

Distribution of Natural Radionuclides and Minerals in Beach Sediments from North East Coast of Tamilnadu, India

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Abstract: Thirty five sediment samples were collected from different beaches from north east coast of Tamilnadu starting from Port novo to Marina beach (Chennai) covering a length of about 200km. Gamma ray and Fourier transform infrared (FTIR) spectroscopic technique were used to identify the natural gamma emitting radionuclides (²³⁸U, ²³²Th and ⁴⁰K) and minerals respectively. The absorbed dose rate are calculated to understand the radiation hazard and compared with internationally recommended values. The major minerals such as quartz, feldspar in different structures and kaolinite and some trace minerals are identified. The relative distribution of major minerals is determined by calculating extinction co-efficient and results are discussed. The correlation analysis is carried out between the radioactivity parameter and extinction coefficient of major minerals. The obtained correlations are presented and discussed.

Key words: Natural Radionuclides • Absorbed dose rate • Minerals • Beach sediments

INTRODUCTION

The natural environmental radioactivity depends mainly on local geological and geographical conditions. Uranium (²³⁸U), thorium (²³²Th) and potassium (⁴⁰K) are the main elements that contribute to the natural terrestrial radioactivity. All of the ²³⁸U and ²³²Th decay series elements are incompatible in the major rock-forming minerals, such as quartz, feldspar, olivine and pyroxenes. There are, however, less incompatible in some trace or accessory phases or minerals [1]. Beach sediments are mineral deposits formed through weathering and erosion of either igneous or metamorphic rocks, these rocks may be rich in U and Th-bearing accessory minerals. These minerals migrate during the weathering and erosion of such rocks and precipitate and enrich in sediments [2]. Natural radioactivity studies of beach sediments provide a better understanding for mineralogy, geochemical, economical implications and radiological significance [3].

The Fourier Transform Infrared (FTIR) spectroscopy is most powerful research tool in soil and sediment mineralogy. It is used by mineralogist and sedimentary petrologists in the aspect of mineralogical applications. The principal constituents of most of the sediments are quartz, feldspar, carbonates and clay minerals [4].

Ramasamy *et al.* [5] showed that mineral characterization of some sediments from Palaru River, Tamilnadu in India.

During the last few decades, the coastal environment of north east coast of Tamilnadu in India has experienced intense developments in industry, tourism, transport, urbanization and aquaculture. Only limited investigations have been carried out in east coast of Tamilnadu to trace the enhanced level of natural radiation due to presence of minerals in beach sediments. Hence, the present study is focused to determine the activity concentrations of natural radionuclides (²³⁵U, ²³²Th and ⁴⁰K) and relative distribution of minerals such as quartz, feldspar and kaolinite through the Gamma ray and FTIR spectroscopic techniques. Ultimate aim of the study is to correlate the activity concentration of radionuclides and absorbed dose rate with relative distribution of major minerals.

MATERIALS AND METHODS

Location and Sample Collection: This study took place in north east coast of Tamilnadu, is bordered on the east by the Bay of Bengal. The total study area spread over from Port novo (Lat: 11° 30' 59"N; Long: 79° 46' 18"E) to Marina beach of Chennai city (Lat: 13° 03' 55"N; Long: 80° 17' 24"E), which covers an area about 200km. Some famous

beaches (Marina, Kovalam, Arovil and Silver), historical place (Mahabalipuram) and Chemical Industries (SIPCOT) are located in this study area.

Beach sediment samples were collected during April 2008. The total study area covers about 200km, from which 35 sampling locations are selected at the interval of 5-6km. The exact position of each sampling site was recorded using Hand held GARMIN GPS (Global Positioning System, Model no 12). The samples were collected from 10-20m away from the high tide, when it makes towards the road side.

Sample Preparation

Gamma-ray Analysis: The collected samples were dried in an oven at 100-110°C for about 24h and sieved through a 2-mm mesh-size sieve to remove stone, pebbles and other macro-impurities. The homogenized sample was placed in a 500g airtight PVC container. The inner lid was placed in and closed tightly with outer cap. The container was sealed hermetically and externally using cellophane tape and kept aside for about a month to ensure equilibrium between Ra and its daughter products before being taken for gamma ray spectrometric analysis.

FTIR Analysis: Wet grinding was carried out by placing 30 to 50 mg of the sample in an agate mortar along with 20 to 25 drops of ethanol. The ground samples were dried in a hot air oven at 110°C to remove the moisture content. Using KBr pellet technique, samples were mixed with KBr at various ratios viz., 1:10, 1:20, 1:30, 1:40 and 1:50. The mixture was then pressed into a transparent disc in an evacuable dye at sufficiently high pressure. The samples in the ratio 1:30 was taken for further analysis, since it gives rise to maximum transmittance and observable peaks.

Instrument

Gamma Spectroscopic Analysis: To estimate the activity levels of the ^{238}U , ^{232}Th and ^{40}K in the samples, a gamma ray spectrometer in the laboratory of Health and Safety Division, Indira Gandhi Centre for Atomic Research, Kalpakkam was used in the present investigations. NaI (TI) crystal detector of size 3" X 3" along with a 8K multichannel analyzer was used to record the gamma spectra. Standard sources of natural Uranium (1997.56Bq), natural Thorium (1237.28 Bq) and KCl (5181.59 Bq) with a standard 250ml container from International Atomic Energy Agency (IAEA) were used for calibrating the gamma ray spectrometer. These standards were obtained

from environmental survey laboratory, Indira Gandhi Centre for Atomic Research (IGCAR), Kalpakkam. With the counting time of 10,000 seconds for each sample, the below detectable limit (BDL) were 21.2Bqkg⁻¹ for ^{40}K , 5.5 Bqkg⁻¹ for ^{238}U and ^{232}Th .

FTIR: Using Perkin Elmer RX1 FTIR spectrophotometer, the infrared spectra of all the collected sediment samples were recorded in the region 4000-400cm⁻¹. The resolution of the instrument is $\pm 0.001\text{ cm}^{-1}$ and the accuracy is $\pm 4\text{ cm}^{-1}$.

Dose Rate Calculation: The absorbed dose rate is calculated from the measured activities of ^{238}U , ^{232}Th and ^{40}K in the surface sediment samples using the formula [6]

$$D\text{ (nGy h}^{-1}\text{)} = 0.462 C_{\text{U}} + 0.604 C_{\text{Th}} + 0.0417 C_{\text{K}}$$

Where D is the absorbed dose rate (nGy h⁻¹). C_{U} , C_{Th} and C_{K} are the activity concentrations (Bqkg⁻¹) of ^{238}U , ^{232}Th and ^{40}K respectively.

Relative Distribution of Minerals: The relative distribution can be quantified by calculating the extinction co-efficient using the following formula [7]

$$K = DA/m$$

Where K is the extinction co-efficient, A is the area of the pellet and m is the mass of the pellet. D is the optical density.

RESULTS AND DISCUSSION

Activity concentration (Bq/kg) of ^{238}U , ^{232}Th and ^{40}K radio nuclides in beach sediment samples are presented in Table 1. The measured activity concentrations are ranged from BDL to 30.42 \pm 7.90 Bqkg⁻¹ for ^{238}U , BDL to 218.64 \pm 8.02 Bqkg⁻¹ for ^{232}Th and 212.6 \pm 24.68 to 423.43 \pm 26.52 Bqkg⁻¹ for ^{40}K . Large variation among the radionuclide concentration in different sites has been observed. It may be due to mineralogy and drainage pattern of the study area since five river estuaries are located in this study area.

The maximum activity concentration of ^{238}U (30.42 \pm 7.90 Bqkg⁻¹) and ^{232}Th (218.64 \pm 8.02 Bqkg⁻¹) are observed in Mahabalipuram beach (S₂₂), which is one of the famous historical and tourism place. According to El-Arabi, [8], Uranium and thorium in beach sand are contained mainly in resistant heavy minerals such as

Table 1: Geographical location and activity concentrations of ^{238}U , ^{232}Th and ^{40}K with their uncertainties and absorbed dose rates for beach sediments

Site Number	Geographical Location		Activity concentration (Bq/kg)			Absorbed Dose rate (nGy/h)
	Latitude	Longitude	^{238}U	^{232}Th	^{40}K	
S ₁	11°30'.953	79° 46'.279	7.94±5.58	12.33±4.33	243.74±25.54	21.28±6.26
S ₂	11°32'.991	79°45'.595	7.28±4.38	10.58±4.63	224.48±24.62	19.11±5.85
S ₃	11°36'.233	79°45'.505	6.46±4.62	11.24±4.74	214.36±24.86	18.71±6.03
S ₄	11°41'.017	79°46'.378	6.31±4.42	9.46±4.34	236.68±24.71	18.50±5.69
S ₅	11°43'.279	79°46'.952	15.16±5.4	96.42±6.6	242.27±24.76	75.34±7.51
S ₆	11°44'.317	79°47'.221	26.99±7.23	185.46±7.68	238.32±34.21	134.43±9.41
S ₇	11°47'.356	79°47'.799	11.42±5.62	10.69±4.64	224.46±24.38	21.09±6.42
S ₈	11°49'.612	79°48'.312	6.37±4.47	7.8±4.05	238.33±24.50	17.59±5.53
S ₉	11°53'.614	79°49'.659	6.24±4.46	6.72±4.08	216.51±24.64	15.97±5.55
S ₁₀	11°55'.391	79°50'.043	BDL	BDL	212.60±24.68	15.63±3.50
S ₁₁	11°57'.442	79°50'.452	6.33±4.39	8.34±4.28	239.44±24.72	17.95±5.64
S ₁₂	12°01'.660	79°52'.030	6.51±4.3	6.65±4.06	226.67±24.74	16.48±5.47
S ₁₃	12°04'.543	79°53'.565	6.68±4.4	5.98±4.08	234.91±24.74	16.49±5.53
S ₁₄	12°06'.697	79°54'.931	6.34±4.52	7.36±4.62	222.62±25.64	16.66±5.95
S ₁₅	12°09'.735	79°56'.894	6.42±4.4	6.98±4.48	232.47±25.04	16.88±5.78
S ₁₆	12°11'.901	79°58'.358	6.54±4.57	7.04±4.56	218.72±24.34	16.14±5.89
S ₁₇	12°16'.269	80° 00'.980	6.44±4.56	6.84±4.24	213.36±24.18	16.00±5.68
S ₁₈	12°17'.962	80°01'.915	7.02±4.6	7.94±4.84	226.74±25.43	17.49±6.11
S ₁₉	12°20'.965	80°04'.104	6.98±4.84	7.26±4.26	246.40±26.40	17.88±5.91
S ₂₀	12°23'.976	80°06'.652	6.51±4.72	7.42±4.17	241.59±24.36	17.56±5.72
S ₂₁	12°16'.734	80°08'.566	8.96±5.26	10.94±4.92	246.54±25.06	21.03±6.45
S ₂₂	12°35'.657	80°11'.359	30.42±7.9	218.64±8.02	254.62±26.08	156.73±9.58
S ₂₃	12°37'.208	80°11'.905	7.36±5.02	19.79±4.48	335.29±26.58	29.34±6.13
S ₂₄	12°38'.746	80°12'.385	6.89±4.51	16.63±4.62	324.65±24.67	26.77±5.90
S ₂₅	12°41'.244	80°13'.307	6.36±4.48	12.61±4.48	294.65±25.12	22.84±5.82
S ₂₆	12°42'.292	80°13'.654	6.49±4.57	9.45±4.69	304.72±24.69	21.41±5.97
S ₂₇	12°43'.778	80°14'.220	5.21±4.48	16.28±4.76	326.24±24.96	25.84±5.99
S ₂₈	12°45'.332	80°14'.707	6.06±4.42	18.73±4.62	348.61±24.89	28.65±5.87
S ₂₉	12°47'.451	80°15'.111	8.78±5.06	24.33±4.75	423.43±26.52	34.76±6.39
S ₃₀	12°49'.899	80°14'.851	7.84±5.02	17.68±4.64	376.45±27.96	30.00±6.29
S ₃₁	12°53'.068	80°15'.112	7.04±5.51	13.92±4.4	353.49±26.86	26.40±6.32
S ₃₂	12°55'.138	80°15'.425	6.38±4.61	14.67±4.31	341.44±25.49	26.05±5.80
S ₃₃	12°57'.617	80°15'.839	BDL	15.48±4.24	348.03±25.80	26.75±5.69
S ₃₄	12°59'.918	80°16'.742	6.34±4.49	12.37±4.8	364.82±25.68	25.61±6.04
S ₃₅	13°03'.974	80°17'.362	BDL	8.56±4.94	422.43±26.00	25.29±6.22
		Average	8.35±4.88	24.53±4.73	276.00±25.58	30.18±6.11
		Maximum	30.42±7.9	218.64±8.02	423.43±26.52	156.73±9.58
		Minimum	BDL	BDL	212.6±24.68	15.63±3.50

monazite, zircon and xenotime. In the present study, higher values of ^{238}U and ^{232}Th could be due to the presence of black sands, which are enriched in the mineral monazite [2].

The highest activity concentration of ^{40}K ($423.43\pm26.52\text{ Bqkg}^{-1}$) is found in Kovalam beach (S₂₉). This highest activity concentration of ^{40}K may be due to the higher amount of clay [9]. The lowest concentration of all radio nuclides is found at Pudhucherry beach (S₁₀) (Table 1). Sediments collected from Pudhuchery beach (S₁₀) had higher coarser and sandy in nature. Marija Jankovic *et al.* [10] reported that the lowest activity concentration of the radionuclides in sediments is may be due to the high composition of quartz. Lower

concentration of radionuclides in site number S₁₀ (present study) may be due to the presence of higher amount of quartz.

The activity concentration of ^{238}U and ^{232}Th was not uniform in the entire study area. The activity concentration of ^{40}K is increased towards the northern side of the study (S₂₂ to S₃₅) area. This may be due to the increasing amount of feldspar and clay minerals. According to El-Arabi *et al.* [8], clay minerals are mainly composed of plate-like secondary aluminium silicates with small grain size and have a negative charged surface. Therefore, clay particles have the ability to absorb cations on their surface. This is the reason for the highest radionuclide activity concentrations occur in clays.

Table 2: The observed absorption wave numbers and corresponding minerals from FTIR spectra of beach sediments

Sl. No.	Name of the minerals	Site Number	Observed wave numbers (cm ⁻¹)
1	Quartz	S ₁ -S ₃₅	458-462
		S ₁ -S ₃₅	510-514
		S ₁ -S ₃₅	690-694
		S ₁ -S ₃₅	776-780
		S ₁ -S ₃₅	796-800
		S ₁ -S ₃₅	1080-1084
		S ₁ -S ₃₅	1160-1164
		S ₁ -S ₃₅	1610-1614
2	Microcline Feldspar	S ₁ -S ₃₅	581-585
		S ₁ -S ₃₅	1870-1874
3	Orthoclase Feldspar	S ₁ -S ₄ , S ₆ -S ₁₇ , S ₁₉ -S ₃₅	646-650
4	Gibbsite	S ₁ -S ₃₅	666-670
5	Kaolinite	S ₁ -S ₃₅	3396-3400
		S ₁ -S ₃₅	3621-3625
6	Calcite	S ₃₂	1425
7	Albite	S ₁ , S ₁₂ , S ₁₄ -S ₁₈ , S ₂₀ -S ₃₀ , S ₃₂ -S ₃₅	1008-1012
8	Organic carbon	S ₁ -S ₃₅	2854 and 2924

FTIR spectra of beach sediments

UNSCEAR [11] has published the world average data for ²³⁸U, ²³²Th and ⁴⁰K in coastal sediments as 25 Bqkg⁻¹, 25 Bqkg⁻¹ and 370 Bqkg⁻¹ respectively. If one compares these values with the present study, the mean activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K are 0.334, 0.981 and 0.746 times the world average values. In the same way, present values of ²³⁸U, ²³²Th and ⁴⁰K are 0.292, 0.384 and 0.842 times the Indian average values (28.67 Bqkg⁻¹ for ²³⁸U, 63.83 Bqkg⁻¹ for ²³²Th, 327.6 Bqkg⁻¹ for ⁴⁰K) respectively.

The calculated values of absorbed dose rate are presented in Table 1. It is ranged from 15.63 nGy h⁻¹ (S₁₀, Pudhucherry beach) to 156.73 nGy h⁻¹ (S₂₂, Mahabalipuram beach) with an average value of 30.18 nGy h⁻¹. The mean absorbed dose rate is 0.592 times the world average value (51 nGy h⁻¹; [11]).

From the recorded FTIR spectra, the observed absorption wave numbers are tabulated with corresponding minerals in Table 2. The minerals such as quartz, feldspar in different composition, kaolinite, gibbsite, calcite, albite and organic carbon were identified by comparing with available literatures [12, 13 and 6]. Relative distributions of Quartz, Feldspar and Kaolinite: With reference to the number of peaks and intensity, the minerals such as quartz, microcline feldspar and kaolinite are consider as major. The others are trace minerals. The relative distribution of major minerals can be quantified by calculating the extinction co-efficient for the characteristic peaks of quartz, microcline feldspar and kaolinite at

around 776 cm⁻¹, 581 cm⁻¹ and 3400 cm⁻¹ respectively for all sites. The calculated values are tabulated in Table 3. From this, it is observed that the maximum extinction co-efficient value for quartz, microcline feldspar and kaolinite are 231.64, 34.42 and 42.58 in the site no. S₁₀, S₃₃ and S₂₉ respectively. In overall view, the amount of kaolinite is lesser than microcline feldspar and very much lesser than quartz (Table 3).

Correlation Analysis: The correlation analysis is carried out between the radioactivity parameter and extinction coefficient of major minerals (using SPSS 16.00 statistical software) and correlation coefficients are presented in Table 4 as Pearson correlation matrix. From this table, strong correlation (r = 0.97) is observed between ²³⁸U and ²³²Th. Mohanty *et al.* [14] have fixed the strong correlation (r = 0.98) between ²³⁸U and ²³²Th for the sediments of Chhatrapur beach, India. Also they were reported, this strong correlation indicates the presence of significant amount of monazite minerals in the sediments. In the present study, existing strong correlation between ²³⁸U and ²³²Th may be due to the presence of monazite mineral in the study area. The same strong correlation was fixed by many authors [2, 15, 16] in various beach sediments. A poor correlation (r = -0.06) exists between ²³²Th and ⁴⁰K, again poor correlation is obtained between the ²³⁸U and ⁴⁰K with correlation coefficient of -0.13, which indicates that ⁴⁰K concentrations may not be related with the presence of ²³²Th and ²³⁸U bearing mineral [14].

Table 3: Extinction Co-efficient of quartz, microcline feldspar and kaolinite in the beach sediments

Site Number	Extinction Co-efficient		
	Quartz	Microcline Feldspar	Kaolinite
S ₁	101.06	19.86	6.79
S ₂	117.83	14.36	6.98
S ₃	112.23	16.02	7.36
S ₄	108.79	13.69	5.49
S ₅	77.34	18.41	4.4
S ₆	86.51	19.68	23.02
S ₇	131.69	18.36	13.24
S ₈	195.91	24.08	4.67
S ₉	206.02	27.96	3.96
S ₁₀	231.64	22.46	5.19
S ₁₁	123.83	21.83	4.88
S ₁₂	142.96	17.94	7.31
S ₁₃	140.68	14.35	5.79
S ₁₄	134.27	15.76	5.37
S ₁₅	144.16	20.46	6.04
S ₁₆	135.86	23.78	7.84
S ₁₇	118.01	30.16	10.31
S ₁₈	128.6	22.09	7.41
S ₁₉	139.42	14.37	4.68
S ₂₀	147.02	12.07	6.08
S ₂₁	143.79	15.86	4.37
S ₂₂	93.69	19.67	6.44
S ₂₃	136.59	21.34	7.27
S ₂₄	145.12	23.67	8.31
S ₂₅	128.46	25.86	8.69
S ₂₆	148.69	21.78	10.09
S ₂₇	129.24	29.36	12.2
S ₂₈	140.87	25.74	13.6
S ₂₉	124.38	24.67	41.58
S ₃₀	131.26	28.79	21.03
S ₃₁	104.41	27.92	15.74
S ₃₂	109.86	32.08	9.06
S ₃₃	142.46	34.42	12.36
S ₃₄	137.73	27.66	15.67
S ₃₅	130.81	30.92	13.9

Table 4: Correlation matrix of radioactivity parameters and relative distribution of quartz, microcline feldspar and kaolinite.

	²³⁸ U	²³² Th	⁴⁰ K	Absorbed Dose rate	Ex-Q	Ex-MF	Ex-K
²³⁸ U	1.00						
²³² Th	0.97	1.00					
⁴⁰ K	-0.14	-0.06	1.00				
Absorbed Dose rate	0.97	0.98	0.01	1.00			
Ex-Q	-0.45	-0.47	-0.12	-0.48	1.00		
Ex-MF	-0.19	-0.10	0.62	-0.06	0.12	1.00	
Ex-K	0.16	0.16	0.65	0.21	-0.23	0.36	1.00

Ex-Q: Extinction co-efficient of Quartz; Ex-MF: Extinction co-efficient of Microcline Feldspar; Ex-K: Extinction co-efficient of Kaolinite.

The correlation coefficient between ^{40}K and extinction co-efficient of kaolinite and microcline feldspar are 0.62 and 0.65 respectively, which indicate that, these two minerals are increased the level of activity concentration of ^{40}K . The extinction co-efficient of quartz is negatively (significant) correlated with absorbed dose rate, activity concentration of ^{238}U and ^{232}Th .

CONCLUSION

The activity concentrations of ^{238}U , ^{232}Th and ^{40}K and absorbed dose rate in beach sediments are studied and are lower than the world and Indian average values. However, the activity concentration values are higher in S_6 and S_{22} . The major minerals such as quartz, microcline feldspar and kaolinite and trace minerals such as orthoclase feldspar, gibbsite, calcite, albite and organic carbon are identified. The calculated extinction coefficient values show that, the amount of quartz is greater than feldspar and very much greater than kaolinite in all the sites. The obtained positive correlation between the ^{238}U and ^{232}Th showed that, heavy mineral (monazite) are present in the study area, whereas ^{40}K is correlated with feldspar and clay mineral (kaolinite). This indicates the sources of enhanced level of natural radioactivity in the beaches of the north east coast of Tamilnadu are mainly found from monazite and to a lesser extent in feldspar and clay minerals.

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