

1 **Rare earth elements in oysters and mussels collected from the**
2 **Chinese coast: Bioaccumulation and human health risks**

3 Puhui Zhao ^{a,b}, Ran Bi ^{a,b,*} Edmond Sanganyado ^{a,c}, Xiangfeng Zeng ^d, Weiwen Li ^e,
4 Zhendong Lyu ^a, Jinyan Liu ^a, Ping Li ^{a,b}, Hong Du ^{a,b}, Wenhua Liu ^{a,b}, Yongfeng Jia ^d

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6 *^a Institute of Marine Sciences and Guangdong Provincial Key Laboratory of Marine*
7 *Disaster Prediction and Prevention, Shantou University, Shantou 515063, China*

8 *^b Southern Marine Science and Engineering Guangdong Laboratory (Guangzhou),*
9 *Guangzhou 511458, China*

10 *^c Northumbria University, Newcastle upon Tyne, NE1 8ST, United Kingdom*

11 *^d Key Laboratory of Pollution Ecology and Environmental Engineering, Institute of*
12 *Applied Ecology, Chinese Academy of Sciences, Shenyang 110016, China*

13 *^e Third Institute of Oceanography, Ministry of Natural Resources, Xiamen 361005,*
14 *China*

15 * Corresponding author: rbi@stu.edu.cn

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17 **Rare earth elements in oysters and mussels collected from the**
18 **Chinese coast: Bioaccumulation and human health risks**

19 **Abstract**

20 Rare earth elements (REEs) are increasingly used in various industries worldwide,
21 resulting in their release into aquatic ecosystems. We evaluated the distribution and
22 bioaccumulation of 14 REEs in marine sediments and biota (oysters and mussels) along
23 the Chinese coasts. The total concentration of REEs (Σ REEs) in sediment samples was
24 41.65-170.94 mg/kg, where Ce concentration was the highest and Tm and Lu
25 concentrations the lowest. The concentration of total light REEs (Σ LREEs) was higher
26 than the concentration of total heavy REEs (Σ HREEs) at all study sites. The
27 concentrations of Σ REEs were 1.97–4.77 mg/kg and 0.62–4.96 mg/kg dry mass (DM)
28 for oysters and mussels, respectively. The bioaccumulation of Σ LREEs was higher than
29 Σ HREEs in oysters and mussels. The bioaccumulation factor (BAF) for Σ REE was
30 0.34–1.49 and 0.25–1.10 for oysters and mussels, respectively, where the BAF was
31 relatively higher in species collected from the south than the north. A positive
32 relationship of REEs was found in bivalves, with a significantly higher correlation of
33 HREEs than LREEs. The correlation between sediment and biotas was higher in
34 mussels than in oysters, showing a good potential for being environmental indicators
35 for REEs. The risk of REEs to humans via bivalve consumption could be negligible
36 based on the estimated daily intake of REEs in oysters and mussels.

37

38 **Keywords:** Rare earth elements, Oysters, Mussels, Bioaccumulation, Health risk

39 **1. Introduction**

40 Rare earth elements (REEs) are emerging pollutants comprising lanthanide series
41 elements (La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu), yttrium
42 (Y), and scandium (Sc). They are used extensively in high technology areas, such as

43 electronics (e.g., luminescent material of display), manufacturing (e.g., metal alloys),
44 medicine (e.g., magnetic resonance imaging), clean energy (e.g., rechargeable batteries
45 in hybrid cars), and agriculture (e.g., fertilizer) (Balaram, 2019). China has the largest
46 reserve of rare earth element resources and the largest production chains in the world.
47 Low-weight REE resources are mainly distributed in the Bayan Obo mining area in
48 Baotou, Inner Mongolia, and their rare earth element reserves account for more than
49 83% of the country's total rare earth reserves. Compared with the northern region, the
50 southern region is rich in high-weight REE resources (Ma et al., 2019).

51

52 The extensive industrial application of REEs has led to their release into aquatic and
53 terrestrial ecosystems (Gu et al., 2020; Wang et al., 2022). Wang et al. (2022) measured
54 the total concentrations of REEs (Σ REEs) that ranged from 1.02 to 178.55 $\mu\text{g}/\text{kg}$ in 14
55 marine wild fish species from the northern coastal region of the South China Sea; a
56 mean Σ REE of 0.35 mg kg^{-1} was reported in shellfish from the southern South China
57 Sea, and a mean value of 0.12 mg kg^{-1} was reported in zooplankton from northwestern
58 Italy (Li et al., 2016; Squadrone et al., 2019). However, REEs have been shown to
59 reduce growth and nutritional quality and impair the metabolic functions of plants
60 (Carpenter et al., 2015); produce genotoxicity and neurotoxicity for biotas (Blinova et
61 al., 2018; Trifuoggi et al., 2017); bioaccumulate across food chains, are chronically and
62 acutely toxic to soil organisms (Gardon et al., 2018); and cause nephrogenic systemic
63 fibrosis, dysfunctional neurological disorder, fibrotic tissue injury, and male sterility in
64 humans (Prince et al., 2008; Thomsen, 2017).

65

66 Marine filtering species, such as oysters and mussels, are exposed to various
67 contaminants, including REEs, hence their wide use in the biomonitoring of
68 environmental contaminants (Briant et al., 2021; Schaefer et al., 2022). As filter-feeding
69 animals and primary consumers, oysters and mussels constitute major components of
70 coastal trophic networks and ecosystem functioning (Briant et al., 2018). They can
71 accumulate pollutants in their tissues at elevated levels related to pollutant availability
72 in the marine environment (Beyer et al., 2017). On the other hand, oysters and mussels

73 have high protein and amino acid contents consumed by humans worldwide (Venugopal
74 and Gopakumar, 2017). Therefore, the accumulation of REEs in oysters and mussels
75 poses a health risk to humans *via* dietary intake (Adeel et al., 2019). Studies on the
76 accumulation of REEs in oysters and mussels and the corresponding human health risks
77 posed by their consumption remain unclear. Therefore, this study was designed to (i)
78 determine the REE concentrations in oysters and mussels along the Chinese coastline
79 and determine the bioaccumulation factor of REEs from sediment to biological tissues;
80 (ii) evaluate the possibility of using oysters and mussels as an indicator for REE
81 concentrations in the local environment; and (iii) assess the possible human health risk
82 of REEs via oyster and mussel intake.

83 **2. Materials and Methods**

84 *2.1. Study site and sample collection*

85 Sediment and bivalve samples were collected at six sites from four provinces (Liaoning,
86 Jiangsu, Zhejiang and Guangdong) along the Chinese coastline between March and
87 May 2021 (Fig. 1). Surface sediment samples were collected using a grab sampler, at
88 about top 10 cm depth, the sediment was homogenized and divided into three portions
89 for chemical analysis. The bivalve samples were collected with the help from local
90 fishermen with fishing cages. Bivalves sample with similar sizes were chosen for the
91 experiment, about five individuals were used for one mixed sample, and three mixed
92 samples were used for chemical analysis. All collected samples were kept in clean
93 polyethylene bags, sealed, and transported to the laboratory with ice. They were stored
94 in the refrigerator at -20 °C in the laboratory.



95

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Fig. 1. Study area and location of the sampling sites of the samples.

97

2.2. Chemicals and reagents

98

All reagents were purchased from Sigma Aldrich, Fisher Chemical, and Merck.

99

Solutions were prepared in deionized water (resistance >18 MΩ·cm) using a Milli-Q

100

water purification system (Millipore). The rare earth element mix for ICP (Trace

101

CERT®, Sigma–Aldrich, Switzerland) was used as a calibration standard for total

102

elemental analysis. Nitric acid (65% HNO₃, Trace Metal Grade, Fisher Chemical,

103

Canada), hydrogen peroxide (30% H₂O₂, Merck, Germany), hydrochloric acid (37%

104 HCl, Merck), and hydrofluoric acid (30% HF, Merck, Germany) were used for sample
105 treatments.

106

107 *2.3. Sample preparation and determination*

108 Each sample was washed with tap water and rinsed with ultrapure water. The edible
109 tissue was harvested and freeze-dried for 96 hours. After freeze-drying, samples were
110 ground to powder using a mortar and ball mill. Approximately 0.1 ± 0.01 g dry weight
111 (dw) oyster/mussel powdery samples were digested with 3 ml HNO₃ and 1 ml H₂O₂
112 mixture in a Teflon® TFM-lined digestion vial using Mars Xpress™ (CEM, USA).
113 The temperature programmed in Mars Xpress was set to 20 min ramps to 190 °C and
114 held for 20 min at 190 °C (Lin et al., 2021). Sediment samples were digested in 3 ml
115 HNO₃, 1 ml HCl and 1 ml HF mixture in the digestion vessel using Mars Xpress™
116 (CEM, USA). The temperature programmed in Mars Xpress was set to 20 min ramps
117 to 210 °C and held for 20 min at 210 °C (Zhao et al., 2022). All the digested samples
118 were filled to 50 ml in polyethylene centrifugation tubes with ultrapure water. The
119 concentrations of REEs in sediments, oysters, and mussels were determined using ICP–
120 MS/MS (iCAP TQ, Thermo Fisher, Germany) using a standard calibration curve and
121 internal standards (10 ppb) Sc, In, Rh, and Ru for mass correction. The KED and O₂
122 modes were used for element analysis in the ICP–MS/MS, and the detailed operating
123 and reaction modes are shown in the supplemental information (Table S1). All sediment,
124 oyster, and mussel samples were run in batches with standards inserted every ten
125 samples, method blanks, and certified reference materials. Analysis precision and
126 accuracy were ensured using the National Center for Material Standards, near-shore
127 marine sediment certified reference material (GBW 07314), GBW10024 (GSB-15,
128 Scallop) from the Institute of geophysical and geochemical exploration, Chinese
129 Academy of Geological Sciences (IGGE). The analyzed results were within $\pm 5\%$ of the
130 certified value.

131

132 *2.4. Accumulation of REEs in oysters and mussels*

133 The REEs in sediments to bioaccumulate in oysters and mussels were determined using

134 the bioaccumulation factor (BAF), which is the ratio of the concentration of REEs in
135 oyster or mussel tissue to the concentration of the REEs in sediments assuming that the
136 organism and the sediment are in equilibrium (Mackay et al., 2018).

$$137 \quad BAF = \frac{C_{oyster \text{ or } mussel}}{C_{sediment}}$$

138

139 *2.5 Human dietary risk*

140 The EDI of REEs through fish consumption was calculated using the following
141 equation (Xu et al., 2020):

$$142 \quad EDI = \frac{C \times IR}{BW}$$

143 where C is the concentration of REEs in oysters or mussels expressed as wet weight
144 (mg/kg, ww) and IR is the average daily ingestion rate (g/day) of oysters or mussels.
145 The IR was set at 25 g/day for children and 50 g/day for adults based on the
146 questionnaire survey for traditional residents near Jiaozhou Bay, China (Zhang and
147 Zhang, 2015). BW is the average human body weight (kg), which is considered to be
148 33 kg for children and 63 kg for adults (Xu et al., 2020).

149 *2.6 QA/QC of rare earth elements determination*

150 All the sample was prepared in the clean chamber in the inorganic chemistry lab, the
151 certified reference material (CRM) for sediment and biological tissues were used for
152 ICP-MS/MS determination verification. The detected CRM values were within $\pm 5\%$ of
153 the certified value for the REEs. The LOD for the rare earth elements ranged 1.0×10^{-5}
154 to 1.0×10^{-4} $\mu\text{g/L}$, and LOQ was calculated as three times concentration of LOD. All the
155 detected REEs concentrations were above the minimum limits of quantification.

156 *2.7 Statistical analysis*

157 One-way ANOVA (analysis of variance) and nonparametric tests were performed to
158 find significant variation between oysters and mussels at different sites using SPSS 25.0.
159 Prior to analysis, all data were checked for normality using the Shapiro–Wilk test.
160 Nonparametric tests such as Kruskal–Wallis were used for the data displaying
161 nonnormality. A significance level of $p < 0.05$ was accepted for all statistical analyses.

162 **3. Results and Discussion**

163 *3.1. Concentrations of REEs in sediments*

164 The concentrations of REEs in sediment samples collected at different study sites along
165 the Chinese coastline are shown in Table 1. The total concentration of REEs (Σ REEs)
166 in sediment samples was 41.65-170.94 mg/kg. The concentration of Ce (15.02 ~ 80.59
167 mg/kg) was the highest, while that of Tm (0.07 ~ 0.34 mg/kg) and Lu (0.07 ~ 0.35
168 mg/kg) was the lowest. The Σ LREEs were higher than the Σ HREEs at all the study sites
169 from the Bohai Sea to the South China Sea, while the Σ LREEs/ Σ HREEs ratio ranged
170 from 9.28 to 10.76, which indicated that LREEs were more easily enriched than HREEs
171 in sediments. The REE content of Jiangsu in the North Yellow Sea was lower than that
172 in other locations because the North Yellow Sea is dominated by Yellow River
173 discharges carrying loess with a low REE content in the sediment (Jiang et al., 2008).
174 The REE concentrations found in Liaoning and Zhejiang in the present study were
175 similar to those obtained in a previous study, suggesting that there have been no changes
176 in the discharge or removal of the REEs in this region (Mi et al. (2020)). The Σ REEs in
177 Guangdong in this study were higher than those in the southern South China Sea (2.62-
178 3.15 μ g/kg) due to the differences in anthropogenic activities Li et al. (2016). Moreover,
179 terrestrial weathering and anthropogenic activities will lead to a higher REE input to
180 the local environment, where the concentration of REEs in coastal wetland sediment in
181 the Vembanad estuary (29.55-229.67 mg/kg) was higher than that in our study (Manoj
182 et al., 2016). Alhassan and Aljahdali (2021) found that the total concentration of REEs
183 in sediment on the Red Sea coast was 12.2-108.98 mg/kg, which was lower than our
184 study due to an underdeveloped high-tech industry and fewer human activities in Africa.
185 Owing to rapid economic development, high-density industrial distribution, and
186 abundant anthropogenic activities along the Chinese coastline, we are concerned that
187 excessive contaminants in the sediments may elevate the bioaccumulation of such
188 contaminants in the associated biota.

189

190 Table 1 Concentration of REEs in the sediments (In this study, n=3).

REEs	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	L/H	Σ REEs
	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg		mg/kg
This study																
LN	23.36	70.77	7.97	24.95	5.05	0.92	4.19	0.58	3.41	0.67	1.91	0.27	1.47	0.28	10.41	145.81
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±
JS	2.61	7.92	0.89	2.79	0.56	0.10	0.46	0.06	0.38	0.07	0.21	0.03	0.16	0.03	1.16	16.33
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±
ZJ 1	13.66	15.02	1.75	5.90	1.25	0.44	1.04	0.14	0.84	0.16	0.47	0.07	0.77	0.07	10.55	41.65
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±
ZJ 2	1.57	1.72	0.20	0.67	0.14	0.05	0.12	0.01	0.09	0.02	0.05	0.01	0.08	0.01	1.21	4.79
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±
GD 1	25.67	52.45	6.08	20.32	4.19	0.87	3.37	0.46	2.61	0.51	1.46	0.21	1.31	0.21	10.76	119.79
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±
GD 1	2.90	5.92	0.68	2.29	0.47	0.09	0.38	0.05	0.29	0.05	0.16	0.02	0.14	0.02	1.21	13.53
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±
GD 1	4.12	46.08	5.34	17.93	3.76	0.13	3.01	0.43	2.42	0.48	1.35	0.19	0.25	0.19	9.28	85.72
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±
GD 1	0.37	4.14	0.48	1.61	0.33	0.01	0.27	0.03	0.21	0.04	0.12	0.01	0.02	0.01	0.83	7.71
	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±
GD 1	28.08	80.59	8.99	30.57	6.37	0.97	4.94	0.72	4.22	0.80	2.19	0.34	1.75	0.35	10.14	170.94

	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±
	3.37	9.67	1.08	3.66	0.76	0.11	0.59	0.08	0.51	0.09	0.26	0.04	0.21	0.04	1.21	20.51
	17.35	67.12	7.55	25.11	5.29	0.57	4.26	0.61	3.53	0.69	1.99	0.29	1.08	0.30	9.61	135.80
GD 2	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±	±
	1.94	7.51	0.84	2.81	0.59	0.06	0.47	0.06	0.39	0.07	0.22	0.03	0.12	0.03	1.07	15.21
Other studies																
The Southern South China Sea	0.73	0.62	0.13	0.60	0.14	0.03	0.17	0.03	0.16	0.03	0.10	0.01	0.08	0.01	3.72	2.88
	2.26	5.45	0.66	2.84	0.61	0.17	0.58	0.09	0.52	0.10	0.25	0.04	0.26	0.03	5.60	13.85
The Central Red Sea	~	~	~	~	~	~	~	~	~	~	~	~	~	~	~	~
	16.54	37.36	4.73	21.60	4.65	1.34	4.45	0.74	4.09	0.79	2.28	0.37	2.25	0.36	7.13	101.53
	6.25	13.33	1.46	5.41	0.91	0.22	0.72	0.09	0.50	0.08	0.26	0.03	0.26	0.03	11.08	29.55
The Vembanad estuary	~	~	~	~	~	~	~	~	~	~	~	~	~	~	~	~
	50.55	104.62	10.61	41.58	7.53	1.76	6.65	0.91	5.25	0.97	2.83	0.37	2.43	0.35	14.00	229.67

191 Note: LN: Liaoning, JS: Jiangsu, ZJ: Zhejiang, GD: Guangdong

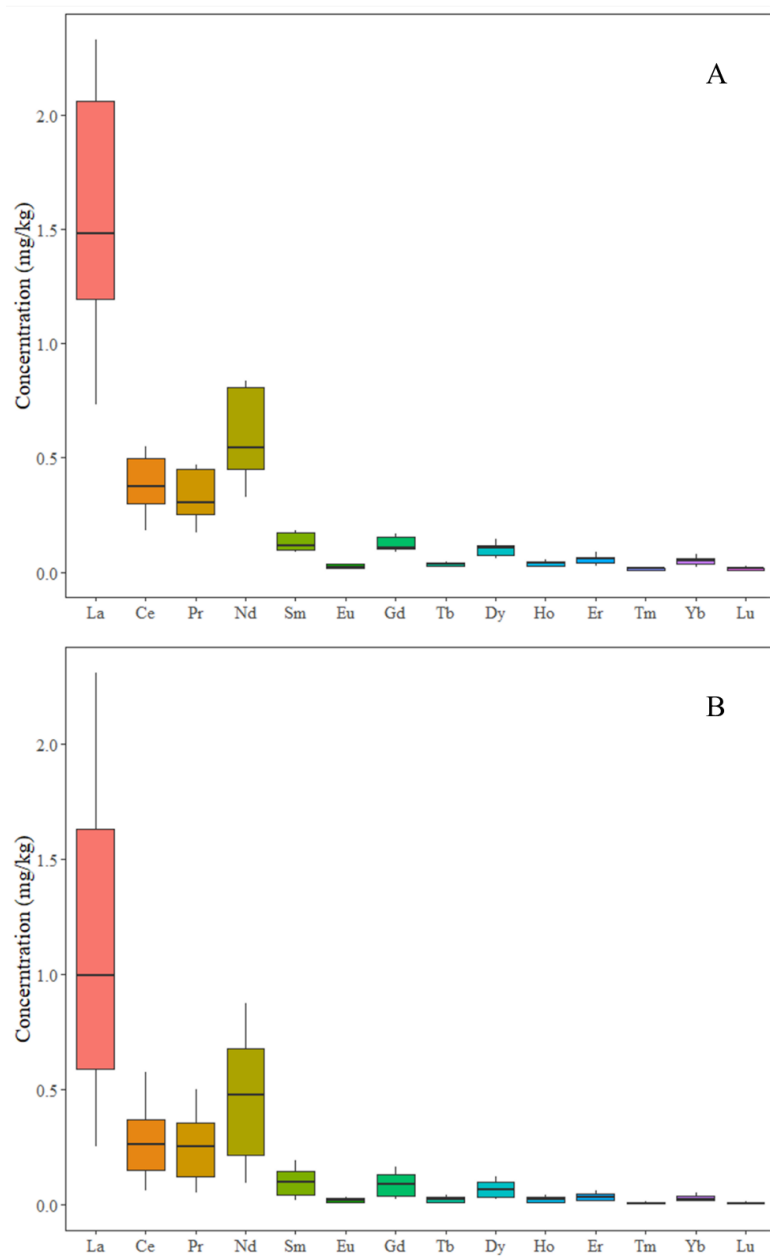
193 3.2. Concentrations of REEs in oysters and mussels

194 Marine filtering species have an abundant capacity to filter pollutants from the
195 surrounding seawater (Briant et al., 2021; Schaefer et al., 2022). In the present study,
196 the Σ REE concentrations were 1.97 to 4.77 mg/kg dry mass (DM) and 0.62 to 4.96
197 mg/kg DM for oysters and mussels, respectively (Fig 2, Table S2). A few studies
198 reported the REE concentrations in various marine biota, where the Σ REEs ranged from
199 0.16-9.10 mg/kg DM for oysters and 0.16-5.00 mg/kg for mussels in French
200 Metropolitan Coasts (Briant et al., 2021), Canadian Arctic (MacMillan et al., 2017),
201 Ligurian Sea, Italia (Squadrone et al., 2019), Zhuhai, China (Briant et al., 2021; Ma et
202 al., 2019). Habitats may have a great influence on the REE concentrations in oysters
203 and mussels. Oysters (*Crassostrea gigas*) collected in the Gironde River mouth estuary
204 had a higher concentration of Σ REEs (10.94 mg/kg) than Persuel Bay (0.29 mg/kg) in
205 French Metropolitan Coasts. The Σ REE concentration in mussels (*Mytilus edulis* and
206 *Mytilus galloprovincialis*) collected in the littoral zone was only 0.18-2.33 mg/kg from
207 the same region (Briant et al., 2021). For filtering species, oysters and mussels tend to
208 have higher capacities for accumulating pollutants in their tissues than fish and
209 crustaceans. Li et al. (2016) found concentrations of Σ REEs in fishes (0.004-0.045
210 mg/kg) and crustaceans (0.10-1.95 mg/kg) from the southern South China Sea. Wang
211 et al. (2022) reported that the average concentration of Σ REEs in 14 fish species from
212 the northern South China Sea was 0.003-0.093 mg/kg, which was significantly lower
213 than the REE concentrations in the filtering species.

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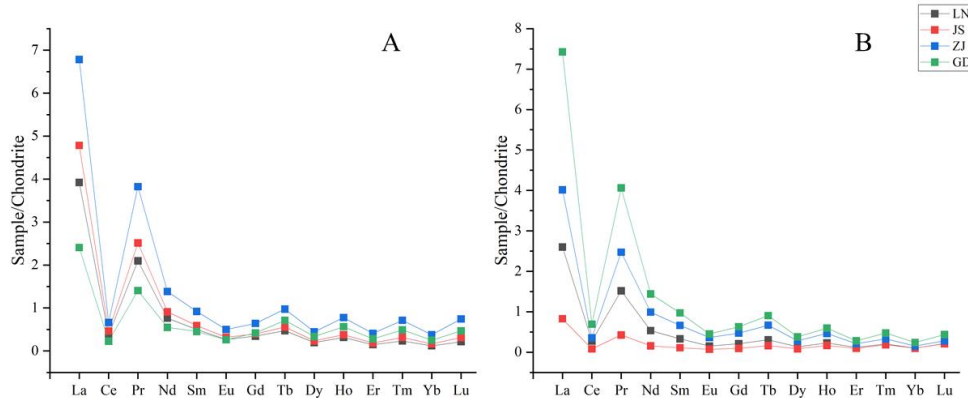
215 The chondrite-normalized patterns (REE_{CN}) of oysters and mussels from different study
216 sites were similar, featuring undulating shapes with a maximum at La and a minimum
217 at Ce (Fig. 3). The chondrite-normalized patterns (REE_{CN}) of oysters in the present
218 study were different from those of French *Crassostrea gigas* characterized by a constant
219 increase from La to Lu with slight Ce depletion (Briant et al., 2021). We implied that
220 the difference was attributed to the seawater environment and differences in REE
221 availability between sites because this pattern was similar to the chondrite-normalized

222 plot in oysters in the Pearl River Estuary except Ce (Ma et al., 2019). The REE_{CN} of
223 mussels in this study was different from that of mussels from the Peace Lagoon on the
224 north beach of Eilat (Benaltabet et al., 2021). Filtering organisms feed on suspended
225 particles and microalgae; thus, the REEs concentration in those species may correspond
226 to the surrounding environment. Although the REE concentrations varied among the
227 different study sites, the individual REEs showed a similar enrichment pattern.
228



229
230

Fig. 2. Concentration of REEs in the oysters (A) and mussels (B).



231

232 Fig. 3. Chondrite-normalized plots for the REEs in the soft tissue of oysters (A) and mussels (B).

233 The chondrite values are from (Boynton, 1984).

234 3.3. Bioaccumulation factors of REEs in oysters and mussels.

235 In general, the bioaccumulation factor (BAF) of Σ REEs was less than one, except the

236 BAF for the oysters (JS) and mussels (ZJ) were 1.49 and 1.10 (Fig. 4, Table S3). The

237 BAF of Σ REEs from sediment to oysters and mussels varied among the sampling sites.

238 The lowest BAF of Σ REEs for oyster and mussels was in Liaoning (LN) at the

239 temperate zone, whereas a trend of increased BAF was found in other sampling sites

240 with lower latitude, except the BAF for oyster in Guangdong (GD). The accumulation

241 characteristics were similar for both species, except a higher BAF of Tb was found,

242 possibly owing to the low Tb concentration in the sediment (Table 1, Table S3).

243

244 The total bioaccumulation factor of REEs was lower than one, except for oysters (JS)

245 and mussels (ZJ), which were 1.49 and 1.10, respectively. The bioaccumulation factor

246 of Σ REEs from sediment to oysters and mussels varied among the species and site

247 locations, where the total BAF was higher in the oysters than in mussels (Fig 4). The

248 total BAFs for the oysters and mussels ranged from 0.34 to 1.49 and 0.25 to 1.10,

249 respectively. The BAF was element specific and site specific. The total BAFs of LREEs

250 (LBAF) and the total BAFs of HREEs (HBAF) were higher in Jiangsu and lower in

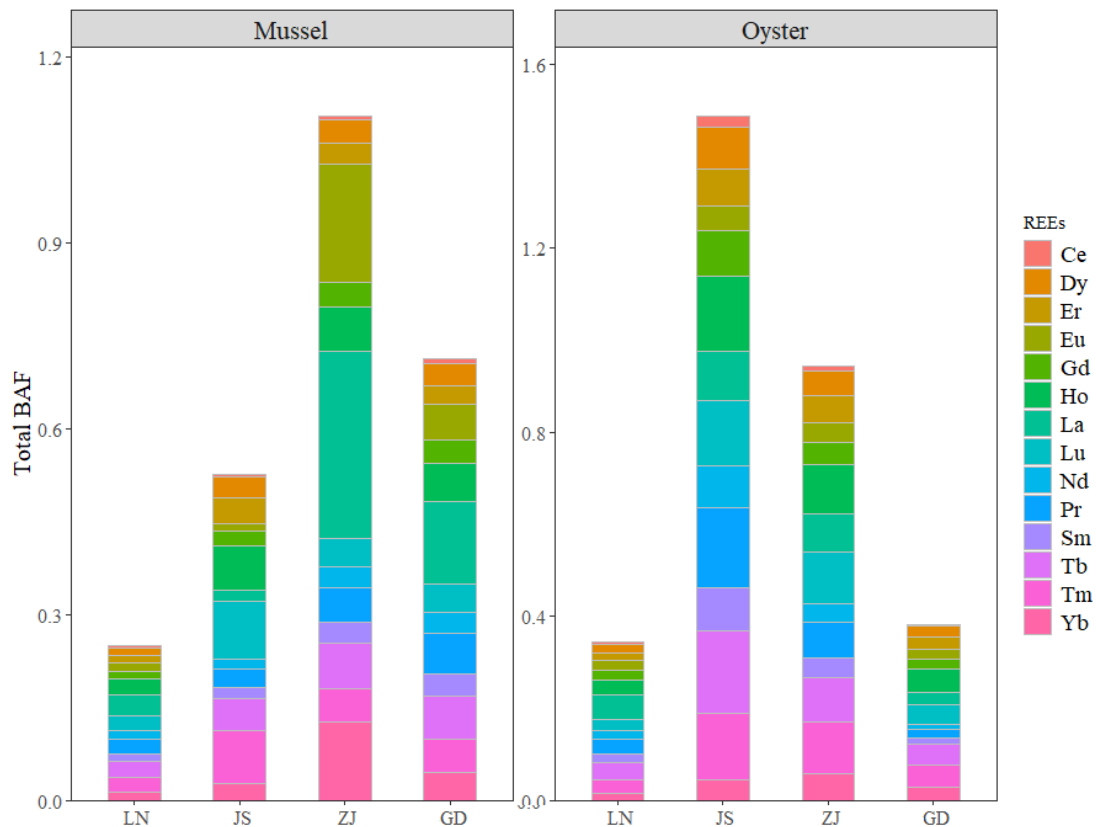
251 Guangdong in oysters. LBAF and HBAF were higher in Zhejiang and were lower in

252 Liaoning in mussels. LBAF was lower than HBAF in oysters and mussels, except

253 LBAF was similar to HBAF in Zhejiang in mussels. The accumulation characteristics
254 were different among species, where the Tb was highest in oysters and La was the
255 highest in mussels. Interestingly, a previous study showed that La was more toxic to
256 mussel embryos than other REEs (Mestre et al., 2019).

257 Marine biota, such as bivalves and fish, may constitute a pathway for human or animal
258 dietary exposure (Squadrone et al., 2019). REE bioaccumulation patterns appear to be
259 species- and tissue specific, and analysis of variance between taxa from the same
260 ecosystem showed that biota at the base of the food web (vegetation, invertebrates) had
261 significantly higher \sum REE concentrations than vertebrate muscle samples from the
262 same ecosystem (MacMillan et al., 2017). Previous studies on aquatic vertebrates have
263 shown that REEs are more concentrated in internal organs (liver, kidney, intestine, gills)
264 than in muscle (Amyot et al., 2017; Copetti et al., 2016). Amyot et al. (2017) found that
265 fish muscles are the edible parts consumed by human beings, and the bioaccumulation
266 of REEs is very low. Although relatively higher bioaccumulation of REEs was found
267 in bivalves, the potential bioaccumulation for REEs was limited (0.002-0.507) in
268 oysters and mussels in the present study.

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270

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Fig. 4. BAF of REEs in oysters and mussels.

272 3.4. Relationship of the REEs between biotas and sediments

273 REEs are usually divided into LREEs and HREEs for analysis because their atomic,
 274 physical, and chemical properties vary gradually along the series. In this study, we
 275 investigated the relationship of LREEs and HREEs between sediment and biotas (Fig
 276 5). The relationships presented a positive correlation, and the trend of HREEs showed
 277 a higher positive correlation. We deduced that this phenomenon was linked with the
 278 preferential scavenging of LREEs in waters and stronger complexation of HREEs in
 279 seawater (Deng et al., 2017; Elderfield and Greaves, 1982), so the filtering biotas could
 280 better absorb HREEs and present a more positive correlation with HREEs. In addition,
 281 we found the trend of mussels was more positive than oysters, and we thought it might
 282 be attributed to biological specific within the oysters and mussels.

283

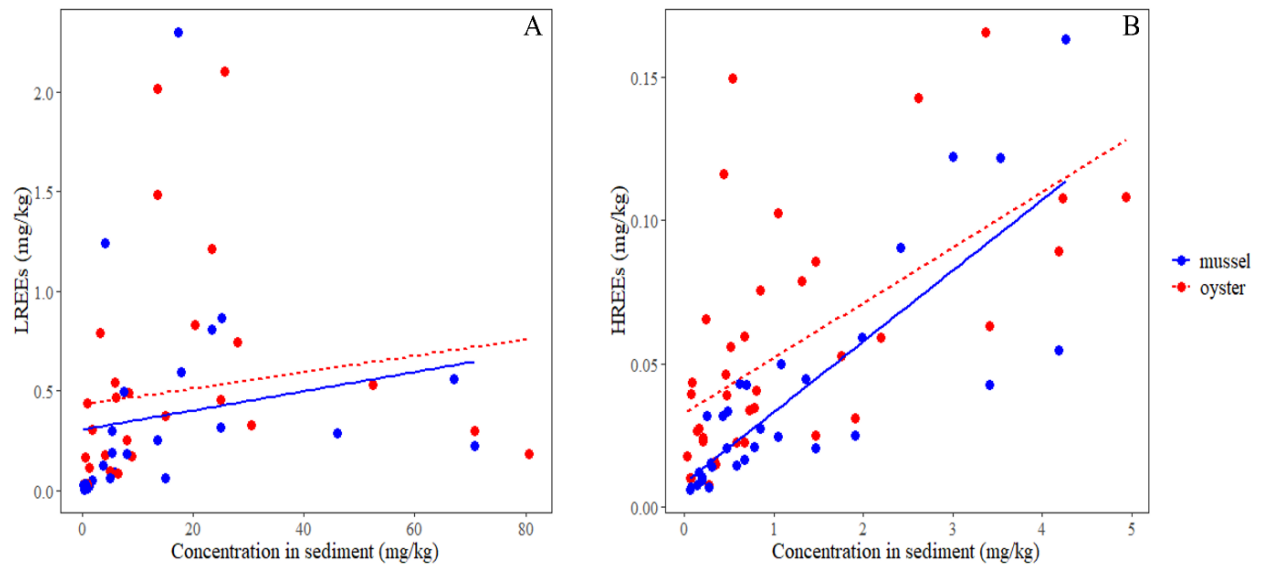


Fig. 5. Relationship of LREEs (A) and HREEs (B) between sediment and biotas.

3.5. Human dietary risk

REEs accumulate in marine sediments and biotas and ultimately enter the food chain. The risks, impacts, and chronic toxicity of widespread REE sediments are of concern due to their environmental persistence (Charalampides et al., 2016). Exposure to REEs may cause dysfunctional neurological disorders such as a reduced intelligence quotient (IQ) in children (Gwenzi et al., 2018). Bone alteration, genotoxicity, and fibrotic tissue injury were reported to be associated with several REEs (Chen and Zhu, 2008; Jenkins et al., 2011). Moreover, Marzec-Wroblewska (2015) showed anti-testicular effects and male sterility after REE exposure. The recommended threshold level of REEs is 70 $\mu\text{g}/\text{kg}/\text{day}$, based on an extensive human health survey in REE mining areas together with animal experiments (Zhu et al., 1997). In the present study, we tested the ΣREEs of oysters and mussels from different sites and calculated the EDI, which was divided between adults and children (Table 2). The EDIs of ΣREEs from oyster and mussel consumption were significantly lower than the recommended EDI threshold proposed by Zhu et al. (1997), indicating that the risk of REE exposure to humans via oyster and mussel consumption was negligible. However, populations living along the coast generally consume more seafood than those living inland, and the consumption of seafood may be significantly higher, thus leading to increased exposure to REEs (Wang

305 et al., 2022).

306

307 Table 2 Estimated daily intake (EDI) of Σ REEs through oyster and mussel consumption
308 by the general population of China.

		Σ REEs	IR (g/day)		BW (kg)		EDI (μ g/kg/day)	
			adults	children	adults	children	adults	children
Oyster	LN	0.654	50	25.1	63	33	0.52	0.50
	JS	0.793	50	25.1	63	33	0.63	0.60
	ZJ	1.193	50	25.1	63	33	0.95	0.91
	GD	0.493	50	25.1	63	33	0.39	0.37
Mussel	LN	0.450	50	25.1	63	33	0.36	0.34
	JS	0.155	50	25.1	63	33	0.12	0.12
	ZJ	0.740	50	25.1	63	33	0.59	0.56
	GD	1.239	50	25.1	63	33	0.98	0.94

309

310 Conclusion

311 By investigating the distribution of REEs in marine sediments and marine biota, this
312 study expands our understanding of the sources, bioaccumulation, and food safety risks
313 of REEs in marine ecosystems. It also provided critical data useful for assessing
314 effectiveness of mitigation efforts, comparing REE occurrences with other regions, and
315 evaluating human and environmental health effects in future. Sediment analysis
316 confirmed that LREEs are preferentially sequestered into marine sediments than
317 HREEs. Ce and Nd concentrations in sediment were an order or two orders of
318 magnitude higher than the other 11 REES. The Σ REEs in marine sediments in the
319 Chinese coasts were relatively higher than those reported in other regions probably due
320 to the extensive industrial development and urbanization in China. However, additional
321 studies are required to better understand the role of biogeochemical characteristics of
322 the sediments on the REEs sequestration. Our findings showed that while the

323 bioaccumulation of HREEs in mussels and oysters was positively correlated to marine
324 sediment concentrations, no significant relationship was observed for LREEs.
325 Determining the BAF of REEs in mussels and oysters revealed that REEs
326 bioaccumulation was element-, species-, and site-specific. For example, oysters and
327 mussels, generally, had higher HBAFs than LBAFs but HBAFs and LBAFs in oysters
328 from Jiangsu were higher than those from Guangdong. At elemental level, Tb had the
329 highest BAF in oysters while La had the highest in mussels. This is concerning because
330 La has been shown to be highly toxic to developing mussel embryos than the other
331 REEs. However, studies on the toxicity of REEs to different marine organisms remain
332 scarce. Although the EDIs of Σ REEs from oyster and mussel consumption were
333 significantly lower than the recommended EDI threshold value, populations frequently
334 consuming oysters and mussels may be exposed to substantially higher levels of REEs.
335 There is need for further investigation on the trophic transfer of REEs in various
336 organisms along the marine food web, and at varying temporal and spatial scales to
337 better understand their ecological and human health risks.

338

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