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Human-induced changes in sediment properties and amplified endmember differences: Possible geological time markers in the future

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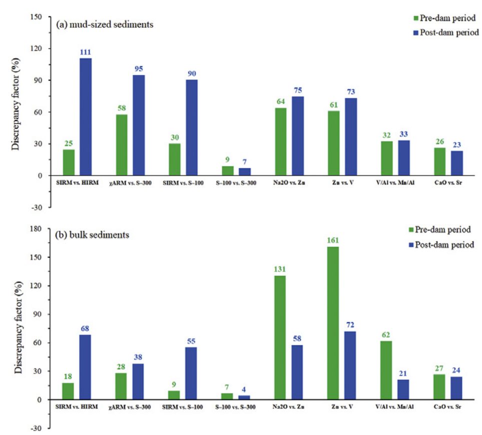
HIGHLIGHTS

- Large differences in sediment properties exist between pre- and post-dam periods.
- An approach of using multiple independent optimum tracers is proposed.
- Human activities increased the differences between the sediments from the two rivers.
- Amplified endmember differences may provide a future geological time marker.

GRAPHICAL ABSTRACT



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ABSTRACT

Many rivers are facing human-induced system regime shifts that have great environmental, ecological and social implications, necessitating an increasing need to quantify the human influence on sediment properties and their impacts on the source-to-sink system of marginal seas. The Huanghe and Changjiang Rivers have experienced a dramatic reduction in sediment flux in recent decades, typifying the human influence on sediment properties of global large rivers. Sediment samples from the two rivers were analyzed to obtain grain size, magnetic and geochemical data. The results show a large difference in sediment properties between pre- and post-dam periods. We applied a discrepancy factor to re-examine the magnetic and geochemical tracers that were previously used in the two rivers. The discrepancy factors of most magnetic and geochemical tracers in the mud-sized sediments of the two rivers increased by an average of about 109% after dam construction. This suggests that human-induced changes in sediment properties have greatly improved the discriminatory ability between the sediments from the two rivers. The results also raise the uncertainty of using previous tracers to distinguish between sediments from the two rivers after damming. Furthermore, significant changes in sediment properties that happened in a relatively short time may provide future geological time markers for sedimentary records with a temporal resolution of 10^0–10^1 years. For marine environments, an approach for identifying sediment sources based on multiple independent optimum tracers is also proposed, with composite magnetic (SIRM vs. HIRM) and geochemical (Na_2O vs. Zn) tracers being considered. The results of this work can advance our knowledge

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of how human activities alter river systems, and identify a sustainable development model under system regime shifts for areas of high-intensity human activity.

1. Introduction

Rivers provide terrestrial materials such as freshwater, sediment and nutrients to the ocean, thus play a key role in controlling the physical and biogeochemical features of global marginal seas (Walling and Fang, 2003; Liu et al., 2009; Wang et al., 2011). In recent years, human activities, e.g., dam building, soil conservation and water diversion, have become a dominant force affecting most river systems. As such, many rivers are facing human-induced system regime shifts (Syvitski et al., 2005; Wang et al., 2010a; Jia et al., 2018), the most direct effect being the reduction of sediment flux and the modifications to the nature of riverbed sediments. In rivers such as the Mississippi, Huanghe, Changjiang, Ebro and Nile rivers, sediment flux is presently below their pre-human levels (Milliman and Farnsworth, 2011; Wang et al., 2011; Yang et al., 2015). Despite many studies of anthropogenic changes in the flux of fluvial sediments, the human influence on sediment properties (e.g. grain size, magnetic and geochemical composition) is poorly understood. Knowledge of this influence is important for understanding the source-to-sink system of marginal seas in the context of human activities, and providing a scientific basis for identifying sustainable development models in coastal areas that response to human-induced system regime shifts.

The Huanghe (Yellow River) and Changjiang (Yangtze River) are among the world's largest river systems in terms of water discharge and sediment yield. They supply sediment to a heavily populated coastal region that is undergoing change. In recent years, these two rivers have experienced dramatic reduction in sediment flux, especially in the period of 1999 to 2003 (Yang et al., 2006; Wang et al., 2007; Gao et al., 2015a). Thus they provide excellent cases of how human activities alter riverine sediment. Sediment flux in these two rivers has decreased from previous estimates of 1080 $\times$ 10^6 t/yr and 478 $\times$ 10^6 t/yr (e.g. Milliman and Meade, 1983; Milliman and Syvitski, 1992) to the present ~150 $\times$ 10^6 t/yr and 147 $\times$ 10^6 t/yr (Yang et al., 2006, 2015; Wang et al., 2007, 2011), respectively. Several studies have attempted to quantify the relative importance of climatic change and human activities on the sediment flux changes (e.g. Dai et al., 2014; Zhao et al., 2015, 2017; Yang et al., 2018a). For the Huanghe River, human activities are responsible for 70% of recent decrease in sediment flux (40% by soil conservation and 30% by reservoir retention), and 30% by decreased precipitation (Wang et al., 2007). Decrease in precipitation is responsible for 14% of the decrease in sediment flux of the Changjiang River, while the remainder i.e. 86% is ascribed to human activities in the river basin, of which the Three Gorges Dam (TGD) contributed 65%, and other dams and soil conservation accounted for 21% (Yang et al., 2015). The construction of the Xiaolangdi Reservoir in 1999 in the middle Huanghe and the TGD in 2003 in the upper Changjiang appear to be responsible for the dominant human influence (Wang et al., 2011, 2017a; Yang et al., 2011, 2014). The drastic reduction in sediment flux and unprecedented human activities have led to a human-induced system regime shift in the two river systems (Wang et al., 2010a; Jia et al., 2018), and corresponding changes in sediment properties (Yang et al., 2007, 2018b; Yu et al., 2013; Gao et al., 2017). In terms of the Huanghe and Changjiang Rivers, the decline in sediment flux is mainly ascribed to human activities, so that human activity is the main cause of changes in sediment properties. In addition, significant changes in sediment properties can leave different markers in coastal-shelf sedimentary records, which may provide a future geological time marker for sedimentary records affected by the Huanghe and/or Changjiang Rivers.

In recent years, investigations have been carried out into the variations in grain size and sediment composition of sediments delivered to the sea by the two rivers (e.g. Wang et al., 2010a; Gao et al., 2015b; Luo et al., 2017; Yang et al., 2018b), and significant sediment coarsening was found in response to dam construction. However, to date there have been few studies that have attempted to quantify the human-induced changes in magnetic and geochemical composition of the Huanghe and Changjiang sediments and their impacts on the discrimination of the sediments from the two rivers. Towards this end, we (1) collect and collate the basic grain size, magnetic, and geochemical data that quantify the human influence on the sediment properties of the two rivers from the pre-dam period to the post-dam period; (2) determine the most appropriate tracers for discriminating the sediments from the two rivers in the post-dam period; and (3) re-examine the previously widely used tracers that differentiate the sediments from the two rivers.

2. Materials and methods

For this study, 34 surface sediment samples were collected using a grab sampler between September and October 2012 from the Huanghe and Changjiang Rivers (Fig. 1; Table S1). Of these, sixteen were collected from the lower reach of the Huanghe River, and eighteen from that of the Changjiang River. All the samples were measured for their grain size composition using a laser Malvern Mastersizer 2000 granulometer. To minimize the grain size effect, the sediment samples were separated into two grain size fractions; i.e., the sand fraction (>63 μm) and the mud fraction (<63 μm) by wet-sieving the sediments through a 63 μm nylon mesh. These two sized fractions were used for the magnetic and geochemical analyses.

The magnetic susceptibility (χ) of the sediments was measured using a Bartington Instruments MS2 magnetic susceptibility meter at a low frequency (χ, 0.47 kHz). An anhysteretic remanent magnetization (ARM) was imparted in a 0.04 mT direct current (DC) bias field that was superimposed onto a peak alternating field (AF) of 100 mT using a DTECH 2000 AF demagnetizer and measured using a Molspin magnetometer. ARM is expressed as the anhysteretic susceptibility (χ_{ARM}) by normalizing ARM against the applied DC field. The isothermal remanent magnetization (IRM) was measured using a forward field of 1 T followed by application of back fields of −100 mT and −300 mT that were imparted using an MMPM10 pulse magnetizer. The IRM obtained at 1 T is referred to as the saturation IRM (SIRM) and the back field remanences are referred to as the IRM_{-100mT} and IRM_{-300mT} (Zhang et al., 2012; Ge et al., 2015). The hard IRM (HIRM) was calculated as follows:

$$HIRM = 0.5 \times (SIRM + IRM_{-300\ mT})$$

and the S ratios (S_{-100} and S_{-300}) were calculated as follows:

$$S_{-100} = 100 \times (SIRM - IRM_{-100\ mT})/(2 \times SIRM)$$

and

$$S_{-300} = 100 \times (SIRM - IRM_{-300\ mT})/(2 \times SIRM)$$

(Ge et al., 2017).

Further, major and trace elements analysis were carried out on the mud and sand fractions using an ARL9800XP + X-ray fluorescence spectrometer (XRF). Work curves were established on the basis of the national geological standard samples (GSD9), which indicated that our

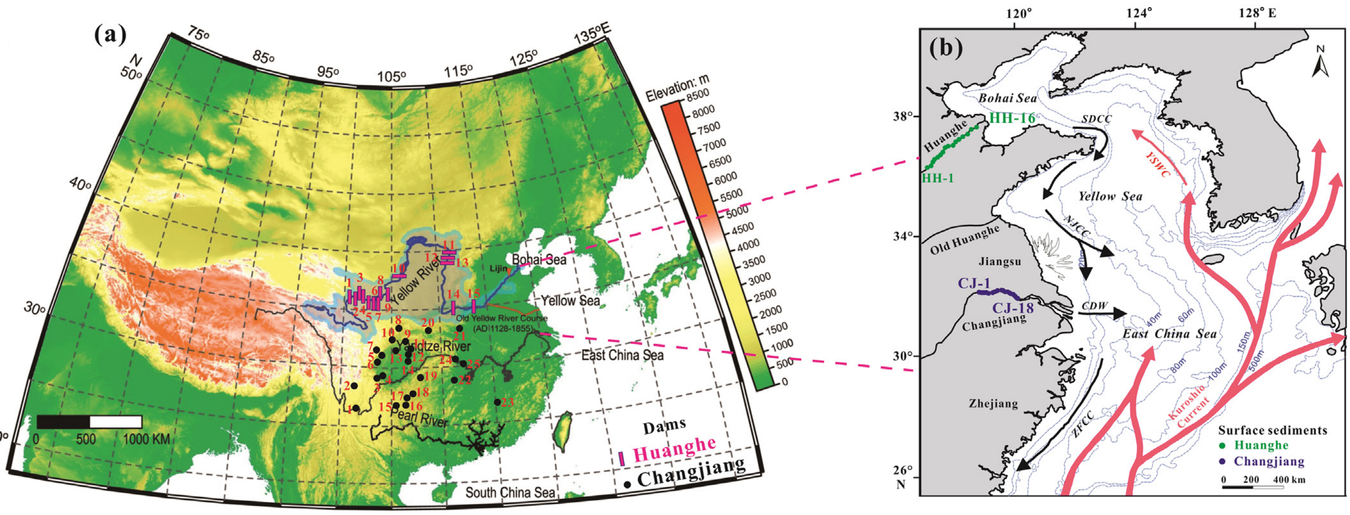

Fig. 1. Map showing the location of the Huanghe and Changjiang Rivers in China (a, modified from Wang et al., 2017a), and sampling locations of surface sediments (b) in these two rivers (HH: Huanghe River and CJ: Changjiang River). Large dams constructed in the two rivers are shown in Fig. 1a and Table S2. The modern-day current systems within the study area are also indicated (modified from Lim et al., 2015): SDCC = Shandong Coastal Current, NJCC = Northern Jiangsu Coastal Current, CDW = Changjiang Diluted Water, ZFCC = Zhejiang Fujian Coastal Current, and YSWC = Huanghe Sea Warm Current.

geochemical analysis of the major and trace elements had a precision better than 10% (Rao et al., 2015). Major and trace element concentrations are expressed as oxide concentrations (%) and μg/g, respectively.

Statistical analysis, such as non-parametric test, discriminant function analysis and principal component analysis (PCA), has previously been successfully used to identify the tracers that were most effective at separating out the potential sources (Collins and Walling, 2002; Walling, 2005; Zhang and Liu, 2016). In the present study, a two-stage statistical selection procedure was used to determine the most appropriate tracers for the two rivers. Firstly, non-parametric test methods (i.e., the Mann-Whitney U and Kolmogorov-Smirnov methods for two independent sets of samples) were used to assess which parameters exhibited a significant difference between the two rivers. Secondly, PCA (Jolliffe, 2002; Yang et al., 2016, 2017) was used to quantify the relative importance of the list of parameters identified in the first stage, and thus the most appropriate tracers could be selected. The relative importance of each parameter (RI) was quantified using the factor loadings and the variance of each PC derived from the PCA, and were expressed as follows (Yang et al., 2016):

$$RI_j = \sum_{i=1}^{n} \left(\lambda_j^2/p\right) * a_i \qquad (1)$$

where i is the component number, n is the total number of components, λ_j is the factor loading of parameter j, p is the eigenvalue, λ_j^2/p is the factor weight, and a_i is the variance of PC$_i$.

3. Results and discussion

3.1. Grain size

The sediments of the Huanghe and Changjiang Rivers show large variations in grain size composition, with the silt and sand fractions being dominant. The sediments consist of 2.5% clay, 51.7% silt, and 45.8% sand in the Huanghe River; and 5.5% clay, 51.8% silt, and 42.7% sand in the Changjiang River. The Huanghe sediments are rich in sand (mean grain size, Mz = 4.3 φ), whereas the Changjiang sediments are finer, with Mz of 4.8 φ (this study, 2012). In comparison, the Mz values of the two rivers in 2013 were reported to be 3.6 φ and 4.5 φ, and analyzed using the same instrument as our study (Rao et al., 2015). Before dams, e.g. 1996–2000, however, the Mz values of the Huanghe and Changjiang sediments were much finer than those of after dams with mean values of 4.8 φ and 6.3 φ (grain size was analyzed by Sedigraph 5100 granulometer, Yang et al., 2004a); e.g. 1997–2000, the Mz values were analyzed by wet sieving and pipette methods with mean values of 5.1 φ and 6.3 φ (Yang et al., 2004b). To reduce the uncertainty caused by spatial variations of samples (most sediment samples were collected from the lower reaches of the two rivers) and laboratory treatments, Mz values of this study and Rao et al. (2015) were averaged as the values of the post-dam period, and those reported by Yang et al. (2004a) and Yang et al. (2004b) as the pre-dam period, respectively. Therefore, the Mz values of the Huanghe and Changjiang sediments can be estimated to be 4.0 φ and 4.7 φ for the post-dam period, and 5.0 φ and 6.3 φ for the pre-dam period (Table 1).

After dams, the Huanghe and Changjiang riverbed sediments exhibited a significant coarsening trend, and the Mz values have been coarsened by about 20% and 25% between approximately 1996 and 2013 (Fig. 3). This coarsening has also been identified by previous studies, and suggests that channel erosion in the lower reaches has become a new sediment source, coarsening the riverbed sediments in the lower reaches of the two rivers (e.g. Wang et al., 2007; Gao et al., 2015b; Yang et al., 2011, 2018a). Because severe downstream erosion has been identified to continue in recent years (Wang et al., 2011; Dai and Liu, 2013), we expect that the coarsening of riverbed sediments in the downstream of the two rivers has most probably continued and has intensified.

3.2. Magnetic properties

For most of the magnetic parameters, the values recorded for the mud- and sand-sized fractions show great differences in these two rivers (Table 1), implying the grain-size effect on magnetic properties. The parameters χ and SIRM reflect primarily the concentration of magnetic minerals in sediments, especially ferrimagnetic minerals (e.g., magnetite; Thompson and Oldfield, 1986; Ge et al., 2015). The Changjiang River sediments have higher mean χ and SIRM values, reflecting their high concentrations of magnetic minerals. S_{-300} serves as a measure of the relative importance of high-coercivity minerals and low-coercivity components (e.g. magnetite and maghemite) in the total magnetic mineral assemblage, whereas S_{-100} reflects the ratio of low-coercivity minerals to medium- and high-coercivity minerals (Robinson et al., 2000). There is a positive correlation between χ and SIRM values in both rivers, together with high S_{-300} values (>90% on average), which indicates the dominance of ferrimagnetic minerals

Table 1
Summary of mean grain size (Mz) and magnetic properties in sediments from the Huanghe and Changjiang Rivers. The number after ± is standard deviation, and RD represents relative deviation.

	Mz (φ)	Samples	χ (10^{-8} m^3/kg)	χ_{ARM} (10^{-8} m^3/kg)	SIRM (10^{-6} Am^2/kg)	HIRM (10^{-6} Am^2/kg)	χ_{ARM}/χ	χ_{ARM}/SIRM (10^{-5} m/A)	S_{-100} (%)	S_{-300} (%)	References	Sampling time
The Huanghe River	4.0	Mud-sized	62.2 ± 32.1	143.6 ± 43.8	8179.4 ± 3930.8	438.2 ± 190.6	2.6 ± 0.8	19.7 ± 6.4	78.0 ± 0.9	94.5 ± 0.7	This study	2012
		Sand-sized	15.0 ± 3.5	42.9 ± 5.9	2128.2 ± 466.1	195.6 ± 33.5	2.9 ± 0.5	20.7 ± 3.4	72.8 ± 1.1	90.7 ± 1.0	This study	2012
		Bulk	43.0 ± 13.0	164.0 ± 80.0	5574.0 ± 1445.0	372.0 ± 101.0	3.8	30.4 ± 13.6	76.4 ± 2.3	93.3 ± 2.3	Zhang et al., 2008	2006
The Changjiang River	4.7	Mud-sized	160.9 ± 29.2	412.5 ± 80.4	22,014.2 ± 3699.5	665.3 ± 73.1	2.6 ± 0.5	19.0 ± 3.3	87.1 ± 1.5	96.9 ± 0.5	This study	2012
		Sand-sized	65.9 ± 15.5	145.0 ± 32.2	8941.9 ± 1362.7	336.8 ± 122.3	2.3 ± 0.8	16.4 ± 3.5	87.4 ± 2.4	96.3 ± 1.3	This study	2012
		Bulk	73.0 ± 20.0	286.0 ± 130.0	10,900.0 ± 2505.0	525.0 ± 111.0	3.9	25.5 ± 8.3	81.8 ± 2.1	95.0 ± 0.7	Zhang et al., 2008	2006
The Huanghe River	5.0	Mud-sized	81.2	254.4	11,318.7	705.2	3.6	21.0	75.6	93.8	Niu et al., 2008	2003
		Sand-sized	20.9	51.9	3264.0	294.4	2.6	15.9	70.1	91.0	Niu et al., 2008	2003
		Bulk	82.2	158.2	11,044.1	883.5	2.4	18.4	74.4	92.3	Wang et al., 2004	2001
The Changjiang River	6.3	Mud-sized	122.6	541.9	16,390.5	675.3	4.4	33.1	87.3	95.9	Niu et al., 2008	2003
		Sand-sized	120.5	337.6	16,050.8	515.2	2.8	21.0	90.0	96.8	Niu et al., 2008	2003
		Bulk	96.7	242.8	11,913.9	639.4	2.6	19.8	82.4	94.8	Wang et al., 2004	2001
The Huanghe River (RD, %)	20.0	Mud-sized	23.4	43.6	27.7	37.9	27.8	6.2	3.2	0.7		Between 2003 and 2012
		Sand-sized	28.2	17.3	34.8	33.6	11.5	30.2	3.9	0.3		Between 2003 and 2012
		Bulk	47.7	3.7	49.5	57.9	59.3	65.2	2.7	1.1		Between 2001 and 2006
The Changjiang River (RD, %)	25.4	Mud-sized	31.2	23.9	34.3	1.5	40.9	42.6	0.2	1.0		Between 2003 and 2012
		Sand-sized	45.3	57.0	44.3	34.6	17.9	21.9	2.9	0.5		Between 2003 and 2012
		Bulk	24.5	17.8	8.5	17.9	48.4	28.5	0.7	0.3		Between 2001 and 2006

in the magnetic mineral assemblage (Zhang et al., 2008; Wang et al., 2010b; Table 1; Fig. 2a). In Fig. 2b, the HIRM and L-ratio [HIRM/(0.5 × (SIRM + IRM_{-100mT}))] are not correlated in the sediments from the Huanghe (two-sized fractions) and Changjiang (only mud-sized fraction), and the relatively stable L-ratios (ca. 0.25 and 0.34 for the mud- and sand-sized fractions) suggest that the variations in HIRM are dominated by changes in the concentration of anti-ferrimagnetic minerals (e.g. hematite, goethite) (Maher, 1988; Liu et al., 2007). In contrast, the relatively strong correlation (R^2 = 0.4166) between the HIRM and the L-ratio in the sand-sized sediments from the Changjiang River indicates that variations in HIRM are caused mainly by changes in the coercivity distribution rather than by the concentration of hematite and/or goethite (Liu et al., 2007). The parameter χ_{ARM} is particularly sensitive to stable single-domain (SSD) ferrimagnetic grains, and higher χ_{ARM} values indicate a great concentration of SSD grains (Maher, 1988; Wang et al., 2010b). χ_{ARM} values are higher for sediments from the Changjiang River, indicate that less SSD grains exist in sediments from the Huanghe River. In addition, the mean χ_{ARM}/SIRM values of the two rivers are almost <30 × 10^{-5} m A^{-1}, and χ_{ARM}/χ values are <5, which reveals the pseudo-single-domain (PSD) and multi-domain (MD) patterns of magnetic grain size (Oldfield and Yu, 1994; Wang et al., 2009).

Most of the magnetic parameters (except S_{-100} and S_{-300}) of the mud-sized (sand-sized) sediments from the Huanghe and Changjiang Rivers showed significant changes with a mean relative deviation (RD) of 27.8% (25.9%) and 29.1% (36.8%) between roughly 2003 and 2012 (Table 1). Further, changes in bulk sediments of the Huanghe River (mean RD of 47.2%) were more drastic than those of the Changjiang River (mean RD of 24.3%) between approximately 2001 and 2006 (Table 1). It is worth mentioning that all the magnetic parameters in Table 1 were measured using the same magnetic system of East China Normal University, China (this study; Wang et al., 2004; Niu et al., 2008; Zhang et al., 2008). In addition, most of the magnetic parameters exhibit relatively small spatial variations, and the mean coefficients of variation of the Huanghe and Changjiang sediments are 23.2% and 18.7%, respectively (Table 1). We believe that the uncertainty caused by instrumental analysis errors and spatial variations has been minimized when comparing the magnetic parameters before and after dams.

After the construction of numerous dams, the concentration of magnetic minerals in the bulk, mud- and sand-sized sediments of the two rivers exhibited a notable decreasing trend, with the exception of the mud-sized sediments of the Changjiang River. The magnetic concentrations (e.g. χ and SIRM values) of the sand-sized sediments from the Huanghe and Changjiang Rivers have been decreased by about 32% and 45% between 2003 and 2012 (Table 1). For the bulk sediments, the concentrations of the two rivers have been decreased by approximately 49% and 17% between 2001 and 2006. The averaged decreased values (~35% and 31% for the Huanghe and Changjiang Rivers; Fig. 3) obtained from the bulk, sand- and/or mud-sized sediments may produce a reasonable estimate of the human influence on the magnetic mineral concentrations.

This decreasing shift has been confirmed by the coarsening of riverbed sediments, as the fine fractions dominate the χ and SIRM values (Zhang et al., 2008; Wang et al., 2009). Although relatively large RDs were found in the χ_{ARM}/χ and χ_{ARM}/SIRM values of the two rivers, most of their values were still <5 and 30 × 10^{-5} m A^{-1}, respectively. Combining the S_{-100} and S_{-300} values, we conclude that human activities have profoundly changed the concentration of magnetic minerals in the sediments of the Huanghe and Changjiang Rivers, but did not systematically changed the magnetic grain sizes and magnetic mineral types. Due to the continued coarsening of riverbed sediments and unprecedented human activities in the coming decades (Wang et al., 2011; Yang et al., 2018a), we expect that the magnetic mineral concentrations of Huanghe and Changjiang sediments will continue to decrease further, and the magnetic grain sizes (more coarse magnetic

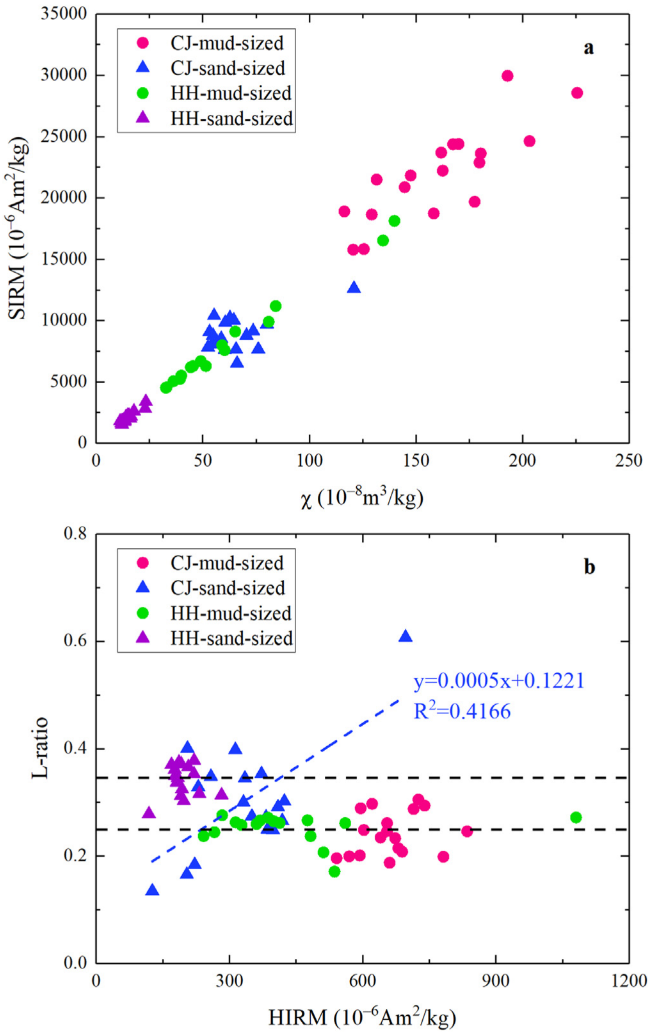

Fig. 2. Correlations between χ and SIRM (a) and HIRM and the L-ratio (b) for mud- and sand-sized sediments from the Huanghe and Changjiang Rivers in the post-dam period. Black dotted lines represent a relatively constant L-ratio, and the blue dotted line indicates the relationship for sand-sized sediments from the Changjiang River. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

particles) and magnetic mineral types (less high-coercivity minerals) have most probably changed.

3.3. Geochemical characteristics

The concentrations of the major and trace elements in the sediments from the Huanghe and Changjiang Rivers are given in Tables 2 and 3, respectively. For these two rivers, the concentrations of most elements are higher in the mud-sized sediments than in the sand-sized sediments, which suggest the presence of a grain-size effect on geochemical properties. Most elements that are concentrated in the sediments of the Changjiang River. Although the major and trace elements in Tables 2 and 3 were determined by three different instruments (e.g. XRF, ICP-AES and ICP-MS), their relative error was <10% (this study; Yang et al., 2002a, 2003a, 2004a; Rao et al., 2015). In addition, most of the elements showed small spatial variability with mean coefficients of variation of 9.5% and 12.5% for the Huanghe and Changjiang Rivers, respectively (Tables 2 and 3). Therefore, the concentrations of major and trace elements between pre-dam and post-dam periods can be reasonably compared.

After dam construction, the concentration of most elements exhibited notable changes, and the mean RDs of the mud-sized (bulk) sediments from the Huanghe and Changjiang Rivers are about 40.2% (35.2%) and 39.4% (33.8%) between roughly 1997 and 2013 (Tables 2 and 3). Changes in geochemical elements were more drastic than changes in magnetic parameters between pre-dam and post-dam periods. The concentration of the major elements showed a significant increasing trend, with the exception of Na_2O and MnO, CaO and TiO_2 in the mud-sized and bulk sediments. The concentrations of most major elements in the Huanghe and Changjiang sediments (bulk and mud-sized) have been increased by about 67% and 61% between approximately 1997 and 2013 (Fig. 3). For the mud-sized sediments in both rivers, the concentrations of Nb, Sr, Y, Hf, Cr and V exhibited an increasing trend, but Th, Ga, Cu, Co and Ni showed a decreasing trend from 1997 to 2012 (Fig. 3). In addition, changes in trace elements in bulk sediments showed great difference between the Huanghe and Changjiang Rivers. The concentrations of Nb, Rb, Th, Zn, Cu, Cr and V and only Sr showed an increasing trend in the bulk sediments of the Huanghe River and Changjiang River, respectively (Fig. 3). Compared with the Huanghe, trace elements are more concentrated in the fine-grained sediments of the Changjiang River (Yang and Li, 1999; Yang et al., 2002a), the coarsening of riverbed sediments leads to a decrease in the concentration of trace elements in the bulk sediments. We expect that the concentrations of major elements in the Huanghe and Changjiang sediments

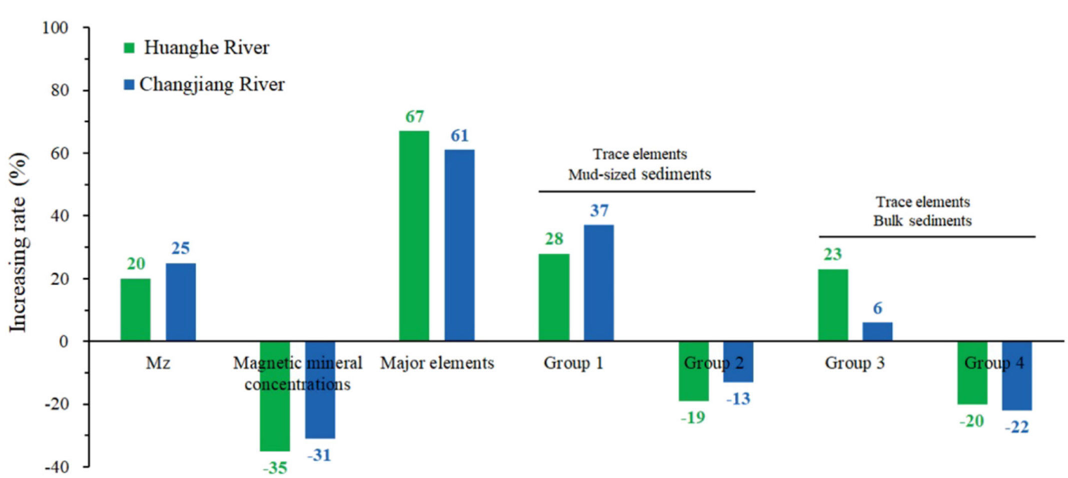

Fig. 3. The increasing rate (%) in mean grain size (Mz), magnetic mineral concentrations (χ and SIRM values), and major and trace elements of the Huanghe and Changjiang sediments from the pre-dam period to the post-dam period. Mud-sized sediments: Group 1 represents Nb, Sr, Y, Hf, Cr and V, and Group 2 represents Th, Ga, Cu, Co and Ni. Bulk sediments: Group 3 represents Nb, Rb, Th, Zn, Cu, Cr and V for the Huanghe River, and only Sr for the Changjiang River; Group 4 represents Pb, Sr, Y, Hf, Co and Ni for the Huanghe River, and all the trace elements with the exception of Sr and Ga for the Changjiang River.

Table 2
Summary of major element concentrations in sediments from the Huanghe and Changjiang Rivers (unit: %). The number after ± is standard deviation, and RD represents relative deviation.

	Samples	SiO_2	Al_2O_3	Fe_2O_3	MnO	CaO	MgO	K_2O	Na_2O	TiO_2	References	Sampling time
The Huanghe River	Mud-sized	61.8 ± 1.4	13.1 ± 0.4	3.6 ± 0.3	0.06 ± 0.01	7.5 ± 0.1	2.2 ± 0.1	2.3 ± 0.1	1.6 ± 0.1	0.8 ± 0.2	This study	2012
	Sand-sized	68.0 ± 1.0	12.1 ± 0.3	2.5 ± 0.1	0.04 ± 0.00	5.5 ± 0.2	1.6 ± 0.1	2.7 ± 0.1	1.8 ± 0.0	0.4 ± 0.1	This study	2012
	Bulk	75.9 ± 1.5	10.7 ± 0.4	3.1 ± 0.3	0.03 ± 0.00	1.4 ± 0.1	1.3 ± 0.2	2.3 ± 0.1	2.2 ± 0.0	0.6 ± 0.1	Rao et al., 2015	2013
The Changjiang River	Mud-sized	59.5 ± 1.0	14.4 ± 0.7	4.9 ± 0.3	0.10 ± 0.01	5.9 ± 0.6	2.5 ± 0.2	2.3 ± 0.1	1.0 ± 0.1	1.1 ± 0.1	This study	2012
	Sand-sized	65.9 ± 1.3	11.8 ± 0.2	3.5 ± 0.2	0.06 ± 0.01	5.5 ± 0.8	2.2 ± 0.2	2.3 ± 0.1	1.5 ± 0.1	0.6 ± 0.0	This study	2012
	Bulk	68.9 ± 3.2	12.6 ± 1.2	4.7 ± 0.7	0.04 ± 0.01	1.8 ± 0.1	2.0 ± 0.2	2.4 ± 0.2	1.7 ± 0.2	0.8 ± 0.1	Rao et al., 2015	2013
The Huanghe River	Mud-sized	–	9.9 ± 0.4	2.4 ± 0.4	0.06 ± 0.00	2.8 ± 0.0	1.1 ± 0.0	1.6 ± 0.0	1.7 ± 0.0	0.6 ± 0.0	Yang and Li, 1999; Yang et al., 2002a	1997–1998
	Bulk	32.3	5.2 ± 0.2	2.2 ± 0.2	0.05 ± 0.00	3.5 ± 0.3	1.0 ± 0.1	1.8 ± 0.1	1.6 ± 0.2	0.5 ± 0.0	Yang et al., 2004a	1997–2000
The Changjiang River	Mud-sized	–	11.6 ± 1.4	3.7 ± 0.4	0.08 ± 0.02	2.1 ± 0.0	1.7 ± 0.0	1.8 ± 0.0	1.1 ± 0.0	1.1 ± 0.3	Yang and Li, 1999; Yang et al., 2002a	1997–1998
	Bulk	28.8	7.2 ± 0.5	4.3 ± 0.5	0.12 ± 0.02	3.3 ± 0.3	1.7 ± 0.1	2.1 ± 0.2	0.9 ± 0.2	1.0 ± 0.1	Yang et al., 2004a	1997–2000
The Huanghe River (RD, %)	Mud-sized	–	151.9	50.0	0.0	167.9	100.0	43.8	5.9	33.3	Between 1997 and 2012	
	Bulk	135.0	105.8	40.9	40.0	−60.0	30.0	27.8	37.5	20.0	Between 1997 and 2013	
The Changjiang River (RD, %)	Mud-sized	–	140.0	32.4	25.0	181.0	47.1	27.8	9.1	0.0	Between 1997 and 2012	
	Bulk	139.2	75.0	9.3	66.7	45.5	17.6	14.3	88.9	20.0	Between 1997 and 2013	

Table 3
Summary of trace element concentrations in sediments from the Huanghe and Changjiang Rivers (unit: μg/g).

	Samples	Nb	Pb	Rb	Sr	Th	Y	Ga	Hf	Zn	Cu	Co	Ni	Cr	V	References	Sampling time
The Huanghe River	Mud-sized	16.4 ± 3.6	20.2 ± 2.5	77.0 ± 7.2	209.5 ± 7.7	12.7 ± 3.1	32.0 ± 9.4	12.0 ± 1.2	12.0 ± 7.3	60.0 ± 6.2	17.7 ± 1.5	9.6 ± 0.8	24.4 ± 2.6	67.6 ± 16.8	127.3 ± 13.5	This study	2012
	Sand-sized	11.7 ± 0.7	14.0 ± 2.5	87.9 ± 7.1	219.5 ± 5.8	3.3 ± 1.4	32.2 ± 2.5	11.1 ± 1.0	110.1 ± 18.9	45.2 ± 5.3	13.6 ± 0.6	5.7 ± 0.4	20.7 ± 1.6	39.9 ± 2.0	90.5 ± 3.2	This study	2012
	Bulk	11.4 ± 1.0	14.9 ± 0.3	78.5 ± 3.6	195.0 ± 8.9	9.2 ± 1.1	21.0 ± 1.8	–	6.5 ± 1.5	49.2 ± 7.0	20.7 ± 2.1	7.5 ± 0.7	18.1 ± 1.9	55.0 ± 9.7	59.3 ± 4.8	Rao et al., 2015	2013
The Changjiang River	Mud-sized	20.8 ± 2.0	34.6 ± 3.2	97.1 ± 8.8	157.7 ± 7.7	15.3 ± 3.0	37.2 ± 7.5	14.9 ± 0.9	13.4 ± 4.8	127.0 ± 8.7	40.9 ± 2.7	14.7 ± 1.1	34.7 ± 2.1	90.9 ± 16.0	171.0 ± 11.2	This study	2012
	Sand-sized	15.9 ± 2.7	14.1 ± 3.8	79.1 ± 5.6	187.1 ± 17.1	4.6 ± 1.6	40.8 ± 8.0	11.6 ± 0.8	133.2 ± 48.4	76.2 ± 9.2	21.9 ± 4.6	10.7 ± 0.9	27.9 ± 2.9	53.9 ± 0.3.5	127.5 ± 6.5	This study	2012
	Bulk	15.3 ± 1.8	25.3 ± 2.3	96.8 ± 8.6	155.7 ± 8.2	12.0 ± 1.6	25.1 ± 2.4	–	4.9 ± 0.6	94.7 ± 28.5	29.3 ± 14.5	12.1 ± 2.1	29.2 ± 5.4	65.4 ± 8.5	90.0 ± 13.5	Rao et al., 2015	2013
The Huanghe River	Mud-sized	15.6 ± 1.8	29.5 ± 2.3	–	186.6 ± 12.9	18.3	20.8 ± 2.7	20.3 ± 2.7	6.9 ± 1.1	60.3 ± 4.6	17.8 ± 2.0	11.0 ± 1.6	26.7 ± 2.1	64.8 ± 10.8	108.9 ± 14.8	Yang and Li, 1999; Yang et al., 2002a	1997–1998
	Bulk	10.1 ± 1.3	17.7 ± 1.3	76.9 ± 5.4	218.7 ± 23.5	8.7 ± 1.3	28.0	11.0	12.0	33.9 ± 4.9	14.2 ± 3.1	8.7 ± 1.0	19.3 ± 3.0	42.3 ± 6.0	50.8 ± 2.6	Yang et al., 2003a	1996–2000
The Changjiang River	Mud-sized	19.3 ± 4.5	20.5 ± 10.4	–	135.5 ± 11.8	16.9	25.0 ± 3.3	19.9 ± 3.3	6.4 ± 1.7	116.2 ± 22.0	47.6 ± 14.7	15.1 ± 2.3	40.9 ± 6.2	78.0 ± 15.4	140.5 ± 28.1	Yang and Li, 1999; Yang et al., 2002a	1997–1998
	Bulk	17.5 ± 1.7	38.5 ± 9.6	114.9 ± 7.6	146.7 ± 3.3	13.4 ± 1.1	28.0	16.0	8.1	106.5 ± 20.4	50.7 ± 13.1	17.3 ± 2.1	40.7 ± 3.9	74.3 ± 6.9	105.5 ± 10.3	Yang et al., 2003a	1996–2000
The Huanghe River (RD, %)	Mud-sized	5.4	31.6	–	12.3	30.8	53.7	40.9	73.8	0.5	0.4	12.5	8.5	4.3	16.9	Between 1997 and 2012	
	Bulk	13.2	15.8	2.1	10.8	6.3	25.0	–	45.8	45.0	45.6	13.8	6.4	29.9	16.7	Between 1996 and 2013	
The Changjiang River (RD, %)	Mud-sized	7.8	68.7	–	16.4	9.7	48.7	25.0	108.6	9.3	14.0	2.5	15.1	16.5	21.7	Between 1997 and 2012	
	Bulk	12.6	34.3	15.8	6.2	10.2	10.4	–	39.5	11.0	42.2	29.9	28.3	11.9	14.7	Between 1996 and 2013	

will increase further, and the concentrations of most trace elements may decrease in the future.

3.4. Distinguishing between the sediments from the Huanghe and Changjiang Rivers

The sediments from the Huanghe and Changjiang Rivers have markedly different magnetic and geochemical characteristics, which can provide a basis for distinguishing between the two rivers. In stage one of the two-stage procedure, the Mann-Whitney U and Kolmogorov-Smirnov tests suggested that most of the magnetic parameters, major and trace elements display significant differences between the two rivers (Tables S3 and S4), except χ_{ARM}/χ, χ_{ARM}/SIRM, K_2O, TiO_2, Nb, Th, Y, Hf and Cr in the mud-sized sediments, and Al_2O_3, CaO, Pb, Rb, Th, Y, Ga and Hf in the sand-sized sediments, which yield P-values >0.05. In stage two, the data are presented only for PC1–PC2 and PC1–PC3, which together explain 95.362% and 96.691% of the variability of the magnetic parameters in the mud- and sand-sized sediments, respectively. The PCA results suggest that SIRM vs. HIRM (mud-sized sediments) and χ_{ARM} vs. S_{-300} (sand-sized sediments) have the highest RI values (Table S5), and were selected as the most appropriate magnetic tracers for the mud and sand fractions, respectively, of the two rivers (Fig. 4a–b). In addition, the values of PC1–PC3 explain most of the variance (93.284% and 91.393% for the mud and sand fractions, respectively) in the geochemical data (Table S6). The RI values suggest that the Na_2O vs. Zn and Zn vs. V diagrams can effectively distinguish the mud- and sand-sized sediments of the two rivers (Fig. 4c–d).

Magnetic parameters can be used as potentially powerful tracers for discriminating sediment provenance in marine environments, especially in relation to rivers (Gyllencreutz and Kissel, 2006; Liu et al., 2010; Wang et al., 2017b). Previous studies showed that a SIRM vs. S_{-100} (or S_{-100} vs. S_{-300}) diagram can be used to discriminate the Huanghe and Changjiang sediments (Zhang et al., 2008; Wang et al., 2009). Here, we re-examine these magnetic tracers by incorporating our newly established tracers (Fig. 5). The results indicate that the SIRM vs. S_{-100} and S_{-100} vs. S_{-300} diagrams can also distinguish the bulk and mud-sized sediments for the two rivers. In addition, the SIRM vs. S_{-100} diagram can also be used to distinguish between aeolian dust and sediments from the Huanghe and Changjiang Rivers (Li et al., 2012). To access the performance of these tracers in both pre- and post-dam periods, we applied a discrepancy factor ($DF = |(CJ/HH-1)| \times 100$) to these tracers of the bulk and mud-sized sediments from the two rivers. The results indicate that diagrams of S_{-100} vs. S_{-300} have the lowest DF values. The DF values of SIRM vs. HIRM, χ_{ARM} vs. S_{-300} and SIRM vs. S_{-100} in bulk and mud-sized sediments exhibited a significant increasing trend from the pre-dam period to the post-dam period, but S_{-100} vs. S_{-300} showed a slight decreasing trend (Fig. 6). The result highlights the effectiveness of SIRM vs. HIRM, χ_{ARM} vs. S_{-300} and SIRM vs. S_{-100} for identifying the Huanghe and Changjiang sediments.

In recent decades, the geochemical composition of the Huanghe and Changjiang sediments has been extensively studied (e.g., Zhao and Yan, 1992; Yang et al., 2002b, 2003b; Jiang et al., 2009; Rao et al., 2015), and some geochemical plots have proved to be effective at separating the two sources, such as V/Al vs. Mn/Al (Yang et al., 2003b) and CaO vs. Sr (Yang et al., 2004b). In addition, the V/Al vs. Mn/Al diagram can be used to distinguish between sediments from Korea and China rivers (Yang et al., 2003b). In this study, we re-examined these geochemical tracers (Fig. 7), and makes a full comparison of the performance of these tracers between pre-dam and post-dam periods (Fig. 6). The results indicate that plots of these geochemical tracers can clearly distinguish the bulk and mud-sized sediments from the Huanghe and Changjiang Rivers, except V/Al vs. Mn/Al in bulk sediments after damming (Fig. 7f). In Fig. 6, the DF values of these geochemical tracers

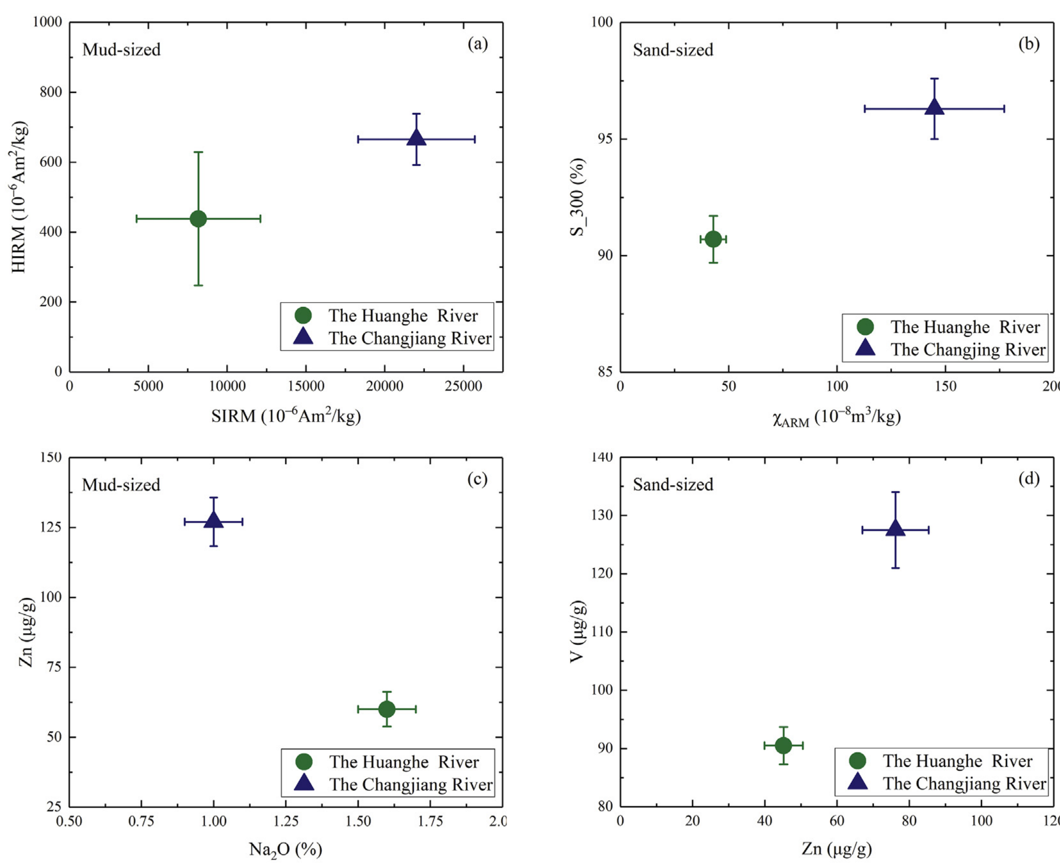

Fig. 4. (a) SIRM vs. HRIM and (b) χ_{ARM} vs. S_{-300} discrimination plots for the mud- and sand-sized sediments from the Huanghe and Changjiang Rivers. Also shown are the discrimination diagrams of geochemical tracers for the mud-sized (c: Na_2O vs. Zn) and sand-sized (d: Zn vs. V) sediments from the two rivers.

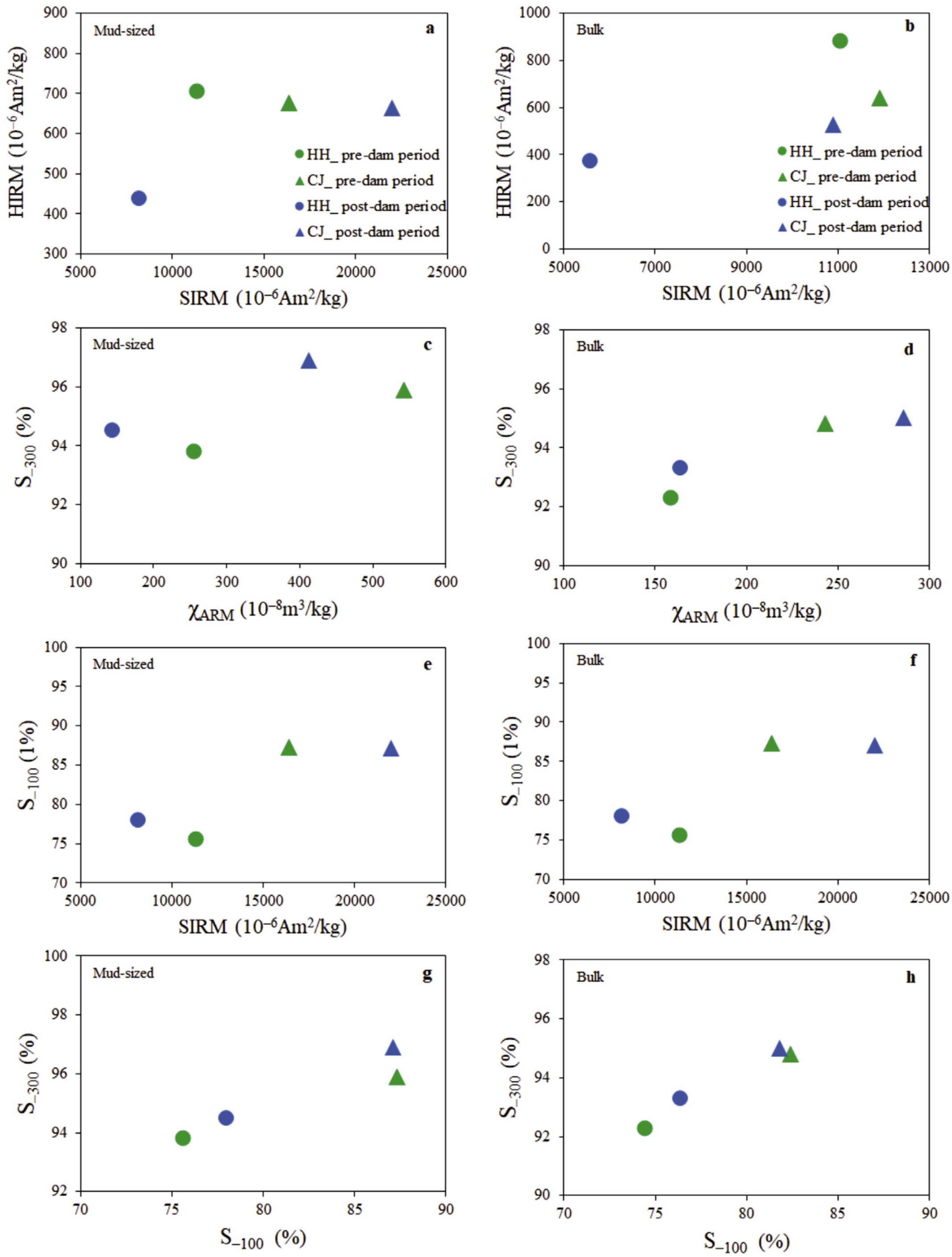

Fig. 5. SIRM vs. HRIM (a and b) and χ_{ARM} vs. S_{-300} (c and d) discrimination plots for the mud-sized and bulk sediments from the Huanghe and Changjiang Rivers in both pre-dam and post-dam periods. Also shown are the SIRM vs. S_{-100} (e and f) and S_{-100} vs. S_{-300} (g and h) diagrams proposed by Zhang et al. (2008) and Wang et al. (2009), respectively.

showed opposite patterns in bulk and mud-sized sediments from the pre-dam period to the post-dam period. In addition, the DF values of our newly established geochemical tracers were >50% in both pre- and post-dam periods, indicating a robust ability of Na_2O vs. Zn and Zn vs. V for discriminating the sediments from the two rivers.

When comparing the DF values between pre-dam and post-dam periods, we found that most of the magnetic and geochemical tracers in the mud-sized sediments have an increase in DF value of about 109% on average after dams (Fig. 6a). This finding suggests that human activities make it easier to distinguish the mud-sized sediments from the Huanghe and Changjiang Rivers (Figs. 5 and 7). The results also demonstrated that tracers from the mud-sized sediments are more favorable for identifying sediment sources than those from bulk sediments. For the bulk sediments, the DF values of the magnetic tracers after dams have been increased by approximately 271%, but the values of geochemical tracers have been decreased by about 46% (Fig. 6b). The DF values indicated that the magnetic tracers have better performance in distinguishing the bulk and mud-sized sediments of the Huanghe and Changjiang Rivers, and demonstrates the robust ability of magnetic methods for tracing sediment sources after dams. The results also raise the uncertainty of using previous tracers to distinguish between sediments from the two rivers after dams, such as diagrams

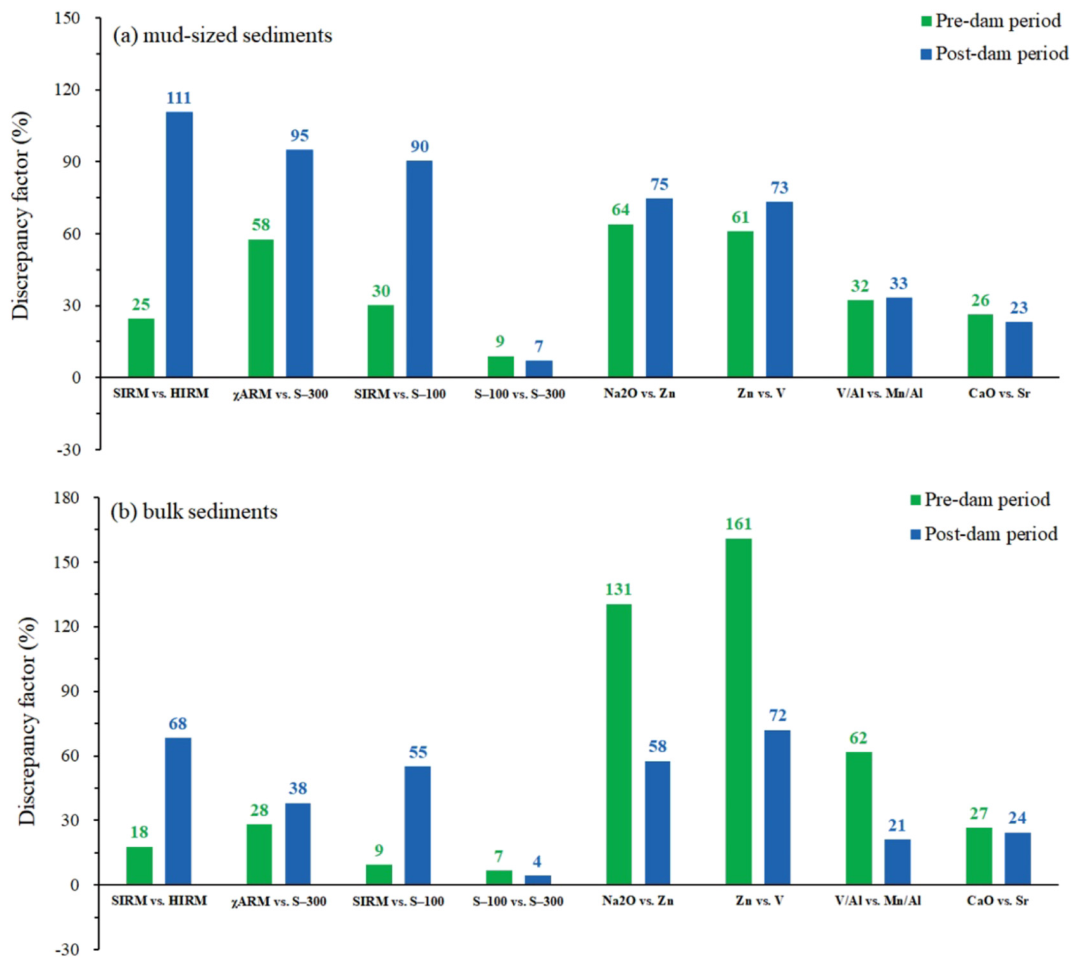

Fig. 6. The discrepancy factor (%) of magnetic and geochemical tracers in the mud-sized (a) and bulk (b) sediments of the Huanghe and Changjiang Rivers between pre-dam and post-dam periods.

of V/Al vs. Mn/Al, S_{-100} vs. S_{-300} and CaO vs. Sr. The impacts of human activities will increase further, and we expect that human activities will make it easier to distinguish between the sediments from the Huanghe and Changjiang Rivers in the future.

A single optimum tracer (e.g. magnetic or geochemical tracer) selected by statistical procedure is widely accepted and used to identify sediment provenance in published literatures (e.g. Yang et al., 2002a; Wang et al., 2009; Li et al., 2014; Hu et al., 2018). However, for an open system like an estuarine-coast system, a single optimum tracer may not be sufficient because sorting effects of both particle size and material composition will occur significantly during sediment transport from the river mouth to the coast-shelf (Ashworth and Ferguson, 1989; Gao, 2003). Consequently, the use of only a single optimum tracer may result in greater uncertainty in the identification of sediment sources. To reduce this uncertainty, the approach of using multiple independent optimum tracers is strongly recommended for estuarine-coast systems, such as composite magnetic (SIRM vs. HIRM) and geochemical (Na_2O vs. Zn) tracers.

3.5. Future geological time markers

Rivers connect continents with oceans in influencing global biogeochemical cycles, and are key pathways for the delivery of terrestrial materials to the oceans (Seitzinger et al., 2005; Syvitski and Milliman, 2007; Milliman and Farnsworth, 2011). Historically rivers have discharged ~20 Gt of terrestrial sediment per year into the global oceans (Milliman and Farnsworth, 2011). However, over the past century, river systems have become increasingly impacted by human activities (e.g. dams and irrigation projects), which collectively have led to dramatic decreases in sediment discharge from rivers on a global scale, e.g., the Huanghe (Wang et al., 2007), Red (Le et al., 2007), Mekong (Fu et al., 2008), Mississippi (Meade and Moody, 2010) and Changjiang (Yang et al., 2015) rivers. In extreme examples, such as the Nile and Colorado rivers, present-day sediment fluxes have decreased to almost nothing (Vörömarty et al., 2003).

Although recent sediment decline is generally attributed to dam construction (e.g. Changjiang and Red rivers), other factors such as climate change, water diversion, and soil conservation can also be important. In the Huanghe and Changjiang Rivers, for example, 30% and 14% of the recent decrease in sediment flux can be ascribed to decreased precipitation throughout the river's watershed (Wang et al., 2007; Yang et al., 2015). In the Nile and Colorado rivers, the dramatic reduction in sediment flux can be explained by water diversion (Wiegel, 1996; Rodriguez et al., 2001). Also, about 50% of the sediment decrease in the Mississippi River can be attributed to land conservation and levee construction (Meade and Moody, 2010). In the Mekong River, river flow is only slightly regulated by dams and reservoirs (Nilsson et al., 2005). When the proposed reservoirs in the lower Mekong River will be completed, the sediment delivery from the Mekong River to the ocean will be greatly reduced (Kummu et al., 2010; Wang et al., 2011). The increasing impact of both climatic change and human activities on global river systems brings severe environmental challenges in the coastal ocean, including the sinking of deltas and declines in coastal wetland areas (Yang et al., 2006; Wang et al., 2011).

Rivers are facing human-induced system regime shifts worldwide due to the decreasing sediment supply (Syvitski et al., 2005; Wang et al., 2010a; Jia et al., 2018), the most direct effect being the

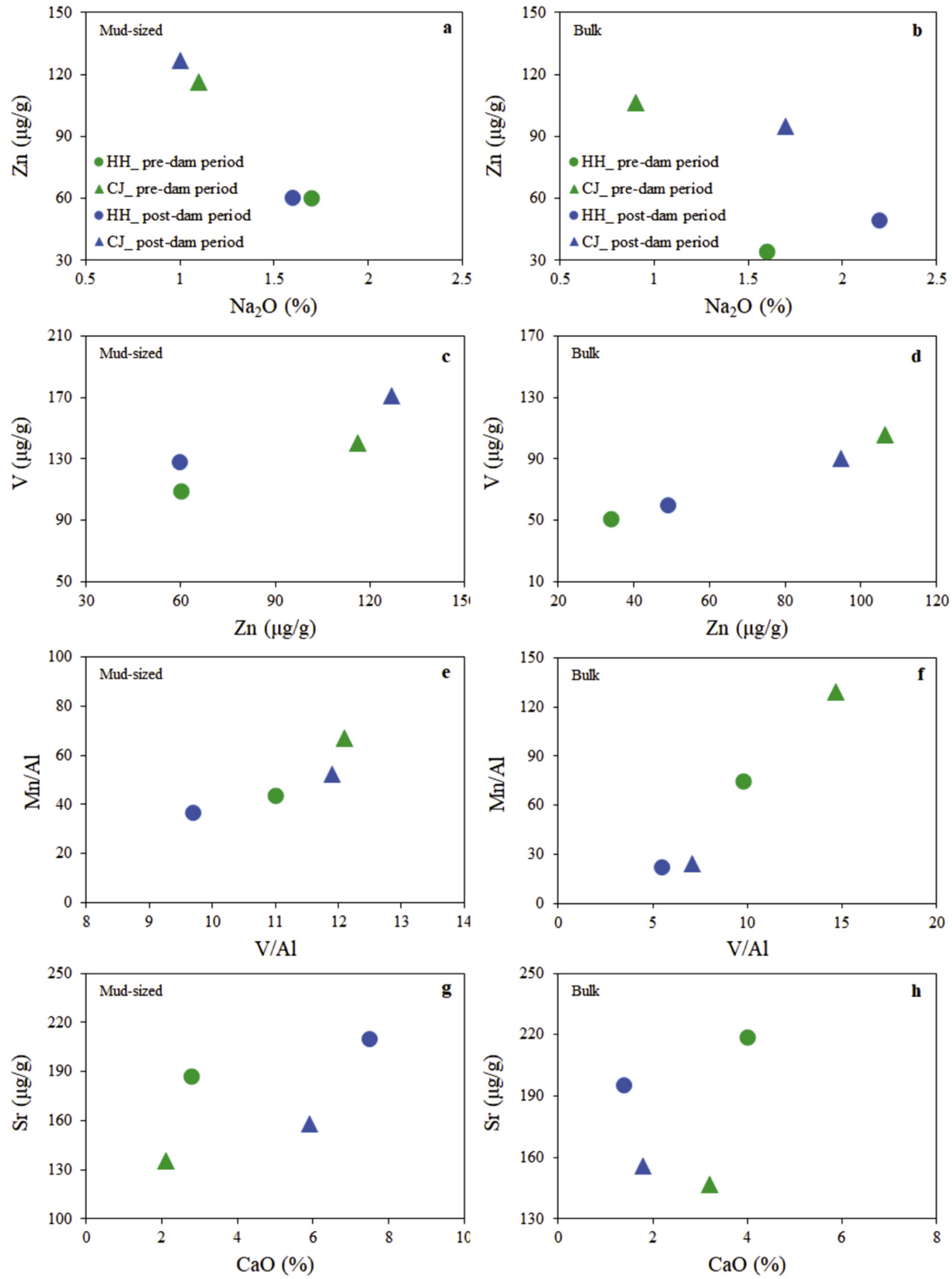

Fig. 7. Na_2O vs. Zn (a and b) and Zn vs. V (c and d) discrimination diagrams for the mud-sized and bulk sediments from the Huanghe and Changjiang Rivers in both pre-dam and post-dam periods. Also shown are V/Al vs. Mn/Al (e and f), and CaO vs. Sr (g and h) diagrams proposed by Yang et al. (2003b) and Yang et al. (2004b), respectively.

modifications to the sediment properties. The latest tens of years saw unprecedented human activities resulting in dramatic changes in sediment properties of the Huanghe and Changjiang Rivers from the pre-dam period to the post-dam period. In addition, the DF values of most magnetic and geochemical tracers in the mud-sized sediments of the two rivers increased by an average of about 109% after dam construction. As a result, these changes can leave different markers in coastal-shelf sedimentary records. The transition point between these significantly increased or decreased values and the under-layer may provide a future geological time marker between 1996 and 2013 for sedimentary records affected by the Huanghe and/or Changjiang Rivers, with a temporal resolution of 10^1 years. As such, the distance between the present-day bed surface and the transition point can be measured, representing the thickness of newly accumulated materials. If the time marker of 1996 or 2013 is known, then the deposition rate can be deduced. This method is similar to estimating deposition rates using a morphological proxy of *Spartina alterniflora* plants, which provide a time marker of the colonization time of *Spartina alterniflora*, e.g. 1993 or 1999 in the Weihai tidal flat, Jiangsu coast in eastern China (Li and Gao, 2013). Future work should test this approach in estuarine-coast systems affected by the Huanghe and/or Changjiang Rivers. The 10^1-year time resolution is sufficient for the millennial-scale sedimentary records, but it is thought to be insufficient for the centennial-scale sedimentary records. One way to solve this problem is to carry out yearly monitoring of the sediments deposited from both of the rivers to analyze changes in sediment composition on an annual basis. Hence, the resolution of the time marker can reach the order of 10^0 years. In addition, we conclude that changes in sediment properties of the Huanghe and Changjiang Rivers are mainly caused by anthropogenic factors, but other factors such as climate change may be important. Such an annual investigation is indeed necessary as it is unclear if some of the changes in the sediment properties result from the human activities or climate change. Analyzing such a change adds a benefit to the proposed use of the changes in sediment properties as a future geological time marker.

Recent global environmental changes suggest that Earth system may have entered a new human-dominated geological epoch, the Anthropocene (Lewis and Maslin, 2015). Actually, the definition and the beginning of Anthropocene have long been contentious and probably remains to be resolved (e.g. Maslin and Lewis, 2015; Williams et al., 2015; Steffen et al., 2016). From a global change perspective, the impact of human activities on the world's large river systems appears to be significant, and thereafter may provide an option to define the Anthropocene. In other words, evidences of beginning of the Anthropocene may be resorted to sedimentary records of significant changes in riverine sediment properties.

4. Conclusions

Analysis of the magnetic properties and geochemical composition of sediments, as well as the grain size, provides the basis for quantifying the human impacts on the sediment properties in the Huanghe and Changjiang Rivers. Specifically, our conclusions are as follows:

(1) The full data set shows a large difference in sediment properties between pre- and post-dam periods. The mean grain size coarsened by about 20% and 25%, the magnetic mineral concentrations decreased by approximately 35% and 31% and the concentrations of most major elements increased by about 67% and 61% in the riverbed sediments from the two rivers, respectively, after damming.

(2) Magnetic (SIRM vs. HIRM and χ_{ARM} vs. S_{-300}) and geochemical (Na_2O vs. Zn and Zn vs. V) tracers were developed to discriminate the mud- and sand-sized sediments from the two rivers. An approach of using multiple independent optimum tracers is most appropriate for marine environments, such as composite magnetic (SIRM vs. HIRM) and geochemical (Na_2O vs. Zn) tracers.

(3) The discrepancy factor values of most magnetic and geochemical tracers in the mud-sized sediments of the two rivers increased by an average of about 109% after dam construction. This finding suggests that the modifications of human activities to the nature of riverbed sediments have greatly improved the discriminatory ability between the sediments from the two rivers, and will become more dramatic in the future.

(4) Significant changes in sediment properties can leave different markers in sedimentary records, which may provide a future geological time marker for sedimentary records affected by the Huanghe and/or Changjiang Rivers with a time resolution of 10^0–10^1 years.

CRediT authorship contribution statement

Yang Yang: Formal analysis, Writing - original draft, Writing - review & editing. **Jianjun Jia:** Conceptualization, Writing - review & editing. **Liang Zhou:** Writing - review & editing. **Wenhua Gao:** Data curation, Writing - review & editing. **Benwei Shi:** Writing - review & editing. **Zhanhai Li:** Writing - review & editing. **Ya Ping Wang:** Writing - review & editing. **Shu Gao:** Conceptualization, Funding acquisition, Writing - review & editing.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.01.115.

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