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Chapter

Investigations on Rare Earth Elements in Sediments from South Vellar Estuary along Bay of Bengal and Palk Strait, Tamil Nadu, India

Puliya kurichi Mookkan Velmurugan, Krishnan Vijayaprabhakaran, Periyathambi Thangappan Devika and Mohammad Suhail Meer

Abstract

The rare earth elements (REEs) in the sediments of the South Vellar estuary on the eastern coast of India were measured using thermal ionization mass spectrometry (TIMS). Lanthanides (La-Lu) and a number of other elements (Cr, Ni, Zn, Pb, Ag, Co, Cd, and V) were studied in bed sediment samples. The estuary’s total REE content (REE) ranges from 73.30 to 360.50 g/g. Continental weathering has a substantial impact on the distribution of REEs. Using factor analysis on the elemental data, two important groups of elements were identified: (a) LREEs, HREEs, Pb, Ag, and Cd, revealing an association with river detritals; and (b) Co, Ni, Zn, Cr, and V, indicating complex estuary processes. Because of the silicate weathering of crustal materials and the resultant increase in LREEs in detritals, LREEs are more enriched than HREEs. We get to the conclusion that 30% of HREE flux and 70% of LREE flux to the Bay of Bengal are provided by the estuarine system.

Keywords: Bay of Bengal, rare earth element, sediment, South Vellar Estuary, TIMS

1. Introduction

Rare earth elements (REEs) are frequently employed as geochemical tracers for provenance studies due to their distinct and chemically coherent behavior [1]. Extensive studies have been reported on the distribution of REE in sediments, suspended particulate matter, and natural waters. Our understanding of REE distribution in the sediments has advanced significantly. For instance, earlier research had found that “REEs as a group are insoluble elements, exhibiting limited fractionation throughout the earth surface processes.” The premise is that the REE are fractionated and mobilized during weathering in all climatic conditions in estuaries [2]. In addition, organic compounds, colloidal particles, and Fe-Mn oxyhydroxides may serve as hosts for REE and their presence in sediments may affect the total concentration of REE.
Rare earth elements and heavy metals have a well-established role in sediments as transporters and potential sources, and when these materials are distributed by geochemical processes in aquatic systems, they have an impact on surface ecosystems. At point inputs, the river mouths, sediments from the land, freshwater, and metals all enter the coastal zone. During transportation in river systems, fractionation of REEs and metals primarily takes place in the solid phase. The degree of REE fractionation in the silt and clay fractions of sediments is also controlled by the type of weathering, the lithology of the rocks, and their susceptibility to erosion [3]. Due to their extensive coverage of vast areas and contribution to a better understanding of the global REE cycle, a number of research investigations have concentrated on the distribution of REE in the sediments of big rivers. Large rivers drain through a variety of rock types under a range of climatic conditions and REEs from different sources are admixed during weathering, erosion, transportation, and deposition, resulting in homogenous REE patterns in large rivers around the world. As a result, it is challenging to identify the source rock characteristics from sediments of large rivers.

Several estuaries had high rates of REE removal to the sediments, but low rates of REE removal in low-salinity areas have also been reported [4]. In recent years, lanthanum to lutetium element groups, which are called rare earth elements, have received a lot of attention [5–8]. The distribution of rare earth elements by geochemical processes in aquatic systems has an impact on surface ecosystems, and sediments represent a well-established role as transports and potential sources of these materials. The coastal zone is impacted at points where metals, freshwater, sediments from the land, and river mouths enter [9, 10]. REE and metal fractionation primarily occur in the solid phase during transportation in river systems. As a result of the discharge of household and agricultural wastes, the intrusion of waste from the metal plating industry, the entry of organic and inorganic chemicals, the leaching of metals from solid waste, and the use of metal and metal components, concentrations of chromium (Cr), cobalt (Co), nickel (Ni), Zink (Zn), lead (Pb), cadmium (Cd), silver (Ag), and vanadium (V) are significantly increasing in coastal environment [11, 12].

The coastal zone eventually receives all of the elements derived from river systems. Additionally, estuaries have a history of regulating the quantity of REEs that enter the coastal ecosystem and eventually the deep ocean [13]. Understanding low-temperature geochemical processes consequently requires an understanding of the distribution and concentration of REEs in estuaries [14]. Estuaries are significant from an ecological and economic perspective since they are the places where the land meets the sea. The marine and freshwater inflows contribute a significant amount of nutrients to the estuary’s water column and sediment [15, 16]. The elemental cycles play a significant role in estuarine environments. Estuaries are important to the world’s biogeochemical cycles, including the carbon cycle, nitrogen cycle, and nutrient cycles, although taking up less than 10% of the ocean’s surface [17, 18]. Insight into the marine cycling of REEs is provided by the quantity of these elements in estuarine sediments, precisely as it is by their abundance in the continental crust [19]. Additionally, it gives more knowledge on the interactions between sediment and water and redox processes that occur in surface environments. The quantity and distribution of REEs in South Vellar estuarine sediments are presented in Table 1, the two classes of REE are light rare earth elements (LREEs) from lanthanum to gadolinium as well as from yttrium to lutetium are the heavy rare earth elements (HREEs).

As compared to HREEs, LREEs are generally richer in all estuarine sediments [20]. The deltaic and estuarine regions of India are not widely recognized. An earlier study [21] looked briefly at the distribution of heavy metals in the Gangetic delta.
The chemical characteristics of suspended sediments in the estuaries of the Narmada, Tapti, Mahanadi, and Godavari rivers were originally studied by Borole et al. in 1982 [22]. Seralathan and Seetharamaswamy (1987) reported on the clay mineralogy and
trace element chemistry of the Cauvery delta based on a single-time sampling (monsoon season) [23]. Later, Ramanathan et al. (1993) investigated the Cauvery delta’s geochemical properties [24]. The majority of these writers’ publications focus on the elemental distribution in these estuaries, while earlier reports did not mention trace metals like V, Ag, Cd, and Co. On the subject of REE distribution in Indian estuaries and deltaic regions, hardly any research has been published. The South Vellar estuary baseline study is comprised of this study (Figure 1). We are providing the distribution of REE in South Vellar estuarine sediments along India’s east coast for the first time, along with its association with heavy metal enrichment.

2. Methodology

2.1 Study area and geologic context

The Palk Strait, a Bay of Bengal strait, is between southern India and northern Sri Lanka. Its boundary is bounded by the Gulf of Mannar, Pamban Island, Adam’s (Rama’s) Bridge, and Mannar Island. The southwest part of the strait is also known as Palk Bay [25]. The strait is less than 100 m deep, 64 to 135 km wide, and 137 km long. The South Vellar River, a non-perennial river, flows into the Bay of Bengal along with Palk Strait. The river originates as a cluster of small streams in the Kumirakatti forest reserve, which is located about 9 km west of the village of Karumalai after passing through Tovarankurichchi and Karumalai, flowing to the southeast after receiving the additional flow from the Vembar retention basin, which is located south of Kodumbalur. After passing through Avudaiyarkoil, it turns to the east and flows until Mumpalai village in the Manamelkudi taluk, where it creates an estuary and discharges into the sea northeast of Manamelkudi.

There are three main lithostratigraphic units in Tamil Nadu’s southern region, which contains the coastal areas to be visited. Charnockites, granulitic retrograded products, and basic intrusive and Tertiary calcareous sandstone are layered above Precambrian granitic rocks, as are recent coastal deposits. During the Proterozoic, the granulitic terrain was divided into multiple massive shear zones, which caused the granulitic facies assemblages to retrograde. Based on changes in landforms and the presence of lineaments, seven major blocks have been discovered between Kanyakumari and Koidiyakarai on Tamil Nadu’s southeast coast [26]. According to a theory, the seven blocks from the glaring weaknesses in the lineaments are what keep striking slip faults from continuing. In the shallow marine shelf areas along the Tamil Nadu coast, the mean annual rate of sediment deposition in the Gulf of Mannar, Palk Bay, and Sand Heads are 0.01 m, 0.006 m, and 0.003 m, respectively, according to a research by Chandramohan et al. [27]. There are numerous examples of erosional and depositional landforms along Tamil Nadu’s southeast coast. Beach ridges, spits, swales, and estuaries are important features of depositional landforms. In and around Koidiyakarai, Thondi, Manamelkudi, Rameshwaram, Mandapam, and other surrounding areas, extensive erosional features such as marine terraces, sea caves, cliffs, and wave-cut platforms can be observed [28].

2.2 Sampling technique

Runoff from the land, bank erosion and internal channel reworking are all possible sources of fluvial sediments. In the estuary ecosystem, sediment performs a lot
of significant roles and serves as the substrate for a huge assortment of organisms. Field sampling is carried out using some scientific procedures (Figure 2). During the monsoon period of July 2022, 12 sediment samples were collected from various parts of the South Vellar estuary (Figure 3). Twelve sediment samples (SS) were obtained in the South Vellar Estuary (Figure 1). Nine samples (SS1-SS9) were collected from the lower estuarine parts using Van Veen grabs on small boat (Figure 4.a). The basic concept of all grab samplers is the same: A specialized bucket or scoop is lowered through the water column used with rope until it meets the bed, at which point, a release mechanism enables sample collection and withdrawal. To minimize disturbance at the sediment surface on contact and to reduce the loss of fine-grained material as the grab is raised through the water column, the grab was lowered and raised gradually and vertically while avoiding free fall. The grab sampler was properly cleansed after collecting to remove any additional material that had become adhered to the equipment. The samples are transferred to plastic bags with labels. Three samples (SS10-12) were collected manually from surface of middle estuary (Figure 4.b). Surface sediments are relatively simple to collect because there is no need to know the exact depth from which the sediment was collected or to preserve sediment structure. Where sampling locations are easily and safely accessible by foot or small boat, samples can be collected by hand using a scoop, allowing for very accurate sampling site location with GPS. All sediment samples were collected by labeled and sealed plastic bags and transported to the laboratory, where they were stored at 4°C until analysis [29–31].

2.3 Analytical method

The samples are kept in an air-dried environment for further REE extraction. Using a mortar and pestle, the dehydrated samples were crushed to a homogeneous powder. All samples were prepared in a flux mixture containing 100 mg of each
sample. The samples were then heated in an inert, heat-resistant graphite crucible at 1050°C, past the melting point of the flux. The crucible was regularly agitated and heated until the sample was completely dissolved in the molten flux. The melt was then poured into a 50 ml aqueous solution of 5% HNO3. This solution required progressive agitation via shaking and magnetic stirring to dissolve the flux with the sample in the acidic solution. Each sample was prepared for plasma quad analysis by taking 1.5 ml of this dilution, adding 0.1 ml of 9.997 mg l⁻¹ Indium internal standard, and 8.4 ml deionized water. This procedure produced an aqueous solution with a ~3300 whole sediment sample dilution and a concentration of 99.97 μg/l. VG thermal ionization mass spectrometer was used to analyze the extracted samples for rare

*Figure 3.* Sediment sampling locations of South Vellar estuary.
earth elements [29]. The technology known as Thermal Ionization Mass Spectrometry (TIMS) was primarily created for the analysis of samples of geological sediment. By using isotope dilution analysis, the technique is frequently used to determine rare-earth elements and other specific elements [32]. Nine Faraday cup collectors were installed on a VG Sector 54–30 thermal TIMS (VG Isotopes Ltd., UK) for the isotope dilution analytical procedures to measure rare earth elements.

An important analytical instrument for determining the elemental composition of environmental samples, especially soil and sediments, is energy-dispersive X-ray fluorescence Spectrometer (EDXRF). The instrument used for this experiment is an EDXRF spectrometer, model EX-6600SDD. This technique has a high sample throughput, which is multi-elementary and nondestructive. EDXRF is utilized in the current study for large area screening as it is quick and only requires a little sample amount [33].

2.4 Result and discussion

As with their abundance in the continental crust, REEs are prevalent in estuary sediments, which can shed light on their marine cycling. Additionally, it gives more knowledge on the interactions between sand and water and redox reactions that take place in surface environments [34]. There are two primary categories of rare earth elements: Currently conducted research compares low and high rare earth elements (Table 2). The distribution and quantity of REEs in the estuary sediments in South Vellar Estuary are displayed in Tables 3-5. In comparison with HREEs, LREEs are typically more enriched in all sampling locations of estuary (Figure 5). Based on mean concentration from LREE group, La (37.75 μg/g) is the dominant element, followed by Ce (36.65 μg/g), Pr (21.38 μg/g), Nd (17.17 μg/g), Sm (7.64 μg/g), Gd (5.35 μg/g), and Eu (4.09 μg/g) (Table 3). In addition, Y (34.53 μg/g) is also abundant in HREE followed by Tb (5.68 μg/g), Dy (3.93 μg/g), Er (3.07 μg/g), Ho (2.67 μg/g), Tm (2.11 μg/g), Yb (1.84 μg/g), and Lu (1.45 μg/g) (Table 4). Based on the individual
Rare earth element (REE)

<table>
<thead>
<tr>
<th>Low rare earth element (LREE)</th>
<th>High rare earth element (HREE)</th>
</tr>
</thead>
<tbody>
<tr>
<td>La Lanthanum</td>
<td>Y Yttrium</td>
</tr>
<tr>
<td>Ce Cerium</td>
<td>Tb Terbium</td>
</tr>
<tr>
<td>Pr Praseodymium.</td>
<td>Dy Dysprosium.</td>
</tr>
<tr>
<td>Nd Neodymium.</td>
<td>Ho Holmium</td>
</tr>
<tr>
<td>Pm Promethium</td>
<td>Er Erbium</td>
</tr>
<tr>
<td>Sm Samarium</td>
<td>Tm Thulium</td>
</tr>
<tr>
<td>Eu Europium</td>
<td>Yb Ytterbium</td>
</tr>
<tr>
<td>Gd Gadolinium</td>
<td>Lu Lutetium</td>
</tr>
</tbody>
</table>

Table 2. Type of rare earth element.

<table>
<thead>
<tr>
<th>Concentration of LREE (μg/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sampling location</td>
</tr>
<tr>
<td>-------------------</td>
</tr>
<tr>
<td>SS1</td>
</tr>
<tr>
<td>SS2</td>
</tr>
<tr>
<td>SS3</td>
</tr>
<tr>
<td>SS4</td>
</tr>
<tr>
<td>SS5</td>
</tr>
<tr>
<td>SS6</td>
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<tr>
<td>SS7</td>
</tr>
<tr>
<td>SS8</td>
</tr>
<tr>
<td>SS9</td>
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<tr>
<td>SS10</td>
</tr>
<tr>
<td>SS11</td>
</tr>
<tr>
<td>SS12</td>
</tr>
<tr>
<td>Total</td>
</tr>
<tr>
<td>Average</td>
</tr>
</tbody>
</table>

Table 3. Concentration of low rare earth elements.

rare earth element, the average concentration of La (37.75 μg/g) and Ce (35.65 μg/g) described in this research was higher than Pichavaram Mangrove (La: 1.43 μg/g; Ce: 1.26 μg/g) [7] and Cauvery estuary (La: 19.92 μg/g; Ce: 34.98 μg/g) [35], five times lesser than Yamuna river (156.0 μg/g) [36].

Mean concentration of Dy (3.93 μg/g), Ho (2.67 μg/g), and Er (3.07 μg/g) described in this research was higher than Pichavaram Mangrove (Dy: 130 μg/g; Ho: 1.75 μg/g; Er: 1.06 μg/g) [7], and Cauvery estuary (Dy: 3.43 μg/g; Ho: 0.70 μg/g; Er: 2.19 μg/g) [35]. Distribution of yttrium is 3.22–63.48 μg/g in this research is higher than Tagus estuary (2.4–32.0 mg/kg), which is located in Portugal [37].
determined heavy metal concentration for 12 coastal locations in the South Vellar estuary by EDXRF, as well as the distribution of heavy metals (other elements than REE) in the sediments, is examined (Table 6). When compared to all other heavy metals, Cr is frequently higher concentrated in all estuarine sampling locations.
For Cr, the heavy metal concentration ranges from 16.02 to 186.1 μg/g; for Zn, it ranges from 16.36 to 67.26 μg/g; for Ni, it ranges from 12.13 to 42.16 μg/g; for Pb, it ranges from 4.76 to 60.08 μg/g; for Co, it ranges from 4.64 to 21.08 μg/g; and for Cd, it ranges from 0.10 to 1.12 μg/g. Based on the individual heavy metal, the average concentration of Cr (77.75 μg/g) described in this research was lower than Pichavaram Mangrove (141.2 μg/g) [38], East Coast estuaries 318.0 μg/g [39], and Tamirabharani estuary 97.162 μg/g [40]. Distribution of Ni reported (12.13 to 42.16 μg/g) and mean concentration (24.86 μg/g) in this research is lesser than Pichavaram Mangrove (62.0 μg/g) [38], East Coast estuaries (582.0 μg/g) [39], and Pondicherry estuary (33.51 μg/g) [41], higher than Tamirabharani estuary (11.852 μg/g) [40].

Distribution of Zn reported (16.36 to 67.26 μg/g) and mean concentration (36.07 μg/g) in this research is higher than Cauvery estuary (26.0 μg/g) [42]; and lesser than Pichavaram Mangrove (89.0 μg/g) [38] and East Coast estuaries (125.0 μg/g) [39]. Mean concentration of Pb (19.76 μg/g) in this research is higher than Cauvery estuary (10.0 μg/g) [42], Pichavaram Mangrove (11.2 μg/g), and East Coast estuaries.
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(11.0 μg/g); and lesser than Mahanadi estuary (23.88 μg/g) [43]. Mean concentration of Co (10.34 μg/g) in this research is lesser than Cauvery estuary (64.0 μg/g) and East Coast estuaries (12.0 μg/g). Distribution of Cd reported (0.10 to 1.12 μg/g) and mean concentration (0.42 μg/g) in this research is lesser than Pichavaram Mangrove (6.96 μg/g) and both sites of Uppanur estuary (site 1; 4.99 μg/g & site 2; 4.375 μg/g) [44]. The mean heavy metal concentrations are in the following order: Cr > Zn > Ni > Pb > Co > Cd (Table 6). According to results from other countries around the world, this study’s heavy metal concentration is comparable. Figure 6 shows the distribution of heavy metals in sediment from South Vellar estuarine for all sampling locations.

### Table 6

<table>
<thead>
<tr>
<th>Sampling location</th>
<th>Cr</th>
<th>Ni</th>
<th>Zn</th>
<th>Pb</th>
<th>Ag</th>
<th>Co</th>
<th>Cd</th>
<th>V</th>
</tr>
</thead>
<tbody>
<tr>
<td>SS1</td>
<td>123.16</td>
<td>28.12</td>
<td>42.17</td>
<td>17.14</td>
<td>—</td>
<td>13.06</td>
<td>0.47</td>
<td>—</td>
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<tr>
<td>SS2</td>
<td>154.08</td>
<td>36.14</td>
<td>58.06</td>
<td>23.13</td>
<td>—</td>
<td>18.12</td>
<td>0.57</td>
<td>—</td>
</tr>
<tr>
<td>SS3</td>
<td>40.12</td>
<td>20.06</td>
<td>22.82</td>
<td>21.62</td>
<td>—</td>
<td>07.14</td>
<td>0.32</td>
<td>—</td>
</tr>
<tr>
<td>SS4</td>
<td>36.62</td>
<td>17.02</td>
<td>20.64</td>
<td>12.60</td>
<td>—</td>
<td>08.62</td>
<td>0.35</td>
<td>—</td>
</tr>
<tr>
<td>SS5</td>
<td>52.14</td>
<td>22.17</td>
<td>30.17</td>
<td>08.17</td>
<td>—</td>
<td>03.17</td>
<td>0.24</td>
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</tr>
<tr>
<td>SS6</td>
<td>58.13</td>
<td>26.14</td>
<td>29.06</td>
<td>18.38</td>
<td>—</td>
<td>06.64</td>
<td>0.26</td>
<td>—</td>
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<tr>
<td>SS7</td>
<td>109.04</td>
<td>32.62</td>
<td>58.32</td>
<td>24.16</td>
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<td>12.42</td>
<td>0.56</td>
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<td>SS8</td>
<td>118.11</td>
<td>29.73</td>
<td>49.62</td>
<td>32.17</td>
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<td>12.63</td>
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<tr>
<td>SS9</td>
<td>186.08</td>
<td>42.16</td>
<td>67.26</td>
<td>60.08</td>
<td>—</td>
<td>21.08</td>
<td>1.12</td>
<td>—</td>
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<tr>
<td>SS10</td>
<td>22.14</td>
<td>12.13</td>
<td>20.18</td>
<td>04.76</td>
<td>—</td>
<td>04.64</td>
<td>0.18</td>
<td>—</td>
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<tr>
<td>SS11</td>
<td>18.07</td>
<td>16.24</td>
<td>16.36</td>
<td>08.13</td>
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<td>07.34</td>
<td>0.10</td>
<td>—</td>
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<tr>
<td>SS12</td>
<td>16.02</td>
<td>15.85</td>
<td>18.24</td>
<td>06.80</td>
<td>—</td>
<td>09.18</td>
<td>0.13</td>
<td>—</td>
</tr>
<tr>
<td>Average</td>
<td>77.75</td>
<td>24.86</td>
<td>36.07</td>
<td>19.76</td>
<td>—</td>
<td>10.34</td>
<td>0.42</td>
<td>—</td>
</tr>
</tbody>
</table>

3. Conclusion

Due to the widespread use of REE in daily life, it is imperative that we have a thorough understanding of how these elements behave in estuarine and coastal mixing zones in order to improve our comprehension of REE geochemistry at the continent-ocean interface. The major causes of the minor heavy metal contribution to the estuary sediment may be several anthropogenic sources. Weathering influences REE and metal distribution in the South Vellar estuary along the Bay of Bengal and the Palk Strait in Tamil Nadu, India. The main source of REE and heavy metals in Estuary sediments is continental weathering, but anthropogenic activities and the marine environment also contribute significantly. Because of complex physiochemical processes, the elements are significantly altered in the intertidal ecotonal region of different salinities. The sediments have a REE range of 73.30–360.50 (μg/g) and a mean value of 184.33 (μg/g). The average LREE and HREE values are 129.03 and 55.3 (μg/g), respectively. The South
Vellar river-estuarine systems contribute roughly equal amounts of LREEs (70%) and HREEs (30%) to the Bay of Bengal along with Palk Strait. Therefore, we suggest that it is possible to estimate the processes of continental weathering on the Indian subcontinent using the distribution of REE in these estuaries.

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