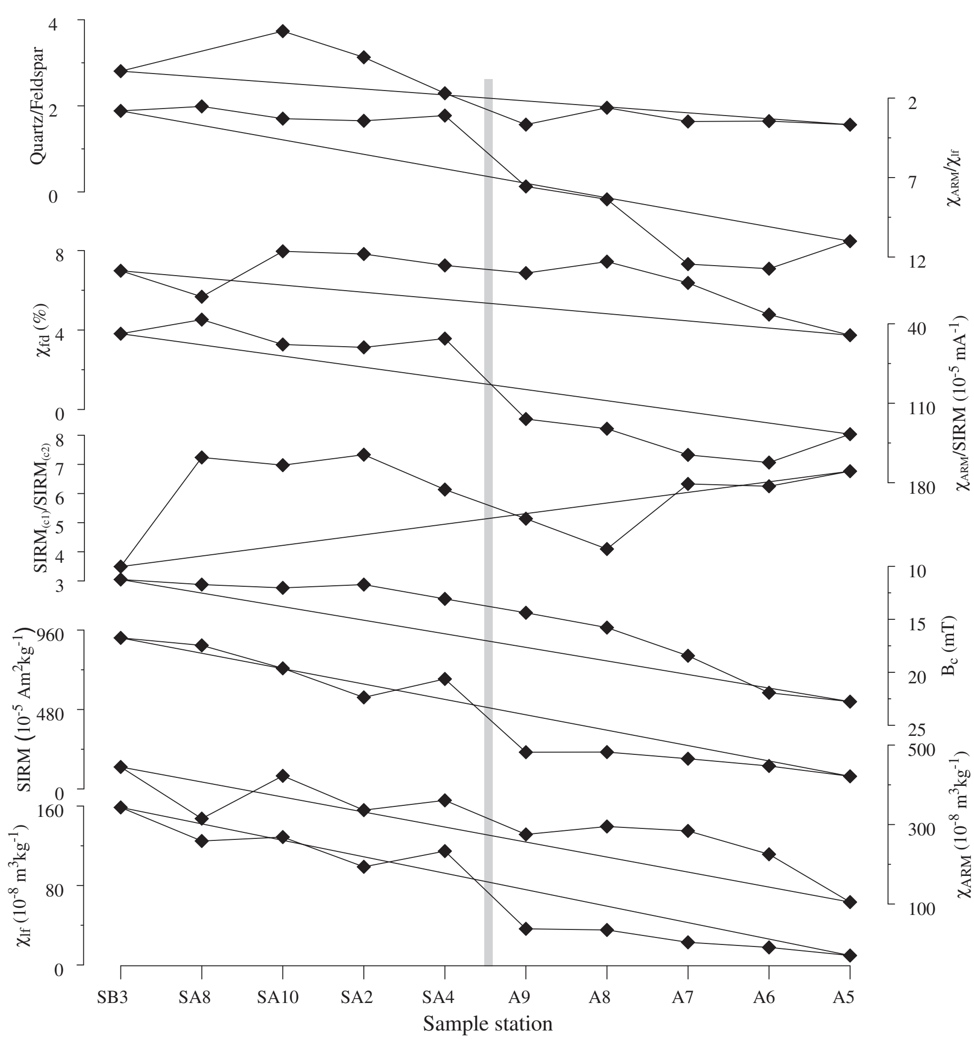

Fig. 6. Variation of magnetic properties and quartz/feldspar content from the PRE (left side of the shaded bar) toward the outer sea (right side of the shaded bar).

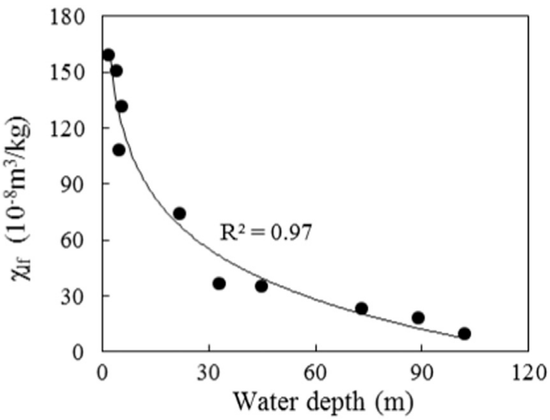

Fig. 7. Relationship between (low frequency) mass-specific magnetic susceptibility and water depth. R^2 refers to a linear correlation of log (χ_{lf}) and water depth.

minerals play a significant role for the sediment magnetic properties. The IRM parameters, the χ_{ARM}/SIRM ratio and χ_{fd} (%) reveal the variation of particle size and relative contents of magnetic minerals. Sediments collected from the PRE and its adjacent seas can be obviously separated by the scatter plot of $B_{1/2}$ of component 1 and χ_{ARM}/SIRM (Fig. 10a). Since sediment deposition dominates the Lingdingyang closed area (Yang, 1986) and sediment particles change from relatively coarse, fine, and gradually coarse from estuary, coastal shallow water area, to the outer waters (Mo and Chen, 1986), sediments from the PRE are represented by a higher soft mineral content and coarser magnetic particles, but also by a larger ultrafine SP fraction. As suggested by Hilton (1987), the main magnetic carriers within the PRE sediments should originate from coarse-grained ferrimagnetic mineral input from the Pearl River. The SP fraction may arise from soil in the catchment. These SP particles are probably largely dissolved in the SEA sediments (Leslie et al., 1990; Grison et al., 2011; Ouyang et al., 2015).

Many previous studies suggested that magnetic measurements are an effective method for identifying sediment sources (Jenkins et al., 2002; Hounslow and Morton, 2004). Recently, some new magnetic parameters were established for provenance research. For example, Q.S. Liu et al. (2007) proposed the concept and application of the L-ratio. Variable L-ratio values were regarded to reflect changes in the provenance or other factors that influence the properties or relative proportions of hematite. However, more study proved that the ratio (HIRM/[0.5 × (SIRM + $IRM_{-100\ mT}$)]) was a suitable substitute for the L-ratio. The samples can be separated into three groups from the scatter plot of SIRM of component 2 and HIRM/[0.5 × (SIRM + $IRM_{-100\ mT}$)] (Fig. 10b). This finding provides an effective method for the study of the provenance of estuarine and offshore sediments, and it is of great significance to the study of marine geology. The PRE sediments are characterized by relatively high SIRM of component 2 and HIRM/[0.5 × (SIRM + $IRM_{-100\ mT}$)]. The ratio HIRM/[0.5 × (SIRM + $IRM_{-100\ mT}$)] value of sediments from sampling sites A8 and A9 was basically consistent with the PRE, indicating the source of this group is the same as for the PRE, i.e. mainly from the PR basin. These results are similar to the results of mineral analysis for this region performed by Liu et al. (2008). However, compared with the PRE, sediments from the sampling sites A8 and A9 have relatively lower SIRM of component 2. The HIRM/[0.5 × (SIRM + $IRM_{-100\ mT}$)] values for sediments from sampling sites A5, A6, and A7 are obviously different from the mentioned two groups, indicating a different sediment source. Moreover, the results of principle component analysis of $B_{1/2}$ of component 1, $SIRM_{(C1)}/SIRM_{(C2)}$, and χ_{lf} indicate that the PRE and A8, A9 sediments are accounted for by PC1 and sediments from sampling sites A5, A6, and A7 are contributed for by PC2 (Fig. 10c). Sampling sites A5, A6, and A7 were located on the N Shelf as described by Liu et al. (2008). According to Liu et al. (2008), the sediments of this area came from three sources, including the Pearl River, Taiwan, and Luzon. At the same time, different contributions of these three sources to A5, A6, and A7 can explain the obvious difference among these three sampling sites.

5. Conclusions

Together with mineral analyses, the rock magnetic investigation conducted in this study relates the magnetic properties of the estuary and its adjacent sea sediments to their provenance. The following conclusions can be deduced:

1) Magnetite and hematite coexist within the sediments from both the PRE and its adjacent seas. Though no obvious difference exists in the composition of the magnetic minerals between the PRE and its adjacent seas, the grain size of magnetite particles in the PRE sediments is coarser than in the adjacent sea sediments, but also an ultrafine SP fraction is observed. From the PRE to its adjacent seas, magnetic parameters reflected by magnetic mineral concentration decrease gradually and change sharply between the PRE and the sea. The parameter ratios related to magnetic particle size indicate that magnetic particles in the MD to PSD range are getting finer from the PRE to its adjacent seas, while the SP fraction is largely dissolved.
2) Though sediments collected from the sampling sites A8, A9 were classified to the SEA due to the triangle plot of clay mineral composition, the scatter plot SIRM of component 2 and HIRM/[0.5 × (SIRM + $IRM_{-100\ mT}$)] and principle component analysis results of $B_{1/2}$ of component 1, $SIRM_{(C1)}/SIRM_{(C2)}$, and χ_{lf} indicate that the sediments at these two sites had partially the same source as the PRE sediments. Therefore, the implication of magnetic parameters for provenance is clearer than the clay mineral analysis.

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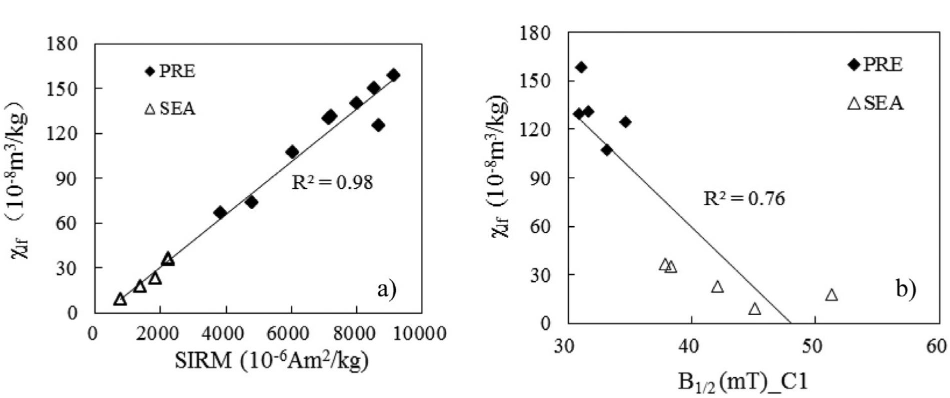

Fig. 8. Relationship between (low frequency) mass-specific magnetic susceptibility and a) SIRM, b) $B_{1/2}$ of component 1.

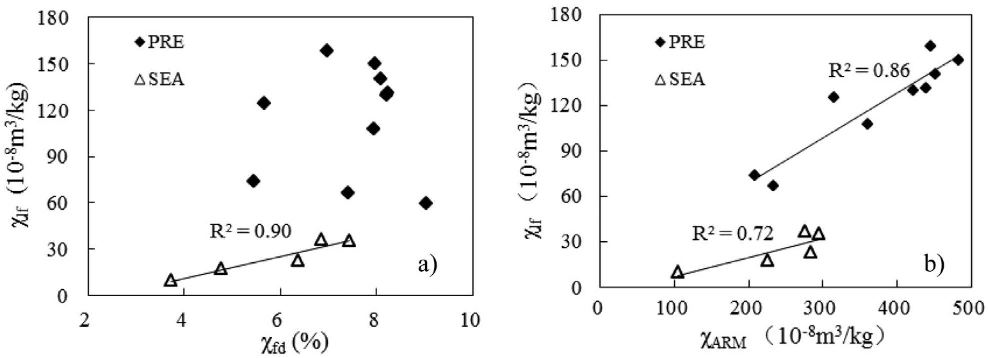

Fig. 9. Relationship between (low frequency) mass-specific magnetic susceptibility and a) χ_{fd}, b) χ_{ARM}; R^2 values are calculated for linear correlation (separately for PRE and SEA samples).

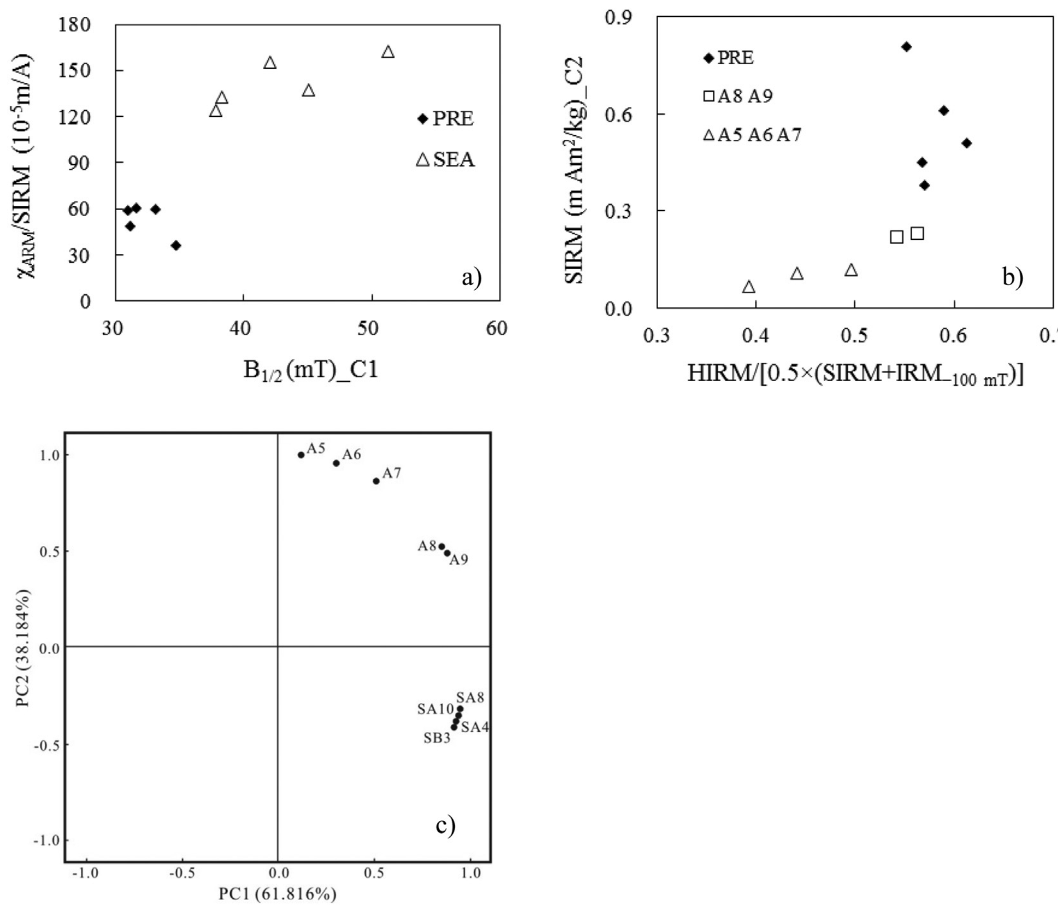

Fig. 10. Scatter plot of a) χ_{ARM}/SIRM and $B_{1/2}$ of component 1, b) SIRM of component 2 and HIRM/[0.5 × (SIRM + $IRM_{-100\ mT}$)], and c) PC1 and PC2 from principle component analysis of $B_{1/2}$ of component 1, $SIRM_{(C1)}/SIRM_{(C2)}$, and χ_{lf}.

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