



Short Communication

Application of multivariate technique to evaluate spatial distribution of natural radionuclides along Tamil Nadu coastline, east coast of India



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Abstract

The study focuses on the spatial mapping of naturally occurring radionuclides along entire Tamil Nadu coast, which constitute nearly 30% of the east coast of India. In view of this, a total of 37 beach sediment samples are collected, and spectral measurement was carried out by High Purity Germanium detector. The average activity concentrations were found to be in the range of 34.33 ± 32.94 Bq kg⁻¹ for ²³⁸U, 51.55 ± 40.39 Bq kg⁻¹ for ²³⁸Th and 282.59 ± 84.45 Bq kg⁻¹ for ⁴⁰K. The contour maps showed the spatial distributions of each radionuclide and were drawn using the kriging method. Box-whisker plot shows the variation of activity concentrations of radionuclides, which follows the order ⁴⁰K > ²³²Th > ²³⁸U in all the sediment samples. To understand the complex relationships among the radioactive variables and its environmental classifications, a multivariate statistical technique such as correlation matrices, and cluster analysis were applied to the radioactive data sets.

Keywords Spatial distribution · Multivariate technique · Natural radioactivity · TN coastline · East coast of India

1 Introduction

Coastal sediments are weathering resistant residues of geological formations, transported to the coast by natural phenomena such as wind, river, and glaciers, and are deposited on the beaches by the coastal process [25, 66]. The naturally occurring radionuclides like ²³⁸U, ²³²Th, and ⁴⁰K, and their varying concentrations in soil and sediment depend upon the local geology of each region in the world [21]. These primordial radionuclides and their daughter products are gamma emitters, which are the main contributors to background radiation. As per the survey, 87% of the dose received by human beings is due to natural radiation sources [32]. The study on distribution of the natural radionuclides is quite helpful in understanding about dredging dispersal, accumulation rate, sediment logical

composition, and also the relation between grain size and density of the sediments [24, 36, 52, 53, 67]. In addition to this, it plays a major role in the adsorption studies with mineralogical, geochemical, and physicochemical parameters [11, 20].

Human beings are constantly exposed to background radiation due to natural radioactivity, which are coming from the different rocks, sand and cosmic rays. The average dose received from background radiation is around 2.4 mSv y⁻¹, which can vary depending on the geology and altitude. Peoples are living safely with a wide range of dose (1–50 mSv y⁻¹) from background radiation. The average background radiation level in Kerala, India, is about 12.5 mSv y⁻¹ due to the monazite sand deposit in the beach. Comparable levels occur in Brazil and Sudan, with average exposures up to about 40 mSv y⁻¹ to many

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people. Taking the individual average dose of 2 mSv y^{-1} , someone who lived to the age of 80 years would have accumulated 160 mSv of radiation from natural sources during their lifetime [60].

Radiological investigations have been carried out by many authors in small stretches along Tamil Nadu (TN) coast [4, 9, 15, 23, 30, 31, 45, 51, 55, 57], as per their specific mandate. In order to have the background data for the entire TN coastline, the current study carried out. This study focused on mapping of the spatial distribution of Naturally Occurring Radioactive Materials (NORMs) such as ^{238}U , ^{232}Th , ^{40}K and their radiological parameters throughout TN coastline, east coast of India, which may serve as baseline and background data for the entire coast. This study also used a multivariate statistical technique to the spatial data set for understanding the distribution pattern of radionuclides and their association with the geological characteristics.

2 Materials and methods

2.1 Study area and sample collection

The study area covers entire TN coastline of 1076 km from Palar of Tiruvallur (Longitude: 80.31849501 (E) Latitude: 13.56020134 (N)) to Thoothoor of Kanyakumari (Longitude: 76.93527495 (E) Latitude: 8.213782474 (N)) districts. The study area is almost like a flat terrain with a gentle slope towards the Bay of Bengal and characterized by alluvial and coastal deltaic plain sediments [33].

Details of samples collected (TN: 01 to 37) with their locations were depicted in Fig. 1. Total of 37 sediment samples (each of about 1 kg) collected from the upper few centimeters of the surface layer in between high tide and low tide line, as per the Guideline of International Atomic Energy Agency (IAEA) Trs No 295 [26] in polypropylene bags were taken immediately to the laboratory. After removing the stones, pebbles, the samples were mixed thoroughly for homogenization and sieved through $< 1 \text{ mm}$ mesh sieve (ASTM Standard C 1402-04). The samples were dried in an oven at $110 \text{ }^\circ\text{C}$ for 24 h until their weight became constant [12]. The dried samples were packed in pre-weighed cylindrical (250 ml capacity) plastic containers. The net weight of the samples was noted. The containers were closed, sealed to prevent the escape of gaseous radioactive daughter products and stored for more than 40 days to achieve secular equilibrium between ^{222}Rn and ^{226}Ra [5, 14, 49].

2.2 Gamma ray spectrometric analysis

All the samples were analyzed using High Purity Germanium (HPGe) detector of 30% relative efficiency and energy resolution of 1.85 keV at 1332.51 keV. Computerized HPGe detector consists of vertical dipstick geometry detector, having a spectroscopy amplifier and a multi-channel analyzer. Lead shielding of 10-cm thickness cover to the detector to reduce the background activities and cooling facility through liquid nitrogen. The certified reference materials sources from IAEA are RGU (Radiometric Reference for Uranium), RGTh (Radiometric Reference for Thorium) and RGK (Radiometric Reference for Potassium) were used for efficiency calculations [14]. The activity contents of the standards in 250 ml geometry are estimated within $\pm 3\%$ accuracy. The sample spectrum and background spectrum were acquired for 10,000 s [37]. The gamma-ray photo peaks at 1460 keV (^{40}K), 1764 keV (^{214}Bi) and 2614 keV (^{232}Th) were used for the estimation of activities of ^{40}K , ^{238}U , and ^{232}Th in samples. The minimum detection limit (MDL) for ^{238}U , ^{232}Th , and ^{40}K was found to be 8.3, 4.6 and 19 Bq/kg respectively for a counting period of 10,000 s.

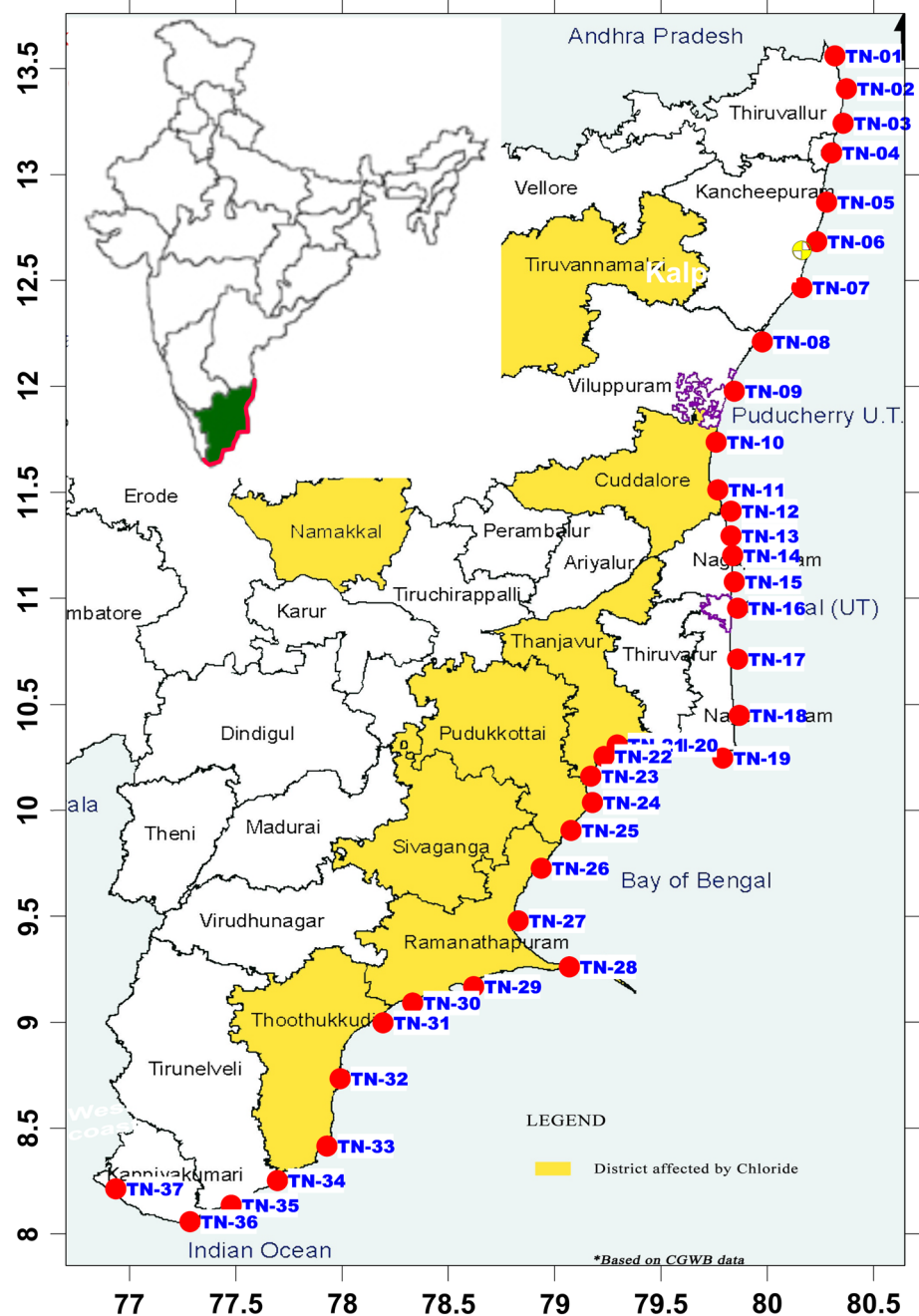
2.3 Spatial interpolation technique

Contouring is a way to visualize the proper spatial distribution of a variable concerning its position [35]. In spite of the availability of many interpolation techniques for plotting contouring map. In this paper, the kriging method used for gridding the data, which is one of the most flexible and accurate technology. Kriging produce visually attractive maps and useful in identifying a trend from irregularly spaced data [14, 37, 61]. This is also an exact or smooth interpolator depending on the user-specific parameter. It incorporates anisotropy and underlying trends efficiently and naturally.

2.4 Statistical analysis

Various multivariate statistical techniques have been used on collected data set from sediment samples, to formulate classifications of environmental variables without losing much information and it is an efficient tool to display complicated relationship among many variables [1, 14, 39]. The data sets of 111 observations including activity concentrations of ^{232}U , ^{238}Th , and ^{40}K as well as their radiological parameters were processed for multivariate statistical analysis, such as Correlation Matrix (CM), and Cluster Analysis (CA) [3, 4, 29, 53, 59]

Fig. 1 Map showing TN coast-line with station locations



using statistical software “Statistical Programme for Social Science” SPSS 19.0.

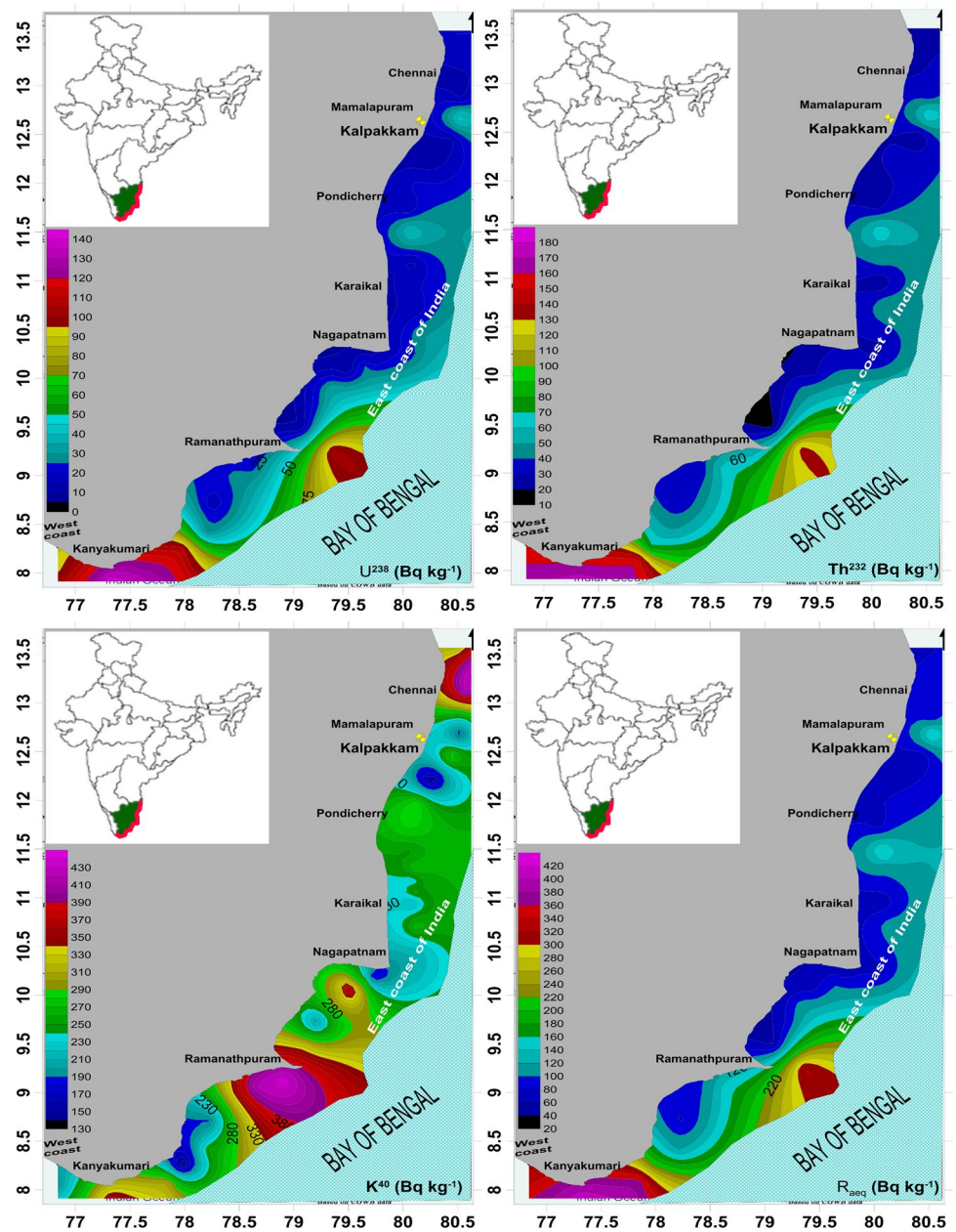
3 Result and discussions

3.1 Spatial distribution

The contour map shows the distribution of the radionuclide’s activity concentrations in the sediment samples analyzed. Golden software surfer 13.0 with the kriging

method was applied for spatial interpolation and gridding the data [37, 38]. The distribution graph of activity concentration of radionuclides is shown in Fig. 2. The radiological indices such as Radium equivalent activity (R_{eq}), Absorbed gamma dose rate (DR), Annual Gonadal Dose Equivalent (AGDE), Annual effective dose rate (HR), Activity utilization index (AUI), external hazard index (H_{ex}), internal hazard index (H_{in}), and Gamma radiation representative level index (RLI) were calculated as per [6, 13, 16, 18, 34, 46, 62, 63, 68].

Fig. 2 Spatial distribution of the ^{238}U , ^{232}Th , and ^{40}K activity concentrations (Bq kg^{-1}) of the sediment from TN coast, East Coast of India



The distribution of average activity concentration of ^{238}U (12.15–138.56) was $34.33 \pm 32.94 \text{ Bq kg}^{-1}$. It has been found that 16% of the samples are higher than the World average of 40 Bq kg^{-1} , and 19% of the samples are higher than the Indian average of 30 Bq kg^{-1} . The average activity concentration of ^{232}Th (15.08–173.03) was $51.55 \pm 40.39 \text{ Bq kg}^{-1}$, where, 27% of the samples were having higher than the world average of 45 Bq kg^{-1} , whereas 19% of the samples remained higher than Indian average of 65 Bq kg^{-1} . The average distribution of the activity concentrations of the ^{40}K (131.56–438.20) was $282.59 \pm 84.45 \text{ Bq kg}^{-1}$ and showed 6% of the samples are higher than the world average of 420 Bq kg^{-1} , whereas

16% of the samples remained higher than Indian average of 400 Bq kg^{-1} . The activity concentrations of ^{238}U and ^{232}Th were relatively high at Kanyakumari coastline, the southern part of TN coast, which could be due to the presence of ilmenite and monazite in the beach and also the presence of granite and granitoid rocks [2, 10]. On the other hand, the ^{40}K activity concentration was relatively high in northern part of the TN coastline and also in Rameswaram coastline, which could be attributed the presence of light minerals and dead shells, mica, and feldspar in the sediment of coastal area [17, 40, 44]. The Ra_{eq} concentrations in the southern part of the TN coast were high, which might be mainly due to high activity

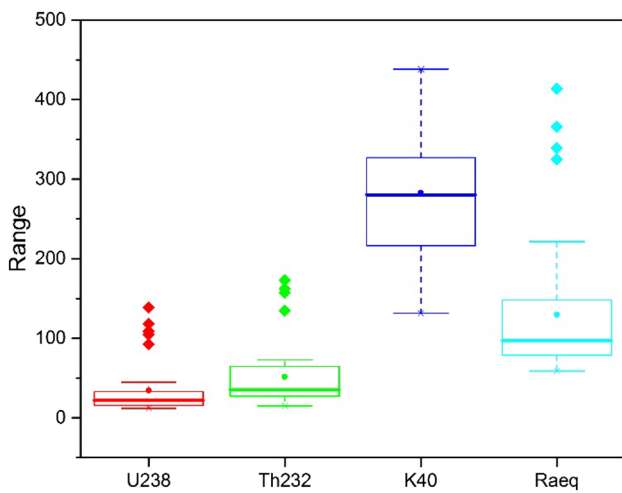


Fig. 3 Box-Whisker plot of the activity concentration of radionuclides in sediment samples (here the minimum and maximum limits, the middle point is median, and the x marks are the outliers of the radionuclide activity concentrations)

concentrations of ^{238}U and ^{232}Th . The distribution of activity concentrations is shown in Fig. 3. The wide range of variation is mainly due to the coastal morphological, lithological, and geological nature of the coastline [7, 8, 28]. In all the samples, the activities concentration was in the order $^{40}\text{K} > ^{232}\text{Th} > ^{238}\text{U}$ and ^{40}K largely dominated the total activity, which might be due to the greater abundance of the particular element in the earth crust and also in the sediments of TN coast [42].

The Box-whisker graph of the activity concentrations of the radionuclides in the TN coastline shown in Fig. 3. From the figure, it observed that the activity concentration of ^{40}K varied widely as compared with the other two radionuclides, whereas activity concentrations of ^{232}Th varied slightly higher than ^{238}U . It could be due to the presence of placer deposits monazite in the sediments of the TN coastline [41, 50, 53].

From the comparison Table 1, the ^{238}U activity was low compared with previous studies by Kannan et al. [30], SureshGandhi et al. [59] and Ajithra and Shanthy [3], whereas the ^{232}Th activity was high from the previous studies by Bukhari [15], Ramasamy et al. [47], Sivakumar et al. [54] and Ravisankar et al. [48]. But the present study ^{40}K activity observed high from the previous studies by Punniyakotti and Ponnusamy [41], Raja [43], Bukhari [15], Ramasamy et al. [47] and Ajithra and Shanthy [3].

Ra_{eq} values varied from 58.88 to 413.57, with an average $129.81 \text{ Bq kg}^{-1}$. As compare with NEA-OECD (1979), the present Ra_{eq} values do not exceed the admissible limit of 370 Bq kg^{-1} . So, it does not pose any negative radiological impact. D_R value ranged in between 53.91 and 346.46 with an average value of $110.90 \text{ nGy h}^{-1}$. Slightly higher D_R value noticed as compared with the world average value of 84 nGy h^{-1} UNSCEAR [62], which may be due to more contribution of ^{232}Th activity in the study area. AEDE ranged in between 0.07 and 0.43 with an average value of 0.14 mSv y^{-1} . As per UNSCEAR [63], the average AEDE for terrestrial radionuclides is 0.46 mSv y^{-1} , whereas, the present study area, the average value is 0.14 mSv y^{-1} ,

Table 1 Comparison of activity concentrations of present work with previous researchers of TN coastline, world, and India average

S/N.	Study area in TN coast	Activity concentration (Bq kg^{-1})			References
		^{238}U	^{232}Th	^{40}K	
<i>Tamil Nadu</i>					
01	South east coast	8.77	76.48	202.87	Punniyakotti and Ponnusamy [41]
02	Gulf of mannar	415	1372	541	Somasundaram [56]
03	Kalpakkam Coast	124	1613	358	Kannan et al. [30]
04	Pichavaram	25.9	65.1	190	Raja [43]
05	Palk strait	14.2	46.4	233.9	Bukhari [15]
06	North east coast	8.4	24.5	274.8	Ramasamy et al. [47]
07	South west coast	18.4	56.5	285	Satheeshkumar et al. [51]
08	North coast of Chennai	8.6	108	372	Inigo Valan et al. [27]
09	Thazhankuda to Kodyakkarai coast	3.67	37.23	387.17	Sivakumar et al. [54]
10	Pattipulam to Devanampattinam coast	BDL	14.29	360.23	Ravisankar et al. [48]
11	North east coast of Tamilnadu	35.12	713.16	349.60	SureshGandhi et al. [59]
12	Kanyakumari district, TN coast	62.90	279.53	108.35	Ajithra and Shanthy [3]
13	India	29	64	400	UNSCEAR [64]
14	Worldwide	35	30	400	UNSCEAR [62]
<i>Present study</i>					
13	Entire TN coastline, East coast of India	34.3	51.6	282.6	Present study

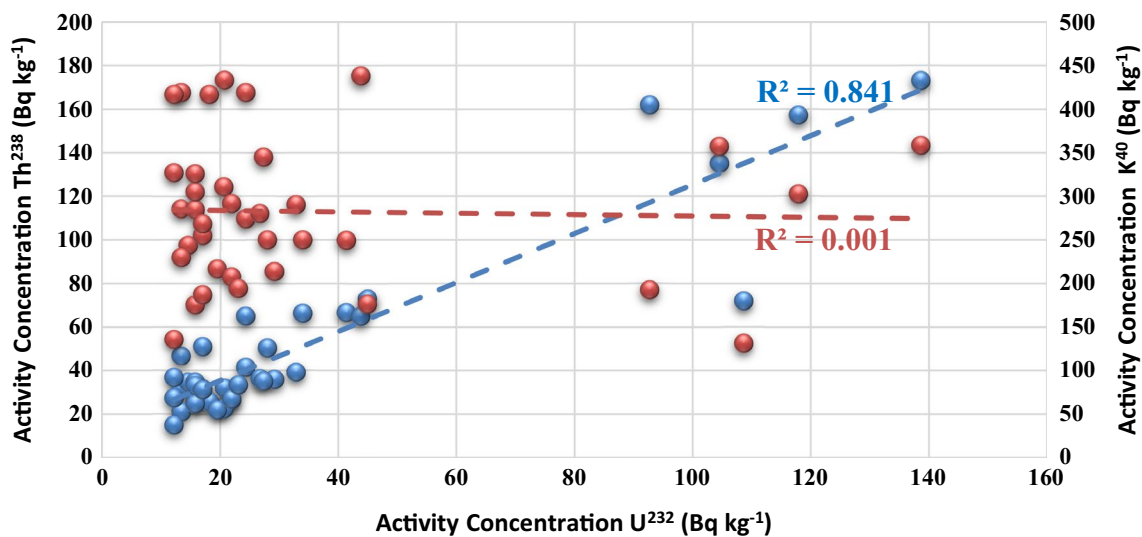


Fig. 4 Scatter plot showing the correlation among ^{238}U with ^{232}Th and ^{238}U and ^{40}K , in blue and red colour dots respectively, and also with R^2 value

which is less than the average world value. So, it satisfies the standards for radiation safety norms. H_{in} value ranged in between 0.19 and 1.49 with an average of 0.44 whereas, H_{ex} values ranged from 0.16 to 1.12 with an average of 0.35. It observed that both H_{in} and H_{ex} values are less than the permissible limit (< 1) as per UNSCEAR [64], which indicates, the study area does not possess any radiological health risk to the people living in nearby.

3.2 Multivariate statistical techniques

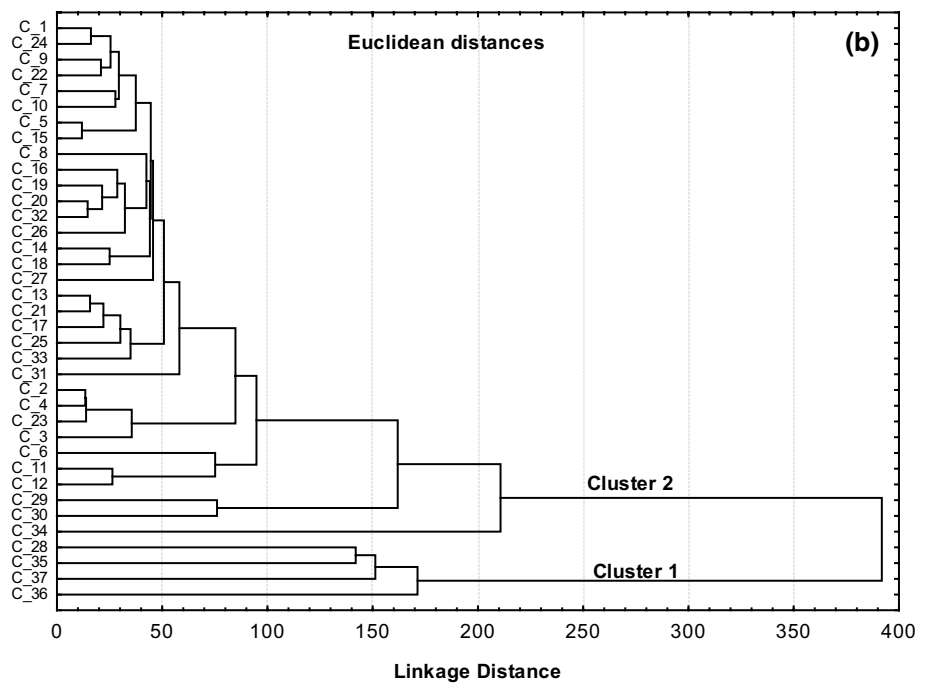
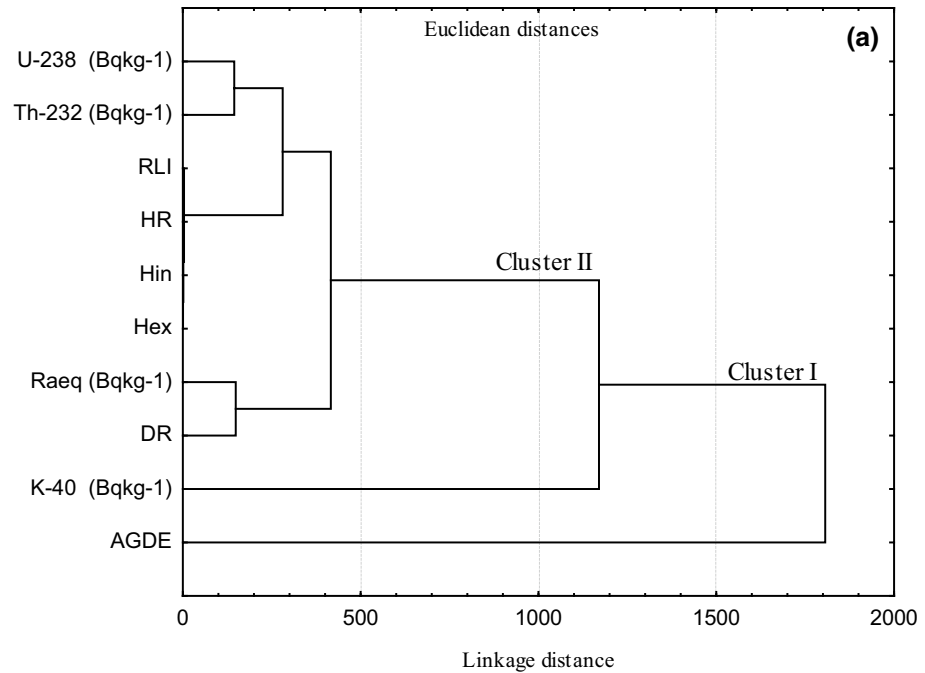
3.2.1 Pearson's correlation coefficient analysis

Pearson correlation analysis has been carried out to determine the inter associations and strength of association among radionuclides and radiological parameters. The results of the Pearson correlation coefficient of variables are given in Fig. 4. A very strong positive correlation ($R^2=0.841$) noticed between the ^{238}U and ^{232}Th . Which indicates a strong relationship between ^{238}U and ^{232}Th decay series in sediment which occur together in nature [1]. A weak correlation ($R^2=0.001$) observed between ^{40}K and uranium/thorium, indicating its origin from a different decay series in nature. The calculated radiological parameters Ra_{eq} , DR , H_{in} , H_{ex} showed a strong positive correlation with ^{238}U and ^{232}Th . It indicates that all these parameters are strongly associated with the activity of ^{238}U and ^{232}Th in the soils, and these radionuclides primarily contribute to the emission of gamma radiation in the study area. The results of the present study are also supported by previous studies [3, 53, 54, 59].

3.2.2 Cluster analysis

Cluster analysis (CA) for classifies a set of data into two or more mutually exclusive un-known groups, called clusters based on their nearness and similar nature [65]. It discovers a system of organized variable data set, into clusters, in which the members of the common share having similar properties and characters [58]. Cluster analysis is a suitable semi-quantitative practice for analyzing the data and determining the linkages [53]. In the present work, CA performed as hierarchical clustering using Ward's method to calculate the Euclidean distance between variables. Figure 5 shows the results of CA as a dendrogram, which was carried out by using (a) ten variables, including activity concentrations as well as radiological indices and (b) 37 variables of all sample locations. All the variables get grouped into two major statistically significant clusters. In Fig. 5a, Cluster I is associated with ^{238}U , ^{232}Th , and other radiological indices, whereas, cluster II is associated with ^{40}K . All of the natural radionuclides were represented as one group, i.e. cluster I with similar characteristics as they originate from ^{232}Th and ^{238}U series. ^{40}K identified in another group far from the other radioisotopes and grouped closely with the group of grain size distribution. The close relation between ^{238}U and ^{232}Th series members but not with ^{40}K was in accordance with the results described by several others [19, 22]. In Fig. 5b, Cluster I is associated with locations 28, 35, 36, and 37, whereas, cluster II is associated other 33 locations. From this, it supports the spatial distribution with high activity concentration in the southern part (cluster 1) of TN coast as compared with northern (cluster 2).

Fig. 5 Hierarchical clustering dendrogram **a** parameter wise and **b** station wise by complete linkage method for coastal sediment samples



4 Conclusion

The study covers the entire coastline of Tamil Nadu, and the data generated will act as the baseline for background radiation level for the entire TN coast. The contour map shows the spatial distribution of activity concentrations of each radionuclide along the coastline. The activity concentration of the ⁴⁰K dominated in the study area as compared with ²³²Th and ²³⁸U, which

might occur due to the greater abundance of potassium in the earth crust. The activity concentrations of radionuclides in most of the sediment samples remained below the world as well as the India average values as per UNSCEAR, which indicates that TN coastline is safe and does not possess any harmful radiological effect to the peoples living around it. The calculated radiological parameters Ra_{eq} , DR, H_{in} , H_{ex} showed that very strong positive correlation with ²³⁸U and ²³²Th, which indicates

that the above parameters are strongly associated with the activity of ^{238}U and ^{232}Th in the sediment.

Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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