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RESEARCH ARTICLE

Gamma Ray Spectroscopy Analysis of Sediments of Coastal Areas in Ennore, Tamil Nadu

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Abstract

Objectives: This research focuses on the determination of the natural radionuclides radium, thorium, and potassium in the twenty-six sediment samples collected at the sea, beach, and creek regions of Ennore Port. **Methods:** The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were determined using gamma ray spectrometry with a high-purity germanium (HPGe) detector. **Findings:** The average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were in the descending order of ^{40}K (397.58 Bq kg^{-1}) > ^{232}Th (65.83 Bq kg^{-1}) > ^{226}Ra (18.28 Bq kg^{-1}). The estimated average values of radiological parameters such as radium equivalent activity (143.04 Bq kg^{-1}), absorbed dose rate (64.91 nGy h^{-1}), annual effective dose equivalent (0.32 mSv y^{-1}), and external hazard index (0.39) were lower than the respective world average values, reported by United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000). Moreover, the representative level index and annual gonadal dose equivalent were slightly higher than the world average value. Hence, this research proved that the study area is radiologically safe for humans and the environment. **Novelty:** A location and sample collection-based novelty is approached to carried out the work. Sea sediments were also collected along with samples from creek and beach regions in order to examine the dispersion of natural radionuclides from land to marine environments. The samples from the beach and creek regions were collected using a Peterson grab sampler. Especially in the sea region, the samples were collected using a Van Veen grab sampler at a depth of 4 m and a distance of 10 m parallel to the shoreline.

Keywords: Natural radioactivity; Sediment; Ennore; Gamma ray spectrometry; HPGe detector; Radiological parameters

1 Introduction

Research on radioactivity measurements in coastal areas is pivotal to comprehending the prospective environmental effects of both natural and artificial sources of radioactivity. The primordial radionuclides (naturally occurring radioactive materials, NORM) such as ^{226}Ra, ^{232}Th, and ^{40}K have spanned throughout the earth's crust, rocks, and soils at various concentrations during the nucleosynthesis process, which have half-lives of more than 10^9 years, equivalent to the earth's half-life[1–3]. The offspring radionuclides in ^{238}U series are disregarded as ^{226}Ra and its offsprings provide approximately 98.5 % of the external gamma radiation[4,5]. These natural radionuclides cause alpha, beta, and gamma-ray exposure to every living creature on earth. Since significant contribution comes from natural sources, substantially, artificial radionuclides such as ^{137}Cs, ^{95}Zr, and ^{90}Sr, technologically produced by humans (technologically enhanced naturally occurring radioactive materials, TENORM) are attributed to the gamma radiation[6,7]. The spatial distribution sequences of radium and thorium and their posterities were influenced by physical, chemical, geo-chemical, and biological interactions, which ultimately depended on the processes involved in the formation and movement of soil and sediment, rocks, and higher altitudes[8,9]. Sediments play a crucial role in the transit and storage of potentially harmful radioactive elements in aquatic ecosystems, as they are the primary source and sink of radioactive substances and pollutants[8,10].

Knowledge of environmental radioactivity is crucial for assessing the distribution of natural radionuclides, sources of radionuclides, effective dose received by humans, effects of human exposure, and level of natural radioactivity. The radiation exposure of humans can occur either externally (dermal contact) or internally (ingestion and/or inhalation). As well as the radionuclides, they can be transferred through the translocation of soil to plants and animals and, thereupon, to humans[7,11]. In India, many radioactivity measurements have been carried out on sediments from coastal regions like Kerala, Tamil Nadu, and Kalpakkam because of the deposition of zircon, monazite, and rare-earth-bearing minerals[8].

The drastic implementation of industrialization and urbanization causes the rapid development of contaminants[12]. Ennore is an industrial region where a number of enterprises have the potential to release radioactive elements into nearby bodies of water. Analyzing radioactivity contributes to determining the level of pollution brought on by industrial activities. The North Chennai thermal power plant, the North Chennai harbour, and the Manali industrial complex are significant economic resources in Ennore. Although they play a major economic role in Ennore, they predominantly contribute to environmental pollution due to the large quantity of toxic discharges. As these thermal power plant, harbour, and industries in Ennore are located near coastal areas, their effluents are discharged without proper waste management to nearby water bodies. Notably, fly ash and bottom ash can release NORMs during the coal combustion process in a coal-fired thermal power plant. These NORMs can have concentrations up to ten times higher than their initial value. The filtration system can only control the emission of fly ash by around 95%, the remaining are gravitated to the ground due to its higher density than air[13]. In addition to that, chemical elements, toxic heavy metals, non-volatile nuclides and volatile pollutants such as CO_2, SO_X, and NO_X are also released into the environment[14]. So that these discharged effluents can contribute to the sediment contamination and variation in the geochemical properties of sediments, in addition to that, increases the level of NORMs in the aquatic environment and then circulate to the aquatic biota and eventually enter the food chain[15–17]. In order to comprehend the natural background levels of radioactivity in the Ennore region, it is imperative to establish baseline data via radioactivity analysis. Future evaluations and comparisons might use this baseline as a point of reference. Therefore, the purpose of this work is (i) to evaluate the activity concentrations of naturally occurring radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in the sea, beach, and creek regions of Ennore, Tamil Nadu, India, using a high-purity germanium detector (HPGe), (ii) to assess the radiological impact of natural radionuclides on humans and environment in the study area through the calculation of radiological parameters, and (iii) to examine the relationship between radionuclides and radiological parameters.

2 Methodology

2.1 Geology of the Study Area

Ennore is situated along the Coromandel Coast in northeastern region of Chennai. Its proximity to the Bay of Bengal renders it a highly advantageous site for maritime operations. Ennore Creek inlet to the south and Pulicat Lake inlet to the north define the study region, which is a 25 km long length of coastal regions[12,18]. The convergence of fresh and saltwater occurs at the creek's mouth, causing semi-diurnal tidal activity in this area. In the creek arm that leads to Pulicat Lake, mangroves are also present in the area. Due to the abundance of oysters in the area, shellfishing is practiced there[19]. Ennore port has strong maritime activity and exists on the east coast of the Indian peninsula in the Bay of Bengal, 2.5 kilometres north of Ennore Creek[18]. As well, this port has various anthropogenic activities such as handling various types of cargo, including containers, coal, iron ore, and petroleum products, plastic and cane basket industries, and so forth. Numerous enterprises, such as manufacturing facilities

and thermal power plants, are located in Ennore. One significant power generating plant in the area that helps meet the region's energy needs is the Ennore Thermal Power Station (ETPS). Additionally, the Manali, Ennore–Tiruvottiyur, and Ambattur–Padi complexes, three industrial belts, encircle the research area[19]. Ennore estuary is surrounded by the Korattaliyar River, Kosasthaliyar River, Red Hills surplus canal, and Buckingam canal. Deltaic alluvial plains, cheniers, paleo lagoonal plains and strandlines, coastal sand dunes, beaches, beach cliffs, paleo barriers, paleo tidal flats, and mud flats, river mouth bars, abandoned river channels, and natural levees make up the area between S1 and C13. This area is also covered by Cretaceous to modern sedimentary strata[16,20].

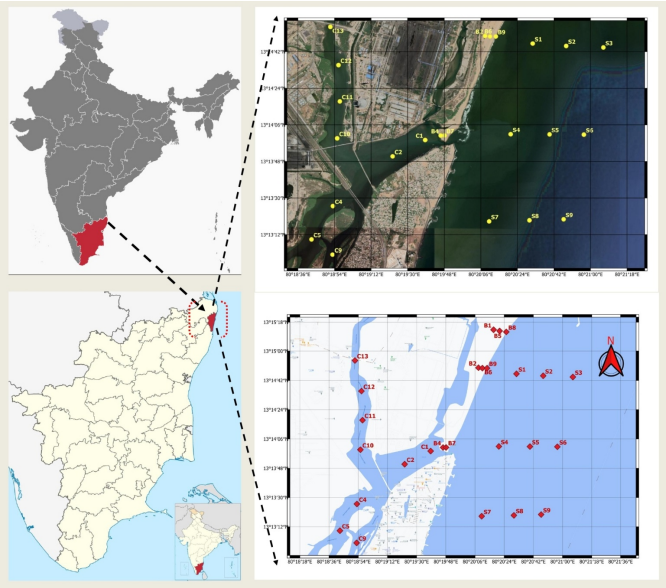

Fig 1. Location map of the Study area

2.2 Sample Collection and Preparation

Figure 1 depicts the sample collection sites (13°13'18.50" N, 80°20'10.10" E to 13°15'13.33" N, 80°20'17.43" E), while Table 1 provides geographical information for the locations. In the course of the pre-monsoon season, 26 sediment samples were collected from the three different regions of Ennore such as sea (S), beach (B), and creek (C) regions on the east coast of Tamil Nadu using a Peterson grab sampler. The sediment samples were collected from a distance of 10 m parallel to the shoreline and 4m depth in the sea region (9 samples) and also beach (8 samples) and creek regions (9 samples) of Ennore to examine the divergence of natural radionuclides from land to marine environment. The sediment samples were transferred to polythene bags and labelled properly with the corresponding sampling site. Thereupon these samples were transported to the laboratory and subsequently air-dried before larger stone shards or shells were manually picked out. Finally, samples were oven-dried for 2 hours at 105 °C to achieve a consistent weight before being ground into a fine powder with an agate mortar and pestle. A 250 μ mesh laboratory test sieve was deployed to sift sediment samples. Each sample was finally relocated in a volume of 100 cm^3 to a radon-indestructible PVC cylindrical container with a diameter of 6.5 cm and a height of 7 cm. To prevent radon gas from fleeing, containers were firmly sealed with vinyl tape around the screw neck. After that, samples were held for four weeks to allow radioactive equilibrium to be achieved between ^{238}U (^{226}Ra) and ^{232}Th (^{228}Ra) and their posterities[20].

Table 1. Latitude and Longitude values of the collected sediment samples in Ennore, Tamil Nadu, India

S. No	Sample ID	Latitude	Longitude
1	S1	13°14'46.02" N	80°20'31.56" E
2	S2	13°14'44.83" N	80°20'48.04" E
3	S3	13°14'44.09" N	80°21'60.35" E
4	S4	13°14'10.40" N	80°20'20.71" E
5	S5	13°14'10.32" N	80°20'39.85" E
6	S6	13°14'10.20" N	80°20'56.76" E

Continued on next page

Table 1 continued

7	S7	13°13'18.50" N	80°20'10.10" E
8	S8	13°13'19.04" N	80°20'29.96" E
9	S9	13°13'19.54" N	80°20'46.77" E
10	B1	13°15'13.33" N	80°20'17.43" E
11	B2	13°14'49.77" N	80°20'80.12" E
12	B4	13°14'00.75" N	80°19'46.26" E
13	B5	13°15'12.59" N	80°20'21.18" E
14	B6	13°14'49.51" N	80°20'10.55" E
15	B7	13°14'00.79" N	80°19'48.15" E
16	B8	13°15'12.01" N	80°20'25.27" E
17	B9	13°14'49.51" N	80°20'13.40" E
18	C1	13°13'58.55" N	80°19'38.62" E
19	C2	13°13'50.50" N	80°19'22.59" E
20	C4	13°13'26.04" N	80°18'53.16" E
21	C5	13°13'90.64" N	80°18'42.67" E
22	C9	13°13'20.15" N	80°18'52.99" E
23	C10	13°13'59.38" N	80°18'55.29" E
24	C11	13°14'17.55" N	80°18'56.60" E
25	C12	13°14'35.42" N	80°18'55.95" E
26	C13	13°14'54.22" N	80°18'51.95" E

2.3 Gamma Ray Spectrometry

Gamma-ray spectroscopy has been used to evaluate the activity content of sediment samples primarily because it is a less time consuming and non-destructive technology. Using a combination of a high-resolution gamma ray spectrometer of the model GEM25P ORTEC and a coaxial hyper-pure germanium (HPGe) detection system (Nuclear Research Lab, Department of Physics, DAV College, Amritsar, Punjab 143001, India), the gamma ray spectra of each sample were scrutinized individually. To lower the system's background level, the detector is shielded by 4" lead on all sides. The secondary standard source of radium ore in the geometry accessible for sample counting is used to calibrate the system's efficiency. The 1.33 MeV gamma ray line of ^{60}Co has an energy resolution of 1.85 keV FWHM and a photo peak efficiency of the detector of roughly 30 %. Sources of ^{60}Co and ^{137}Cs were used to perform the calibration. Following that, the samples were tallied using an HPGe detector that is based on a high-resolution gamma spectrometry device. It has been possible to measure the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in sediment, from the 1764 keV gamma line of ^{214}Bi, the 2610 keV gamma line of ^{208}Tl, and the 1460 keV photo peaks, respectively [9,21]. On account of accuracy, each sample was counted for 10,000 secs in three trails on the high purity HPGe detector. The minimum detectable activity for the radionuclides ^{226}Ra, ^{232}Th, and ^{40}K are 3 Bq kg^{-1}, 3 Bq kg^{-1}, and 30 Bq kg^{-1}, respectively, were calculated using the below equation.

$$MDA = \frac{L_D}{TI\varepsilon M} \tag{1}$$

Where, MDA is minimum detectable activity, L_D is detection limit ($L_D = L_C + K\,\sigma_D$), L_C is the critical level below which no signal can be detected, σ_D is standard deviation, K is error probability, T is counting time, I represent the emission probability of gamma ray photo peak, ε is efficiency of the detector, and M is the mass of the sample (dry mass in kg) [22].

2.4 Statistical Analysis

The statistical analyses such as Pearson correlation analysis (PC), Cluster analysis (CA) and Principal component analysis (PCA) were performed for the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K and radiological parameters such as Ra_{eq}, D_R, AEDE, I_γ, AGDE, and H_{ex}. These analyses help to examine the contribution of natural radionuclides to the radiological human exposure.

3 Results and Discussion

3.1 Activity Concentration in Sediment Samples

The natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K and their activity concentrations presented in the collected sediment samples were determined using the photopeaks obtained from the gamma ray spectrum. The conventional formula to calculate the

activity concentration of radionuclides using HPGe spectrometry was given below.

$$A\ (Bq\ kg^{-1}) = \frac{cps}{E \times I \times W} \quad (2)$$

Where, A stands for the activity concentration in Bq kg^{-1}, cps denotes the net counts per second, E is the efficiency of the detector, I stand for the intensity of the gamma ray, and W refers to the weight of the sample. The uncertainties due to counting errors occurring while the measurements were calculated using following equation.

$$\sigma = \sqrt{\frac{N_t}{T_t^2} + \frac{N_b}{T_b^2}} \quad (3)$$

where, σ refers the standard deviation, N_t is the sample counts, T_t is the counting time for the sample, N_b is the background counts, and T_b is the counting time for the background counts [23].

From **Supplementary table I**, the results of activity concentrations were corroborated the random dispersion of naturally occurring radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in the studied sea, beach and creek region of Ennore. Regional comparisons revealed that the sea and creek regions had higher mean activity concentrations of ^{226}Ra (26.75 and 16.50 Bq kg^{-1}) and ^{232}Th (96.43 and 64.40 Bq kg^{-1}) than the beach region (10.76 and 33.02 Bq kg^{-1}). In contrast, the beach region (423.93 Bq kg-1) exhibits a higher mean concentration of ^{40}K than the creek and sea regions (420.72 and 352.68 Bq kg^{-1}). The overall average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the sea, beach, and creek regions were 18.28, 65.83, and 397.58 Bq kg^{-1}, respectively. Correspondingly, they have the range 3.09±1.5 to 56.0±1.2, 08.06±1.9 to 317±3.3, and 216.56±5.4 to 541.85±10.2 Bq kg^{-1}. The activity concentrations of ^{226}Ra radionuclide were lower than the world average value of 35 Bq kg^{-1}, reported by, UNSCEAR (2000) [24,25], in all sampling locations, except S2, S4 (sea region), and C10 (creek region). While for ^{232}Th, with the exception of S5, S8 (sea region), B2, B4, B7 to B9 (beach region), and C1, C2, C9, C11, and C12 (creek region), the remaining samples have activity concentrations higher than the world average value of 45 Bq kg^{-1} [24]. The abundant radionuclide ^{40}K has a higher activity concentration in most of the samples than the world average value of 420 Bq kg^{-1}, with the exclusion of S2, S4, S7 to S9 (sea region), B5, B9 (beach region), and C2, C10 and C13 (creek region). Overall, the order of natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in the sediment samples was ^{40}K > ^{232}Th > ^{226}Ra. Comparatively, the high concentration of ^{40}K in the sediment samples was due to the abundance of ^{40}K in the earth crust, continental rocks, and many light minerals as well as the fact that the average concentration of ^{40}K was lower than the world average value of 420 Bq kg^{-1} [24]. The content of ^{232}Th was higher than the ^{226}Ra in all sampling locations. This variation in the activity concentration of natural radionuclides was due to the presence of an aquatic environment, the solubility and mobility of natural radionuclides, and the physical, chemical, and geo-chemical properties of sediment. Figure 2 shows the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the sediment samples.

Table 2. Comparison of activity concentration of ^{226}Ra, ^{232}Th, and ^{40} K in the present work with similar works around the world

Countries		No. of Samples	^{226} Ra	^{232} Th	^{40} K	References
Port Said, Egypt		44	12.37	6.16	223.30	[26]
Yemen		23	16.25 ± 0.63	23.80 ± 1.46	518.54 ± 45.84	[27]
Ghana		19	43 ± 6	22 ± 1	393 ± 74	[28]
Egypt		84	23.8	19.6	374.9	[29]
North Sulawesi		6	5.9	5.2	238.8	[30]
Namibia		20	175.59 ± 0.92	40.17 ± 27	349.66 ± 8	[31]
India	Bangladesh	10	94.39 ± 8.05	121.9 ± 6.2	498.0 ± 7.4	[32]
	Kerala	39	170.4 ± 8.4	547.3 ± 10.9	117.2 ± 16	[33]
	North Chennai to Pondicherry, Tami Nadu	21	50	32	543	[34]
	East Coast of Tamil Nadu	20	36.82	50.11	320.38	[7]
	Southeast Coast of India	79	75	182	298	[8]
	Ennore, Tamil Nadu	26	18.28	65.83	397.58	Present work
World average value		-	35	45	420	[24]

Table 2 represents the comparison of activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in the present work with the similar works around the world. From this, the mean activity concentration of ^{226}Ra in the present work is lower compared with the results reported in East Coast of Tamil Nadu, Southeast Coast of India, Ghana, Egypt, Namibia, Bangladesh, Kerala, and North Chennai to Pondicherry (Tamil Nadu),[7,8,28,29,31–34]. Comparatively, the activity of ^{232}Th is higher than the existing reports in East Coast of Tamil Nadu, Port Said (Egypt), Yemen, Ghana, Egypt, North Sulawesi, Namibia, and North Chennai to Pondicherry (Tamil Nadu)[7,26–31,34], but lower than Southeast Coast of India, Bangladesh, and Kerala[8,32,33]. The activity of ^{40}K is higher than the results of East Coast of Tamil Nadu, Southeast Coast of India, Port Said (Egypt), Ghana, Egypt, North Sulawesi, Namibia, Kerala[7,8,26,28–31,33], while lower than Yemen, Bangladesh, and North Chennai to Pondicherry (Tamil Nadu)[27,32,34].

3.2 Radiological Parameters

3.2.1 Radium Equivalent Activity (Ra_{eq})

The natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K are not evenly distributed in sediments. Regardless of whether they are present in an identical amount, discrepancies are observed in the emission of gamma dose rates[35]. Radium equivalent activity (Ra_{eq}) is a frequently used radiological hazard indicator that describes the combined effect of ^{226}Ra, ^{232}Th, and ^{40}K in sediments. It relies on the premise that a similar gamma radiation dose rate is yielded by 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th, and 4810 Bq kg^{-1} of ^{40}K. According to Beretka and Mathew (1985)[35], Ra_{eq} is calculated using the below equation.

$$Ra_{eq}\ (Bq\ kg^{-1}) = A_{Ra} + 1.43\,A_{Th} + 0.077\,A_K \tag{4}$$

Where, A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th,and ^{40}K, respectively, in Bq kg^{-1}.

As can be seen from **Supplementary table I**, the radium equivalent activity (Ra_{eq}) values ranged from 39.31 (B9) to 525.94 Bq kg^{-1} (S2), with an average of 143.04 Bq kg^{-1}. All samples in three regions did not exceed the world average value of 370 Bq kg^{-1}, excluding S2 (sea region) and C10 (creek region) locations. However, natural radionuclides do not pose any radiological hazards. Figure 2 shows the radium equivalent activity (Ra_{eq}) of different locations in the study area.

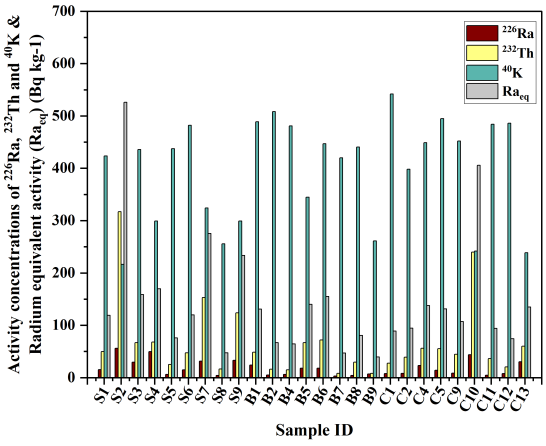

Fig 2. Sample ID versus Activity concentrations of ^{226}Ra, ^{232}Th,and ^{40}K & Radium equivalent activity (Ra_{eq}), (Bq kg^{-1})

3.2.2 Absorbed Gamma Dose Rate (D_R)

The absorbed gamma dose rate is the radiation exposure, which represents the amount of radiation energy absorbed per unit mass by the body, organ, or tissue[36]. The absorbed dose rate of gamma radiation is calculated by the following equation.

$$D_R\ (nGy\ h^{-1}) = 0.462\,A_{Ra} + 0.604\,A_{Th} + 0.042\,A_K \tag{5}$$

Where, A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th,and ^{40}K, respectively, in Bq kg^{-1}; 0.462, 0.604 and 0.0417 are the conversion factors which converts the activity concentration (in Bq kg^{-1}) to absorbed dose rate (in nGy h^{-1}), reported by UNSCEAR, 2000[24].

In this study, the calculated absorbed gamma dose rate (D_R) values given in **Supplementary table I** ranged from 19.37 nGy h^{-1} (B9) to 226.41 nGy h^{-1} (S2), with an average of 64.91 nGy h^{-1}. In some samples, in sea regions S2, S7, and S9, and in creek regions C10, the values were higher than the world average value of 84 nGy h^{-1} reported by the organization UNSCEAR, 2000 [24]. On the other hand, no samples in the beach region exceeded the reported value. Overall, the average value of the absorbed dose rate does not exceed the reported value and does not pose any hazardous effects on humans, animals, or plants in the study region. Figure 3 shows the absorbed gamma dose rate (D_R) of different locations in the study area.

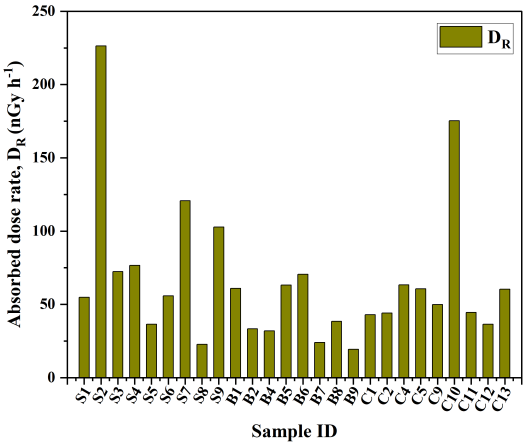

Fig 3. Sample ID versus Absorbed gamma dose rate (D_R) (nGy h^{-1})

3.2.3 Annual Effective Dose Equivalent (AEDE)

The annual effective dose equivalent was used to assess the radiological hazard to human health. It stands for the absorption of gamma dose rate per annual by the human. According to UNSCEAR (2000) [24], people spent their time 80% in indoor and 20% in outdoor. The absorbed dose rate is converted into annual effective dose equivalent by using the below equation [36–38].

$$AEDE\ (mSv\ y^{-1})\ =\ D_R\ (nGy\ h^{-1})\ x\ 8760\ h\ x\ 0.2\ x\ 0.7\ SvGy^{-1} x\ 10^{-6} \tag{6}$$

Where, AEDE (mSv y^{-1}) is the annual effective dose equivalent, D_R (nGy h^{-1}) is the absorbed dose rate in outdoor, 8760 is the exposure time for one year in hours (~365 days' x 24 hours), 0.2 is the outdoor occupancy factor and 0.7 $SvGy^{-1}$ is the dose conversion coefficient.

The calculated annual effective dose equivalent values, given in **Supplementary table I**, for the sediment samples were ranged between 0.02 and 0.28 mSv y^{-1}, with an average of 0.08 mSv y^{-1}. Majority of the samples have the lower AEDE value than the reported value of 0.07 mSv y^{-1}, by UNSCEAR (2000) [24], with the exception of S2-S4, S7, S9 (sea region), B5, B6 (beach region) and C4, C10 (creek region) samples. This result reflects the sediment do not pose any significant radiation hazards and safely used for construction purposes. Figure 4 shows the annual effective dose equivalent (AEDE) of different locations of the study area.

3.2.4 Representative Level Index (I_γ)

The radiological parameter, "Representative level index", is used to assess whether the materials are safe or not for construction purposes. It is the level of gamma radiation associated with the activity concentrations of natural radionuclides ^{226}Ra, ^{232}Th,and ^{40}K [39]. The below equation is used to determine the representative level index (I_γ),

$$I_\gamma = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \tag{7}$$

Where, A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th,and ^{40}K, respectively, in Bq kg^{-1}. According to European Commission (1999) [40], $I_\gamma \leq 1$ is the limit for the material to be used for construction purpose without posing any radiological

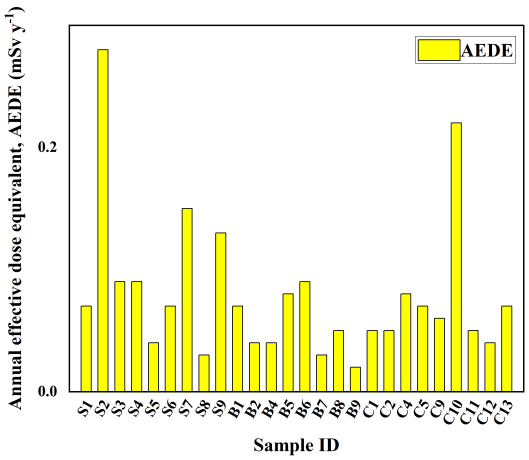

Fig 4. Sample ID versus Annual effective dose equivalent (AEDE) (mSv y^{-1})

hazards, while for the materials which has $I_\gamma \leq 2$ increases the annual effective dose by 0.3 mSv and for $2 \leq I_\gamma \leq 6$, cause the increase of annual effective dose by 1 mSv.

In this present study, I_γ values are obtained low values than 1 in the most of the samples. However, the samples S2 (3.69), S3 (1.16), S4 (1.21), S7 (1.96), S9 (1.66) (sea region), B5 (1.02), B6 (1.14) (beach region) and C4 (1.01), C10 (2.85) (creek region) have the I_γ values more than 1 which denote that these samples cause the increase of annual effective dose rate. The overall average value of I_γ (1.05) in three regions slightly excelled the reported value of 1, with a range of 0.31 to 3.69 for the collected sediment samples. The calculated values of the external hazard index for the studied samples were presented in **Supplementary table I**. Figure 5 shows the gamma representative level index (I_γ) of different locations of the study area.

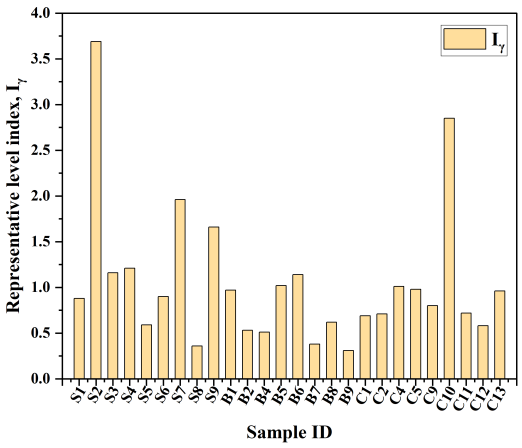

Fig 5. Sample ID versus Representative level index (I_γ)

3.2.5 Annual Gonadal Dose Equivalent (AGDE)

It is the genetic measurement of such organs as bone marrow, bone surface cells, and reproductive organs (gonads) of the population that received the yearly dose equivalent from the exposure to gamma radiation due to the natural radionuclides ^{226}Ra, ^{232}Th,and ^{40}K UNSCEAR, 2000 [24]. Therefore, the annual gonadal dose equivalent (AGDE) was calculated using the

following formula [7].

$$AGDE\ (\mu Sv\ y^{-1})\ =\ 3.09\ A_{Ra} + 4.18\ A_{Th} + 0.314\ A_K$$

The AGDE values for collected sediment samples, given in **Supplementary table I**, were ranged from 139.33 to 1565.92 μSv y^{-1}, with an average of 456.51 μSv y^{-1}. Unless the samples, S5, S8 (sea region), B2, B4, B7 to B9 (beach region) and C12 (creek region), all other samples were higher than the permissible limit, 300 μSv y^{-1} which is reported by UNSCEAR, 2000 [24]. This elevation of AGDE values is due to the variation in the activity concentrations of natural radionuclides ^{226}Ra, ^{232}Th,and ^{40}K in the study area. Figure 6 shows the annual gonadal dose equivalent (AGDE) of different locations of the study area.

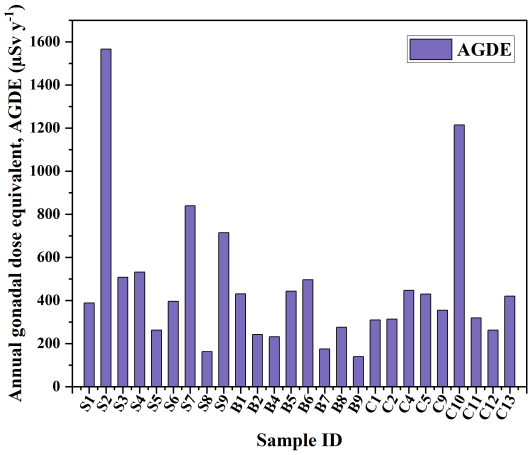

Fig 6. Sample ID versus Annual gonadal dose equivalent (AGDE) (μSv y^{-1})

3.2.6 External Hazard Index (H_{ex})

Another criterion used to determine a material's radiological compatibility is the external hazard index. That is, the measured activities in building materials are used to predict the radiation dosage that is projected to be supplied externally if a building is built using these materials. This index can be evaluated using the following equation [37].

$$H_{ex}\ =\ \frac{A_{Ra}}{370\ Bq/kg} + \frac{A_{Th}}{259\ Bq/kg} + \frac{A_K}{4810\ Bq/kg} \tag{9}$$

To keep the radiation, hazard insignificant, the value of H_{ex} must be less than one. The permissible limit of 370 Bq kg^{-1} for Ra_{eq}, is comparable to the highest possible value of unity for H_{ex} [6,7]. **Supplementary table I** shows the estimated amount of the external hazard index for the collected samples. The H_{ex} values varied from 0.11 (B9) to 1.42 (S2), with an average of 0.39, indicating that the samples satisfy the $H_{ex} < 1$ requirement. This means that processes involving the use of sediment samples are safe and do not expose people to high amounts of radiation. These sediments may not be harmful to the region's workers. Figure 7 depicts the external hazard index (H_{ex}) of different locations in the study area.

3.3 Statistical Analysis

The relationship between natural radionuclides and radiological parameters were identified using IBM Statistical Package for the Social Sciences version 23. Pearson correlation analysis is used to identify the interrelation between the variables whether strong or weak and positive or negative.

Pearson correlation analysis exhibits ^{226}Ra and ^{232}Th are mainly contributing in the exposure of gamma radiation than ^{40}K. The strong positive correlation coefficients (>0.7) (given in bold, in Table 3) between ^{226}Ra, ^{232}Th and radiological parameters revealed that ^{226}Ra and ^{232}Th are the predominant radionuclides which causing the radiation hazards to the humans and other living things; and ^{40}K has a negative correlation with the radionuclides and radiological parameters [34].

Hierarchical cluster analysis was performed using ward linkage method and squared Euclidean distance to get to know the grouping of radionuclides and radiological parameters [41]. From dendrogram (Figure 8), Cluster-I comprised of ^{232}Th and

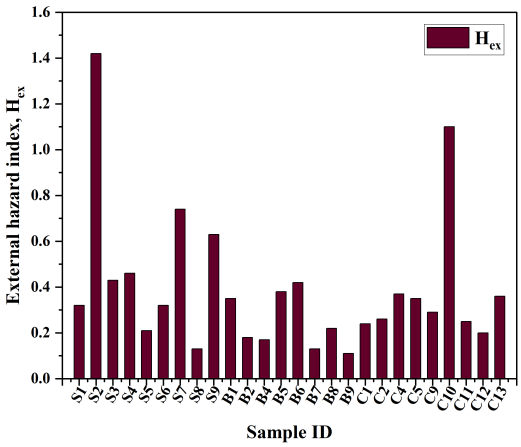

Fig 7. Sample ID versus External hazard index (H_{ex})

Table 3. Pearson correlation matrix for radionuclides and radiological parameters

Variables	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	D_R	AEDE	I_γ	AGDE	H_{ex}
^{226}Ra	1								
^{232}Th	**0.825**	1							
^{40}K	-0.606	-0.575	1						
Ra_{eq}	**0.857**	**0.997**	-0.545	1					
D_R	**0.857**	**0.996**	-0.533	**1.000**	1				
AEDE	**0.858**	**0.996**	-0.536	**1.000**	**1.000**	1			
I_γ	**0.852**	**0.996**	-0.532	**1.000**	**1.000**	**1.000**	1		
AGDE	**0.855**	**0.995**	-0.527	**1.000**	**1.000**	**1.000**	**1.000**	1	
H_{ex}	**0.856**	**0.997**	-0.548	**1.000**	**1.000**	**1.000**	**1.000**	**1.000**	1

^{226}Ra with all radiological parameters, and Cluster-II has ^{40}K, which was singly grouped. This demonstrated that ^{232}Th was the natural radionuclides which has more contribution in the gamma radiation exposure to humans followed by ^{226}Ra.

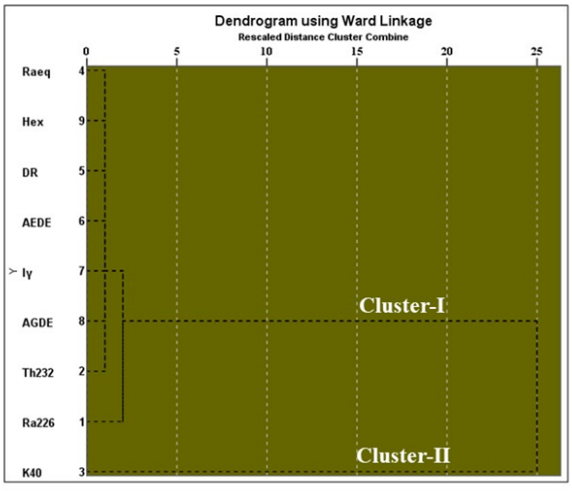

Fig 8. Hierarchical cluster analysis - Dendrogram using Ward linkage method

Principal component analysis used to deduct the dimension of set of variables significantly. By using varimax rotation method and principal component extraction, one component with a total variance of 89.832% was occurred [42]. This principal component has ^{226}Ra, ^{232}Th, Ra_{eq}, D_R, AEDE, I_γ , AGDE, and H_{ex}, which has eigen value more than 1, illustrated in Figure 9. These three analyses revealed the same results that ^{226}Ra and ^{232}Th are the main cause for the exposure of gamma radiation than ^{40}K in the study area.

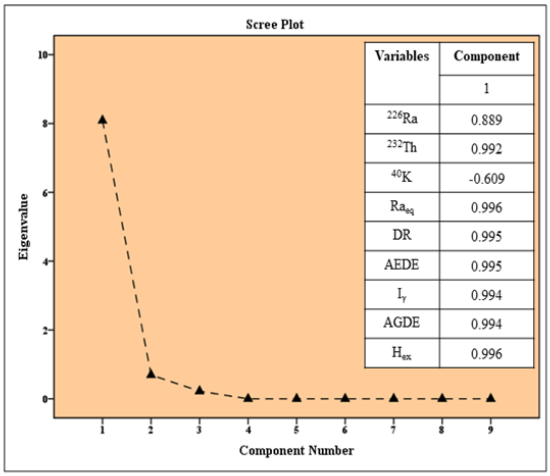

Fig 9. Scree plot with extracted component

4 Conclusion

The radiological investigation was done on the sediments of sea, beach and creek region in Ennore, East coast of Tamil Nadu, India using high purity germanium (HPGe) detector based on high-resolution gamma spectrometry system. The activity concentrations of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K presented in the sediment samples were determined. The abundance of these radionuclides in the present study area is in the form of ^{40}K (397.58 Bq kg^{-1}) > ^{232}Th (65.83 Bq kg^{-1}) > ^{226}Ra (18.28 Bq kg^{-1}). The average concentration of ^{226}Ra and ^{40}K were lower than the world average value while ^{232}Th was slightly elevated than the limit. The average value of radium equivalent activity in the samples (143.04 Bq kg^{-1}) was roughly as low as world average value. The average annual gonadal dose equivalent (456.51 μSv y^{-1}) was exceeded the world average value. The calculated average values of absorbed dose rate (64.91 nGy h^{-1}), annual effective dose equivalent (0.32 mSv y^{-1}), and external hazard index (0.39) were considerably lower than the world average value while average representative level index (1.05) was approximately equal to the reported limit. Comparatively, the results of radiological parameters exhibit the radiation risk in the order of Sea > Creek > Beach regions. Eventually, the findings reveal that the study area does not cause harmful radiological effect to the sailors, fishers, public people and tourists involved along this study area. This research serves as the foundation for more in-depth analysis that will be conducted in Ennore.

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