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SEDIMENTS, SEC 1 • SEDIMENT QUALITY AND IMPACT ASSESSMENT • RESEARCH ARTICLE

Distribution and provenance implication of rare earth elements and Sr-Nd isotopes in surface sediments of Jiulong River, Southeast China

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Abstract

Purpose The purposes of this paper are to investigate the geochemical characteristics of rare earth elements (REEs) in the surface sediments of Jiulong River, southeast China, to probe the provenance compositions of the sediments, and to analyze the potential anthropogenic influence on REEs in the sediments. REEs and Sr-Nd isotopes were selected as the tools because REEs can be used to identify the anthropogenic effects on sediments and Sr-Nd isotopes have been widely known as powerful tracers for provenance analysis.

Materials and methods Fifty-three samples of surface sediments (0~5 cm) were collected from Jiulong River. The concentrations of REEs and Sr-Nd isotopic compositions in the surface sediments were determined by inductively coupled plasma mass spectrometry (ICP-MS) and thermal ionization mass spectrometry (TIMS), respectively. The chondrite-normalized and WRAS-normalized REEs patterns, enrichment factor, plots of La-Th-Sc and La/Yb-∑REE, and plots of $\varepsilon_{Nd(0)}$ vs $^{87}Sr/^{86}Sr$ and $\varepsilon_{Nd(0)}$ vs δEu are presented.

Results and discussion The mean concentration of ΣREEs in the surface sediments of Jiulong River was 254.25 mg kg^{-1}. The mean values of ΣLREEs (227.6 mg kg^{-1}), ΣHREEs (26.64 mg kg^{-1}), and $(La/Yb)_N$ ratios (9.24) suggested an enrichment of LREEs compared to HREEs. Negative Eu anomalies were observed in the surface sediments. The distribution patterns of REEs in the surface sediments from different areas of Jiulong River were remarkably similar. The values of $^{87}Sr/^{86}Sr$, $^{143}Nd/^{144}Nd$, and $\varepsilon_{Nd(0)}$ were 0.714091~0.733476, 0.511875~0.512271, and − 14.88~− 7.16, respectively. The plots of $\varepsilon_{Nd(0)}$ vs $^{87}Sr/^{86}Sr$, $\varepsilon_{Nd(0)}$ vs 1/[Nd], and $\varepsilon_{Nd(0)}$ vs δEu indicated that the sediments in Jiulong River were mainly derived from natural geological processes and the REEs might be also influenced by anthropogenic activities such as Fujian Pb-Zn deposit, coal ash, and industrial sludge.

Conclusions The REEs in the surface sediments at different sites are similar in geochemical characteristics, with a right-inclined distribution pattern and higher enrichment of light REEs (LREEs) compared to heavy REEs (HREEs), and a negative Eu anomaly but no evidence of Ce anomaly. The sediments in Jiulong River were mainly derived from natural geological processes (granite and magmatic rocks), and the REEs in the sediments were also influenced by anthropogenic activities (Fujian Pb-Zn deposit, coal ash, and industrial sludge).

Keywords Jiulong River · Provenance implication · Rare earth elements · Sr-Nd isotopes · Surface sediment

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1 Introduction

Rare earth elements (REEs) are a series of elements with the atomic numbers of 57–71 in group IIIB of the periodic table, including light REEs (LREEs: from La to Eu) and heavy REEs (HREEs: from Gd to Lu), which have similar electronic structures and chemical properties (Haskin and Haskin 1966; Taylor and Mclennan 1995; Chang et al. 2016). In fact, REEs are not rare but abundant in the Earth's crust (Wang et al. 1998), and they have been found in more than 200 minerals all over the world (Henderson 1984). China is abundant in REE mineral resources, and REEs were applied in Chinese industry and agriculture for more than 40 years (Cao et al. 2000; Loell et al. 2011). The concentration of REEs has greatly increased in the environment as a result of their heavy use in the industrial production and agricultural production and the exploitation activities of REE resources (Essington and Mattigod 1990; Gonzalez et al. 2014). The concentrations and environmental effects of REEs have received increasing attention (Yan et al. 2012; Tripathee et al. 2016; Alfaro et al. 2018). In the past decades, REEs have been widely used to trace material sources due to their geochemical characteristics of weak mobility and fractionation during earth surface processes (Piper 1974; Ramesh et al. 1999; Munksgaard et al. 2003; Yan et al. 2012). REEs can also be used as a tool for identifying anthropogenic effects on sediments whose geochemical characteristics were modified by unnatural liquid or solid input (Borrego et al. 2004; Liu et al. 2015; Kulaksız and Bau 2013).

Radiogenic isotopes have been widely known as powerful tracers for provenance analysis. Nd isotopic compositions can be used as tracers to analyze the provenance of detrital sediments since Nd isotopic compositions are primarily controlled by the source rocks and nearly unaffected by intracrustal processes during weathering and transport (Goldstein and Jacobsen 1988; Najman 2006; Wu et al. 2010). Sr is more mobile than Nd, and Sr isotopic compositions are also affected by the processes of weathering, erosion, and transportation (Goldstein and Jacobsen 1987). In addition, Walter et al. (2000) suggest that Sr isotopic compositions are also useful tracers in analyzing the material sources of sediments. So far, the Sr-Nd isotopic compositions have been successfully applied to analyze the provenance of sediments in the Eastern Mediterranean Sea (Weldeab et al. 2002), Tibetan Plateau (Wu et al. 2010), South China Sea (Wei et al. 2012), Amazon (Roddaz et al. 2014), and Yellow Sea (Lim et al. 2015).

Jiulong River is the second largest river in Fujian province, Southeast China, with a total drainage basin area of 1.47×10^4 km^2 and a total flow of 12.4×10^9 m^3 year^{-1}. It flows through Longyan, Zhangzhou, and Xiamen regions and acts as the important source of drinking, industrial and agricultural water for the three regions (Lin et al. 2016). There were more than 3.8 million of people living in the eight cities within the catchment basin (Chen et al. 2015). The safety of the water environment in Jiulong River basin is of great significance to the economic zone on the west side of Taiwan Straits. The research on geochemical characteristics of trace elements in sediments is an ideal means to assess the environmental quality since the sediments can serve as both a source and sink for trace elements (Sun et al. 2010). Therefore, a lot of investigations on heavy metal contamination in sediments have been carried out in this river. It has been reported that Jiulong River has been polluted by heavy metals as a consequence of human activities (Wang et al. 2014; Zhang et al. 2014a, b). But there are few data about the provenance compositions of the sediments and geochemical characteristics of REEs in the sediments of Jiulong River (Jin et al. 2010).

In this study, the concentrations, distribution patterns of REEs, and the isotopic compositions of Sr and Nd in the surface sediments of Jiulong River were analyzed with the following objectives: (i) to investigate the concentrations and geochemical characteristics of REEs in the surface sediments of Jiulong River, (ii) to probe the provenance compositions of the surface sediments of Jiulong River, and (iii) to analyze the potential anthropogenic influence on REEs in the sediments.

2 Materials and methods

2.1 Sampling and preparation

Fifty-three samples of surface sediment (0~5 cm) were collected from Jiulong River in October 2012 (Fig. 1), including North River (sites 1~13), West River (sites 14~17), Coastal wetland (sites 18~32), and Estuary (sites 33~53). After being transported to the laboratory, the sediment samples were freeze-dried to a constant weight. The samples were ground lightly with a wood stick and passed through a 2 mm sieve to remove rocks, roots, and other large particles. The samples were then ground with an agate pestle and mortar and sieved with a 63 μm nylon sieve due to strong association of metals with fine-grained sediments (Horowitz and Elrick 1987). The sediment under the sieve was kept in a sealed plastic vessel at 4 °C for further analysis.

In order to trace the sources, some potential anthropogenic source samples (Pb-Zn deposit, coal ash, atmospheric dust, and industrial sludge) were also collected (Fig. 1). Fujian Pb-Zn deposit samples were collected from Pb-Zn deposits in Longyan City. Coal ashes were collected from coal-fired power plants and some other coal-fired enterprises in the basin. Atmospheric dusts were collected from the main cities (Longyan, Zhangzhou, and Xiamen) in the basin. Industrial sludge samples were collected from sewage treatment plants in the basin.

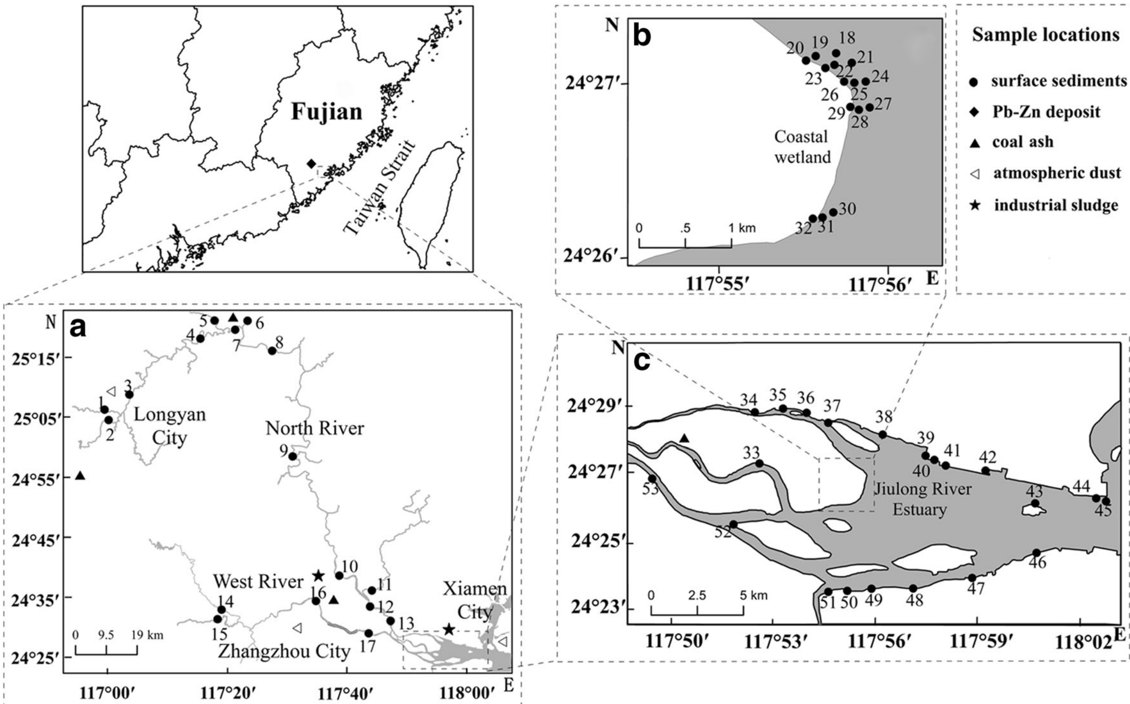

Fig. 1 Sampling sites of the surface sediments in Jiulong River. **a** North River and West River. **b** Coastal wetland. **c** Estuary

2.2 Analysis method and quality assurance

For the analysis of trace elements (including REEs), about 100 mg dry sediment was put in a Teflon vessel; then, a mixture of 2 mL HCl, 6 mL HNO_3, and 2 mL HF was added. The Teflon vessel was tightly closed and digested on a hot plate at 120 °C for 24 h. Then, the solution was evaporated to dryness and dissolved in 5% HNO_3 solution spiked with an internal standard Rh (5 μg L^{-1}) for analysis. The concentrations of trace elements in the digestion solutions were determined by inductively coupled plasma-mass spectrometry (ICP-MS) (ELAN9000, Perkin-Elmer, USA) in Analytical Laboratory, Beijing Research Institute of Uranium Geology. Reagent blanks and sample replicates were included throughout the analysis, and the analytical precisions were better than 10%.

For the analysis of Sr and Nd isotope ratios, the separation and purification for Sr and Nd isotopes were processed according to the analytical procedures for determinations for isotopes of lead, strontium, and neodymium in rock samples (GB/T 17672-1999, issued by the State Bureau of Quality and Technical Supervision, China). The $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios were determined using VG354 thermal ionization mass spectrometry (TI-MS). Solutions of reference materials (NBS987 for Sr isotope and JMC for Nd isotope) were determined to check the reproducibility and accuracy of the analyses of Sr and Nd isotopes. The measured $^{87}Sr/^{86}Sr$ of NBS987 was 0.710220 ± 0.000015, and $^{143}Nd/^{144}Nd$ of JMC was 0.511986 ± 0.000022. In this study, 29 sediment samples were analyzed for $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios.

2.3 Characteristic parameters of REEs

Characteristic parameters of REEs, including ΣREEs, LREEs/HREEs, $(La/Yb)_N$, $(La/Sm)_N$, $(Gd/Yb)_N$, δCe, and δEu, can be employed to investigate the geochemical characteristics of REEs. ΣREEs is the total concentration of REEs. LREEs/HREEs is the ratio of ΣLREEs to ΣHREEs. The three parameters of $(La/Yb)_N$, $(La/Sm)_N$, and $(Gd/Yb)_N$ represent the normalized ratios of a LREEs to a HREEs and were commonly used to characterize the differentiation of LREEs to HREEs. The subscript N stands for the Chondrite-normalized value (Taylor and Mclennan 1995). δCe and δEu represent the anomaly values of Ce and Eu, respectively. The values of δCe and δEu were calculated as follows (Aide and Aide 2012):

$$\delta Ce = \frac{(Ce)_N}{(La_N \times Pr_N)^{0.5}} \quad (1)$$

$$\delta Eu = \frac{(Eu)_N}{(Sm_N \times Gd_N)^{0.5}} \quad (2)$$

where subscript N stands for the Chondrite-normalized value. δCe and δEu values greater than 1.05 indicate positive anomalies, and less than 0.95 indicates negative anomalies (Yao et al. 2010).

2.4 Enrichment factors of REEs

The enrichment factor (EF) represents the normalization of heavy metal concentrations in sediments relative to a reference element (N'Guessan et al. 2009). In the past decades, EF was widely used to evaluate the anthropogenic impact on sediments. Generally, Al, Cs, Sc, Fe, and even organic matter content can be used as reference elements (N'Guessan et al. 2009). Considering the lower coefficient of variation (0.19) and higher correlations with REEs, Sc was chosen as the reference element. The values of EF were calculated by Eq. (3) (N'Guessan et al. 2009):

$$\mathrm{EF} = \frac{(\mathrm{E/Sc})_{sample}}{(\mathrm{E/Sc})_{background}} \tag{3}$$

where $(\mathrm{E/Sc})_{\mathrm{sample}}$ is the ratio of the concentration of an element to that of Sc in the sediment sample, and $(\mathrm{E/Sc})_{\mathrm{background}}$ is the same ratio in Fujian soil (Chen et al. 1992). According to the EF values, the enrichment level of elements can be classified into six classes (Sutherland 2000): non-enriched ($\mathrm{EF} < 1$), slightly enriched ($1 \le \mathrm{EF} < 2$), moderately enriched ($2 \le \mathrm{EF} < 5$), significantly enriched ($5 \le \mathrm{EF} < 20$), strongly enriched ($20 \le \mathrm{EF} < 40$), and extremely enriched ($40 \le \mathrm{EF}$).

3 Results and discussion

3.1 Concentrations and characteristic parameters of REEs in the surface sediments

Distributions of REEs in the surface sediments of Jiulong River are shown in Fig. 2. It is interesting that the REEs showed considerable variation in North River and West River, but stabilization in Coastal Wetland and Estuary. The concentrations of REEs were higher at sites of 5, 8, 14, and 17 owing to the flourishing agriculture and pig-breeding industry around these sites, while they were lower at sites 2, 6, 15, and 16. The distributions of all REEs were similar, suggesting that all the REEs in the surface sediments were of similar geochemical characteristics. Concentrations of REEs in the surface sediments of Jiulong River are listed in Table 1. The mean concentrations of REEs in the surface sediments of Jiulong River followed a generalized sequence of Ce > La > Nd > Pr > Sm > Gd > Dy>Er > Yb > Eu > Ho > Tb > Lu > Tm, following the Oddo-Harkins rule (Wei et al. 2001). The coefficient of variation (CV) values were 0.19~0.25 with the mean of 0.21, suggesting small variations of REE concentrations in the sediments. The concentrations of REEs in sediments at different sites were similar in geochemical characteristics. It is worth noting that the concentrations of REEs in the surface sediments of Jiulong River were all higher than the background values in Fujian soil (Chen et al. 1992).

The results of characteristic parameters of REEs in the surface sediments of Jiulong River are listed in Table 2. The mean concentrations of REEs (ΣREEs) were 244.85 mg kg^{-1} in North River, 233.57 mg kg^{-1} in West River, 241.01 mg kg^{-1} in Coastal wetland, and 273.46 mg kg^{-1} in Estuary area. The variations of ΣREEs in the surface sediments had a small CV value of 0.21. The total mean ΣREEs in the surface sediments of Jiulong River was 254.25 mg kg^{-1}, which was higher than those in Fujian soil (198.37 mg kg^{-1}) (Chen et al. 1992), the sediments of Yangtse River (167.1 mg kg^{-1}), the sediments of Minjiang River (171.18 mg kg^{-1}), and the sediments of Fujian coastal area (171.05 mg kg^{-1}) while lower than that in deep-sea sediments (411 mg kg^{-1}) (Shen 1990). The mean values of ΣLREEs and ΣHREEs were 227.6 mg kg^{-1} and 26.64 mg kg^{-1}, respectively. The mean value of LREEs/HREEs was 8.56, suggesting that LREEs were enriched compared to HREEs. The $(\mathrm{La/Yb})_N$ ratios were 5.99~15.27 with a mean of 9.24. It has been reported that the values of $(\mathrm{La/Yb})_N$ ratio can indicate the erosion rates in the river system (Longjiang et al. 2010). The higher values of $(\mathrm{La/Yb})_N$ ratio were observed at sites 1, 13, and 45, suggesting that the erosion rates were higher at these sites because of human activities (such as anthropogenic REEs input). The REEs typically exhibit trivalent oxidation states, except Eu and Ce, which may also occur as Eu^{2+} and Ce^{4+}, respectively (Aide and Aide 2012). The values of δCe ranged from 0.81 to 1.13 with the mean of 1.01, which showed no evidence of Ce anomalies. The values of δEu ranged from 0.38 to 0.66 with the mean of 0.55, which showed negative Eu anomalies. The negative Eu anomalies indicated that Eu^{3+} was reduced to Eu^{2+} under reduction conditions and might be associated with the process of strong leaching which easily occurs on warm and humid subtropical regions.

3.2 REE distribution patterns in the surface sediments

REE distribution patterns were mainly associated with their sources because of the characteristic that they seldom fractionate during transportation (Taylor and Mclennan 1995; Plank and Langmuir 1998). Therefore, REE distribution patterns can be used to trace the source components of sediments (Munksgaard et al. 2003). It is difficult to directly compare graphically REE abundance because REEs with even atomic numbers are usually more abundant than their adjacent REEs, which is called "Oddo-Harkins effect" (Zhang et al. 2014a, b). In order to eliminate the effect, the concentrations of REEs were normalized to mean chondritic meteorites (Taylor and Mclennan 1995) and World River Average Silt (WRAS) (Bayon et al. 2015) in this study.

The chondrite-normalized REE patterns in the sediments and in some potential sources are illustrated in Fig. 3a. This normalization pattern can be used to compare the REE abundance in geological materials and to identify the enrichment or

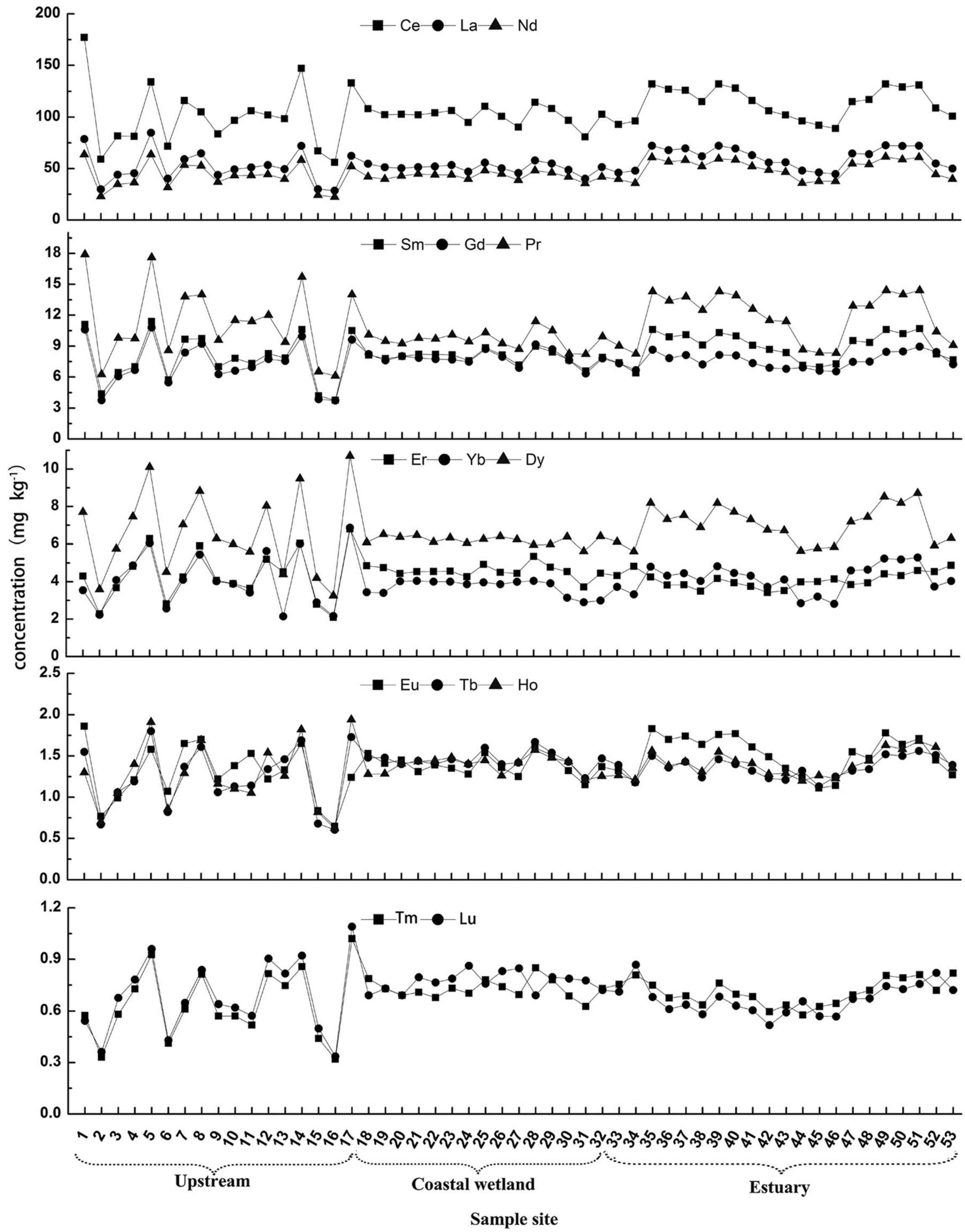

Fig. 2 Distribution of REEs in the surface sediments of Jiulong River (mg kg^{-1})

deficiency of elements (Lin et al. 2013). As shown in Fig. 3a, the distribution patterns of REEs in the surface sediments from different areas (North River, West River, Coastal Wetland, and Estuary) were remarkably similar, which suggested that all the REEs in the surface sediments were of similar origin. The normalized patterns were also similar to those of Fujian soils, Fujian Pb-Zn deposit, industrial sludge and coal ash with LREE enrichment, flat HREE patterns, and a negative Eu anomaly, while different from atmospheric dusts. These distribution patterns are typical characteristics of REEs in terrigenous sediments (Prego et al. 2009). The consistency of REE distribution pattern between surface sediments and Fujian soils indicated that the sediments were mainly derived from the local soil.

Table 1 Concentrations of REEs (mg kg^{-1}) in surface sediments of Jiulong River and Fujian soil, Chondrite and World River Average Silt

		La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
North River $n = 13$	Max	84.70	177.0	17.90	63.60	11.40	1.860	10.80	1.800	10.10	1.910	6.290	0.927	6.050	0.960
	Min	29.80	59.10	6.240	23.00	4.370	0.768	3.770	0.669	3.580	0.702	2.280	0.331	2.140	0.362
	Mean	53.35	101.0	11.66	43.55	7.969	1.347	7.388	1.246	6.557	1.254	4.279	0.631	3.996	0.676
West River $n = 4$	Max	72.00	147.0	15.70	58.20	10.60	1.650	9.940	1.730	10.70	1.940	6.800	1.020	6.870	1.090
	Min	28.60	56.00	6.100	22.30	3.760	0.646	3.740	0.603	3.250	0.622	2.090	0.319	2.160	0.336
	Mean	48.23	100.8	10.58	39.18	7.263	1.093	6.793	1.175	6.908	1.299	4.430	0.659	4.485	0.711
Coastal wetland $n = 15$	Max	57.90	114.3	11.40	48.05	8.980	1.600	9.160	1.670	6.520	1.572	5.350	0.850	4.040	0.862
	Min	40.10	80.70	8.21	35.54	6.580	1.150	6.330	1.230	5.610	1.207	3.710	0.627	2.900	0.690
	Mean	50.97	101.6	9.62	42.75	7.976	1.388	7.849	1.455	6.211	1.387	4.573	0.727	3.703	0.768
Estuary $n = 21$	Max	72.50	132.0	14.40	61.40	10.70	1.830	8.940	1.560	8.710	1.680	4.870	0.819	5.290	0.868
	Min	44.60	88.90	8.24	35.64	6.380	1.110	6.540	1.130	5.600	1.197	3.420	0.577	2.810	0.518
	Mean	60.52	113.5	11.83	50.12	8.926	1.512	7.598	1.360	7.040	1.403	4.092	0.708	4.172	0.667
Total mean		55.13	106.1	11.07	45.60	8.297	1.405	7.557	1.345	6.677	1.354	4.300	0.691	4.020	0.701
CV		0.22	0.21	0.25	0.22	0.21	0.19	0.19	0.19	0.22	0.19	0.20	0.19	0.25	0.20
Fujian soil		41.10	89.8	9.40	33.1	6.12	1.21	5.21	0.72	4.60	0.91	2.84	0.39	2.60	0.37
Chondrite		0.315	0.813	0.115	0.597	0.192	0.0722	0.259	0.0473	0.325	0.0723	0.213	0.0333	0.208	0.0323
WRAS		37.8	78	8.77	32.7	6.15	1.19	5.19	0.82	4.95	1.02	2.97	–	3.01	0.46

Note: The data of Fujian soil, Chondrite, and World River Average Silt were cited from Chen et al. (1992), Taylor and Mclennan (1995), and Bayon et al. (2015), respectively

The WRAS-normalized REE patterns of sediments are illustrated in Fig. 3b. This normalization pattern provided a new estimate for the mean composition of the weathered and eroded upper continental crust and could reflect the effects of mixing, homogenization, and differentiation in sediment deposition (Bayon et al. 2015). As shown in Fig. 3b, the distribution patterns of REEs in different area were remarkably similar with flat patterns, implying similar origin. Figure 3a, b shows that the distribution patterns of LREEs were similar, while HREEs were higher in the surface sediments than those in Fujian soil. This might be because HREEs in the soil were more easily delivered into the sediments by runoff. The results indicated that the REEs in the surface sediments in Jiulong River were mainly the result of natural geological processes and also might be impacted by Pb-Zn deposit and coal combustion.

Table 2 Characteristic parameters of REEs in surface sediments of Jiulong River

		ΣREE	LREE	HREE	LREE/HREE	$(La/Yb)_N$	$(La/Sm)_N$	$(Gd/Yb)_N$	δCe	δEu
North River $n = 13$	Max	380.27	350.16	38.84	11.63	15.27	4.53	2.84	1.1	0.66
	Min	137.2	123.28	13.92	6.48	6.16	3.73	1.1	0.81	0.44
	Mean	244.85	218.82	26.03	8.47	9.29	4.07	1.57	0.95	0.55
West River $n = 4$	Max	341.93	305.15	39.77	8.95	8.74	4.64	1.39	1.12	0.64
	Min	130.53	117.41	13.12	6.87	5.99	3.62	1.08	0.99	0.38
	Mean	233.57	207.11	26.46	8.08	7.37	4.19	1.23	1.05	0.51
Coastal wetland $n = 15$	Max	271.51	242.23	29.28	8.37	11.35	4.09	2.1	1.13	0.58
	Min	194.67	172.28	22.39	7.39	7.56	3.71	1.38	1.04	0.5
	Mean	241.01	214.33	26.67	8.03	9.16	3.89	1.72	1.07	0.54
Estuary $n = 21$	Max	323.98	292.68	32.34	10.12	11.12	4.58	1.95	1.13	0.63
	Min	210.94	187.92	23.02	7.66	8.17	3.74	1.3	0.93	0.51
	Mean	273.46	246.42	27.04	9.08	9.62	4.13	1.49	1	0.57
Total mean		254.25	227.6	26.64	8.56	9.24	4.05	1.55	1.01	0.55
CV		0.21	0.21	0.19	0.11	0.18	0.06	0.21	0.08	0.09

Note: $\delta Eu = Eu_N/(Sm_N \times Gd_N)^{1/2}$; $\delta Ce = Ce_N/(La_N \times Pr_N)^{1/2}$; subscript N stands for chondrite-normalized value

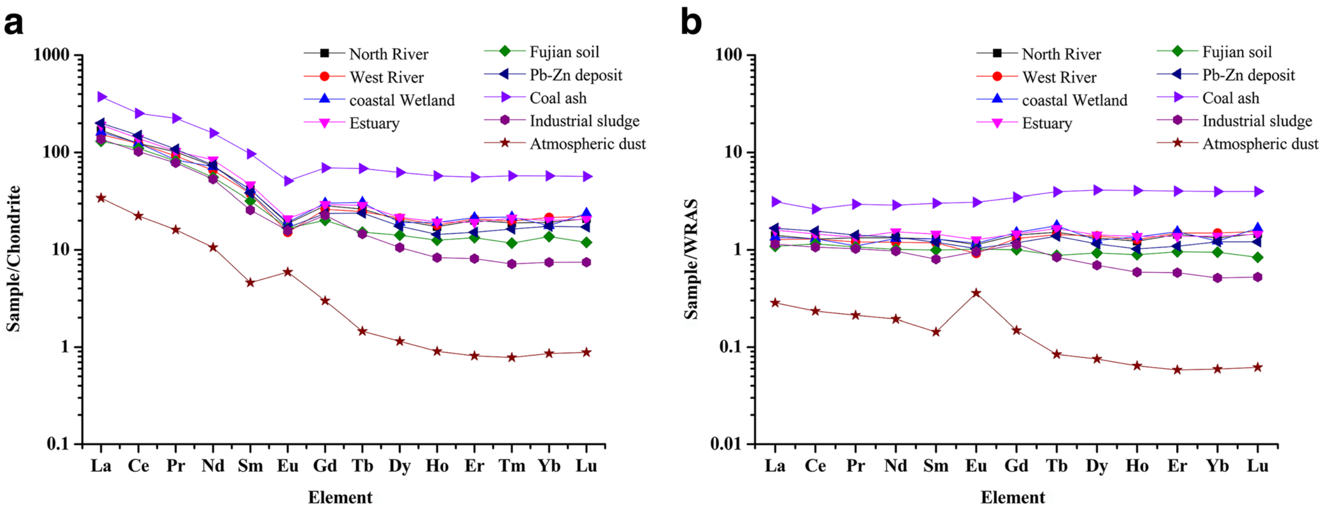

Fig. 3 REE distribution patterns of surface sediments in Jiulong River. **a** Chondrite-normalized. **b** WRAS-normalized

3.3 Enrichment factors of REEs in sediments

The calculated EF values of the REEs in this study are listed in Table 3. The mean EF values of 14 rare earth elements ranged from 1.08 to 1.80, indicating that all the rare earth elements were slightly enriched ($1 \leq EF < 2$).

According to the above discussion, the concentrations of REEs in surface sediments of Jiulong River Basin (245.25 mg kg^{-1}) were significantly higher than those in Fujian soil background (198.37 mg kg^{-1}) (Chen et al. 1992), Yangtse River (167.1 mg kg^{-1}), Minjiang River (171.18 mg kg^{-1}), and Fujian coastal area (171.05 mg kg^{-1}), indicating that the REEs were slightly enriched in surface sediments. These results might be related to one factor or a combination of several factors. Two possible factors were listed as follows: (1) the sediments themselves might derive from parent materials with high REE concentrations; (2) the elements might be added by anthropogenic activities such as coal combustion and industrial sludge, etc. Therefore, we would like to use La-Th-Sc diagram, La/Yb-∑REEs diagram, and plot of $\varepsilon_{Nd(0)}$ vs $^{87}Sr/^{86}Sr$ to probe the provenance compositions of the sediments, and use the plot of $\varepsilon_{Nd\ (0)}$ vs 1/[Nd] and $\varepsilon_{Nd\ (0)}$ vs δEu to analyze the potential anthropogenic influence on REEs in the sediments in the following paragraphs.

Table 3 Enrichment factors of REEs in surface sediments of Jiulong River

	La	Ce	Pr	Nd	Sm	Eu	Gd
North River	1.47	1.26	1.4	1.48	1.46	1.25	1.6
West River	1.46	1.39	1.4	1.47	1.45	1.13	1.61
Coastal Wetland	1.05	0.96	0.87	1.09	1.1	0.97	1.27
Estuary	1.23	1.06	1.05	1.27	1.22	1.05	1.23
Mean	1.26	1.11	1.11	1.28	1.27	1.08	1.36
	Tb	Dy	Ho	Er	Tm	Yb	Lu
North River	1.95	1.63	1.57	1.72	1.85	1.78	2.1
West River	1.99	1.81	1.72	1.88	2.04	2.08	2.32
Coastal Wetland	1.71	1.15	1.29	1.36	1.58	1.21	1.77
Estuary	1.6	1.29	1.3	1.23	1.55	1.35	1.54
Mean	1.75	1.37	1.4	1.44	1.67	1.47	1.8

3.4 Provenance compositions of the sediments

According to the discussion of REE distribution patterns, the distribution patterns of REEs in the surface sediments were similar to those of Fujian soils and Pb-Zn deposit with LREE enrichment, flat HREE patterns, and negative Eu anomalies. It is reported that REE composition of soils is inherited from parent rocks because they seldom fractionate during transportation (Taylor and Mclennan 1995; Plank and Langmuir 1998) and were with low solubility and relative immobility of REEs in the upper crust (Ross et al. 1995).

La-Th-Sc diagram and La/Yb-∑REEs diagram were used to discriminate felsic and basic provenance of surface sediments (Nyakairu and Koeberl 2001). As shown in Fig. 4, the data of surface sediments especially the sediments of North River were closer to the typical granitic gneiss sources and the clays, silts, sands, and gravels from mixed sources. This result indicated that the surface sediments might be derived from granite weathering and some other mixed sources of felsic and basic rocks. The La/Yb-∑REEs diagrams of the sediments, Fujian soil, and some potential source rocks are illustrated in Fig. 5. The data of surface sediments fell within the range of granite and were close to the range of alkaline basalt, indicating that the sediments might derive from granite and alkaline basalt. It is reported that the rocks in the Jiulong River basin were mainly Mesozoic magmatic rocks, and the soil-forming rocks were mainly granites (Hong et al. 2009).

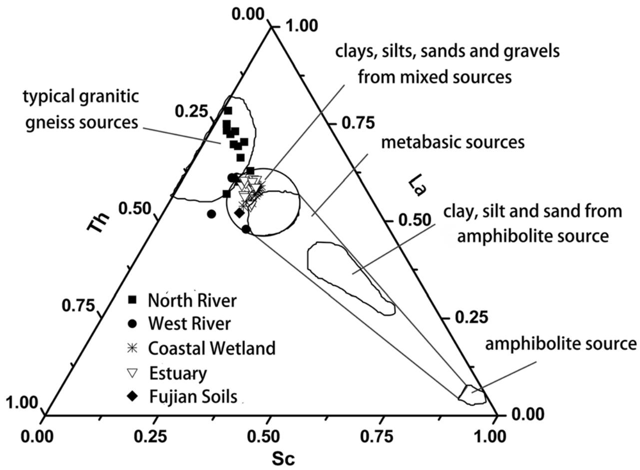

Fig. 4 Ternary plot of La-Th-Sc for the surface sediment samples in Jiulong River. The data were cited from Nyakairu and Koeberl, (2001)

Therefore, the surface sediments were mainly derived from local granite with minor contributions from basic rocks.

The Sr and Nd isotopic compositions in surface sediments of Jiulong River are listed in Table 4. The values of $\varepsilon_{Nd(0)}$ were calculated using Eq. 4 (Weldeab et al. 2002):

$$\varepsilon_{Nd(0)} = \left[\frac{\left({}^{143}Nd/{}^{144}Nd\right)_{measured}}{\left({}^{143}Nd/{}^{144}Nd\right)_{CHUR(0)}} - 1\right] \times 10^4 \quad (4)$$

where $({}^{143}Nd/{}^{144}Nd)_{measured}$ is the ratio of ${}^{143}Nd/{}^{144}Nd$ in the surface sediments and $({}^{143}Nd/{}^{144}Nd)_{CHUR(0)}$ is the ratio in the CHUR reference reservoir as 0.512638 (Jacobsen and Wasserburg 1980).

As shown in Table 4, the mean ${}^{87}Sr/{}^{86}Sr$ ratio in the surface sediments was 0.721930 with the range of 0.714091~0.733476, showing considerable variation; the mean ${}^{143}Nd/{}^{144}Nd$ ratio was 0.512089 with the range of 0.511875~0.512271, and the mean $\varepsilon_{Nd(0)}$ value was − 10.71 with the range of − 14.88~− 7.16. The surface sediments from Estuary show higher ${}^{143}Nd/{}^{144}Nd$ ratios and lower ${}^{87}Sr/{}^{86}Sr$ ratios, while the surface sediments from Coastal Wetland show lower ${}^{143}Nd/{}^{144}Nd$ ratios and higher ${}^{87}Sr/{}^{86}Sr$ ratios, implying that the sediments in Coastal Wetlands were more affected by continental crust than estuarine sediments.

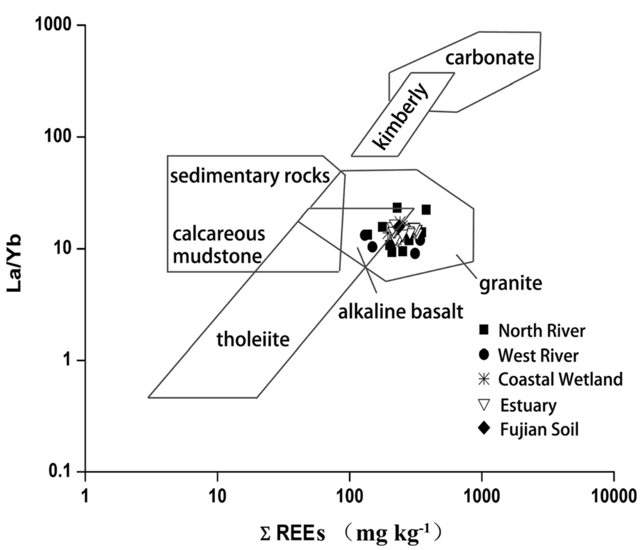

Fig. 5 Plots of La/Yb vs ΣREEs for the surface sediments in Jiulong River. The data were cited from Yan et al. (2012)

Compared with the Sr and Nd isotopic compositions in the sediments of major rivers over the world, the ${}^{87}Sr/{}^{86}Sr$ ratios and the $\varepsilon_{Nd(0)}$ values in the surface sediments in Jiulong River fell within the range of major rivers over the world (0.70435~0.80016 and − 42.6~+ 7.1, respectively) (Goldstein and Jacobsen 1988). The mean $\varepsilon_{Nd(0)}$ value in the surface sediments in Jiulong River (− 10.71) was higher than the mean upper continental crust (UCC) value (− 17) (Goldstein and Jacobsen 1988) and similar with the mean weathered crust value (− 11.4) (Goldstein et al. 1984). Compared with the Sr and Nd isotopic compositions in sediments of the local rivers, the mean ${}^{87}Sr/{}^{86}Sr$ ratio in the surface sediments of Jiulong River (0.721930) was higher than those in the sediments of the Minjiang River (0.720171), the Qiantang River (0.720950), and the western Taiwan island rivers (0.718804), while lower than the Yangtse River (0.722479) and the Oujiang River (0.723757) (Mi et al. 2017). The mean $\varepsilon_{Nd(0)}$ value in the surface sediments in Jiulong River (− 10.71) was higher than those in the sediments of the Minjiang River (− 12.3), the Qiantang River (− 11.9), the western Taiwan island rivers (− 12.4), the Yangtse River (− 11.6), and the Oujiang River (− 11.9) (Mi et al. 2017).

In order to probe the provenance compositions of the sediments, $\varepsilon_{Nd(0)}$ vs ${}^{87}Sr/{}^{86}Sr$ of the surface sediments and some potential sources are plotted in Fig. 6. The values of εNd(0) and ${}^{87}Sr/{}^{86}Sr$ in the surface sediments of Jiulong River (− 14.88~− 7.16 and 0.714091~0.733476, respectively) were relatively similar with the values of Fujian granite (− 16.37~− 4.00 and 0.706390~0.731931, respectively) and Western Fujian magmatic rock (− 13.33~− 10.11 and 0.713340~0.741210, respectively) (Huang et al. 1986; Ling and Shen 1999), indicating that the sediments in Jiulong River were mainly derived from local (specially upstream) granite and magmatic rocks with a result of natural geological processes.

3.5 Anthropogenic influence on REEs in the sediments

According to the above discussion, the sediments of Jiulong River were mainly derived from local granite and magmatic rocks with a result of natural geological processes. However, the total REE concentrations of Fujian granites (162.04 mg kg^{-1}) (Huang and Lin 2017) were lower than those in the sediments, indicating that the REEs in the sediments might also be affected by anthropogenic activities which contributed higher REE concentrations.

Table 4 Sr and Nd isotopic compositions in surface sediments of Jiulong River

	Sample	$^{87}Sr/^{86}Sr$	$^{143}Nd/^{144}Nd$	$\varepsilon_{Nd(0)}$		Sample	$^{87}Sr/^{86}Sr$	$^{143}Nd/^{144}Nd$	$\varepsilon_{Nd(0)}$
North River	1	0.721421	0.511905	− 14.3	Estuary	33	0.719973	0.512099	− 10.51
	2	0.7219	0.512118	− 10.14		34	0.718844	0.512033	− 11.8
	4	0.721071	0.512218	− 8.19		35	0.714091	0.512271	− 7.16
	5	0.720728	0.512061	− 11.26		37	0.7189	0.512131	− 9.89
	6	0.730552	0.511978	− 12.87		42	0.7166	0.512143	− 9.66
	8	0.725406	0.511968	− 13.07		43	0.7163	0.512095	− 10.59
	9	0.722626	0.512069	− 11.1		44	0.720953	0.512129	− 9.93
	12	0.722382	0.512128	− 9.95		45	0.717687	0.512073	− 11.02
	13	0.720989	0.512088	− 10.73		46	0.720669	0.51211	− 10.3
West River	14	0.721603	0.512067	− 11.14		48	0.7176	0.51226	− 7.37
	15	0.721475	0.512112	− 10.26		51	0.72001	0.512181	− 8.91
Coastal wetland	18	0.732106	0.511875	− 14.88		52	0.718415	0.512105	− 10.4
	21	0.722523	0.512204	− 8.47		53	0.719992	0.512141	− 9.69
	24	0.724917	0.512074	− 11	Max		0.733476	0.512271	− 7.16
	27	0.733476	0.512017	− 12.11	Min		0.714091	0.511875	− 14.88
	30	0.732769	0.511926	− 13.89	Mean		0.72193	0.512089	− 10.71

It is reported that the correlation coefficient between isotopic composition and the concentration can be used to identify the characteristics of the sources (N'Guessan et al. 2009; Lin et al. 2016). The plot of $\varepsilon_{Nd\ (0)}$ vs 1/[Nd] is illustrated in Fig. 7. It was found that $\varepsilon_{Nd\ (0)}$ was not well related to 1/[Nd] with the square of correlation coefficient of 0.0017. This result indicated that the sediments in Jiulong River were controlled by more than two sources with different isotopic compositions (Farmer et al. 2006).

In this study, the correlation coefficient between δCe and δEu was 0.0608, δCe, and ΣREEs was − 0.294, and δEu and ΣREEs were 0.0191, suggesting the weak relationship between REE parameters (δCe, δEu, and ΣREEs) and the REE characteristics can be used to determine sediment provenance since the sediments retained the REE characteristics of their source rocks when correlation coefficients were low (Shields and Stille 2001).

Considering that the isotopic compositions might be affected by the processes of weathering, erosion, and transportation (Goldstein and Jacobsen 1987), the combination of $\varepsilon_{Nd\ (0)}$ and δEu was taken for further analysis (Mclennan 1989; Wei et al. 2012). As shown in Fig. 8, the values of $\varepsilon_{Nd(0)}$ and δEu in the surface sediments of Jiulong River (− 14.88~− 7.16 and 0.44~0.66, respectively) were relatively similar with the values of coal ash (− 14.01~− 9.52 and 0.51~0.63, respectively), industrial sludge (− 11.59~− 7.84 and 0.47~0.55, respectively) and Fujian Pb-Zn deposit (− 13.15~− 16.72 and 0.44~0.56, respectively), while clearly different from those

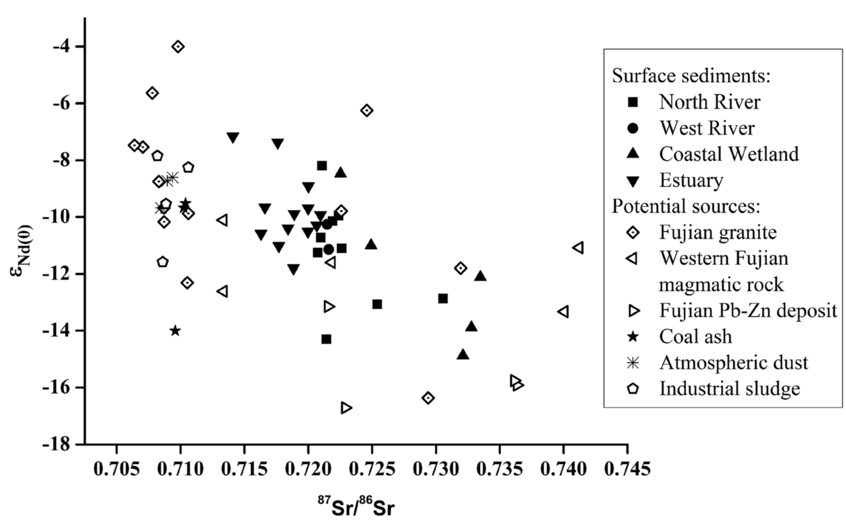

Fig. 6 Plots of $\varepsilon_{Nd(0)}$ vs $^{87}Sr/^{86}Sr$ in the surface sediments of Jiulong River and in some potential sources. The data of surface sediments, Fujian Pb-Zn deposit, coal ash, atmospheric dust, and industrial sludge were analyzed in this study; the data of Fujian granite are from Ling and Shen (1999), and the data of Western Fujian magmatic rock are from Huang et al. (1986)

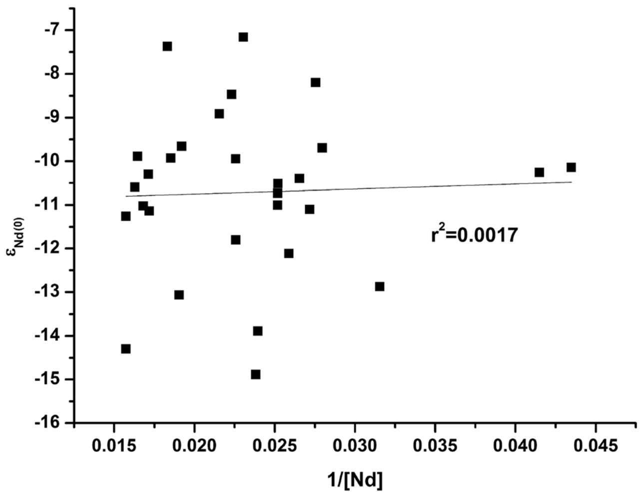

Fig. 7 Plot of $\varepsilon_{Nd(0)}$ vs 1/[Nd] in the surface sediments of Jiulong River

of atmospheric dust (− 9.67~− 8.60 and 1.09~2.06). In addition, the concentrations of REEs in the surface sediments of Jiulong River were all higher than those in Fujian soil (Chen et al. 1992). Furthermore, the distribution patterns of REEs in the surface sediments were similar to those of Fujian Pb-Zn deposit, coal ash, and industrial sludge. These results indicate that the REEs in the sediments of Jiulong River might be also influenced by anthropogenic activities such as Fujian Pb-Zn deposit, coal combustion, and industrial sludge.

4 Conclusions

The REE concentrations in surface sediments of Jiulong River were analyzed with the aim to investigate the geochemical characteristics of REEs, to probe the provenance compositions of the sediments, and to analyze the potential anthropogenic influence on the REEs in the sediments. Concentrations of REEs in the surface sediments of Jiulong River were all higher than those in Fujian soil, and the REEs showed a generalized sequence of Ce > La > Nd > Pr > Sm > Gd > Dy>Er > Yb > Eu > Ho > Tb > Lu > Tm, meeting the Oddo-Harkins rule. The mean ΣREEs in the surface sediments of Jiulong River (254.25 mg kg^{-1}) were higher than that in Fujian soil (198.37 mg kg^{-1}), while lower than deep-sea sediments (411 mg kg^{-1}). The mean values of ΣLREEs (227.6 mg kg^{-1}), ΣHREEs (26.64 mg kg^{-1}), and $(La/Yb)_N$ ratios (9.24) suggested an enrichment of LREEs compared to HREEs. Negative Eu anomalies were observed in the surface sediments. The distribution patterns of REEs in the surface sediments from different areas of Jiulong River were remarkably similar, which suggested that all the REEs in the surface sediments were of similar origin. The REE distribution patterns were mainly the result of natural geological processes. The plots of La-Th-Sc and La/Yb-∑REEs suggested that the surface sediments were mainly derived from local granite and with minor contributions from basic rocks. The values of $^{87}Sr/^{86}Sr$, $^{143}Nd/^{144}Nd$, and $\varepsilon_{Nd(0)}$ were 0.714091~0.733476, 0.511875~0.512271, and − 14.88~− 7.16, respectively, and these values were similar with the range of major rivers over the world. The plots of $\varepsilon_{Nd(0)}$ vs $^{87}Sr/^{86}Sr$ in the surface sediments and some potential sources indicated that the sediments in Jiulong River were mainly derived from granite and magmatic rocks as a result of natural geological processes. The results of the enrichment factor indicated that all the REEs were slightly enriched (1 ≤ EF < 2). The plots of $\varepsilon_{Nd(0)}$ vs 1/[Nd] and $\varepsilon_{Nd(0)}$ vs δEu indicated that the REEs in the sediments of Jiulong River might be also influenced by anthropogenic activities such as Fujian Pb-Zn deposit, coal combustion, and industrial sludge. In conclusion, the REEs in sediments of Jiulong River were mainly derived from natural geological processes (granite and magmatic rocks) and were influenced by anthropogenic activities (Fujian Pb-Zn deposit, coal ash, and industrial sludge).

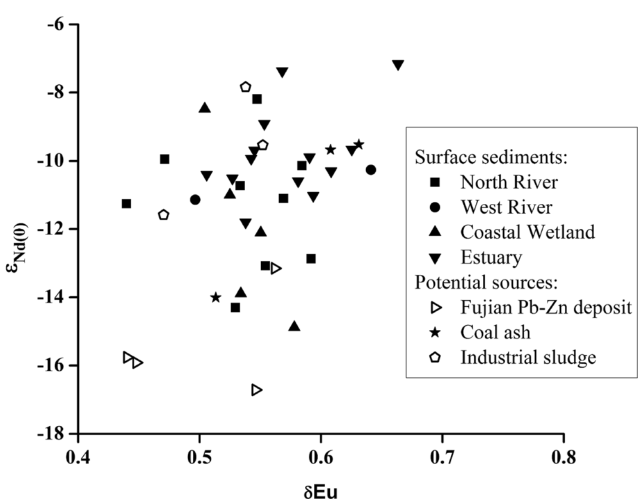

Fig. 8 Plots of $\varepsilon_{Nd(0)}$ vs δEu in the surface sediments of Jiulong River and in some potential sources

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