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Compositional characteristics of sediment from Jiaozhou Bay in North China and the implication to the provenance*

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Abstract Rare earth elements (REEs) can be used to trace source materials and identify their provenances, because of significant conservation and immobility during chemical alteration processes after erosion of materials from the provenance. This study focused on the temporal variation of REEs for columnar sediments from the mouth of Jiaozhou Bay in North China to understand the potential controls for the geochemical variations of sediments. Through extraction experiments, we identified that the residual fraction is the main host for REEs compared with other fractions (i.e., exchangeable and carbonate fraction, easily reducible oxides fraction, reducible oxides fraction, magnetite fraction). REE ratios (e.g., La_N/Sm_N and La_N/Yb_N; N: normalized by chondrite) lack correlations with grain size or the chemical index of alteration (CIA), which is correlated with major elements. All these indicate that these REE variations reflect the varying contribution of source materials from different provenances instead of grain size or chemical weathering effects. REE ratios (e.g., La_N/Sm_N and La_N/Yb_N) remain relatively constant until the depth of roughly 40 cm (equivalent to the year 1995), and show obvious changes beyond this depth. Compared REE characteristics of Jiaozhou Bay with those of neighboring rivers and bedrocks, the relative contributions of Dagu River-Jiaolai River, and Licun River may have been increased during the sedimentary processes, which could be caused by the construction of reservoir and related change of aquaculture (e.g., rapid accumulation of organic materials).

Keyword: Jiaozhou Bay; sediments; rare earth elements (REEs); sequential extraction; provenance

1 INTRODUCTION

As the product of various natural chemical and physical processes, fine-grained sediments record important geological and environmental information (Guo et al., 2017), such as source materials from the provenance (Bhatia, 1985; Cullers, 1994; Hoskin and Ireland, 2000; Gürsu et al., 2017), tectonic settings (Bhatia, 1985; Etemad-Saeed et al., 2015; Khan and Khan, 2015), and sedimentary processes (Taylor and McLennan, 1981; Goodfellow, 1983; Blair, 1987). However, complex physical and chemical processes after being eroded from the provenance can alter the geochemical compositions

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of source materials (Rashid and Ganai, 2015; Ma et al., 2017). For example, chemical weathering can cause the replacement of feldspars in source rocks by secondary clay minerals, leading to the selective leaching of Ca^{2+} and Na^{+} (Roy et al., 2006; Roy and Smykatz-Kloss, 2007). As a result, these alterations can also influence the geochemical variation of fine-grained sediments. Hence, it is important to identify different geological processes and environmental effects by selecting appropriate geochemical indicators.

Studies on trace elements, which is resistant to secondary processes (e.g., diagenesis), are important to better constrain the provenance (Taylor and McLennan, 1985; Cullers, 1994; Mabrouk et al., 2015). Rare earth elements (REEs) with similar ionic sizes and the same charge (except Eu and Ce) share similar geochemical behaviors, and can be greatly conserved during most chemical alteration processes (Taylor and McLennan, 1985; Coldstein and Jacobsen, 1988; Elderfield et al., 1990). In addition, geochemical behaviors of various elements in nature mainly depend on their hosted fractions, which is important to further reveal different sedimentary and biogeochemical processes (Chester and Green, 1968; Claff et al., 2010; Kiczka et al., 2011; Zhu et al., 2012). Sequential extraction experiments show that REEs are mainly hosted in the residual fraction, which are mainly composed of silicate minerals and represent the component of source materials from the provenance (Zhu et al., 2012; Burdige and Komada, 2020; Jung et al., 2021). Thus, REEs are the ideal candidate for tracing source materials and identifying their provenances (Khan et al., 2017; Noa Tang et al., 2020).

Jiaozhou Bay is regarded as a typical gulf area of the Chinese marginal sea. Many previous studies have focused on Jiaozhou Bay in different aspects for studying the environmental change of embayment in response to different anthropogenic and natural factors (Peng et al., 2019; Wang et al., 2021a), including the sedimentary dynamics (Zhang et al., 2019; Yuan et al., 2021), the nutrient structure (Yuan et al., 2018; Li et al., 2020a), the phytoplankton community compositions (Liu et al., 2020b; Wang et al., 2021b), the heavy metal pollution (Xu et al., 2017; Kang et al., 2018; Liang et al., 2018), and the ecosystem (Peng et al., 2019; Cao et al., 2020; Liu et al., 2021; Li et al., 2022). The sediments from the mouth of Jiaozhou Bay are located at the intersection of the inner and outer bays, receiving source materials from the inner and outer bays, which can better represent the sedimentary information of the Jiaozhou Bay. This study focuses on the temporal variations of REEs for columnar sediments from the mouth of Jiaozhou Bay. Together with other geochemical indicators, we aim to identify the controls on REE characteristics of sediments from Jiaozhou Bay and reveal their provenance.

2 MATERIAL

Jiaozhou Bay is located on the southern shore of the Shandong Peninsula, an inlet of the Yellow Sea in eastern China (Fig.1). It is a typical semi-closed

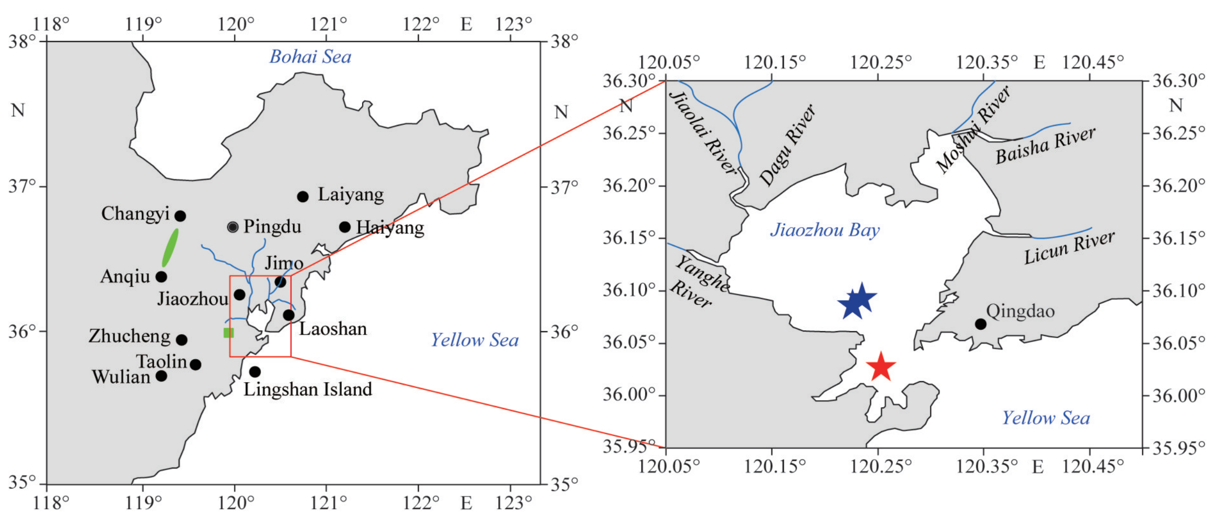

Fig.1 The area (red rectangle) and sampling site (red star) of this study, and the sampling sites L1 and L2 of Wang et al. (2003) (blue stars)

Modified after Yang et al., 2003a; Li et al., 2014; Liu et al., 2019a.

bay with abundant inputs of wastewater and sediments from more than ten rivers, including the Baisha River, Yanghe River, Dagu River, and Licun River (Wang et al., 2017). It is an excellent area for studying the coastal sea with strong influences from both terrestrial and human activities (Dai et al., 2007). A continuous surface sediment column lifted by a gravity tube through R/V *Science* with an approximately 60 cm long was sampled in 2019 from Jiaozhou Bay, China (120°22′48″E; 36°02′01″N; water depth: 15 m; Fig.1), which is very close to the sampling location of a ca. 79-cm long sediment column named D4 obtained in 2003 (120° 15′55″E; 36° 01′14″N; Li et al., 2011). By using ^{210}Pb dating, Li et al. (2011) acquired the depositional rates before 1992 and after 1992 were 3.96 cm/a and 1.63 cm/a, respectively. In this study, we also used these depositional rates, and calculated the depositional age of our sediment column, which can be dated back to ca. 30 years ago. To identify the compositional variation, the sediment column was separated into 1–2-cm long slices, and 38 sample slices were obtained in total. All the samples were stored in polyethylene for further testing. Dried sediments were carefully hand-crushed to fine-grained powders using an agate mortar and dried at 105 °C in oven before analysis.

3 ANALYTICAL METHOD

3.1 Grain size analysis

Granulometric analysis was done by using the Cilas 1190L instrument after the ultrasonic dispersion. The measurement range was 0.04–2 500 μm, and the grain sizes were <4 μm for clay, 4–63 μm for silt, and >63 μm for sand (Wentworth, 1922; Liu et al., 2019b).

3.2 Sequential extraction

The modified sequential selective extraction procedure was operated to separate five operationally defined fractions (Poulton and Canfield, 2005; Henkel et al., 2016): exchangeable and carbonate fraction (F1), easily reducible oxides fraction (F2), reducible oxides fraction (F3), magnetite fraction (F4), and residual fraction (F5).

Briefly, about 200 mg of homogenized sediment samples were weighed and loaded into a 15-mL centrifuge tube, and F1 fraction was extracted with 10 mL of 1-mol/L Na-acetate (adjusted to pH=4.5 with acetic acid), shaken for 24 h at 22 °C. F2 fraction was extracted with 10 mL of 1-mol/L hydroxylamine-HCl in 25% v/v acetic acid for 48 h. F3 fraction was extracted with freshly-prepared citrate-buffered sodium dithionite (50-g/L Na-dithionite+0.02-mol/L sodium citrate buffer solution at pH=4.8) for 2 h. Extraction of F4 fraction was completed by leaching with 0.2-mol/L ammonium oxalate/0.17-mol/L oxalic acid for 6 h. Finally, the residual fraction contained primarily silicate minerals, the residue from the fourth step was dissolved with mixed acid (HCl-HNO_3-HF). After each extraction step, solution was centrifuged and the supernatants filtered through 0.2-μm polyether sulfone filters into 15-mL metal-free tubes.

3.3 Element analysis

The geochemical analysis for this study was performed at the Laboratory of Oceanic Lithosphere and Mantle Dynamics, Institute of Oceanology, Chinese Academy of Sciences. Major elements and trace elements were analyzed using Agilent-5100 inductively coupled plasma optical emission spectrometer (ICP-OES) and Agilent-7900 quadrupole inductively coupled plasma mass spectrometer (ICP-MS), respectively. For major element analysis, the alkali fusion method was adopted. Then, 40–50-mg dried powder sample and 250 mg of metaboric acid were mixed in a platinum crucible and heated in a muffle furnace at 1 050 °C for 0.5 h for melting. After quickly being transferred to 5% nitric acid, the sample solution was thoroughly mixed using ultrasonic. Finally, the uniform sample solution was diluted to be roughly 2 000 times the sample weight (Kong et al., 2019). The international standards used were GSP-2, AGV-2, BCR-2, W-2a, and BHVO-2, indicating both the accuracy and precision of major element analysis better than 5%. For trace element analysis, anti-aqua regia was mixed with 50-mg dried sample powder in high-pressure bombs (Teflon cups jacketed by stainless vessels) for dissolution (Chen et al., 2017). The monitoring standards used were GSP-2, AGV-2, BCR-2, W-2a, and BHVO-2. The precision of trace element analysis is better than 5%, and the accuracy is better than 10%. The test results of major elements and trace elements are given in Supplementary Table S1.

4 RESULT

4.1 Grain size

The sediment types in Jiaozhou Bay were mainly sandy silt and silt. The silt content was relatively high, ranging from 65% to 74%, with an average content of 68%; while the clay content was 20%–29%, with an average of 23%; the content of sand was the lowest, between 2%–14%, with an average of only 8% (Fig.2; Supplementary Table S2).

4.2 Major elements and REEs

The analyzed sediments in Jiaozhou Bay were characterized by high SiO_2 (61.86%–70.88%), followed by intermediate Al_2O_3 (12.10%–15.27%), Fe_2O_3 (3.93%–5.73%), and other elements were less than 3% (Supplementary Table S1). Total REE (ΣREE) concentrations ranged from 160×10^{-6} to 209×10^{-6} with an average value of 180×10^{-6} (Table 1; Supplementary Table S1).

4.3 REEs in different fractions

The results of sequential extraction of sediment samples from Jiaozhou Bay show that the residual fraction is the dominant hosted fraction of REEs, and the proportion in this fraction exceeds 50% (Fig.3; Supplementary Table S3), reflecting the relatively simple sedimentary environment in this area, mainly from the supply of terrigenous, and the contribution of other sources is relatively low (Zhu et al., 2012; Burdige and Komada, 2020; Jung et al., 2021).

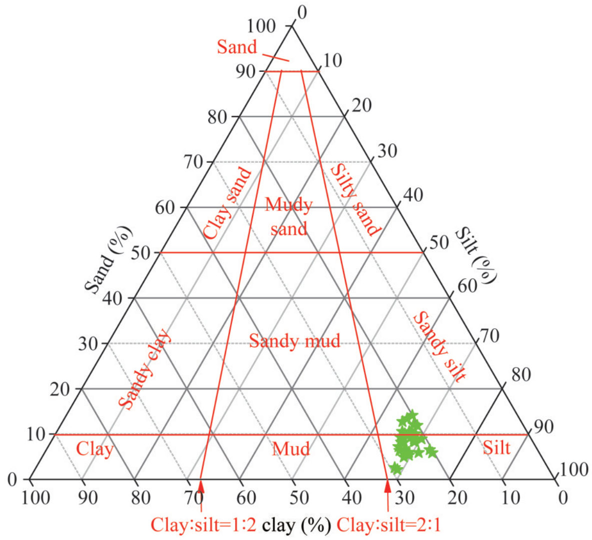

Fig.2 The ternary diagram of grain size classification for Jiaozhou Bay sediments (green stars)

Table 1 The comparison of average REE contents in the sediments from different areas ($\times10^{-6}$)

Element	JZB	JZB upper	JZB lower	CJR	YR	BHS	YS	ECS	HR	SDL	UCC
La	38.58	38.87	38.02	53.40	33.50	31.60	33.51	33.53	76.90	31.43	30.00
Ce	78.48	78.81	77.83	98.80	54.10	62.72	66.89	64.67	141.00	65.76	64.00
Pr	8.28	8.28	8.29	11.00	6.47	7.56	7.99	7.82	16.50	6.50	7.10
Nd	30.96	30.83	31.21	34.40	21.20	28.24	29.77	29.19	44.20	26.59	26.00
Sm	5.82	5.78	5.90	6.22	3.96	5.12	5.68	5.43	7.50	4.97	4.50
Eu	1.19	1.18	1.20	1.47	0.85	1.07	1.14	1.11	1.55	1.08	0.88
Gd	4.67	4.63	4.77	5.68	3.68	4.43	4.86	4.88	6.15	4.31	3.80
Tb	0.75	0.71	0.77	0.84	0.55	0.68	0.74	0.73	0.90	0.67	0.64
Dy	4.22	4.16	4.33	4.64	3.04	3.82	4.04	4.09	4.83	3.94	3.50
Ho	0.87	0.86	0.89	0.94	0.61	0.77	0.86	0.79	0.92	0.75	0.80
Er	2.50	2.46	2.59	2.67	1.67	2.18	2.29	2.34	2.53	2.69	2.30
Tm	0.38	0.38	0.40	0.39	0.25	0.34	0.35	0.34	0.36	0.34	0.33
Yb	2.54	2.50	2.61	2.38	1.50	2.22	2.22	2.19	1.99	2.22	2.20
Lu	0.38	0.37	0.39	0.37	0.26	0.35	0.35	0.34	0.34	0.36	0.32
$\sum REE_{average}$	179.61	179.83	179.20	223.20	131.64	151.10	160.69	157.45	305.67	151.61	146.37
Ce/Ce*	1.08	1.08	1.08	1.00	0.90	0.99	1.00	0.98	0.97	1.13	1.08
Eu/Eu*	0.70	0.70	0.69	0.76	0.68	0.69	0.66	0.66	0.70	0.71	0.65
La_N/Sm_N	4.28	4.34	4.16	5.55	5.46	3.98	3.81	3.98	6.62	4.08	4.31
La_N/Yb_N	10.91	11.16	10.44	16.10	16.02	10.21	10.82	10.98	27.72	10.14	9.78

JZB: Jiaozhou Bay; CJR: Changjiang River (Yang et al., 2003b); YR: Yellow River (Yang et al., 2003b); BHS: Bohai Sea (Mi et al., 2020); ECS: East China Sea (Mi et al., 2020); YS: Yellow Sea (Mi et al., 2020); HR: Hanhe River (Yang et al., 2003b); UCC: upper continental crust (Taylor and McLennan, 1995); SDL: Shandong Loess (Jia et al., 2016). La_N/Sm_N and La_N/Yb_N (N: normalized by chondrite which is from Sun and McDonough, 1989). The sediments in the upper part of Jiaozhou Bay are from 1995 to 2019; the sediments in the lower part of Jiaozhou Bay are from 1988 to 1995.

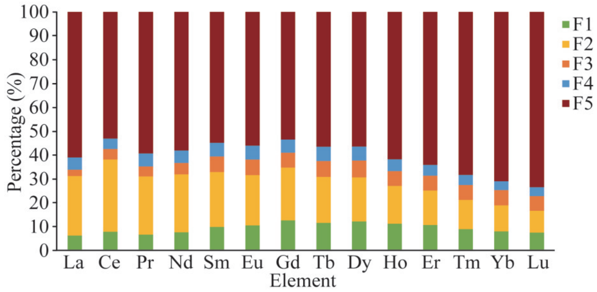

Fig.3 The average percentage of REE contents in the five fractions

The contents of each fraction are the average analysis results of multiple samples, and the percentage of each fraction is its percentage of the five fractions. F1: exchangeable and carbonate fraction; F2: easily reducible oxides fraction; F3: reducible oxides fraction; F4: magnetite fraction; F5: residual fraction.

5 DISCUSSION

5.1 The conservation of REEs during the sedimentary processes

Due to complex sedimentary processes, geochemical compositions of sediments are likely affected by other factors, including the influences of grain size and chemical weathering. Thus, it is necessary to exclude the influences of other factors to better constrain the provenance.

The grain size of Jiaozhou Bay sediments in this study did not change significantly (Fig.2). Compared with major element contents, REE contents are poorly correlated with the grain size (Supplementary Table S4), and the REE ratios (e.g., La_N/Sm_N and La_N/Yb_N, N: normalized by chondrite which is from Sun and McDonough (1989)) have no correlation with the grain size (Fig.4a–b; Supplementary Table S4). Thus, it reflects that REE contents, especially REE ratios, have not been affected by varying grain size.

To identify the effects of chemical weathering on geochemical compositions of sediments from Jiaozhou Bay, the CIA value proposed by Nesbitt and Young (1982) is used, which is expressed as $[Al_2O_3/(Al_2O_3+Na_2O+K_2O+CaO^*)]\times100$, where CaO* is the amount of CaO incorporated in the silicate fraction of the rock and all values are molecular proportions. The method to correct the CaO content proposed by McLennan (1993) is used, i.e., $n(CaO^0)=n(CaO)-10\times n(P_2O_5)/3$. If $n(CaO^0)<n(Na_2O)$, then $n(CaO^*)=n(CaO^0)$; otherwise $n(CaO^*)=n(Na_2O)$, where "*n*" represents molecular proportions.

In Fig.5, the weathering trend of sediments from

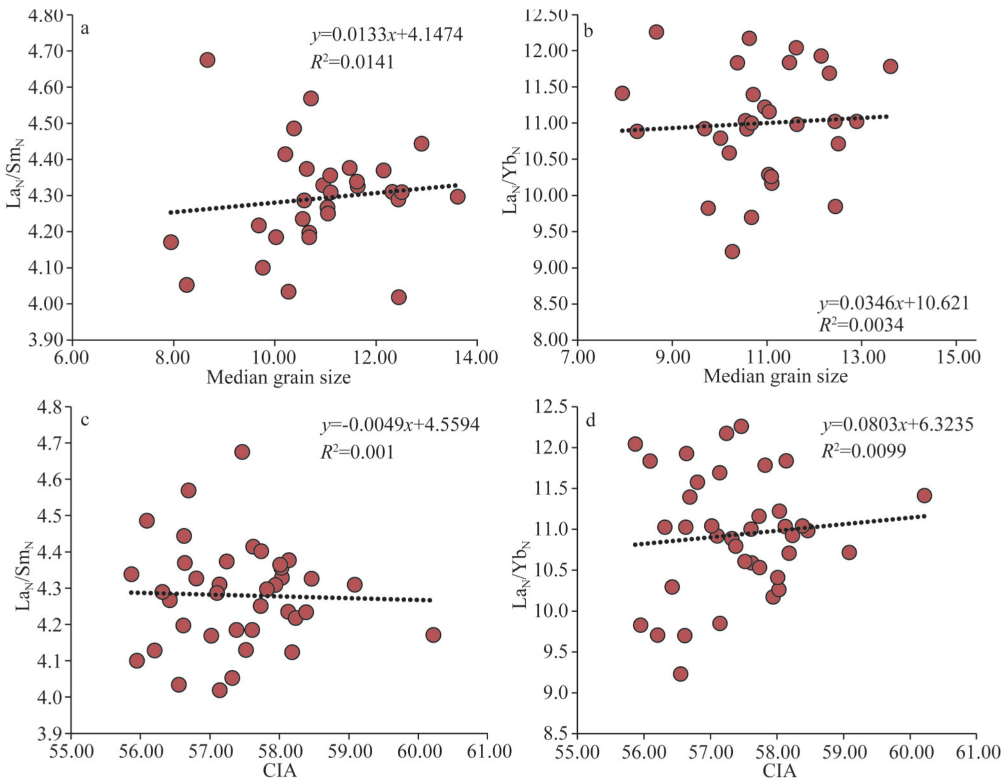

Fig.4 Co-variation diagrams of median grain size with La_N/Sm_N and La_N/Yb_N for sediment samples from Jiaozhou Bay (a & b); co-variation diagrams of CIA with La_N/Sm_N and La_N/Yb_N for sediment samples from Jiaozhou Bay (c & d)

CIA: the chemical index of alteration.

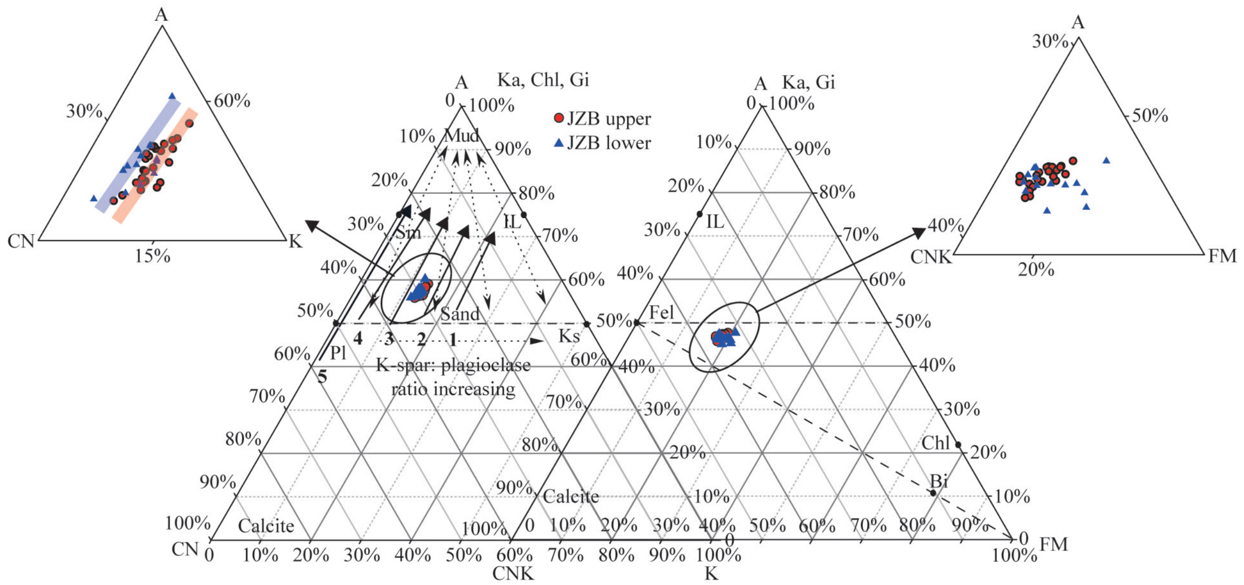

Fig.5 The chemical weathering trends of A-CN-K and A-CNK-FM

Red circles: the upper Jiaozhou Bay sediments from 1995 to 2019; blue triangles: the lower Jiaozhou Bay sediments from 1988 to 1995. Ka: kaolinite; Chl: chlorite; Gi: gibbsite; Sm: smectite; IL: illite; Pl: plagioclase; Ks: K-feldspar; Fel: feldspar; Bi: biotite; A: Al_2O_3; CN: CaO^*+Na_2O; K: K_2O; CNK: $CaO^*+Na_2O+K_2O$; FM: FeO(T)+MgO. Black solid line: the ideal theoretical weathering trend of different parent rocks. 1: average granite; 2: average adamellite; 3: average granodiorite; 4: average tonalite; 5: average gabbro.

Jiaozhou Bay is parallel to the ideal theoretical weathering trend (Nesbitt and Young, 1989), which indicates the insignificant influence of K metasomatism (Fedo et al., 1995). Furthermore, as the lack of correlation between CIA and the grain size (Supplementary Table S4) (Bouchez et al., 2010), these CIA values can effectively represent the intensity of chemical weathering for Jiaozhou Bay sediments. The CIA values of the Jiaozhou Bay sediments in this study (CIA values are 55–60; Supplementary Table S1) are lower than those of average shale (about 65–70; McLennan, 1993), Bohai Sea (about 52–78; Liang et al., 2020) and the Yellow Sea (about 65–86; Cao et al., 2021; Chen et al., 2022). According to the classification of weathering degree by Nesbitt and Young (1982), the Jiaozhou Bay sediments experienced a low degree of chemical weathering (Nesbitt and Young, 1982; Silva et al., 2016; Blake et al., 2017; Pei et al., 2020). In addition, the REE ratios (e.g., La_N/Sm_N and La_N/Yb_N) are not correlated with CIA (Fig.4c–d; Supplementary Table S4). This further indicates these REE ratios have not been affected by chemical weathering during the sedimentary processes.

Because REEs are resistant to chemical weathering and not affected by grain size during the sedimentary processes, REEs can greatly inherit from the provenance. This is also supported by the dominance of residual fraction for the conservation of REEs among all the five fractions (Fig.3). Therefore, REEs can be convincingly used to trace the sediment provenance.

5.2 Provenance of Jiaozhou Bay sediments

The geochemical characteristics of sediments from Jiaozhou Bay are further compared with those from neighboring rivers and seas. In Fig.6a, the sediments from Jiaozhou Bay in this study have low $(La/Sm)_{UCC}$ and $(Gd/Yb)_{UCC}$ ratios (UCC: normalized by upper continental crust which is form Taylor and McLennan (1995)), which geochemical characteristics are highly consistent with those of the Shandong loess (Fig.6a–b). Considering that coastal rivers (such as the Dagu River and Baisha River) are the main input of sediments to Jiaozhou Bay (Fig.1) (Fu et al., 2007; Chen et al., 2019; Liu et al., 2020a), we further compared geochemical characteristics of sediments from Jiaozou Bay with bedrocks in the drainage basin of rivers entering the bay. The geochemical characteristics of Jiaozhou Bay sediments are similar to those of Dagu River and Changyi-Anqiu and Pingdu (Dagu River-Jiaolai River), followed by Laoshan (Licun River) (Fig.6c). As REEs are mainly hosted by the residual fraction, the similarity of REE ratios between the residual fraction and those of Jiaolai River-Dagu River (Fig.6c) also supports that sediments of Jiaozhou Bay are mainly derived from Jiaolai River-Dagu River.

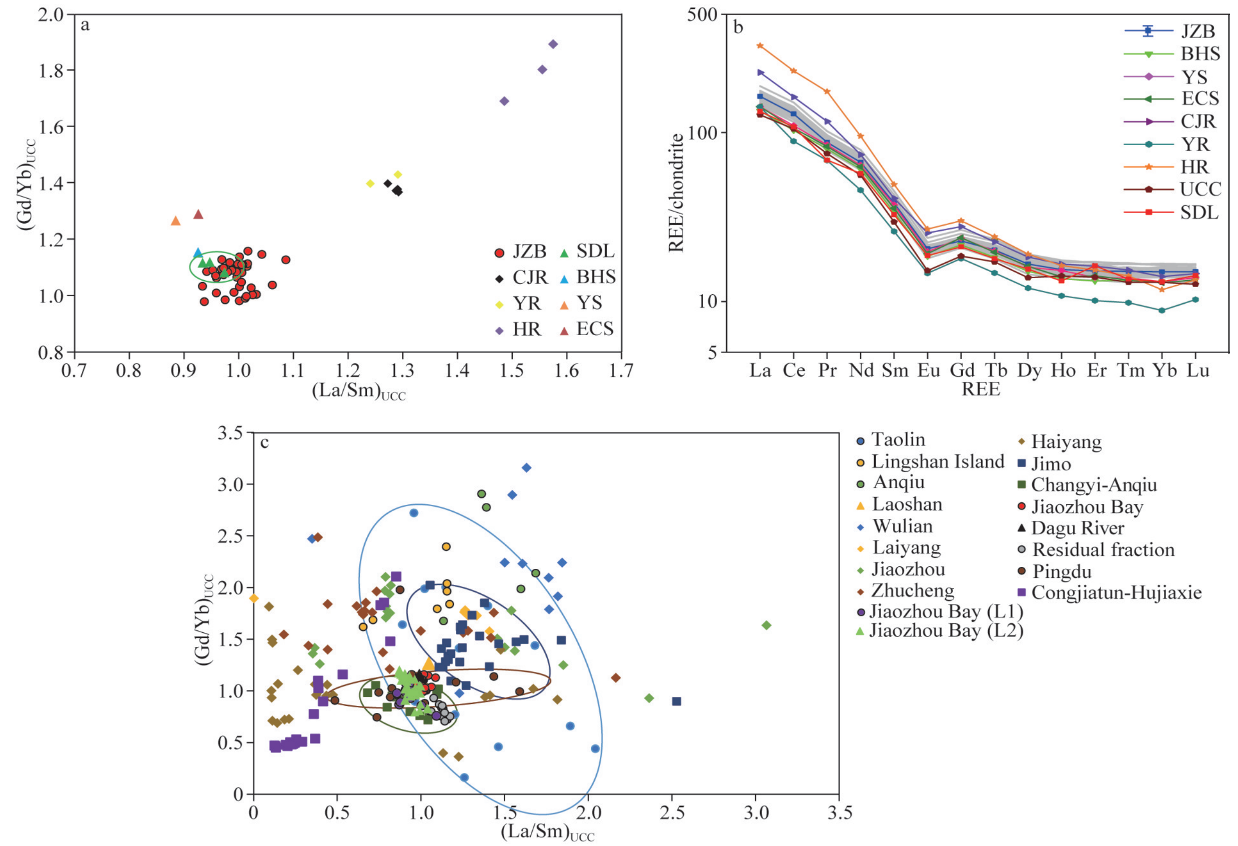

Fig.6 Discrimination diagram of $(Gd/Yb)_{UCC}$ vs. $(La/Sm)_{UCC}$ (a & c), and chondrite-normalized REE pattern of sediments of Jiaozhou Bay and other sources (b)

a & c is modified after Rao et al., 2017 and Zhao et al., 2018, and chondrite used to normalize in (b) is from Sun and McDonough, 1989. UCC: normalized by upper continental crust (Taylor and McLennan, 1995); JZB: Jiaozhou Bay; residual fraction: the residual fraction of sediment in Jiaozhou Bay; CJR: Changjiang River (Yang et al., 2003b); YR: Huanghe River (Yang et al., 2003b); BHS: Bohai Sea (Mi et al., 2020); ECS: East China Sea (Mi et al., 2020); YS: Yellow Sea (Mi et al., 2020); HR: Hanhe River (Yang et al., 2003b); SDL: Shandong Loess (Jia et al., 2016); UCC: upper continental crust (Taylor and McLennan, 1995); Changyi-Anqiu (Li et al., 2020c); Jimo (Pang, 2015; Hou et al., 2016; Cao, 2018; Gu, 2019; Li et al., 2020b; Zhu et al., 2021); Wulian (Chen et al., 1993; Qiu and Wang, 1999); Zhucheng (Su et al., 1997; Meng et al., 2006; Cao, 2018); Congjiatun-Hujiaxie (Song et al., 2020); Haiyang (Guo et al., 2002; Wang et al., 2018; Ma et al., 2021); Jiaozhou (Meng et al., 2006; Kuang et al., 2012; Cao, 2018); Dagu River (Zhang, 2014); Laiyang (Pang, 2015); Pingdu (Meng, 2016); Laoshan (Gu, 2019); Anqiu (Cao, 2018); Taolin (Zhang, 2017); Lingshan Island (Zhang, 2017); and L1 and L2 in Jiaozhou Bay (Wang et al., 2003).

Furthermore, we classified data of the collected bedrocks into four groups based on their relevant distributions, i.e., Moshui River-Baisha River, Licun River, Dagu River-Jiaolai River, Yanghe River, and calculated the contribution of different areas to Jiaozhou Bay sediments. The provenance index (PI) (Yang et al., 2000) is expressed as $PI=\Sigma|C_{ix}-C_{i1}|/(\Sigma|C_{ix}-C_{i1}|+\Sigma|C_{ix}-C_{i2}|+\Sigma|C_{ix}-C_{i3}|+\Sigma|C_{ix}-C_{i4}|)$, where i represents a ratio of two elements, C_{ix} stands for the sediments in Jiaozhou Bay, C_{i1}, C_{i2}, C_{i3}, and C_{i4} are Moshui River-Baisha River, Licun River, Dagu River-Jiaolai River, and Yanghe River, respectively. The PI reflects the general degree of similarity in chemical composition of different components. The smaller the PI value is, the larger the contribution is. We used the LREE/HREE ratio (e.g., La/Yb) to calculate different areas contribution in this study. According to the sediment samples from Jiaozhou Bay and the residual fraction (Table 2), it can be found that the Dagu River-Jiaolai River accounts are the highest proportion, followed by Licun River, Moshui River-Baisha River, and Yanghe River, which is generally consistent with the order sediments loaded in the four areas (Sheng et al., 2014; Bi et al., 2015).

With increasing depths, La_N/Sm_N and La_N/Yb_N ratios generally remain constant until the depths of roughly 40 cm, and show an obvious change beyond this depth (Fig.7), reflecting a slightly stronger fractionation of LREE from M-HREE (11 vs. 10 for the average La_N/Yb_N ratio, respectively; Table 1). This change likely reflects that the relative

Table 2 The provenance index (PI) of different areas

Areas	Licun River	Moshui River-Baisha River	Dagu River-Jiaolai River	Yanghe River
Jiaozhou Bay	0.125	0.390	0.004	0.482
Residual fraction of Jiaozhou Bay	0.155	0.356	0.063	0.426
Jiaozhou Bay upper	0.114	0.389	0.011	0.485
Jiaozhou Bay lower	0.137	0.376	0.027	0.459
L1, L2	0.150	0.361	0.054	0.434

The smaller the PI value is, the larger the contribution is.

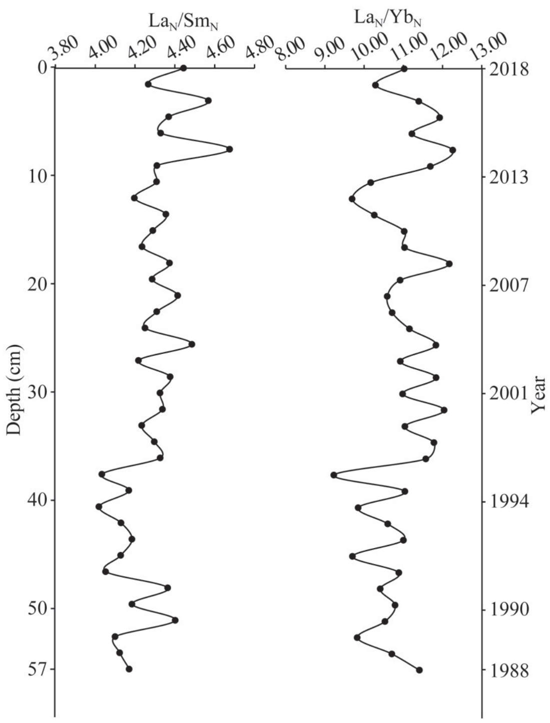

Fig.7 Variations in La_N/Sm_N and La_N/Yb_N of sediment samples from Jiaozhou Bay with increasing depths

The age models were estimated by previously reported study (i.e., 3.96 cm/a before 1992 and 1.63 cm/a after 1992 as proposed by Li et al. (2011) using ^{210}Pb to dating D4).

contributions of different source materials varied at the depths of ca. 40 cm. The upper and lower parts of the Jiaozhou Bay sediments have obvious parallel distribution in Fig.5, which also indicates the variation of mineral proportions.

We compared the upper and lower parts (with a limit of roughly 40 cm) with the four areas. For the composition change of the sampling profile roughly 1995, it is speculated that it may be caused by the increased contribution of Dagu River-Jiaolai River and Licun River. However, this change is also influenced by other factors. Compared with HREEs, L-MREEs are more enriched in active fraction (non-residual fraction) (Fig.3), which may be caused by the adsorption of carbonates and iron-manganese oxides. Since the early 1990s, because of the construction of reservoirs and the related evolution of aquaculture, the amounts of sediments transported by rivers have been greatly reduced, and solid waste discharge has become an important source of sediment injected into Jiaozhou Bay (Li et al., 2006; Wang, 2009; Xing et al., 2017). Furthermore, LREEs are preferentially absorbed by organic matter (He et al., 2004; Hathorne et al., 2015; Liu et al., 2019a; Yu et al., 2021). Thus, the increase of La_N/Yb_N ratio with depths may also reflect the effect of rapid increasing organic matter in the bay after the 1990s (Fig.7) (Wang et al., 2017, 2021b; Yuan et al., 2018; Cao et al., 2020).

We also compared with the sediments from other locations of Jiaozhou Bay. The locations of sediment columns L1 and L2 previously reported (Wang et al., 2003) are close to our sampling location. They have low $(La/Sm)_{UCC}$ and $(Gd/Yb)_{UCC}$ ratios, which geochemical characteristics are also similar with those of the Jiaozhou Bay sediments in this study and the Dagu River-Jiaolai River (Fig.6c). Combined with their consistent values of provenance index, sediments from L1 and L2 could be also mainly derived from the Dagu River-Jiaolai River, with some contributions of Licun River, Moshui River-Baisha River, and Yanghe River (Table 2).

6 CONCLUSION

In this study, the REEs in a newly collected sediment column from Jiaozhou Bay were systematic analyzed. During the sedimentary processes, the ratios of REE in Jiaozhou Bay sediments was not affected by grain size or chemical weathering, and the information characteristics of the provenance area were completely preserved. By further comparing the REEs of the total and residual fractions of the sediments in Jiaozhou Bay with the bedrock of the main river input areas, it was found that Dagu River-Jiaolai River is the main source materials input of Jiaozhou Bay, followed by Licun

River, Moshui River-Baisha River, and Yanghe River. Compared with the systematic variations of element ratios along the sampling profile, the change of source materials and related compositions around 1995 (roughly 40-cm depth) may be caused by the increased contribution from Dagu River-Jiaolai River and Licun River due to the construction of reservoir, and related change of aquaculture (e.g., rapid accumulation of organic materials).

7 DATA AVAILABILITY STATEMENT

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

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Electronic supplementary material

Supplementary material (Supplementary Tables S1–S4) is available in the online version of this article at https://doi.org/10.1007/s00343-022-2019-z.