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Original Research

Geochemical, mineralogical and textural nature of beach placers, north-east Sri Lanka: Implications for provenance and potential resource

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A B S T R A C T

The Pulmoddai placer sands in Sri Lanka (9°3′23.65″–8°51′38.83″N and 80°55′22.91″–81°3′32.65″E) is considered to be one of the major rare earth element (REE) prospects world-wide. This deposit has a global significance in terms of strategic economic resources and can provide valuable insight for resource estimation and for its economic use. A detailed study of the geochemical, mineralogical, and textural properties of the heavy minerals of this region is undertaken applying the scanning electron microscopy –electron dispersive X-ray spectroscopy (SEM–EDS), inductively coupled plasma–optical emission spectroscopy (ICP–OES), X-Ray diffraction (XRD), and X-Ray fluorescence (XRF) techniques. The results from these analyses are modeled to delineate the source(s) which contribute to the formation of the mineral deposits and their subsequent enrichment. The beach sediment from this region, exhibited elevated concentrations of major elements like silicon (Si) and manganese (Mn) and high abundance of minor and trace elements like yttrium (Y), strontium (Sr), lanthanum (La), barium (Ba), which indicates that the provenance is similar to calcium-silicate rocks. The highest (total) concentration of Y, La, scandium (Sc) and cerium (Ce) are mainly associated to fine grained sediment, signifying that their occurrences are governed by the grain size distribution and ambient hydrodynamic conditions primarily from lagoonal input. Monazite, zircon, ilmenite, rutile, and xenotime are more abundant than garnet in the study area, as is evident from the higher abundance of thorium (Th) and uranium (U) and the enrichment of zirconium (Zr), Ce, vanadium (V), chromium (Cr), and Y. Textural analysis of sediment shows sediment particles are poorly sorted and have more angular grain boundaries, indicating their lower textural maturity. This suggests that the sediment is fluvially derived from a close source rock of charnockite specific to the hinterland lithology. The sediment transported by perennial rivers and the hydrodynamic conditions of the Kokkilai Lagoon estuary, are the two primary sources of heavy mineral enrichment in the study area, which is quite unique in terms of the placer formation. Subsequently, these sediment particles formed the beach placer deposits enriched in REE–Th rich heavy minerals, which are of immense strategic importance.

1. Introduction

Rare earth elements (REEs) are group of seventeen elements (fifteen lanthanoids + yttrium + scandium) which are very crucial in our technology driven world for their unique chemical, physical, catalytic, phosphorescent, electrical, magnetic, and optical properties (Balaram, 2019). The average concentration of the REEs is about 130–240 μg/g in the Earth's crust which is much higher when compared to chondritic abundances (Zepf, 2013). Therefore, in spite of the name, REEs are more abundant than precious metals, like gold and platinum, in the upper continental crust (Haxel et al., 2002). However, REEs are dispersed extensively in the crust and

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rarely form mineable deposits, making their extraction more difficult (Kim & Jariwala, 2021). REEs have huge applications in super magnet, metallurgy, electronic, petroleum, automobile, glass, and ceramic industries. They are also called green elements because of their indispensable use in many green energy applications. The growing worldwide demand for these critical elements, has generated a considerable economic interest towards exploration for REEs and their subsequent extraction, in association with recent developments in technological applications.

REE-enriched beach sands are associated with various types of placers (Khan et al., 2021), laterite, and bauxite, which are formed due to the weathering and erosion of the host rock(s). Consequently, various factors such as lithology with structural aspects, weathering and erosion, coastal morpho-dynamics, drainage network, and climatic conditions favor the origin of rich placer deposits in coastal environment specially enriched in thorium. The transportation and deposition of the minerals enriched in REE and radioactive elements, is controlled by the proximity of physical, chemical, and biogenic inputs where fresh water and seawater interact (Ladipo et al., 2011). The major and trace elements in the sediment, are transported by perennial rivers through partitioning between the particulate and dissolved phases (Ip et al., 2007). These transported sediment particles are again reworked, as coastal zones are dynamic regions with littoral currents and tidal flow playing a significant role in the distribution of sediment.

The REE potential in Sri Lanka is in the form of carbonatite, pegmatite, beach, and alluvial placer deposits. The main REE bearing minerals in the beach placers of Sri Lanka are monazite and xenotime. The heavy mineral prospects of the north-eastern coast of Sri Lanka are high, with a heavy mineral content of about 70%–85% in the Pulmoddai deposit and 45%–50% in the Verugal deposit (Amalan et al., 2018). In comparison, the sediment on southwest coast of Sri Lanka from the region close to the mouth of the Gin River, have an average heavy mineral content of about ~2% in offshore sediment and ~10% in onshore sediment (Amalan et al., 2018). The sediment in Trincomalee Bay and Koddiyar Bay, about 60 km from Pulmoddai beach and nearer to the Mahaweli River, has similar geochemical composition to the sediments which are supplied by the river. This indicates that the reworking along the northeast coastal areas of Sri Lanka increases the REE content (Young et al., 2014). The Pulmoddai deposit lies over the Wanni Complex rocks, Sri Lanka and corresponds to the age of Precambrian crystalline rocks (He et al., 2018). This deposit contains 6 million tons of heavy mineral sands consisting of 70%–72% ilmenite, 8% rutile, 8%–10% zircon, 0.3% monazite, and 1% sillimanite (Amalan et al., 2018; Geological Survey and Mines Bureau, 2014; Ismail et al., 1983).

The primary source of the Pulmoddai deposit has been attributed to the weathered and eroded Highland Complex rocks transported by the Mahaweli River, one of the longest rivers (335 km) in Sri Lanka, according to previous studies (Batapola et al., 2020; Udarika et al., 2016; Weththasinghe et al., 2020). The offshore sediment could be a secondary source of these heavy minerals, and it is surmised that the sediment mechanically accrued along the northeast coast as a result of ocean currents and waves (Amalan et al., 2014), especially during the monsoonal season. Amalan et al. (2014) suggested that the differential oceanic circulation in the Central Indian Ocean aided by regional monsoon related energy and wave circulation has resulted in proportionally abundant heavy mineral resources in the north-east region, primarily Pulmoddai, when compared to the south-western part of Sri Lanka. The placer deposits are unique in South Asia as this area is the only deposit which has both a fluvial contribution as well as a lagoonal input nearby as most of the other deposits are primarily formed due to perennial rivers (Sengupta & van Gosen, 2016).

Several investigations have been done to classify the chemical and mineralogical characteristics of the Pulmoddai placer deposit in order to understand the mechanisms of formation and sources of the deposit (Amalan et al., 2014; Udarika et al., 2016). A detailed mineral geochemistry (e.g., REEs and major elements) of the heavy minerals is required to delineate the provenance of this beach placer. Since mineral content changes over time, a chemical, mineralogical, and mineral morphological study was recently done using the inductively coupled plasma–optical emission spectroscopy (ICP–OES), scanning electron microscopy (SEM), and X-ray diffraction (XRD) analysis to provide new data on the source, distribution, and transport of sediment. The current results also will provide important information for the future management of the coastal deposit as an economic resource and for sustainable mining, as well as to provide insight into the origin and subsequent enrichment processes of the deposit. This is the only beach placer deposit in South Asia and plausibly one of the few worldwide placer deposit where both perennial rivers and lagoonal input contribute to its formation.

2. Study area

2.1. Location

The study area is situated at the eastern part of Sri-Lanka, between 9°3′23.65″–8°51′38.83″N and 80°55′22.91″–81°3′32.65″E (Fig. 1). The samples which are taken from the Pulmoddai region are denoted P01–P10 and from Thiriyai region are denoted T01–T10. The samples denoted U01–U10 are related to the Kokkilai lagoon. The samples ME1 and ME2 are taken from the Mee Oya River, MA1 and MA2 are taken from the Maa Oya River, and CH1 and CH2 are taken from the Churiyan Aru River. The sampling locations are shown in Fig. 1c.

2.2. Geology

The lithology of the study area comprises about 90% high-grade metamorphic rock, 9% limestone, and 1% igneous rocks as intrusions such as pegmatite and dolerite. The metamorphic portion is divided into subgroups, namely, Kadugannawa Complex (KC), Highland Complex (HC), Vijayan Complex (VC), and Wanni Complex (WC) based on geological structure, metamorphic grade, geochemistry, isotope composition, and petrology (Cooray, 1994). The Pulmoddai region is located in the Wanni Complex, which is about 25 km north of the HC-WC boundary of northeastern Sri Lanka. The Wanni Complex consists mainly of high grade orthogneisses and a relatively lower abundance of metasediments in the eastern regions of the country (Pohl & Emmermann, 1991). The basement rock of the Pulmoddai region consists of undifferentiated charnokitic biotite gneisses with boudinaged mafic layers of orthopyroxene (He et al., 2018). The HC–WC boundary is rich in undifferentiated tectonically intercalated metasedimentary and metaigneous rocks (Fig. 1b). The most common boundary rocks are quartz-feldspar-garnet granulite, garnet-sillimanite gneisses, quartzite, and garnet biotite gneisses (He et al., 2018). The basement of Pulmoddai region is covered with alluvial and lagoonal sediment that originated in the rivers and basically flow along the rocks present in the Wanni Complex.

2.3. Geomorphology

The Thiriyai sampling area is a curved beach covered by a cuspate foreland on southern side. The 140 km long Yan Oya River is the fifth longest river in Sri Lanka and it enters the sea about 3 km north of the Thiriyai sampling locations. The Pulmoddai sampling

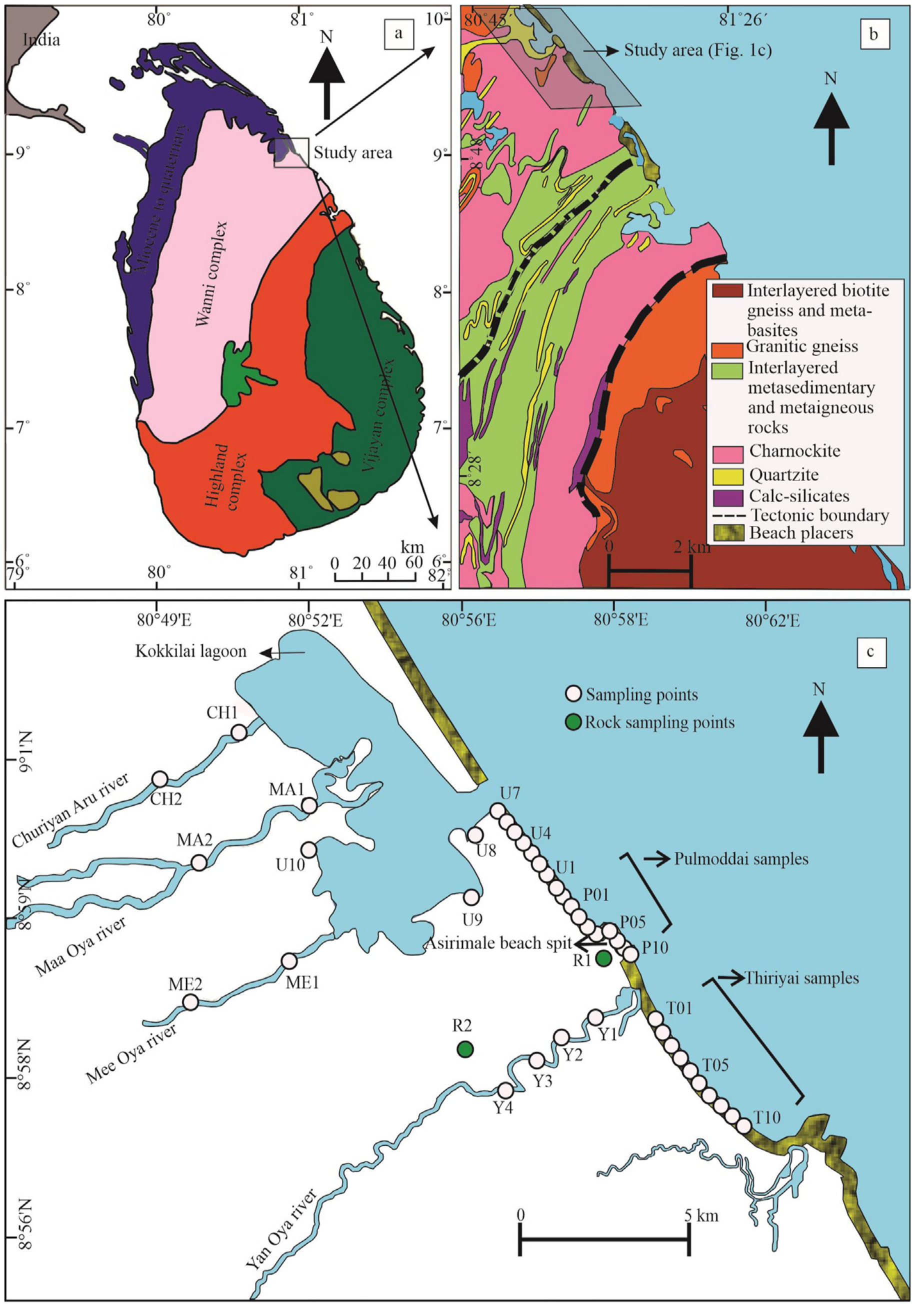

Fig. 1. (a) Geological framework of Sri Lanka showing the major crustal blocks (modified after Cooray, 1994; He et al., 2018; Rajapaksha et al., 2012). (b) Detailed geological map of the study area. (c) Map showing sampling locations and the presence of Kokkilai Lagoon, Churiyan Aru River, Maa Oya River, Mee Oya River, and Yan Oya River near the study area. Reproduced with permission from Cooray, 1994, © Elsevier B.V. 1994; He et al., 2018, © Walter de Gruyter GmbH, Berlin/Boston, 2018.

area is bounded by the Asirimale beach spit on the southern side and extends to the Kokkilai Lagoon on the northern side. This lagoon is a shallow inland body of water of about 13 km in length and parallel to the coastline. The areal distribution of the lagoon is greater along the southern part compared to the northern part. The Kokkilai Lagoon is a coastal aquatic environment which is located at the northern part of the Pulmoddai deposit. It is linked to the Indian Ocean by a narrow inlet that is 100 m wide. The input of fresh water to the lagoon is contributed by the Churiyan Aru River, Mee Oya River, and Ma Oya River. However, the inlet has gradually become narrower with the passage of time. The water body has gradually been occupied with land-sourced sediment (probably) through the natural evolution and sediment mainly is supplied by rivers like the Ma Oya and Churiyan Aru. The catchment area of the perennial rivers of the Kokkilai Lagoon lies on the Wanni Complex rocks of Sri Lanka, where the Pulmoddai sand deposit is located.

3. Materials and methods

3.1. Sample collection and preparation

A total of 27 samples (200 gm each) were collected from the coastal region (Pulmoddai-P01, P02, P03, P04, P05, P06, P07, P08, P09, and P010; Thiriyai-T01, T02, T03, T04, T05, T06, T07, T08, T09, and T010; Kokkilai beach samples (U1, U2, U3, U4, U5, U6, and U7). Also, three samples (200 gm) were collected from the lagoon

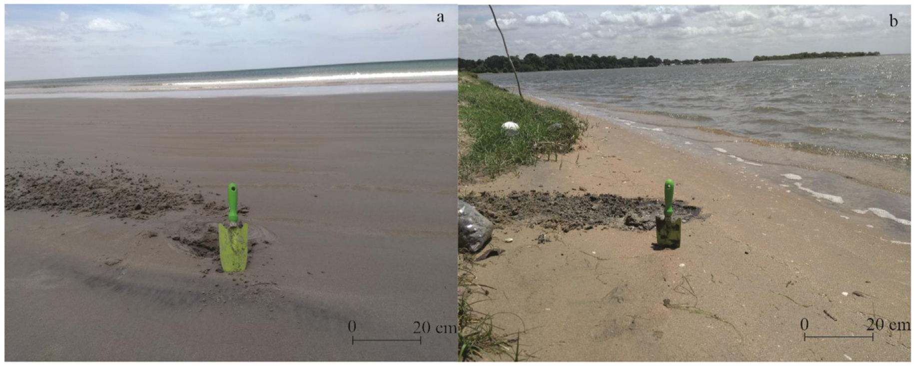

Fig. 2. Field photographs showing the locations of samples collected from beach and river sites.

environment (U8, U9, and U10) and six samples (200 gm) from riverside (CH1, CH2, MA1, MA2, ME1, and ME2) in the study area. These samples were collected using the coning and quartering methodology for reliability and homogeneity. The end margin of the berm region was selected for sampling to avoid the effect of the disturbances caused by tides because it was well beyond the high tide zone (Fig. 2) and highly humid land. The distance of the sample location to the coast varies with the distance from the berm. Generally, the berm region is around 5 m inward from the coastline of north-eastern Sri Lanka. The sand samples were collected at a depth of 30 cm keeping a 1 km distance between each sampling point. The samples were washed and dried at 60 °C in the oven, to obtain moisture-free samples for subsequent analysis.

3.2. Analytical procedures

3.2.1. Scanning electron microscopy—electron dispersive X-ray spectroscopy (SEM—EDS) analysis

SEM is an important observational tool to examine the micro-characteristics of the sample in the field of geological scientific research, and the EDS attachment with SEM allows for composition analysis during morphological observation. Heavy minerals were separated from a few grams of the collected sand samples using density separation techniques, using bromoform, and followed by cleaning of the heavy minerals with ethyl alcohol for SEM analysis. The cleaned and separated heavy minerals were kept to form an epoxy mount with Araldite. The mounts were coated with carbon, prior to SEM—EDS analysis. The sand samples from the study area were studied using the SEM—EDX microanalyzer at the Department of Geology and Geophysics, Indian Institute of Technology (IIT) Kharagpur.

3.2.2. X-ray diffraction (XRD) analysis

XRD analysis was used to identify the minerals (heavy minerals) present in the sample set that consisted of five Pulmoddai samples, five Thiriyai samples, seven Kokkilai beach samples, three Kokkilai Lagoon samples, and the six river samples. A small quantity of each powder sample was sent for XRD analysis using an X-ray diffractometer (Rigaku Ultima IV at Uva Wellassa University, Sri Lanka). The heavy minerals present in the sample were identified using related reference peaks by comparing the *d*-spacing and integrated intensities of the standards compiled on the Joint Committee on Powder Diffraction Standards (JCPDS) cards of the same minerals published by the JCPDS (König & Verryn, 2021).

3.2.3. X-ray fluorescence (XRF) analysis

To emphasize the importance of river and lagoon contributions to the distribution of beach placers in the study area, XRF analysis was done to determine the major elements in all the river samples, lagoon samples, and a few beach samples.

3.2.4. Inductively coupled plasma—optical emission spectroscopy (ICP—OES) analysis

Sixty elements were estimated in the samples using ICP—OES. A 0.5 g split of each sample was fused at 750 °C with sodium peroxide and the fusion cake was dissolved in a dilute nitric acid. The resulting solution was then analyzed using ICP—OES.

4. Results

4.1. Mineralogy

Tables 1 and 2 list the mineral accumulations and their abundances that were calculated using the XRD data of ten selected samples from the Pulmoddai and Thiriyai areas, six river samples, and ten samples from the Kokkilai Lagoon. The XRD data revealed that the Pulmoddai samples are composed of ilmenite and zircon, with minor amounts of rutile and monazite, while the Thiriyai samples consisted of quartz with lesser amounts of ilmenite, zircon, and monazite. There is a lack of heavy minerals especially in the river samples as well as the samples from the Kokkilai Lagoon. However, the samples are rich in quartz, hematite, corundum, bornite, eskolaite, and calcite (Table 1). The crystalline phases of heavy minerals in

Table 1
Mineral assemblages and abundances in percent by weight for six river samples and ten Kokkilai lagoon samples.

Sample	CA1	CA2	MA1	MA2	ME1	ME2	U1	U2	U3	U4	U5	U6	U7	U8	U9	U10
Quartz	18	23	16	20	21	6	95	75	84	83	81	85	90	80	73	17
Fluorite							5	3	1							
Calcite	15	13	15	9	13	71		21	4	5	7	10	4	15	6	11
Hematite	50	43	46	11	22	11		1	1	2	1	2	2	1	6	20
Corundum				52	22				4	5	5	3	4	3	10	40
Eskolaite	12	15	10	8	21	11			4	5	6				4	9
Bornite	5	8	12			1										
Total	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100

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Table 2
Mineral assemblages and abundances in percent by weight for ten selected samples in the Pulmoddai and Thiriyai areas.

Sample	P01	P03	P05	P07	P09	T01	T03	T5	T07	T09
Monazite	3.11	4.77	4.51	7.69	7.98	3.81	4.58	4.55	5.04	5.10
Zircon	25.77	33.13	19.72	17.44	21.81	4.76	5.34	31.82	14.28	9.18
Rutile	16.58	3.81	3.13	1.54	0	0	0.76	1.82	0	4.08
Ilmenite	38.39	11.02	11.88	4.62	8.51	8.57	13.74	0	3.36	4.08
Quartz	16.1	48.04	61.21	68.72	61.70	82.86	75.57	61.82	77.31	77.55
Total	100	100	100	100	100	100	100	100	100	100

the samples obtained from the Pulmoddai and Thiriyai areas that were identified by XRD are shown in the diffractograms (Fig. 3).

Further analysis of the Pulmoddai and Thiriyai samples was done for confirmation of the presence of heavy minerals. Analysis of heavy mineral samples revealed a variety of minerals, dominantly ilmenite and zircon (Figs. 4 and 5). The next most heavy minerals were sillimanite, monazite, and rutile. Apart from the heavy minerals, calcium and chromium minerals also were observed. The heavy mineral

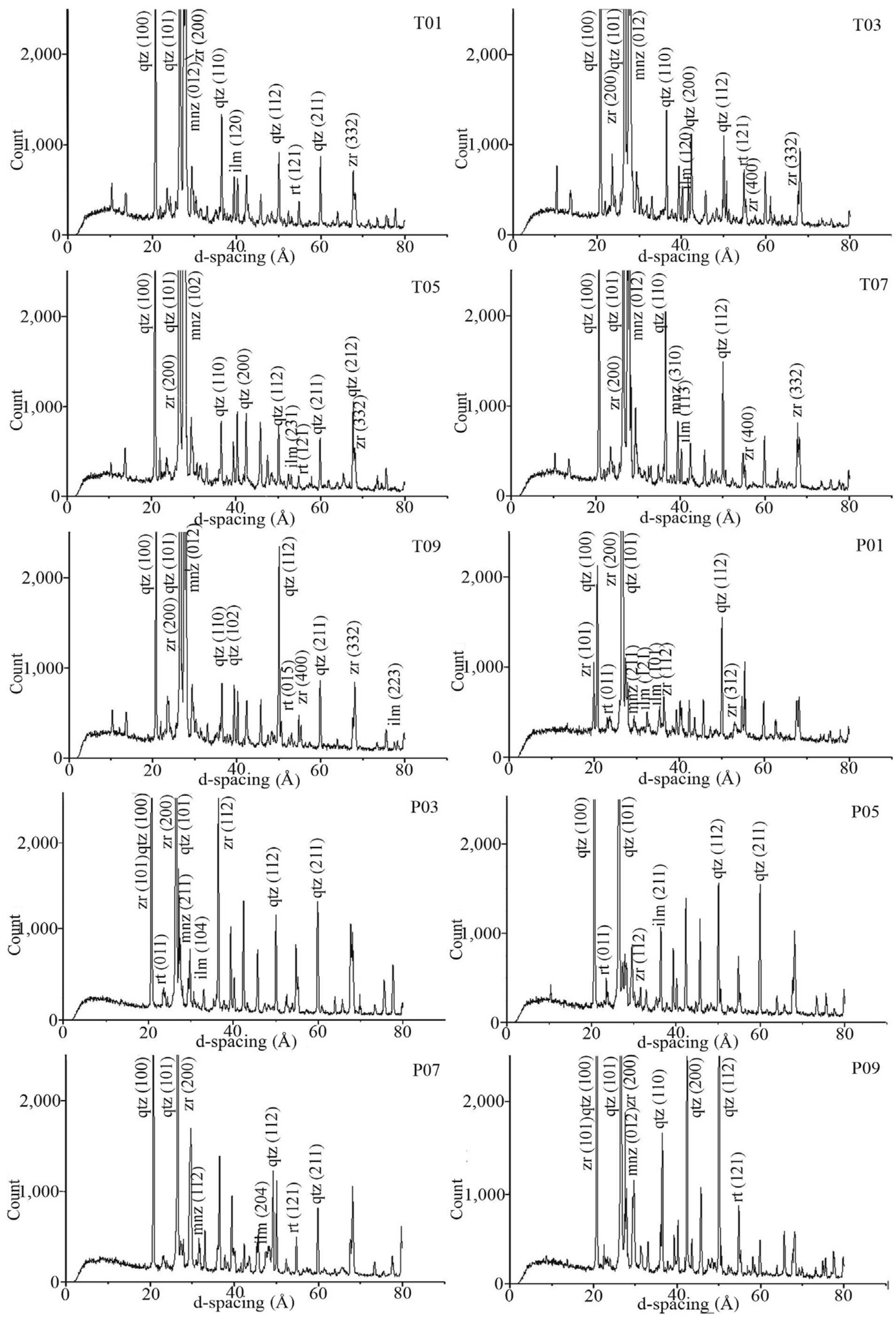

Fig. 3. XRD spectra showing the composition of samples T01, T03, T05, T07, and T09 from the Thiriyai area and P01, P03, P05, P07, and P09 from the Pulmoddai area, respectively.

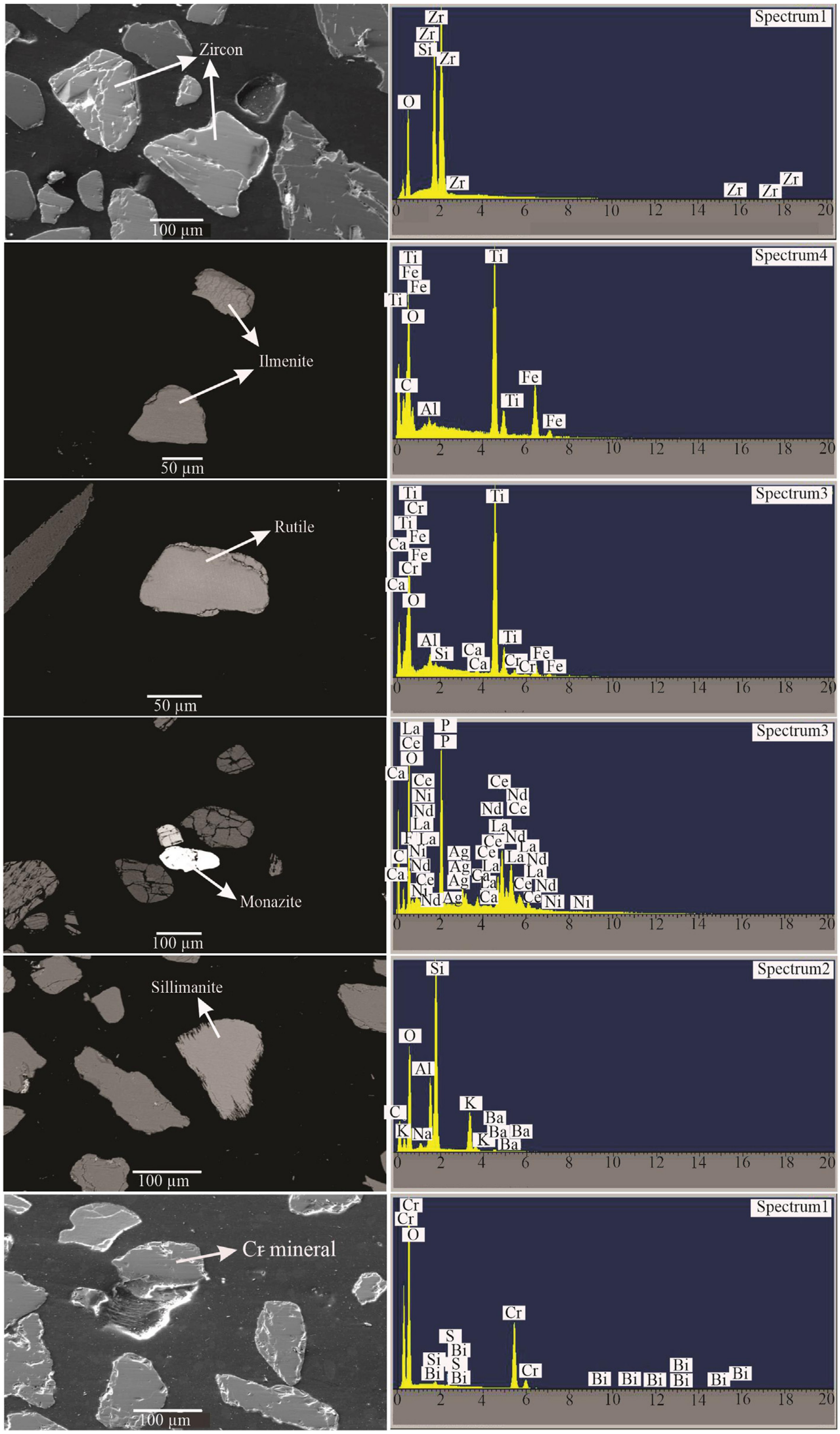

Fig. 4. SEM micrograph of heavy mineral assemblages (zircon, ilmenite, rutile, monazite, sillimanite, and Cr-mineral), based on their chemical composition, from the Pulmoddai beach sands.

sands obtained from the Yan Oya River have been analyzed using SEM-EDS and ilmenite, zircon, garnet, rutile, and sillimanite were identified (Fig. 6). Thin sections of all the major lithologies near the study area were investigated using SEM for the presence of heavy minerals (Fig. 7). The WC granite gneiss (sample R1) appears to preserve monazites and zircons without much alteration. The pyroxene bearing quartzo-feldspathic gneiss domains (sample R2) appears to preserve many ilmenite, rutile, and zircon grains.

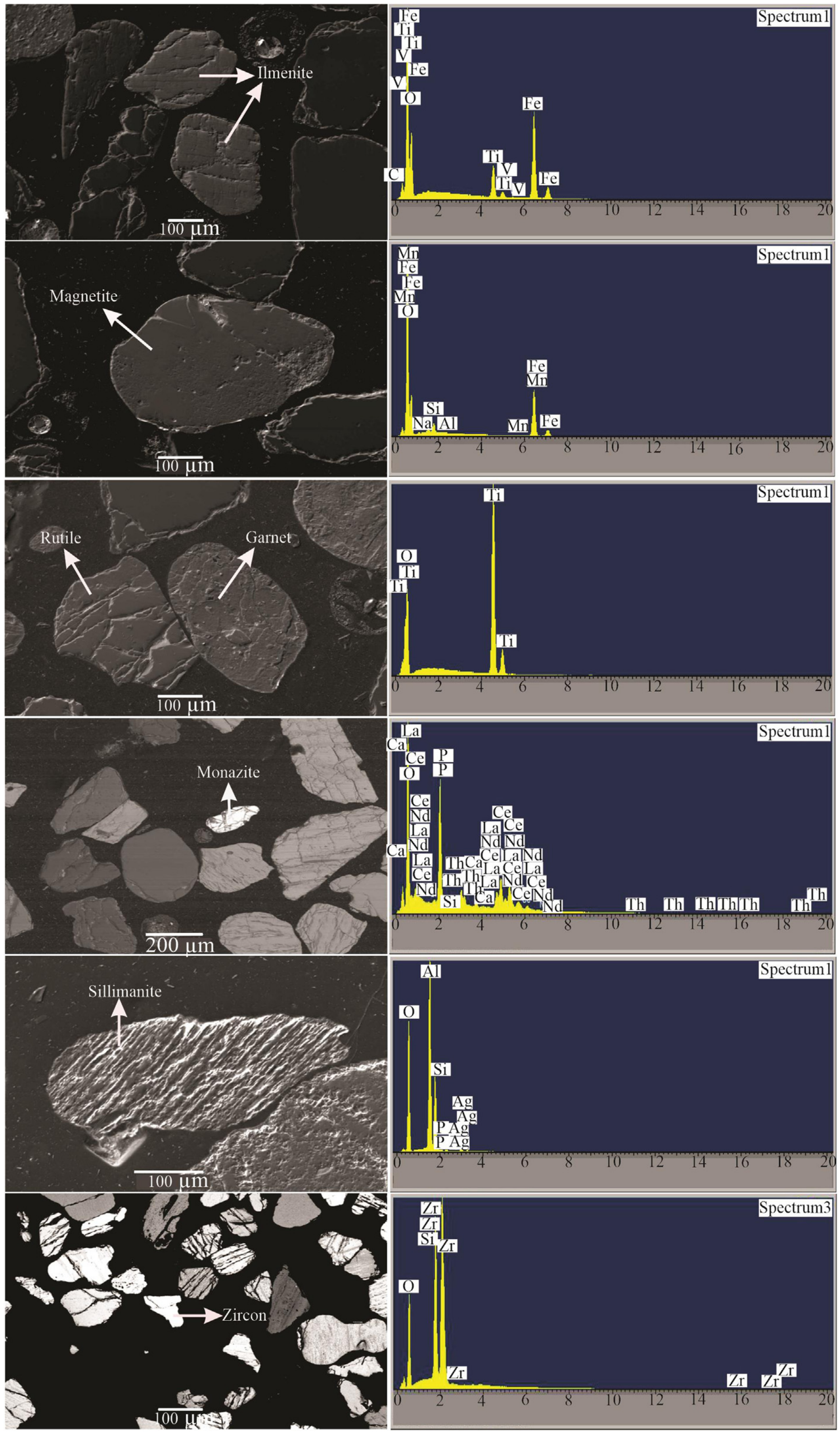

Fig. 5. SEM micrograph of heavy mineral assemblages (ilmenite, magnetite, rutile, garnet, monazite, sillimanite, and zircon), based on their chemical composition, from the Thiriyai Beach sands.

4.2. *Geochemistry*

4.2.1. *Major element concentrations*

The major elemental concentrations of the river and Kokkilai Lagoon samples detected by XRF are listed in Table 3. The silicon dioxide (SiO_2) content of the river samples varies from 31 to 35 wt% whereas the Kokkilai Lagoon samples have SiO_2 content that varies from 20 to 51 wt% (Table 3). The titanium oxide (TiO_2) concentration somehow shows the same trend, and has lower abundance in river samples than in the Kokkilai Lagoon samples. In samples U7

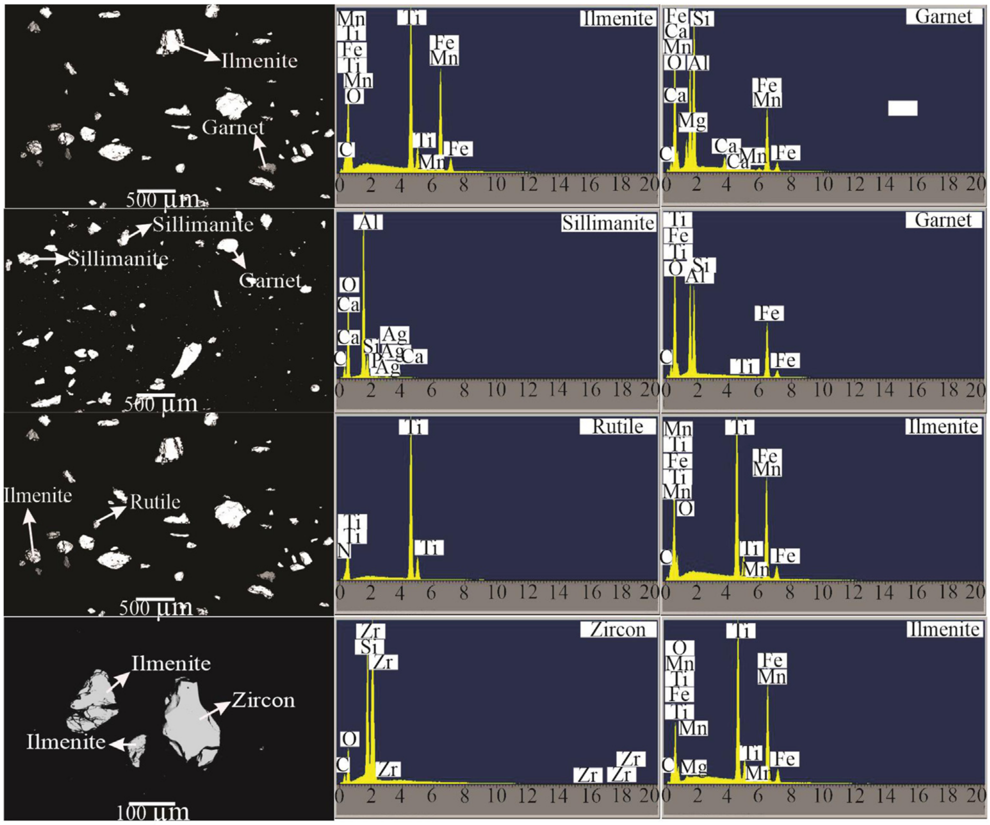

Fig. 6. SEM micrograph of heavy mineral assemblages (ilmenite, garnet, sillimanite, rutile, and zircon), based on their chemical composition, from the Yan Oya River.

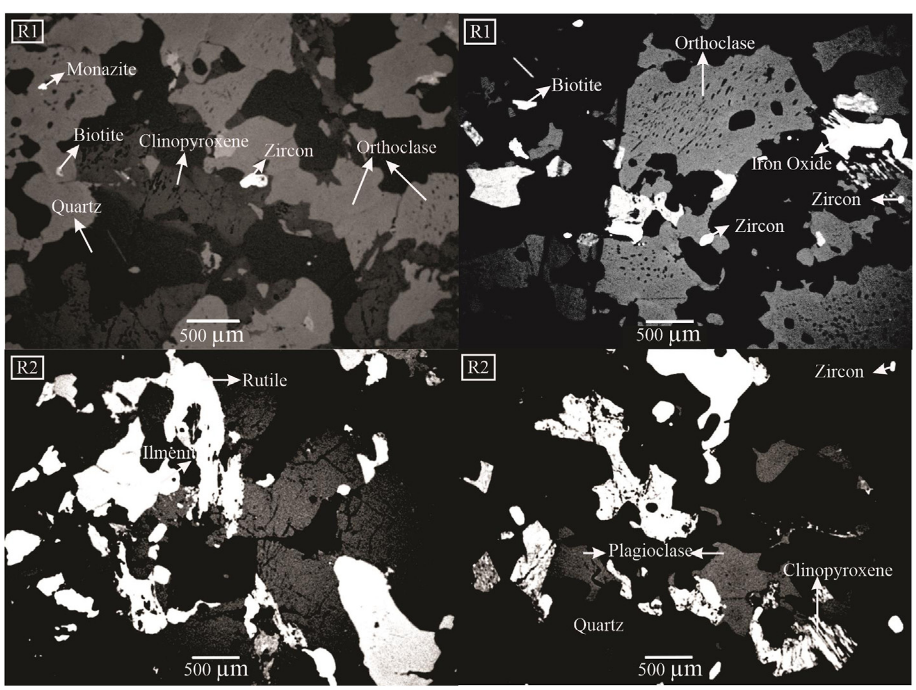

Fig. 7. Identification of heavy minerals associated with granite gneiss (sample R1) and pyroxene bearing quartzo-feldspathic rock (sample R2).

and U8, the TiO_2 concentration is higher away from the lagoon mouth. The aluminum oxide (Al_2O_3) content varies from 8 to 12 wt% in the river samples and 2–9 wt% in the Kokkilai Lagoon samples. The average concentrations of aluminum oxide (Al_2O_3) and iron oxide (Fe_2O_3) are higher in the river samples as compared to Kokkilai Lagoon samples. In addition, the calcium oxide (CaO) content varies

Table 3
Major elements found from the Kokillai Lagoon and river samples in percent by weight (wt%).

Oxide	CH 01	CH 02	MA 01	MA 02	ME 01	ME 02	U1	U2	U3	U4	U5	U6	U7	U8	U9	U10
Al_2O_3	11.2	11.9	10.7	8.32	10.6	9.14	4.1	3.74	3.69	3.13	3.32	3.08	2.95	3.15	7.66	9.23
CaO	4.45	2.08	2.66	4.1	3.62	3.14	3.69	4.44	5.87	6.41	8.05	14.2	7.07	11.6	3.98	4.44
Cr_2O_3	0.12	0.08	0.07	0.09	0.06	0.08	0.15	0.19	0.09	0.07	0.04	0.03	0.07	0.03	0.10	0.09
Fe_2O_3	20	21.3	17.5	25.6	12.5	13.3	24.8	22.2	9.78	11.7	4.56	3.74	12.5	4.24	7.93	12.7
K_2O	0.58	0.65	0.65	0.6	0.85	0.88	0.17	0.17	0.26	0.23	0.25	0.28	0.25	0.29	0.77	0.47
MgO	3.09	3.17	3.43	3.19	3	2.34	2.59	2.02	1.77	1.46	1.61	2.01	1.43	1.76	5.93	8.5
MnO	0.56	0.67	0.30	0.40	0.18	0.21	0.51	0.46	0.20	0.25	0.08	0.06	0.26	0.07	0.13	0.35
Nb_2O_5	0.05	0.04	0.04	0.06	0.04	0.05	0.09	0.10	0.06	0.07	0.04	0.04	0.08	0.04	0.04	0.04
SiO_2	31.4	31.8	31.7	33.8	35.4	33.5	20.2	27.6	45.8	42.5	51.6	48.6	35	44.4	36	33
TiO_2	6.3	6.86	6.06	13.5	6.81	7.73	28.4	26.3	11.7	14.2	7.2	6.23	17	6.72	6.3	6.64
V_2O_5	0.05	0.05	0.05	0.09	0.05	0.06	0.16	0.15	0.08	0.09	0.05	0.05	0.10	0.05	0.05	0.05
ZrO_2	0.55	0.60	0.54	0.85	0.78	0.81	1.52	3.03	1.53	2.06	0.70	0.63	1.12	0.54	0.78	0.78
CeO_2	0.02	0.02	0.02	0.02	0.02	0.02	0.01	0.01	0.02	0.02	0.02	0.02	0.01	0.02	0.02	0.02
P_2O_5	0.05	0.04	0.05	0.06	0.05	0.05	0.08	0.11	0.08	0.09	0.07	0.05	0.06	0.05	0.06	0.06
Na_2O	0.14	0.14	0.19	0.02	0.23	0.28	0.23	0.20	0.23	0.21	0.24	0.25	0.19	0.21	0.51	0.36
SO_3	0.04	0.03	0.14	0.07	0.04	0.06	0.10	0.10	0.09	0.10	0.09	0.12	0.09	0.09	0.39	0.09
HfO_2	0.02	0.01	0.01	0.01	0.01	0.01	0.02	0.03	0.01	0.01	0.00	0.00	0.01	0.00	0.01	0.01

Note: Al_2O_3 = aluminum oxide, CaO = calcium oxide, Cr_2O_3 = chromium oxide, Fe_2O_3 = iron oxide, K_2O = Potassium oxide, MgO = magnesium oxide, MnO = manganese oxide, Nb_2O_5 = niobium (V) oxide, SiO_2 = silicon dioxide, TiO_2 = titanium oxide, V_2O_5 = vanadium pentoxide, ZrO_2 = zirconium oxide, CeO_2 = cerium (IV) oxide, P_2O_5 = phosphorus pentoxide, Na_2O = sodium oxide, SO_3 = sulfur trioxide, HfO_2 = hafnium oxide.

from 2 to 14 wt% due to the presence of calcite. The samples from the study area, had a high silica content and low abundance of Ca and Fe.

4.2.2. Trace element abundances

The profusions of major elements, REEs, trace elements, and radioactive elements were found in selected sediments (P01–P08) because of the high proportion of heavy minerals. The standard deviation (SD), mean concentration, and relative standard deviation (RSD) of minor and trace elements found in the study samples and their comparison with upper continental crust (UCC) concentrations (Rudnick & Gao, 2003; Taylor & McLennan, 1985, 1995; Wedepohl, 1995) are summarized in Table 4. The mean concentrations of Hf, V, Zr, and Cr are relatively higher than the UCC values, whereas the mean concentrations of other elements are relatively lower than the UCC values.

The REE concentrations indicate enrichment, when compared to the UCC values (Table 5). Cerium (Ce) shows the highest value followed by La, Nd, Y, Pr, Sm, Gd, Dy, Yb, Er, Tb, Ho, Eu, Lu, and Tm. The measured concentrations of the sum of ($\sum$) REEs in the Pulmoddai Beach placers vary from 79 to 4099.18 ppm, with an average concentration of 686.83 ppm; these concentrations are about 7.1–4.1 times higher (enriched) than those in the UCC, average shale value (ASV), sedimentary rock, and post archean Australian shale (PAAS). The light REEs (LREEs) contribute 96.5% of the total REE budget while the heavy REEs (HREEs) contribute 3.5% of the total REE budget. The LREE/HREE values range from 9.02 to 34.6 with an average value of 18.39 which is compared in Table 6. The LREEs show an enhancement of 3.8–5.6 ppm, and the HREEs show an enhancement of 1.6–3.1 ppm compared to the crustal values with yttrium (Y) being the most abundant. These data indicate an enrichment of about 4.68 times in the total REE concentration. The La/Lu ratio is about 135.27, which is greater than the crustal average of 100 (Rudnick & Gao, 2003). This indicates that the study area has an enriched concentration of LREEs as compared to HREEs with a negative Eu anomaly (Fig. 8), i.e., the REEs indicate less Eu content than Sm and Gd content.

4.2.3. Radioactive elements specifications

Th/U ratios ranged from 4.24 to 8.49 (average Th/U ratio = 7.90, number of samples (n) = 8); however, the Th/U ratio for the UCC is 3.89 (Rudnick & Gao, 2003) (Table 6). The mean abundance of Th is 74.26 (Table 7), which is 7.07 times enriched compared to the UCC, whereas the U mean abundance is 9.39 (Table 7), which is 3.47 times higher compared to the UCC (Rudnick & Gao, 2003). Thus, thorium abundance is about four times higher than uranium abundance in these samples, which indicates that the Th-rich minerals, such as monazite [(La, Ce, Nd, Th) PO_4] are more predominant as compared to the U-rich mineral such as zirconium silicate ($ZrSiO_4$) (Ndjigui et al., 2020).

4.2.4. Role of heavy minerals in sediment geochemistry

The occurrence of heavy minerals has some influence on the geochemistry of the sediment obtained from the beach placer (He et al., 2015). Figures 9a and 9c shows the connection between heavy mineral concentration as presented by Th/Al and LREE and HREE enrichment as proxied by La/Sm and Tb/Yb, respectively. Garzanti et al. (2011) described that monazite and allanite are very rich in LREE elements, whereas xenotime and zircon are very rich in HREEs. REE compositions are significantly influenced by the HREE-enriched heavy minerals which ultimately control the provenance (e.g., Rollinson, 1993). There is a positive correlation of heavy minerals with LREE and HREE, which suggests that the presence or absence of key heavy minerals partially controls the chemistry of LREEs and HREEs, especially in monazite or zircon. These two minerals are controlled by both provenance and current sorting. Figure 9b establishes a positive correlation between the enrichment in High Field Strength Elements (HFSE) and abundance of heavy minerals. Figure 9d shows a plot of Th/Al against Zr/Al. Here, Al is used as a normalizing element, while Th and Zr are accommodated in heavy minerals, mainly in monazite and zircon, respectively (Ni et al., 1995). Figure 9d shows a positive correlation between Zr/Al and Th/Al, signifying that the sediment enriched in monazite also are rich in zircon, as expected due to marine processes concentrating the heavy minerals (He et al., 2015).

4.3. Textural analysis of sediments

Generally, the heavy resistant minerals are found as beach placers and these are derived from source rock(s) (through rivers) due to weathering and chemical erosion. Here it has been observed that the boundary of major heavy minerals is more angular (Figs. 10a–10d) suggesting that the provenance is closer to the placer deposits (Ndjigui et al., 2014). Some of the heavy minerals

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Table 4
Concentration of minor and trace elements (ppm) composition from the beach sand samples and its comparison with upper continental crust (UCC) derived from surface exposure, sedimentary and loess data.

Element	Taylor and McLennan (1985, 1995) (ppm)	Wedepohl (1995) (ppm)	Rudinick and Gao (2003) (ppm)	This study (mean in ppm)	SD (standard deviation) (ppm)	RSD (relative standard deviation) (%)
Aluminum (Al)	—	—	—	14,587.5	1,511.33	10.36
Calcium (Ca)	—	—	—	62,612.5	38,246.17	61.08
Iron (Fe)	—	611	557	27,012.5	33,440.29	123.79
Potassium (K)	—	—	—	4,937.5	1,208.23	24.47
Magnesium (Mg)	—	—	—	4,050	1,942.75	47.97
Phosphorus (P)	—	—	—	350	307.06	87.73
Sulfur (S)	—	953	621	1,000	0	0
Silicon (Si)	—	—	—	302,625	60,944.79	20.14
Titanium (Ti)	—	—	—	31,912.5	42,749.25	133.95
Silver (Ag)	50	55	53	1.125	0.35	31.42
Arsenic (As)	1.5	2	4.8	6.25	2.76	44.23
Boron (B)	15	17	17	10.25	0.7	6.89
Barium (Ba)	550	668	624	321.75	67.31	20.92
Beryllium (Be)	3	3.1	2.1	5	0	0
Bismuth (Bi)	1.3	1.23	1.6	0.1	1.48	1.48
Cadmium (Cd)	0.09	0.1	0.09	0.2	2.96	1.48
Cobalt (Co)	17	12	17.3	7.5	8.92	118.74
Chromium (Cr)	85	35	92	117	111.33	95.16
Cesium (Cs)	4.6	5.8	4.9	0.1	0.03	31.42
Coper (Cu)	25	14	28	6	2.82	47.14
Erbium (Er)	—	—	—	4.9	7.38	149.79
Gallium (Ga)	17	14	17.5	7.6	7.07	92.94
Gadolinium (Gd)	—	—	—	12.4	22.26	178.22
Germanium (Ge)	1.6	1.4	1.4	1.2	0.7	56.56
Hafnium (Hf)	—	—	—	266.3	572.98	215.1
Indium (In)	0.05	0.061	0.056	0.2	0.03	16.63
Lithium (Li)	20	22	21	10	0	0
Manganese (Mn)	—	—	—	609.8	690.52	113.22
Molybdenum (Mo)	1.5	1.4	1.1	2.6	1.76	67.34
Niobium (Nb)	12	26	12	72.8	106.18	145.85
Neodymium (Nd)	—	—	—	118.4	238.13	201.01
Nickel (Ni)	44	19	47	5.5	1.06	19.43
Lead (Pb)	17	17	17	18.3	19.27	104.88
Rubidium (Rb)	112	110	84	11.2	2.55	22.66
Antimony (Sb)	0.2	0.31	0.4	0.47	0.43	92.62
Scandium (Sc)	13.6	7	14	11.12	11.9	107.05
Selenium (Se)	—	—	—	5	0	0
Tin (Sn)	5.5	2.5	2.1	5.25	5.47	104.2
Strontium (Sr)	350	316	320	463.88	227.84	49.11
Tantalum (Ta)	1	1.5	0.9	4.48	6.52	145.51
Terbium (Tb)	—	—	—	1.52	2.48	163.32
Tellurium (Te)	—	—	—	0.5	0	0
Thallium (Tl)	0.75	0.75	0.9	0.5	0	0
Vanadium (V)	107	53	97	146.75	199.68	136.07
Tungsten (W)	2	1.4	1.9	3.5	5.5	157.23
Zinc (Zn)	71	52	67	36.25	49.75	137.24
Zirconium (Zr)	190	237	193	11,139	23937.83	214.9

(Figs. 10e–10h) show cracks and fractures, suggesting the energy required for sediment transport is sufficient to crack the grains. Consequently, the sediment particles were probably subjected to high-energy surf zones as indicated by conchoidal to sub-conchoidal fractures that developed due to wave action. However, the cracks may form due to radiation damage in monazite and zircon. The geomorphic processes that played a major role in sediment transportation should have high velocity and turbidity. Recent data have been obtained to revise the concept of heavy minerals abundance as a result of a simple cycle of marine transgression and regression (Hou et al., 2003). Subsequent to the marine transgressions and regressions, other factors such as storm surges and cyclones have also enhanced the abundance of heavy minerals present (Mohanty et al., 2021b).

5. Discussion

The current study has confirmed that the Pulmoddai region, consists of abundant heavy minerals such as zircon, ilmenite, and monazite, when compared to the southern (Thiriyai region) and the northern coastal zones (Kokkilai lagoon region). In addition, the Pulmoddai region is enriched in Hf, V, Y, Zr, and Cr. V can plausibly be associated with ilmenite (Grammatikopoulos et al., 2002). Hf and Zr show a greater abundance due to the presence of zircon, in these beach placers. The REEs, Hf, Y, Zr, and Th are immobile in nature (Rollinson, 1993) and their mobility is controlled by mineralogical changes that take place during weathering. Immobile elements such as REEs, Zr, Hf, and Sn may be mechanically distributed according to grain size. Low field strength elements (LFSEs) such as Fe, Mn, and Pb are more mobile during diagenesis (Rollinson, 1993). All the studied samples have a high silica content and relatively low Ca and Fe content. However, calc-silicate rocks have comparatively high manganese concentrations (437–754 ppm) and high Y, La, Sr, and Ba content (He et al., 2018). Elevated concentration of LREEs and comparatively lower value of HREEs confirm the presence of monazite and the depletion of xenotime in the sediment (Bern et al., 2016). The depleted HREE content indicates that the primitive magma had plausibly generated from deeper subsurface depths

Table 5
Concentrations of REEs (ppm) from the analyses of beach sand samples and comparison with upper continental crust (UCC) values derived from surface exposure, sedimentary, and loess data.

Element	Taylor and McLennan (1985, 1995) (ppm)	Wedepohl (1995) (ppm)	Rudinick and Gao (2003) (ppm)	This study (Mean) (ppm)	SD (ppm)	RSD (%)
Lanthanum (La)	30	32.3	31	170.45	348.44	204.42
Cerium (Ce)	64	65.7	63	308.8	633	204.98
Praseodymium (Pr)	7.1	6.3	7.1	34.14	70.04	205.17
Neodymium (Nd)	26		27	118.46	238.13	201.01
Samarium (Sm)	4.5	4.7	4.7	17.11	32.94	192.5
Europium (Eu)	0.88	0.95	1	1.41	1.55	110.05
Gadolinium (Gd)	3.8	2.8	4	12.49	22.26	178.22
Terbium (Tb)	0.64	0.5	0.7	1.52	2.48	163.32
Dysprosium (Dy)	3.5	2.9	3.9	7.34	10.74	146.32
Holmium (Ho)	0.8	0.62	0.83	1.43	2.08	145.21
Erbium (Er)	2.3		2.3	4.93	7.38	149.79
Thallium (Tm)	0.33		0.3	0.89	1.42	158.84
Ytterbium (Yb)	2.2	1.5	2	6.57	10.71	162.93
Lutetium (Lu)	0.32	0.27	0.31	1.26	2.22	175.5
Yttrium (Y)	22	21	21	39.11	55.3	141.39

Table 6
Elemental abundances in sediments from the study area compared with those in literature data (UCC (Rudinick & Gao, 2003); ASV (average shale value): Turekian & Wedepohl (1961); sedimentary rock (S.Rock): Yudovich & Ketris (2009); Post Archean Australian Shale (PAAS): Taylor & McLennan, 1985).

Sample	∑REE	∑LREE	∑HREE	∑LREE/∑HREE	Th/U	Eu/Sm	Ce/La	La/Yb
P01	4,099.2	3,984.1	115.2	34.6	8.4	0.05	1.81	31.3
P02	201.2	190.9	10.2	18.8	6.3	0.14	1.75	18.29
P03	99.8	92.8	6.9	13.3	6.3	0.21	1.74	13.05
P04	198.3	188.1	10.3	18.3	7.3	0.16	1.77	18.42
P05	280.6	265.7	14.9	17.8	7.1	0.12	1.77	17
P06	424.6	405.9	18.7	21.7	7.1	0.1	1.86	19.43
P07	79.6	71.69	7.9	9.1	4.2	0.27	1.84	9.58
P08	111.4	103.9	7.5	13.7	5.9	0.22	1.76	16
Mean	686.8	662.8	23.9	18.4	6.6	0.2	1.8	18
SD	1,294.0	1,259.4	34.6	7.2	1.2	0.07	0.04	6
Median	199.7	189.5	10.2	18.1	6.675	0.15	1.77	17.6
Min	79.63	71.7	6.98	9.02	5.97	0.05	1.74	9.6
Max	4,099.18	3,984.04	115.2	34.6	8.93	0.22	1.86	19.43
UCC	106	99	7.21	13.7	3.89	4.7	2.03	15.5
ASV	166	157	8.9	17.7	3.24	6.4	0.64	35.4
S.Rock	96.5	89.5	6.98	12.8	2.26	5.85	1.63	16
PAAS	132	124	8.73	14.2	4.84	5.09	2.11	13.6

where the residual phases were dominated by plagioclase rather than garnet. The highest (total) concentrations of Ce, La, Sc, and Y are mostly related to fine grained sediment, signifying that the distributions of these elements are controlled by the Kokkilai Lagoon hydrodynamics (Martin et al., 2016). The concentrations of radioactive elements (U and Th) and REE are concordant in terms of the observed enrichment pattern. Th and REE enrichment depend on the mineralogy of the beach placers. The heavy minerals are derived from the WC charnockite–granite gneiss zone, that lies adjacent to the coastal area.

In Table 8, an intercomparison of the mineralogy and the parent rock composition of different Th-REE-rich heavy mineral/monazite beach placer deposits around the Indian subcontinent is listed. A striking similarity has been observed in the radionuclide concentration, mineralogy and rock composition of the Sri Lankan beach placers (Withanage & Mahawatte, 2013) with the beach placers along the eastern part of the Indian peninsula (Ghosal et al., 2017; Kannan et al., 2002; Khan et al., 2021; Mohanty et al., 2021a; Palaparthi et al., 2017; Sengupta & van Gosen, 2016) and the Bangladesh coast (Sasaki et al., 2015; Zaman et al., 2012). As Sri Lanka was connected to the eastern side of India prior to the rift between India and Antarctica at > *ca.* 130 Ma (Katz, 1978; Ratheesh Kumar et al., 2020), a few heavy mineral grains may be the product of charnockite and khondalite present along the eastern ghat of India. Further geochemical and specifically geochronological studies based on the foregoing observation can provide insight about the debatable paleo-position of Sri Lanka with respect to India.

The angularity of sediment samples confirms that the source rock is close to the current placer deposits. Heavy minerals show a much poorer roundness due to river deposition than that observed in texturally matured grains from aeolian and coastal environments (Dill, 2007). The Yan Oya River, Mee Oya River, Churiyan Aru River, and Maa Oya River which run close to the Pulmoddai area and their perennial nature due to specific monsoonal patterns (Amalan et al., 2014; Mallik et al., 1987), account for the transportation and subsequent enrichment of heavy minerals in the study area (Fig. 1). Monsoon rainfall in north-eastern Sri Lanka has a direct effect on the sediment flux of the river. The river basin is covered by the Wanni Complex that is dominated by charnockitic gneisses, granitoid gneisses, and granites with rare intercalations of metasediments, mainly calc-silicate rocks and quartzites (Bataploa, 2020). Monazite, which occurs as placer deposits on the beach, generally appears as an accessory mineral in granites, gneisses, and other igneous and metamorphic rocks (Wall, 2014). The charnokites found near the study area consist mainly of Orthopyroxene (Opx) – Clinopyroxene (Cpx) – Plagioclase (Pl) – Hornblende (Hbl) –Quartz (Qtz) and Garnet (Grt) – Biotite (Bt) – Cpx – Pl – Qtz along with minor amounts of magnetite, ilmenite, apatite, rutile, and zircon (Fernando et al., 2003). The rock sample R1 which is

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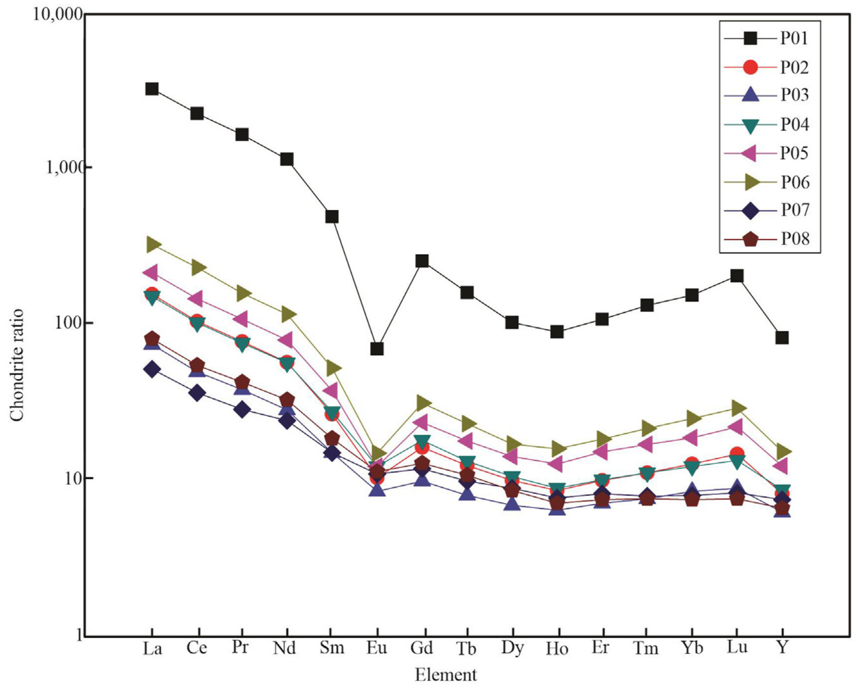

Fig. 8. REE chondrite plot of the samples studied (note: La = lanthanum, Ce = Cerium, Pr = praseodymium, Nd = neodymium, Sm = samarium, Eu = Europium, Gd = gadolinium, Tb = terbium, Dy = dysprosium, Ho = holmium, Er = erbium, Tm = thulium, Yb = ytterbium, Lu = lutetium, Y = yttrium).

Table 7
Measured concentrations of radioactive elements (ppm) from the beach sand samples and comparison with upper continental crust (UCC) values derived from surface exposure, sedimentary, and loess data.

Element	Taylor and McLennan (1985, 1995)	Wedepohl (1995)	Rudinick and Gao (2003)	This study (Mean)	SD	RSD
Uranium (U)	2.8 ppm	2.5 ppm	2.7 ppm	9.39 ppm	17.72 ppm	188.55%
Thorium (Th)	10.7 ppm	10.3 ppm	10.5 ppm	74.26 ppm	150.48 ppm	202.63%
Potassium (K)				0.49%	0.12%	24.47%

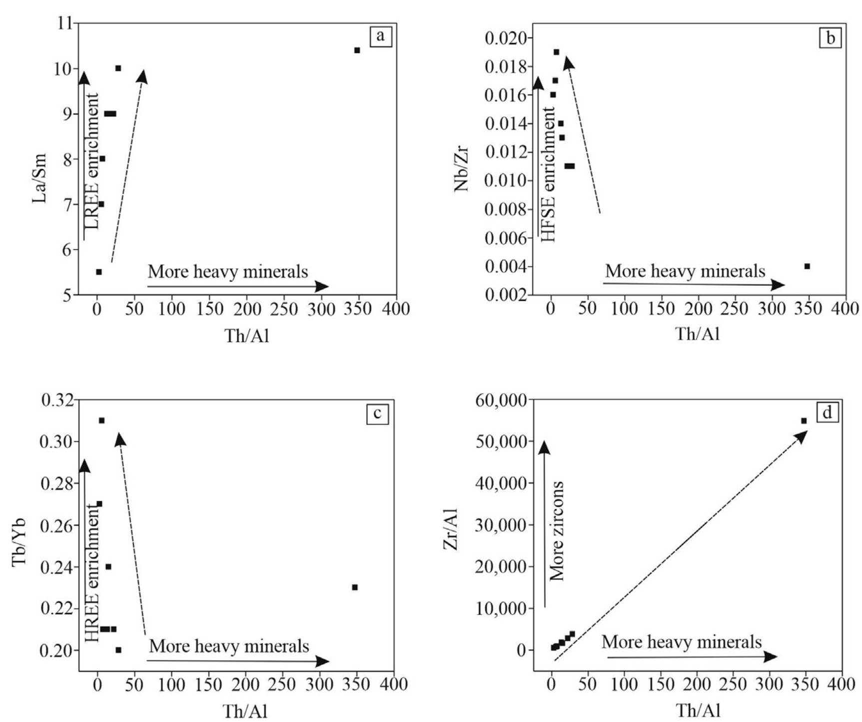

Fig. 9. Plots showing deviations of trace element ratios of the beach sediment. (a) Th/Al vs. La/Sm representing the relation between LREE enrichment and heavy mineral accumulation. (b) Th/Al vs. Nb/Zr showing HFSE enrichment and heavy mineral accumulation. (c) Th/Al vs. Tb/Yb presenting HREE enrichment compared to heavy mineral accumulation. (d) Zr/Al vs. Th/Al presenting enrichment of zircon compared to heavy mineral accumulation.

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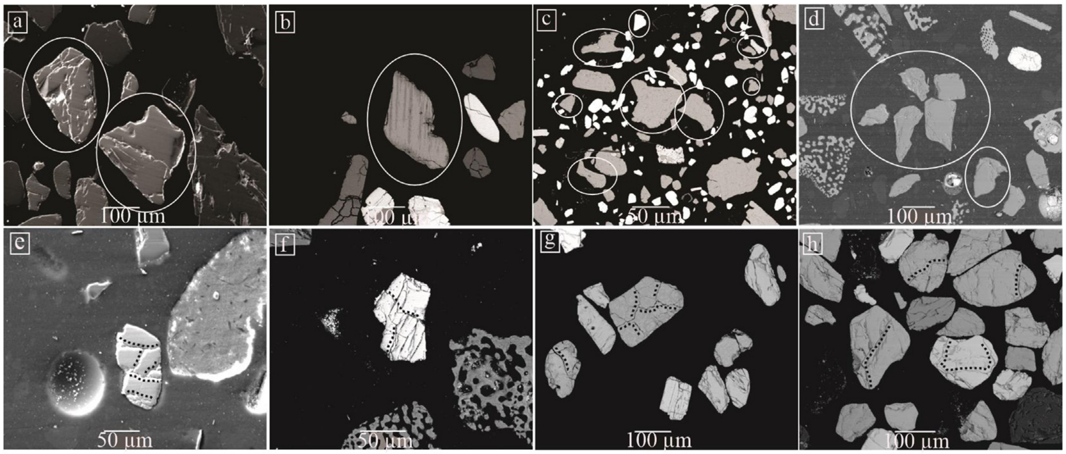

Fig. 10. Textural characteristics of heavy minerals from the coastal sands of Sri Lanka.

Table 8
Comparison of radioactive elements found in beach placers in north-east Sri Lanka to worldwide conditions (note: Mnz = monazite, Zr = zircon, Bst = bastanasite, Alnt = allanite, Rtl = rutile, Xnt = xenotime, and HM-heavy metals).

Location	Mineralogy	Lithology	Reference
Malaysia-Black sand	Mnz and other HM	Granitoids and Basalts	Khandaker et al. (2019)
Bangladesh-Cox Bazar	Zr, Mnz, Rtl	Granites, Basalts (kinberlite), Amphibolite schists	Zaman et al. (2012)
South East Bangladesh (Cox Bazar)	Zr, Mnz, Rtl	Granites, Basalts (kinberlite), Amphibolite schists	Sasaki et al. (2015)
India-Odisha	Mnz, Xnt, Rtl, Zr	Khondalites, Charnockites, Migmatites, Anorthosites, Granites, and Pyroxene granulites	Ghosal et al. (2017)
India-Andhra Pradesh	Mnz, Xnt, Zr, Alnt, Bst	Khondalites, Charnockites, Granitic gneiss	Palaparthi et al. (2017)
India-Tamil Nadu (north)	Grt, Mnz	Charnockites, Pyroxene granulites, Granites, Garnetiferous gneisses	Kannan et al. (2002)
Srilanka(SW coast)	Mnz (Ce,Nd), Xnt, Bst, Alnt, rinkite	Charnockite, Garnetiferous Granulite and Gneiss, Granite	Withanage and Mahawatte. (2013)
North-East Sri Lanka	Mnz, Zr, Ilm. Rt, Xnt	Charnockite, Calc-silicate rocks	This study

collected from the granite gneiss zone (Figs. 1b and 1c), mainly consists of Qtz–Cpx-Orthoclase with minor amounts of zircon, biotite, monazite, and iron oxide (Fig. 10). Another rock sample denoted R2 from the charnockitic zone near the Yan Oya River has been collected (Figs. 1b and 1c) and it is mainly pyroxene bearing quartzo-feldspathic rock with minor amounts of rutile, ilmenite, and zircon (Fig. 10). It is evident that the heavy minerals may be the product of weathering of charnockite and granite gneiss present near the source area and the Yan Oya River (Fig. 6). The other secondary rivers such as the Mee Oya, Churiya Aru, and Maa Oya of the Kokkilai Lagoon are the primary means of transport of heavy minerals, resulting in the greater enrichment observed in the beach sands of the Pulmoddai region when compared to the Thiriyai region.

6. Conclusions

The current study undertaken in the Pulmoddai area, north eastern part of Sri Lanka, has great significance in delineating the favorable depositional environment and the role of the geomorphic processes responsible for its formation and the subsequent enrichment of heavy minerals. The current study would also help in exploring suitable regions for rare earth-enriched beach placers. The main findings of the current study are highlighted as follows.

1) The heavy minerals in the study area are primarily monazite, zircon, rutile, and ilmenite, and they are mainly present in the surface sediment in the Pulmoddai and Thiriyai areas in Sri Lanka.
2) The enrichment of Hf, V, and Zr as compared to the mean value of the UCC suggests the abundance of zircon and ilmenite. The REE and HFSE elements exhibited a positive correlation with heavy mineral abundance.
3) The majority of the heavy mineral grains which were angular to sub-angular in shape and reasonably homogenous in composition with minor variations plausibly are the result of short distance fluvial transport. The angular nature of the sediment grains indicates that the source rock is proximal and the distribution of the detritus is mostly controlled by the regional monsoon related waves and the associated energy conditions.
4) The concordant geochemical nature of radioactive minerals and REEs, suggest that the heavy minerals are derived from charnockite–granite gneisses whereas the calc-silicate rocks are secondary source rocks, as evidenced by the presence of higher Mg.
5) The current study indicates that the coastal sediments along the north-east part of Sri Lanka are primarily transported by the Yan Oya River and the input of the Kokkilai Lagoon opening to the sea, thereby providing data representing the hinterland lithology. The relative abundance and lateral distribution of heavy minerals of economic importance in the study area are mainly controlled by the hydraulic effects of the lagoon and fluvial transportation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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