Magnetic fingerprints of surface sediment in the Bohai Sea, China

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ABSTRACT

The understanding of the surrounding river impacts on the sediment supply to the Bohai Sea, northern China, is insufficient. In this study, magnetic measurements on surface sediment from the Bohai Sea and surrounding rivers were performed to interpret the distribution and origin of sediment in the Bohai Sea. Different magnetic properties in the analyzed material correspond to seven provenance areas. The magnetic mineral content decreases gradually toward the central Bohai Sea from its marginal areas. Sediment in the central and northern parts contains higher concentrations of finer magnetic minerals than the southern Bohai Sea. The western Bohai Sea (western Bohai and Liaodong bays) has high contents of magnetic minerals because its provenance areas contain abundant old metamorphic rocks with magnetite. The Yellow River, with the most elevated sediment flux, contributes with fine, high-coercivity magnetic minerals (e.g., hematite) to the southern Bohai Sea (mainly in the northwestern Laizhou Bay). Some local rivers (e.g., Fuzhou River) are also important contributors to the coastal waters, due to the magnetic similarities. Sediment from the north Yellow Sea is magnetically similar to the Yellow River detritus. The results highlight the use of the rapid magnetic method in provenance tracing.

1. Introduction

Intracontinental seas surrounded by continents are connected to open oceans with straits. The seas play a significant role in terrigenous sediment dispersal since they are mainly fed with fluvial material with a composition that is affected by land weathering, human activities, oceanic circulations, and continental-oceanic interactions (Imai et al., 2006; Qiao et al., 2010; Zalasiewicz et al., 2016). The Bohai Sea in northern China, connected with the Yellow Sea by the Bohai Strait, is a nearly-closed inland sea encircled on three sides by the Liaodong Peninsula, the Northern China Plain, and the Shandong Peninsula (Fig. 1). It is composed of the Liaodong Bay, the Bohai Bay, the Laizhou Bay, the Central Basin, and the Bohai Strait (Fig. 1; Qin et al., 1985). The sea is supplied by dozens of rivers flowing through regions with disparate lithology and human activity intensities. Among these rivers, the Yellow River (Huanghe), one of the worldwide largest rivers, is thought to be the primary source of the Bohai Sea and other east China seas, especially for suspended load (Qin et al., 1985; Qiao et al., 2017). For instance, it is confirmed that the Yellow River sediment settle in the Laizhou Bay, east side of the river estuary (Fig. 1) (Li et al., 2010; Qiao et al., 2010), but numerical simulations indicate that the Yellow River is responsible for only 30% of the suspended in the northern Bohai Sea (Li

et al., 2010). Thus, studying the sediment contributions from rivers of small sizes is essential for a full understanding of the sediment transport into the Bohai Sea.

Over the past decades, considerable influence from local rivers other than the Yellow River has been revealed by mineralogical and geochemical sediment compositions. As an example, Chen et al. (1980) divided the Bohai Sea into several sub-provenance areas of different fluvial catchments based on detrital mineral distributions. The southeastern Laizhou Bay in the southern Bohai Sea (Fig. 1) is known to be fed by waterways along the south and east coasts, as revealed by heavy minerals, such as amphibole and epidote (K. Wang et al., 2010c). Heavy-mineral distributions indicate that the Yellow River only affects the central part of the Bohai Bay in the west, and that the northern part of the bay instead is mainly fed from the Luanhe River that enters the Bohai Sea in the northwest (Fig. 1; Han et al., 2013). In addition, Dou et al. (2014) revealed that the clay minerals in the northern Liaodong Bay in the northeastern Bohai Sea partly derive from rivers in the north, and that the western Liaodong Bay is supplied by the Luanhe River, although Hou et al. (2017) argued for a possible transport pathway from the north also for the western Liaodong Bay. Besides, Fuzhou River, a small stream entering the eastern Liaodong Bay, is proven to be a contributor there (Chen et al., 1980; Hou et al., 2017; Wang et al.,

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Fig. 1. Topography, sampling sites (coloured and black dots), river systems, and sediment types in the Bohai Sea and adjacent regions. Numbers represent the 34 rivers listed in Table 1. The nine potential provenance regions are marked by nine colors. The solid yellow line and white dotted lines stand for the -20 m isobath and boundaries of different sea regions, respectively. The sediment types are from Liu et al., (2010). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2014). These previous studies only deal with parts of the Bohai Sea, and lack sediment samples from rivers that represent the source regions.

A large-scale sediment-transport perspective is possible by using mineral magnetism, a non-destructive and more rapid method than traditionally mineralogical and geochemical methods. Due to significant magnetic difference for sediment from different source areas (Caitcheon, 1993; Kissel et al., 2016, 2017; Zhang et al., 2008), magnetism is widely used to track transport of fine-grained sediments (Ellwood et al., 2006; Gamboa et al., 2017; Hatfield et al., 2013; Hornig and Huh, 2011; Kim et al., 2013; Kissel et al., 2018; Wang et al., 2017; Xue et al., 2019). For instance, sediment of the Yellow-River and Yangtze-River estuaries can be distinguished by different relative contents of ferrimagnetic and anti-ferromagnetic minerals and saturation isothermal remanent magnetization (Li et al., 2012; Zhang et al., 2008). Furthermore, minerals in Iceland and Greenland sediment sources have different magnetic-crystal grain-sizes, making it possible to discriminate provenance in the northern North Atlantic Ocean (Hatfield et al., 2013). East China Sea sediment from the old Yellow River, the Yangtze River, Taiwan rivers, and Korea rivers have been distinguished by magnetic parameters and Fuzzy c-means cluster analysis (Kim et al., 2013). Variations in relative content of ferrimagnetic and anti-ferromagnetic minerals in a core indicates that the provenance in the northwest slope of South China Sea changed with time over the past 36 kyr (Li et al., 2018).

Magnetic properties for sediment from the Liaodong Bay (Wang et al., 2015), Central Bohai Sea (Wang et al., 2017), Yellow River estuary (Zhang et al., 2008), and mud area (Y. Wang et al., 2010a) has been used to discriminate different sediment sources for Bohai Sea deposits. However, the Yellow River was the only river sourcing in the Bohai Sea itself that were included in the magnetic studies, thus making the understanding of the sediment flux into the Bohai Sea incomplete.

In this study, multiple magnetic parameters of surface sediment collected from the Bohai Sea and its surrounding rivers, as well as the adjacent north Yellow Sea are presented. The results lead us to constrain the transport paths of magnetic sediment particles for the entire Bohai Sea.

2. Regional settings

2.1. Physiography and sediment distributions

The Bohai Sea, connected with the Yellow Sea by the Bohai Strait, is a nearly-closed inland sea encircled on three sides by the Liaodong Peninsula, the Northern China Plain, and the Shandong Peninsula (Fig. 1). The total area is 77,000 km² and the average water depth is 18 m (Xu et al., 1997). The seabed of the three bays slope down to the central basin and the strait, and the deepest point is located in the Laotieshan Waterway with -84 m. (Fig. 1). The average slopes for the Liaodong, Bohai, and Laizhou bays all are 0.2%. The average and deepest water depths in the Liaodong Bay are < 30 m and > 60 m, respectively. Notably, the seabed of the Liaodong Shoal on the north side of Laotieshan Waterway is undulating. The Bohai Bay's maximum and mean water depths are 39 m and about 20 m, respectively. The Laizhou Bay is shallower than 20 m, with an inconspicuous underwater bank slope on the east bank. The water depth of the Bohai Central Basin ranges from 20 to 26 m (Xu et al., 1997). The Bohai Strait, a connection between the Bohai and Yellow seas, is about 115 km in width and is divided into several waterways by the Miaodao Islands (Fig. 1) (Xu et al., 1997).

A “Y-shaped” mud region filled with silt clay distributes in the central Bohai Bay, Central Basin, northern Laizhou Bay, and extends to the Yellow Sea along the south coast (Liu et al., 2010; Qiao et al., 2017). Coast shoals in southern Laizhou Bay, south Bohai Bay, and northern Liaodong Bay are covered by silt. However, sediments in the north Bohai Bay, and the east and west sides of Liaodong Bay are medium to fine sand, and fine sand. Sand-silt-clay sediments distribute in the periphery of mud region. Fine sand and silty sand distribute in areas close to the central and northern Bohai Strait.

2.2. Catchment geology and hydrography of the Bohai Sea

Except for rivers close to Qinhuangdao, such as the Liugu River that in the western Liaodong Bay, almost sediment samples covering all

Table 1
General characteristics of rivers flowing into the Bohai Sea.

River No.	River name	Length (km)	Area (km ²)	Water discharge (10 ⁸ m ³ /a)	Sediment load (10 ⁴ t/a)	Duration	References
01	Dayang	230	1968.4	31	12.58	/	Zhang, 2000
02	Biliu	165	2848	3.78	13.21	1956–2000	Yang, 2011
03	Fuzhou	133.7	1593	1.37	16.8	/	Chen et al., 1998
04	Daliao	97	1962	43.62	193.66	1963–2007	H. Wang, 2010
05	Liao	1396	12.64 × 10 ⁴	31.29	1420	1954–2015	MWRC, 2019
06	Daling	397	23,549	19.63	2740	/	Chen et al., 1998
07	Xiaoling	397.4	23,549	4.03	364	/	Chen et al., 1998
08	Luanhe	877	4.49 × 10 ⁴	46.51	1739	1950–1984	Chen et al., 1998
09	Xiaoqinglong	75	383	/	/	/	/
10	Dou	121.4	1340	13.10	12.1	/	Chen et al., 1998
11	Chaobai	458	14.8	7.30	7.30	1973–1997	Lei et al., 2007
12	Yongding	> 740	5.08 × 10 ⁴	0.85	1.71	1973–1997	Lei et al., 2007
13	Jiyun	144.54	1.1 × 10 ⁴	6.45	6.81	1973–1997	Lei et al., 2007
14	Haihe	1036	8.4 × 10 ⁴	7.921	2540	1960–2015	MWRC, 2019
15	Dagu	80.5	/	/	/	/	/
16	Beipaihe	143.8	1328	0.93	2.67	/	Chen et al., 1998
17	Duliujian	67	511	10.2	/	1950–1999	Lei et al., 2007
18	Ziya New	143.4	5200	3.09	38.6	/	Chen et al., 1998
19	Xuanhui	169.7	3031	0.44	4.24	/	Chen et al., 1998
20	Zhangweixin	202.7	3.70 × 10 ⁴	1.32	Silted	1973–2003	Yu, 2004
21	Majia	448	12,239.2	2.93	76.2	/	Chen et al., 1998
22	Tuhai	446.5	1368	8.97	157	/	Chen et al., 1998
23	Chao	73.5	11,408	/	/	/	Zhao and Yu, 2019
24	Yellow	5465	75.19 × 10 ⁴	292.8	67,400	1952–2015	MWRC, 2019
25	Guangli	47.3	500	/	Silted	/	Yang et al., 2005
26	Zimai	134.55	3356	/	/	/	/
27	Xiaoqinghe	233	10,498.8	8.78	36.9	/	Chen et al., 1998
28	Mihe	206	3847	4.30	84.09	/	Chen et al., 1998
29	Bailang	127	1237	0.39	/	/	Yao et al., 2014
30	Yu	75	890	/	/	/	Yao et al., 2014
31	Wei	242	6367	14.46	92.44	/	Chen et al., 1998
32	Wang	50	449.19	/	/	/	/
33	Jie	45	576.8	/	/	/	/
34	Jia	80	2295.5	6.15	34	1953–1957; 1966–1979	Jin et al., 2007

rivers around the Bohai Sea are investigated in this study (Table 1). Larger rivers flowing into the Bohai Sea mainly include the Yellow, Haihe, Liaohe, and Luanhe Rivers (Fig. 1). The Yellow River is the second-longest river in China, originating from the Tibetan Plateau and flowing through the Loess Plateau and North China Plain (Milliman and Meade, 1983). The annual average runoff is $292.8 \times 10^8 \text{ m}^3$ (MWRC, 2019) and annual mean sediment load is 674 Mt. The Haihe and Luanhe Rivers originate from the mountains on the northwest side of the North China Plain, and flow through large, heavy industry cities, Tianjin and Tangshan, respectively. The Liaohe River passes through the Northeastern China Plain. Due to the regional precipitation being controlled by the monsoon, river sediment is particularly abundant in summer and autumn. For instance, the runoff between June and September accounts for about 3/4 of the annual runoff of the Luanhe River. For the Yellow River, runoff, and sediment load from July to October accounts for 60% and 80% of the annual quantities, respectively (Su and Yuan, 2005).

Based on lithological differences in the catchment areas, the surrounding rivers can be divided into nine possible provenance groups (Fig. 1). The Dayang and Biliu rivers (Region A) originate from the Liaodong Peninsula mountains covered by Proterozoic monzonitic granite and Indosinian to Jurassic monzonitic granite, respectively (Liaoning Bureau of Geology and Mineral Resources, 1989). The Fuzhou River (Region B) in the western Liaodong Peninsula flows through Upper Proterozoic shale, sandstone, and limestone (Liaoning Bureau of Geology and Mineral Resources, 1989). The Liaohe and Daliao rivers (Region C) mainly flow through plains covered by Quaternary sediments. The Daling and Xiaoling rivers charging the western Liaodong Bay (Region D) coast flow through Jurassic and Cretaceous andesite, rhyolite, shale, and sandstone, and Archaean gneissic granite. Archaean gneissic granite is widely distributed in the west side of the Liaodong Bay. The Luanhe-Haihe River System, including the Luanhe, the Xiaoqinglong, the Dou, the Chaobai, the Yongding, the Jiyun, the

Haihe, the Dagu, the Beipai, the Duliujian, and the Ziyaxin rivers (Region E), flows through Archean metamorphic rocks (e.g., amphibolite, gneiss) and Quaternary sediment in the broad northern China plain (Hebei Bureau of Geology and Mineral Resources, 1982). These rivers pass through or close to big cities, including Beijing, Tianjin, and Tangshan (Fig. 1).

The Xuanhui, Zhangweixin, Majia, and Tuhai rivers of the Haihe River System in the southern Bohai Bay and the Yellow River system further east have the same catchment (Region F), as the these rivers were captured by the Yellow River several times over the past thousand years (Ye, 1989a, b). Loess terrain in the middle reaches provides more than 90% sediment to the Yellow River (Ren et al., 1986). Rivers close to the southern Laizhou Bay (Region G) pass through mountains covered by Archaean metamorphic rocks and a plain with the Quaternary sediment. Rivers in the western and northern Shandong Peninsula (Regions H and I) flow through granite regions (Shandong Bureau of Geology and Mineral Resources, 1991).

2.3. Marine hydrodynamic characteristics

Most areas of the Bohai Sea are controlled by regular, semidiurnal tides (Qiao et al., 2008). The tidal range and flow rate in most areas of the Bohai Sea are 2–3 m and 0.5–2.0 m/s, respectively. Waves in the Bohai Sea is mainly caused by wind, with average heights of 1.1–1.7 m in January, 3.5–6.0 m during cold periods, and 6 m during occasional typhoons. The waves have a particularly strong influence in the Bohai Strait and the central sea (Feng et al., 2007). Ocean circulations of the Bohai Sea include the Yellow Sea Warm Current, the Liaonan Coastal Current, the Liaodong Coastal Current, and the southern Bohai Sea Coastal Current (Fig. 2). Surface circulations are controlled by the regional winds with directions of the north in winter and southeast in summer (J. Wang et al., 2010b). In winter, the high-salinity Yellow Sea

Fig. 2. Ocean circulations in the Bohai Sea in summer (a) and winter (b). YSWC = Yellow Sea Warm Current, LNCC = Liaonan Coastal Current, LDCC = Liaodong Coastal Current, BSCC = Bohai Sea Coastal Current (modified after Fang et al., 2000; Guan, 1994; Zhao et al., 1995). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Warm Current enters the Bohai Sea from the Yellow Sea through the northern Bohai Strait (Fig. 2b). In the northwestern Bohai Sea, the current is divided into two branches. One branch moves northward along the west bank of the Liaodong Bay and forms the anti-cyclonic Liaodong Gyre and two weak reversed gyres (Fig. 2b; Fang et al., 2000). Another branch moves into the Bohai Bay and forms a counterclockwise circulation when encountering the diluted northward-directed Yellow River water (Zhao et al., 1995). The Yellow-River water flows via the Laizhou Bay and the southern Bohai Strait into the Yellow Sea (Guan, 1994). Strong winter winds with cold surges lead to a vertical mixture of the water body in the Bohai Sea and drives the currents southward (Su, 1998). In summer, with the weakening or disappearance of the Yellow Sea Warm Current, the cold bottom tongue from the Yellow Sea Cold Water enters the Bohai Sea through the Bohai Strait (Guan, 1994). In addition, the summer winds drive northward boundary currents in the shallow waters near the coast (Dou et al., 2014). Within the Liaodong Bay, the current moves clockwise in the long-term, but in the opposite direction when the Liaohe River discharge increases, and favourable wind prevails (Guan, 1994). Currents of the Bohai and Laizhou bays in summer resemble that in winter (Zhao et al., 1995).

3. Materials and methods

3.1. Sample acquisition

During two survey cruises in 2014 and 2016, the uppermost 10 cm of sediment from 167 sites in the study area was collected with a stainless grab sampler. 121 samples are from the Bohai Sea, 34 from surrounding rivers, and 12 from the north Yellow Sea (Fig. 1). There are 104 marine samples correspond to clayey silt (YT), silt (T), and sand-silt-clay (STY) (Fig. 1), according to the classification of Liu et al. (2010). Each sample was packed into a non-magnetic, 8 cm³ plastic cube and weighed after dried below 40 °C.

3.2. Magnetic parameters and measurements

Volume low- and high-frequency magnetic susceptibility (κ_{lf} and κ_{hf}) were measured at low (976 Hz) and high frequency (15, 616 Hz) under low fields (200 m/A) using a Kappabridge MFK1-FA (AGICO). Mass susceptibilities (χ_{lf} and χ_{hf}) were then obtained as κ_{lf} and κ_{hf} divided with the sample density. Frequency-dependent magnetic susceptibility (χ_{fd}) was calculated from $\chi_{fd} = [(\chi_{lf} - \chi_{hf}) / \chi_{lf}] \times 100\%$, and it serves as an indicator to the relative content of the ultra-fine magnetic particles close to the superparamagnetic (SP)/single domain (SD) boundary of ferrimagnetic minerals (Oldfield, 1991).

Each sample was applied to an alternating field (AF) with a peak field of 100 mT with a direct current (DC) bias field of 0.05 mT by a D-2000 AF demagnetizer to get an anhysteretic remanent magnetization (ARM), which is regarded as a representative of the stable single-domain ferrimagnetic content (Oldfield, 1991). The ARM was measured by a JR-6A Spinner Magnetometer (AGICO). The susceptibility of anhysteretic remanent magnetization (χ_{ARM}) was the result of the ARM divided by the DC bias (0.05 mT). Saturation isothermal remanent magnetization (SIRM) was imparted to the Z-axis for each sample at a DC field of 1 T using an IM-10-30 Impulse Magnetizer and was measured by the JR-6A Spinner Magnetometer (AGICO). The values of SIRM rely on the content of magnetic minerals primarily and magnetic crystal grain size secondarily, but irrelevant with superparamagnetic domains. Then each sample was put into back-fields (−100 mT, −300 mT) and the isothermal remanent magnetizations in back-fields ($IRM_{-100\text{ mT}}$, $IRM_{-300\text{ mT}}$) were measured. The S-ratio (S_{-300}) is often used to show the relative content of ferrimagnetic (e.g., magnetite, and maghemite) and anti-ferromagnetic mineral (e.g., hematite, and goethite) within a sample. It was calculated using the formula $S_{-300} = (SIRM - IRM_{-300\text{ mT}}) / (2 \times SIRM)$ (Bloemendal et al., 1992). High content of ferrimagnetic minerals corresponds to high S_{-300} value close to 1. “Hard” isothermal remanent magnetizations (HIRM) is roughly proportional to the absolute content of anti-ferromagnetic mineral (e.g., hematite and goethite) in a sample and is computed using the formula $HIRM = (SIRM + IRM_{-300\text{ mT}}) / 2$ (Bloemendal et al., 1993). The L-ratio was calculated as L-ratio = $(SIRM - IRM_{-300\text{ mT}}) / (SIRM - IRM_{-100\text{ mT}})$ (Liu et al., 2007). $\chi_{ARM}/SIRM$ was used to reflect magnetic grain sizes because it is not affected by superparamagnetic grains. High $\chi_{ARM}/SIRM$ value reflects high content of single-domain grains, and low $\chi_{ARM}/SIRM$ values reflect coarse magnetic grain sizes (Bloemendal et al., 1992; Robinson, 1986). All of the measured/calculated magnetic parameters are listed in Table 2.

3.3. Diffuse reflectance spectroscopy measurements and calculations

Diffuse reflectance spectroscopy (DRS) is a useful method to quantify both hematite and goethite contents (Deaton and Balsam, 1991) and has been used widely in marine sediment (Li et al., 2019; Zhang et al., 2018). DRS was measured for all samples using a Cary 5000 ultraviolet-visible-infrared spectrometer equipped with BaSO₄ as the white standard. DSR were recorded in the spectrum range from 300 to 2, 600 nm in 0.5-nm steps at a scan rate of 300 nm/min. Detailed DRS methods were introduced by Zhang et al. (2018). Results are shown in forms of the band intensities for hematite and goethite (I_{hem} and I_{goe}), and the Hem/(Hem + Goe) was calculated with the equation: Hem/

Table 2

Magnetic parameters referred in this paper and their meanings (after Liu et al., 2007; Maher, 2007; Oldfield, 1991; Torrent et al., 2007; Zhang et al., 2018).

Magnetic parameter	Abbreviate	Meaning of the parameter
Low-frequency magnetic susceptibility	χ_{lf}	Indicator of the total magnetic mineral concentration, including low-coercivity magnetic/ferrimagnetic minerals (e.g., magnetite, maghemite), high-coercivity magnetic/anti-ferromagnetic minerals (e.g., hematite, goethite), with different magnetic domains (superparamagnetic domain, single domain, “pseudo-single domain,” and multidomain).
Frequency-dependent magnetic susceptibility	χ_{fd}	Contribution of the ultra-fine grains (mainly the superparamagnetic grains) to the total magnetic mineral concentration (χ_{lf}).
Anhyseretic remanent magnetization	ARM	Proportional to the concentration of the stable single domains.
Saturation isothermal remanent magnetization	SIRM	Concentration of remanence-carrying magnetic minerals, but independent with the superparamagnetic grains.
“Hard” isothermal remanent magnetization	HIRM	Indicative of the absolute concentration of the high-coercivity magnetic minerals (e.g., hematite, goethite).
S-ratio	S_{-300}	Ratio of low-coercivity minerals (e.g. magnetite, maghemite) to concentration of remanence-carrying magnetic minerals (SIRM). High S-ratio (close to 1) indicates a high proportion of low-coercivity minerals (e.g., magnetite, maghemite), while low S-ratio (close to 0) indicates a high proportion of high-coercivity minerals (e.g., hematite, goethite).
L-ratio	L-ratio	Judge the availability of HIRM. Changes in HIRM only reflect changes in the absolute concentration of hematite and/or goethite if the L-ratio is relatively constant.
Ratio of anhyseretic remanent magnetization to saturation isothermal remanent magnetization	$\chi_{ARM}/SIRM$	Indicator of grain size of magnetic mineral crystal. High $\chi_{ARM}/SIRM$ value suggests a high content of SSD grains.
Band intensity of goethite	I_{goe}	Proportional to the concentration of goethite.
Band intensity of hematite	I_{hem}	Proportional to the concentration of hematite.
Ratio of hematite to goethite and hematite	$Hem/(Goe + Hem)$	Ratio of hematite concentration to the total concentration of goethite and hematite.

$(Hem + Goe) = -0.133 + 2.871X - 1.709X^2$, where X is the $I_{hem}/(I_{hem} + I_{goe})$ (Torrent et al., 2007).

All of the above-mentioned measurements in this study were completed at the Institute of Geology and Geophysics, Chinese Academy of Sciences (Beijing, China).

3.4. XRD and SEM-EDS analyses for extracted magnetic minerals

Morphology and composition analysis were performed for representative extracted magnetic minerals. Magnetic extraction was performed for ten dried samples, including 6 river and 4 marine samples, using a magnet housed in a glass sheath. All of the extracted samples were performed with X-ray diffraction (XRD) analysis with a X-ray diffractometer (Bruker D8 Advance). The X-ray scans were run from 5° to 90° (2θ). Results are presented after subtraction of the background trend. Seven of the extrated samples, including 6 river samples and 1 marine sample, were observed by the scanning electron microscope with energy dispersive spectrometer (SEM-EDS). Observations of magnetic extracts were made using a Quanta FEG 250 Field-emission SEM with the magnification of $1500 \times -6000 \times$ and an acceleration voltage of 20 kV. Chemical spectra of minerals were obtained using an Oxford Instruments Ltd. INCA X-MAX-50 EDS. Two particles in every sample were photographed, and were scanned with chemical spectra.

4. Results

4.1. Magnetic properties for river sediments

Region D (the Daling and Xiaoling rivers) has the highest magnetic concentration with χ_{lf} values higher than $160 \times 10^{-8} \text{ m}^3/\text{kg}$, followed by Regions E, A, and I with χ_{lf} values around $80 \times 10^{-8} \text{ m}^3/\text{kg}$ (Fig. 3a, Table S1). Regions B and C on the northeast, and Regions F and G on the southwest have the similar χ_{lf} values of $20\text{--}60 \times 10^{-8} \text{ m}^3/\text{kg}$. Magnetic mineral concentrations of the two rivers in Region H are changeable, with χ_{lf} values of 10 and $40 \times 10^{-8} \text{ m}^3/\text{kg}$. The orders of SIRM and ARM in provenance regions resemble to χ_{lf} , with some discrepancies in Regions A, E and I (Fig. 3a-c).

χ_{fd} values are higher than 4% for most river samples, except samples in Region D (Fig. 3d). Samples in Regions A, C, E, F, and G have the similar χ_{fd} values ranging from approximately 4% to 10%. Most sediments in Regions B and H contain higher χ_{fd} values than other regions.

HIRM values in Regions A and I are lower than most samples, except

the two samples with negative values in Regions D and G. In total, HIRM values of Regions B, C, D, and E in the north are higher than Regions F, G, H, I in the south (Fig. 3e).

S_{-300} values are higher than 0.90 for all samples (Fig. 3f). The high S_{-300} values occur in Region A, D, and I, and the low values occur in Regions B, F, and H. Rivers in Region G have evidently changeable S_{-300} values. L-ratio values are mainly concentrated in 0.2–0.4, despite the presence of outliers (Fig. 3g). $\chi_{ARM}/SIRM$ values for 80% samples are less than $30 \times 10^{-5} \text{ m}^3/\text{A}$. Sediments in Region B and part of Region E contain the high $\chi_{ARM}/SIRM$ values (Fig. 3h).

I_{goe} values are changeable in Regions A-C of the Liaodong Peninsula, and increase from Regions D and E on the northwest to Regions F, G, H, I on the south (Fig. 3i). I_{hem} values are lower than 12×10^{-5} for all samples except some samples from Region H (Fig. 3j). I_{hem} values for samples in Regions A-C are changeable, while the I_{hem} values for samples in Regions D-G are restricted in $4\text{--}12 \times 10^{-5}$. $Hem/(Goe + Hem)$ are low in Regions A-B and I, high in Regions C–F and H, and moderate in Region G (Fig. 3k).

4.2. Magnetic properties for the Bohai and North Yellow seas

Values of χ_{lf} and SIRM are high in the western coast of Bohai Sea, especially the northern Bohai and western Liaodong bays, and decrease southeastward (Fig. 4a, b). χ_{lf} and SIRM are correlated to a large extent (Fig. 5b). χ_{lf} and ARM are more correlated with low values than high values (Fig. 5c). ARM presents high values in the mud area of central Bohai Bay to northern Liaodong Bay, similar to the distribution of $\chi_{ARM}/SIRM$. χ_{fd} present high values not only in the mud area, but also the areas close to the estuaries in the southern Bohai Bay and eastern Liaodong Bay.

HIRM is uncorrelated to L-ratio for most samples from the Bohai and north Yellow seas except three abnormal samples from the Laizhou Bay (Fig. 5a), which implies the HIRM is useful to reflect the anti-ferromagnetic (e.g., hematite and goethite) mineral content in the sea (Liu et al., 2007). HIRM resemble to χ_{fd} in the Laizhou and Bohai bays, rather than the Liaodong Bay and the central sea. HIRM presents high values in the west Liaodong Bay, and decreases eastward, consistent with χ_{lf} and SIRM (Fig. 4).

S_{-300} is higher than 0.86 for all marine sediment samples. S_{-300} values of sediment from the northern Bohai Sea are higher than those of sediments from the southern sea (Table S1). L-ratio is the highest in northeastern Laizhou Bay, the central Bohai Bay, and the central sea.

Fig. 3. Magnetic parameters for the nine provenance regions A to I. Outliers are marked with crosses. (a) χ_{lf} , (b) SIRM, (c) ARM, (d) χ_{fd} , (e) HIRM, (f) S_{300} , (g) L-ratio, (h) χ_{lf}/SIRM , (i) I_{goe} , (j) I_{hem} , and (k) Hem/(Goe + Hem).

Fig. 4. Spatial distributions of magnetic properties, including (a) χ_{ir} , (b) SIRM, (c) ARM, (d) χ_{fd} , (e) HIRM, (f) S_{-300} , (g) L-ratio, (h) $\chi_{ARM}/SIRM$, (i) I_{goe} , (j) I_{hem} , and (k) $Hem/(Goe + Hem)$. Solid dots show values for the surrounding rivers. The two mud areas (defined by M_z more than 6ϕ) were delineated by black lines (Qiao et al., 2017).

I_{goe} is the highest in the Laizhou Bay, especially in the southern and northeastern bay. Similar to HIRM, particularly high I_{hem} values are presented in the Laizhou and Bohai bays. $Hem/(Goe + Hem)$ is similar to HIRM and I_{hem} in most regions except Liaodong Bay.

In the north Yellow Sea, values of ARM, HIRM, L-ratio, $\chi_{ARM}/SIRM$, I_{goe} , I_{hem} , and $Hem/(Goe + Hem)$ are higher in the south than the northern areas. Other magnetic parameters are slightly changed in spatial.

4.3. Major features of magnetic minerals from the XRD and SEM-EDS

The extracted samples are mainly consist of magnetite, hematite, and goethite, with the mix of quartz (Fig. 6c, f, i). Magnetic particles present octahedral structure, subangular shape with numerous dissolution pits, and spherical structure (Figs. 6a, d, g, S1). Grain sizes of all observed magnetic particles are nearly 20–70 μm (Fig. S1). Spherical

magnetic particles, which are thought to be from human activities, were found in Rivers 04, 06, 14, 24, and 27 (Fig. S1). In contrast, the octahedral and subangular magnetic minerals are the typical detrital minerals with natural origins that are respectively unground and ground during transport processes (Badesab et al., 2018; Horng and Huh, 2011). In total, magnetic particles of the Daling and Luanhe rivers (Nos. 06 and 08), west coast of the Bohai Sea are coarser than other areas. The 70 μm -size magnetites, the largest of all observed magnetic minerals, were detected in the Luanhe River and its adjacent sea area (Particle 2 of CFD 38, Fig. S1). The scanned minerals are mainly consist of Fe, O, Si, Al, and Ti, which are indicatives of the magnetite/titanomagnetite (Fig. S1). The Ti-contents are changeable in titanomagnetite (Fig. S1), so for simplicity, we prefer to use magnetite in the following discussion.

Fig. 5. Scatter plots of (a) HIRM and L-ratio, (b) χ_{lf} and SIRM, (c) χ_{lf} and ARM, (d) χ_{lf} and S_{-300} , (e) I_{goe} and S_{-300} , (f) I_{hem} and ARM, (g) SIRM and S_{-300} , and (h) χ_{lf} and I_{goe} . Possible provenances are delineated.

5. Discussion

5.1. Controlling factors on the magnetic properties in the source regions

Parent rock plays a significant role in magnetic minerals of source regions, especially for Regions C, E, and F, the most important sediment contributors. The concentration of magnetic minerals for river sediments in Region E is higher than Regions C and F, probably because some metamorphic rocks in Region E have extremely strong magnetism. For instance, the SIRM values of amphibolite in Region E are higher than $10 \text{ Am}^2/\text{kg}$ (Piper and Zhang, 1999), over a thousand times larger than the Bohai Sea sediments (Fig. 4b). The parent rocks of Region E must result in the enrichment of magnetic minerals in rivers, because the number of magnetite particles in the sediments of Luanhe River, Region E was several ten times higher than the sediments of Yellow River, Region F (Chen et al., 1980; Wang et al., 2013). Magnetic minerals in Region F contain more SP particles, and hematite and goethite than Regions C and E (Li et al., 2012; Zhang et al., 2008), which are consistent with the primary source region of the Yellow River, the loess in the river's middle reaches (Maher, 2007). In Region C, the magnetic minerals are mainly derived from the downstream Quarternary

sediments, with the addition of weathered granite and quartz sandstone from the upstream. The substance mixture results in the medium magnetic mineral concentration and S_{-300} and coarse magnetic particles (Fig. 3).

The very high contents of SP particles and goethite in the Regions B and H are probably due to the strong pedogenesis that produces the formations of SP particles and goethite (Torrent et al., 2007). The main covered soils in Regions B and H are alfisols, different to semi-alfisols and semi-hydromorphic soils in other regions (Shi et al., 2004). Higher content of goethite in Regions B and H than other regions can be formed due to the evident eluviation, as iron is released from soil Fe^{II} compounds such as Fe silicates in the parent materials, i.e. granite and sedimentary, and is finally oxidized in an aerobic environment (Cornell and Schwertmann, 2003).

The influences of human activities on magnetic minerals in source regions are evident, due to the ubiquitous spherical magnetite particles within the river samples (Fig. 6a, S1). Especially, magnetic minerals in Region D are mainly from human activities, because the magnetic properties, i.e. very high coarse magnetic mineral concentration with low contents of SP particles and hematite and goethite, seem to be irrelevant to weathering and pedogenesis. The very high content of

Fig. 6. SEM images (a, d, g), EDS (b, e, h), and XRD (c, f, i) of magnetic extracts from surface sediments of Rivers 06 (Daling River) and 08 (Luanhe River), and the Bohai Bay. In the XRD plots, Mt. = magnetite, Gt = goethite, Hm = hematite, and Qz = quartz. Note that, magnetite in the Luanhe River is coarser than other areas.

nearly pure magnetite tends to present in the anthropogenic magnetic minerals, e.g., fly ashes from factories (Jordanova et al., 2004; Veneva et al., 2004; Zhang et al., 2011). The two rivers in Region D flow through Jinzhou (Fig. 1), an industrial city with petroleum processing, ferrous metal smelting, and coking, which probably provides non-negligible anthropogenic magnetic minerals to the rivers.

The possible reductive diagenesis after deposition should be particularly concerned, because once it occurs, the primary iron oxides would be reduced into iron sulfides, including greigite and pyrite (Roberts et al., 1999). The post-depositional influence on magnetic minerals in the source regions and Bohai Sea could be ruled out, because the iron sulfides were not found in the XRD and SEM results. The greigite can be identified by sensitive magnetic changes such as the increased SIRM/ χ_{lf} value (Abrajevitch and Kodama, 2011; Ge et al., 2017; Roberts, 1995). The SIRM/ χ_{lf} value lower than 20 kA/m for all samples closes to the iron oxides (Maher, 2007; Roberts, 1995). This is consistent with previous studies, which demonstrated that terrigenous iron hydroxides/oxides (magnetite, hematite, and goethite) predominate the Bohai Sea surface sediments (Wang et al., 2015; Y. Wang et al., 2010; Wang et al., 2017).

5.2. Sources and pathways of magnetic minerals

The correlations between magnetic-concentration parameters, including χ_{lf} and SIRM, and S_{-300} (Fig. 5d, g) indicate that sediments from Region F are transported to most regions of the Laizhou Bay, southern Bohai Bay, and the central Bohai Sea, and can be carried away to the Liaodong Bay and Yellow Sea (Figs. 5, 7). Materials in the Region E with high values of χ_{lf} , SIRM and S_{-300} were supplied to the northern

Bohai Bay, and were transported northeastward (Fig. 5d, g). Materials from Region C tend to be settled along the eastern Liaodong Bay. Other local rivers, including the Fuzhou River (Region B) and rivers in Region G and H, only supply sediments to the coastal waters (Figs. 5, 7). No contribution from Regions A and I can be confirmed from the magnetic data (Fig. 5).

5.2.1. Laizhou Bay

It can be inferred from the scatter plots of magnetic parameters that most areas of the Laizhou Bay are supplied by Region F (Figs. 5d-h, 7). High contents of fine-grained magnetic particles concentrated in the mud areas are probably due to the input and transport of suspended matters from the most sediment-filled river on Earth, the Yellow River in Region F (Qin et al., 1985; Li et al., 2010). Due to the Bohai Sea Coastal Current with three directions (Fig. 2), the Yellow River sediments tend to be transported southward, eastward and northward (Li et al., 2010; Qiao et al., 2010). As a result, fine magnetic minerals are transported with the suspended matters.

Four samples in the southern Laizhou Bay seem like differ to the Region F, because they have lower magnetic mineral contents and S_{-300} , but high I_{goe} (Fig. 5d-h). In contrast, the magnetic properties of the four samples resemble to some rivers of Region G. This implies Region G also supplies the Laizhou Bay, which is consistent with previous studies based on heavy mineralogy (K. Wang et al., 2010c). The northward residual current in the Laizhou Bay probably results in the sediment transport from Region G to the bay, according to previous study of numerical simulation (Li et al., 2010) (Fig. 7).

For eight samples in northeastern bay, χ_{lf} and SIRM are negative correlated to S_{-300} (Fig. 5d, g), in contrast to Region F. Instead, the

Fig. 7. Transport pathways in the Bohai Sea. Dashed red and dotted white lines represent provenance and geographical boundaries, respectively. Arrows with varied colors stand for sediment input and transport directions, and numbers on the arrows denote the annual fluvial sediment load in 10^4 t/a (Table 1). Red solid lines delineate the regions of mixed sources. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

relation of magnetic parameters is close to Region H (Fig. 5d, g), which implies Region H is a main provenance for the region. This is consistent with previous mineralogical study (K. Wang et al., 2010c).

5.2.2. Bohai Bay

Most areas of the Bohai Bay except the the southern bay is supplied by Region E, according to the comparisons of magnetic parameters (Fig. 5d-h). The formation of coarse magnetite-enriched zone with medium to fine sand (MFS) in the southern Bohai Bay to western Liaodong Bay (Figs. 1, 3) is mainly ascribed to the input of the mountain rivers in Region E. Because the mountain rivers have strong water flow to carry coarse materials to the coastal area (Chen et al., 1980). The water current can hardly move away the sand-size magnetic minerals in the area. The enrichment of magnetic minerals in the western Bohai Bay close to Tianjin and Tangshan Cities is partly due to the human activities, as the spherical magnetite particles were found in River 14 (Haihe River) (Fig. S1). The magnetic minerals tend to be spreaded eastward due to the favourable currents (Fig. 2).

Sediments in the southern Bohai Bay are mainly contributed by the Region F, which can be derived from the magnetic proximity (Figs. 5, 7). The transport pathway is supported by the westward current along the coast in the northern bay (Fig. 2).

5.2.3. Liaodong Bay

It can be speculated from Fig. 5 that materials in the northern and partly central Liaodong Bay mainly originate from Region C. This is consistent with previous studies of mineralogy analysis (Dou et al., 2014; Wang et al., 2014). Materials in the river estuaries of Region C are

transported southward by the Liaodong Gyre and Liaodong Coastal Current (Fig. 2b).

Materials from Region F enter into the southeastern bay, and some areas of the central bay (Figs. 5, 7). Wang et al. (2015) also argued that the Yellow River in Region F is probably an important source of Liaodong Bay, due to the magnetic similarity between the Liaodong Bay and the Yellow River Delta. The viewpoint is not supported by the study of detrital minerals (Wang et al., 2014), because the detrital minerals were only analyzed for sand fraction that probably derived from the relict continental shelf. In contrast, the study of clay mineralogy support our viewpoint (Dou et al., 2014), which implies materials from Region F are likely transported to Liaodong Bay in the form of suspended matters under the favourable currents (Fig. 2; Qin et al., 1985).

χ_{lf} is negative to S_{-300} for three samples in the northwestern bay, different to the Region E-derived samples (Fig. 5d), although the three samples are close to Region E in other scatter plots (Fig. 5e, g). Hou et al. (2017) argued that the region is supplied by Liugu River and the rivers in Region D, based on mineralogical analysis for the same three samples. However, in present study, the lack of samples from Liugu River limits our understanding on the contribution from the important river. The magnetite content of Liugu River is 3 times higher than the Luanhe River in Region E, and about 2 times higher than rivers in Region D (Wang et al., 2013). Hematite contents in the Liugu River and Region D are 2–6 times higher than Luanhe River (Wang et al., 2013). The enrichment zone of magnetic minerals in western Liaodong Bay is hardly affected by Region E, as the transport capability of the coarse magnetic minerals in Region E is limited. Therefore, the Liugu River and rivers in Region D are the possible source (Dou et al., 2014; Hou

et al., 2017; Wang et al., 2014).

The southeastern bay is supplied by local rivers except the Yellow River. Magnetic properties of the Fuzhou River (River 03) in Region B and its adjacent sample are similar, and are evidently different with other sources (Fig. 5d-e, g-h; Fig. 7). This implies the possible contribution from the small river with a limited transport distance. The sample in the southeastern bay close to Laotieshan Waterway has high S_{-300} and low χ_{lf} and SIRM (red cross in Fig. 5), which is quite different with the surrounding rivers. The sample is classified into a mixture of different sources from the Laotieshan Waterway and the adjacent rivers, as the oscillating tide currents control the sedimentary processes (Liu et al., 1998).

5.2.4. Central Bohai Sea and North Yellow Sea

Based on magnetic comparisons, we infer the Central Bohai Sea is mainly supplemented from the Yellow River in Region F (Figs. 5, 7). The Yellow River-derived particles were suspended northward by the residual current and were blocked by the Yellow Sea Warm Current (Fig. 2). Region E provides materials in the northeastern region, according to the similarity of magnetic properties (Figs. 5, 7). There is a mixed region of the two provenances, with indistinguishable magnetic properties (red points in Fig. 5). The mixture of provenances is probably ascribed to the interaction effect of several currents (Fig. 2).

Magnetic properties of the North Yellow Sea sediment are close to the Yellow River System (Region F), rather than the coastal rivers (Regions A and I) (Fig. 5). This implies the north Yellow Sea is mainly supplied by the Yellow River, which is consistent with magnetic study by Wang et al., 2017 that focused on the whole Yellow Sea. The transport process is probably controlled by the southern Bohai Sea Coastal Current, which are supported by Qiao et al. (2017).

6. Conclusions

Surface sediments collected from the Bohai Sea and adjacent rivers were studied to reveal key information about sediment province of the sea. Magnetite, hematite, and goethite are the main magnetic minerals in the studied area, with variable concentrations and grain sizes. Magnetic properties in the river sediments are mainly affected by the combined factors of parent rocks, pedogenesis, and human activities. The integrative effect of the three factors results in the magnetic difference in the source regions, making it possible to track provenance in the sea with the magnetic methods. Based on scatter plots of magnetic parameters, such as χ_{lf} and S_{-300} , the sediment provinces corresponding to different sources were divided. The sea is supplied by both the huge rivers, e.g., the Yellow River, and local small rivers, e.g., Fuzhou River, which are due to the transport of currents. Magnetic properties of the north Yellow Sea is close to the Yellow River, which indicates a transport pathway that the Yellow River-derived sediments are suspended out of the Bohai Sea. This research is helpful to understand the provenance and transport process in the sea using the sensitive, rapid magnetic methods, and provides reference for the further study of provenance in seas.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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