Evaluation of Natural Radionuclide Content in River Sediments and Excess Lifetime Cancer Risk Due to Gamma Radioactivity

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Abstract: The objective of this study is to evaluate the radionuclides (U^{238} , Th^{232} and K^{40}) content and outdoor terrestrial gamma dose rates in the Ponnaiyar river sediments to understand the radiation hazards to mankind. The determined activity concentrations of all sites are fall within the typical world and Indian average values although some extreme values are determined. To assess the radiological hazard of river sediments, the radiological hazard indices such as absorbed dose rate, annual effective dose equivalent (outdoor and indoor) (AEDE), hazard indices (H_{ex} and H_{in}), activity utilization index (I) and excess life time cancer risk (ELCR) are calculated. The radiological hazard indices are below the internationally recommended values. The last eight sampling sites have higher ELCR value.

Key words: Activity utilization index, annual effective dose equivalent, radioactivity, radiological hazard indices, ponnaiyar river

INTRODUCTION

Human beings are always exposed to background radiation that stems both from natural and man-made sources. Natural radioactivity is widespread in the earth environment and it exists in various geological formations such as earth crust, rocks, soils, plants, water and air. Natural radioactive concentration mainly depends on geological and geographical condition and appears at different level in soils of each different geological region (UNSCEAR, 2000). Soil radionuclide activity concentration is one of the main determinants of the natural background radiation. When rocks are disintegrated through natural process, radionuclides are carried to soil by rain and flows (Taskin *et al.*, 2009). In addition to the natural sources; soil radioactivity is also affected by man-made activities.

Radioactivity of various building materials was measured by many authors, including ceramics, gypsum, sand, mosaic tiles, marbles, granites, river sediments, etc., in different parts of the world (Tzortzis and Haralabos (2003); Ramasamy *et al.*, 2002, 2004, 2005(a & b) and 2006).

Among the various building materials, river sediment (sand) is one of the most important and major mixing materials for building construction in India, especially in Tamilnadu (state). The concentration of naturally occurring radionuclides in river sediments is measured in an effort to better understand the spatial distribution of the radionuclides. Naturally occurring radionuclides of terrestrial origin are present in river sediments as well (Krmar *et al.*, 2009). The environmental uranium and partial thorium concentrations are increased due to the fertilizers. Usually fertilizers are considered as technologically enhanced natural radiation (El-Gamal *et al.*, 2007). Considerable amounts of natural radio nuclides can be found in river sediments as the end result of fertilizer washing and industrial activities (Krmar *et al.*, 2009). The long-term exposure to uranium and radium through inhalation has several health effects as chronic lung diseases, acute leucopoenia, anemia and necrosis of the mouth. Radium causes bone, cranial, and nasal tumours. Thorium exposure can cause lung, pancreas, hepatic, bone, kidney cancers and leukaemia (Taskin, *et al.*, 2009). Therefore, gamma dose rates and radionuclides activity concentrations should be monitored.

Knowledge of natural radioactivity present in river sediments (Building materials) enables one to assess any possible radiological hazard to mankind by the uses of such materials. Hence, the objective of this study is to evaluate the radioactivity concentrations as well as the environmental outdoor (observed) gamma dose rates. The absorbed dose rate, annual effective dose rate (indoor and outdoor), hazard indices (external and internal), activity utilization index and excess life time cancer risk are also calculated.

MATERIALS AND METHODS

Study Area: In the present study, sediment samples were collected from various sites of the Ponnaiyar river. It is originated on the hills of Nandidrug in Kolar districts of Karnataka state, and flows south and then east for 400 Km through Karnataka and Tamilnadu, and terminated at Cuddalore, Tamilnadu in Bay of Bengal. It is entered in Tamilnadu at Dharmapuri district. It covers four districts (Dharmapuri, Thiruvannamalai, Villupuram and Cuddalore) in Tamilnadu. A dam is constructed on this river at Satthanur, Chengam taluk, Thiruvannamalai

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Site N o.	Location	Latitude	Longitude	Activity Concentration (Bq/Kg)			
				²³⁸ U	²³² Th	⁴⁰ K	
S1	Satthanur	12¢ ^a 10'606" N	78¢°50'464"E	9.51±4.21	6.11 ± 4.69	201.23 ± 23.96	
S2	Kolanjiyanur	12¢*07'978" N	78¢°53'950"E	11.36±6.13	6.91 ± 9.20	287.81 ± 25.36	
S3	Thiruva dath anoor-1	12¢°06'415"N	78¢°55'087"E	7.89±3.96	BDL	279.10 ± 26.17	
S4	Thiruvadath anoor-2	12¢ ^a 06'414" N	78¢°55'075"E	8.11±4.11	7.21 ± 4.19	282.64 ± 25.11	
S5	Rayendapuram	12¢ ^a 06'416"N	78¢°55'080"E	7.97±3.65	8.76 ± 4.33	305.01 ± 27.28	
S6	Jambai	12¢°00'677" N	79¢°03'273"E	7.32±3.54	8.18 ± 3.95	329.52 ± 25.32	
S7	Mugaiyur	12¢°00'104" N	79¢°04'169"E	8.13±4.21	9.55 ± 4.33	306.83 ± 34.34	
S8	Manalurpettai	12¢ ^a 00'271"N	79¢°05'481"E	7.23±3.95	8.98 ± 4.26	342.63 ± 26.17	
S9	Kuvanur	11¢ ^a 58'786"N	79¢°06'923"E	7.11±3.65	11.88 ± 3.62	384.97 ± 26.42	
S10	Thagadi	11¢°58'007" N	79¢°07'880"E	7.54±3.45	10.99 ± 4.01	378.79±25.96	
S11	Karadi	11¢ ^a 57'860" N	79¢°09'838"E	7.88±3.65	12.35±4.26	385.55±26.44	
S12	Thirukovilur	11¢ ^a 58'400" N	79¢°12'726"E	8.01±4.51	19.32±4.44	363.05±19.90	
S13	Vaddakk u N emeli	11¢°56'876"N	79¢ ^a 14'198"E	7.32±3.11	15.39±4.37	388.55±26.44	
S14	Vadamarudr	11¢°56'734" N	79¢°18'353"E	7.13±2.98	20.94±4.51	381.11±26.98	
S15	T.Pudupalayam	11¢ ^a 54'832" N	79¢ ^a 18'771"E	7.56±3.41	24.26±4.60	354.77±26.14	
S16	Kongerayanur	11¢ ^a 54'402"N	79¢°20'273"E	7.54±3.65	26.25±4.57	382.33±29.11	
S17	Saethur	11¢°55'082" N	79¢°22'377"E	6.98±3.98	25.68±4.59	380.21±25.69	
S18	Enathirimangalam	11¢ ^a 54'172"N	79¢ ^a 24'086"E	7.12±3.48	29.71±4.75	388.97±27.41	
S19	Perangiyur	11¢°52'354" N	79¢ ^a 26'245"E	7.56±3.85	33.95±4.76	388.08±27.39	
S20	Kavanur	11¢°52'065" N	79¢°28'584"E	6.99±3.01	37.90±5.07	397.01±27.78	
S21	Korathirur	11¢°51'688" N	79¢°29'731"E	7.21±3.21	38.2±5.15	385.55±26.44	
S22	Periya kallipattu	11¢ ^a 51'309"N	79¢°30'778"E	7.24±3.26	42.36±5.56	399.32±29.12	
S23	Kandarakuttai	11¢ ^a 50'533" N	79¢°33'457"E	6.96±3.01	48.69±5.87	402.68±27.01	
S24	Palavanur	11¢°50'367"N	79¢°33'916"E	6.71±2.98	58.48±3.80	406.82±30.53	
S25	Meikumercmangalam	11¢ ^a 50'406"N	79¢°35'451"E	6.64±2.69	66.25±4.21	408.10±25.78	
S26	K uppath and ap al ay am	11¢ ^a 49'579" N	79¢°36'488"E	6.98±3.11	59.68±4.44	405.04±25.58	
S27	Paka ndi	11¢ ^a 49'328"N	79¢°37'131"E	6.68±2.96	58.69±5.26	405.11±28.82	
S28	Kilkava rapa ttu	11¢ ^a 49'001" N	79¢°37'273"E	6.97±2.89	68.59±5.01	403.94±32.17	
S29	Male pattampakkam	11¢ ^a 48'090" N	79¢°37'994"E	6.69±2.65	70.96±6.18	418.53±27.18	
S30	Elangikuppan	11¢ ^a 47'853"N	79¢°38'736"E	7.24±3.21	74.52±6.11	430.36±27.16	
S31	V i sh w an at ha pu ra m	11¢ ^a 47'124" N	79¢°39'249"E	6.45±3.14	78.36±6.25	424.09±25.68	
S32	R a m ap ak k am	11¢ ^a 48'021"N	79¢°39'755"E	6.98±2.96	87.23±6.54	420.55±27.72	
S33	V a np ak k am	11¢ ^a 48'464" N	79¢°40'249"E	BDL	74.52±6.01	424.72±32.57	
S34	V e ll ep ak k am	11¢ ^a 47'414" N	79¢ ^a 41'792"E	6.68±2.65	89.65±6.98	434.59±28.69	
S35	Alagiyanallur	11¢ ^a 47'549" N	79¢°42'287"E	6.57±2.56	95.64±6.34	426.24±30.18	
S36	Marudhadu	11¢ ^a 47'482"N	79¢ ^a 42'758"E	BDL	96.64±6.24	437.71±27.81	
S37	Nathapa ttu	11¢ ^a 46'831"N	79¢°43'642"E	6.56±2.68	94.65±6.21	455.89±20.62	
S38	Unnamalai savadi	11¢ ^a 46'821"N	79¢°44'201"E	BDL	97.26±6.98	442.09±25.68	
S39	Cuddalore	11¢ ^a 46'405"N	79¢°45'821"E	6.59±2.87	96.48±6.16	454.21±22.32	
S40	Thalanoadai	11¢ ^a 45'350"N	79¢°47'685"E	6.52±2.98	106.11±7.16	467.71±22.54	
			Average	7.31±3.41	46.85±5.25	384.113±26.82	
			Maximum	11.6±6.13	106.11±9.20	467.71±34.34	
			Minimum	BDL	BDL	20 1.2 3±19.9	

district. Capacity of this dam nearly 4600 M CFT. The sediments of this river are excavated only for building constructions. The small hydraulic structure and barrages were constructed for drinking and agriculture purposes, respectively on the study area. On both side of the bank of this river, so many living residents and some industries are situated. None of the industrials have proper and controlled outlet. The discharge wastes and toxic metals from such industries and living residents are directly let out in to the river. Also along the river, lot of agricultural lands is available, overuse of chemical fertilizers and pesticides are washed into the river. These are all main factors for enrichments of pollutants in the study area.

Sample Collection and Preparation: The present study area (Ponnaiyar river) covers a total length of 200 Km, from which 40 locations were selected. Location of sampling site with their latitude and longitude are given in Table 1. Each location is separated by a distance of 4-5 Km approximately. All sediment samples were collected at 0-10 cm depth during the summer season (April-May 2008). Each sample has a weight of 3-4 kg approximately. The collected samples were dried at room temperature in open air for two days and stored in black polythene bags.

The samples were dried in an oven 110°C till the constant dry weight was obtained, crushed and homogenized. The homogenized samples were packed in a 250 ml plastic container (9cm x 6.5cm: Height x Diameter) to its full volume with uniform mass. These containers shielded hermetically and also shielded externally to ensure that all daughter products of uranium and thorium, in particular, radon isotope formed, do not escape. A time of four weeks was allowed after packing to attain secular equilibrium between Ra-226 and its short-lived daughter products. The net weight of the sample was determined before counting.

Radioactivity measurements: The gamma ray spectrometer with NaI(TI)detector was used to determine the concentration of primordial radionuclides (238 U, 232 Th, 40 K). The detector was shielded by 15 cm thick lead on all four sides and 10 cm thick on top. The energy resolution of 2.0 Kev and relative efficiency of 33% at 1.33Mev was achieved in the system with the counting time of 10000 seconds. The Standard International Atomic Energy Agency (IAEA) sources were used for calibration. From the counting spectra, the activity concentrations of 238 U, 232 Th and 40 K were determined using computer program. The peak corresponds to 1460Kev (K-40) for 40 K, 1764.5

Kev (Bi-214) for ²³⁸U and 2614.5Kev (Ti-208) for ²³²Th were considered in arriving at the activity levels (Bq/kg).

RESULTS

Activity Concentration of ²³⁸U, ²³²Th and ⁴⁰K: The activity concentration of natural radionuclides (²³⁸U, ²³²Th and ⁴⁰K) for all samples is determined and is shown in Table 1. The mean activity concentration ranges for ²³⁸U, ²³²Th and ⁴⁰K are BDL -11.60 \pm 6.13 Bq/kg with an average 7.31 \pm 3.41 Bq/kg, BDL - 106.11 \pm 9.20Bq/kg with an average 46.85 \pm 5.25 Bq/kg and 201.23 \pm 19.90 - 467.71 \pm 34.34 Bq/kg with an average 384.03 \pm 26.82 Bq/kg, respectively.

Dose calculation:

Absorbed and observed dose rate: The mean activity concentrations of Th and K are converted in to dose rate based on the conversion factor given by UNSCEAR (2000) (Table 2).

 $D = (0.462C_{\rm U} + 0.604 C_{\rm Th} + 0.0417 C_{\rm K}) nGyh^{-1}$

Where D is the absorbed dose rate (nGyh⁻¹), $C_{U} U_{Th}$, C_{K}

Ta ble 2: D ose rate, AED E, Hazard ind ices, I and ELC R of all the sites

Th and C_{K} are the activity concentrations (Bq/kg) of ²³⁸U, ²³²Th and ⁴⁰K in river sediments respectively. The range of absorbed dose rates is from 16.48 nGy/h to 86.17 nGy/h with average of 47.07 nGy/h.

The outdoor terrestrial gamma dose rates are measured 1 m above the ground by a portable digital ERDM at all the sampling sites. A total five readings are recorded at each spot and average is taken (Table 2).

The Annual Effective Dose Equivalent (AEDE): The annual effective dose equivalent received by a member is calculated from the absorbed dose rate by applying dose conversion factor of 0.7 Sv/Gy and the occupancy factor for outdoor and indoor was 0.2(5/24) and 0.8(19/24), respectively (Veiga *et al.*, 2006).

The annual effective dose is determined using the following equations

AEDE (Outdoor) (μ Sv/y) = (Absorbed dose) nGy/h x 8760h x 0.7 Sv/Gy x 0.2 x10⁻³

AEDE (Indoor) (μ Sv/y) = (Absorbed dose) nGy/h x 8760h x 0.7 Sv/Gy x 0.8x10⁻³

Site No.	Dose Rate (nGy/h)		AEDE (mSv/	AEDE (mSv/y)		Hazard indices		
	Absorbed dose ra te	Observed dose ra te	Outdoor	Indoor	Hex	Hin	I	ELCR x 10 ⁻³
1	16.48	55	20.21	80.82	0.091	0.117	0.28	0.071
2	21.53	75	26.41	105.64	0.118	0.149	0.41	0.092
3	18.61	65	22.82	91.27	0.101	0.117	0.40	0.080
4	19.89	65	24.39	97.56	0.109	0.125	0.40	0.085
5	21.69	45	26.60	106.41	0.119	0.135	0.43	0.093
6	22.06	60	27.06	108.23	0.120	0.134	0.47	0.095
7	22.32	65	27.37	109.49	0.123	0.139	0.43	0.096
8	23.05	45	28.27	113.08	0.125	0.140	0.49	0.099
9	26.51	50	32.52	130.07	0.145	0.159	0.55	0.114
10	25.92	70	31.78	127.14	0.142	0.157	0.54	0.111
11	27.18	60	33.33	133.32	0.149	0.165	0.55	0.117
12	30.51	75	37.42	149.67	0.172	0.188	0.53	0.131
13	28.88	65	35.42	141.67	0.160	0.174	0.55	0.124
14	31.83	75	39.04	156.17	0.179	0.193	0.56	0.137
15	32.94	90	40.40	161.59	0.188	0.203	0.55	0.141
16	35.28	65	43.27	173.08	0.201	0.216	0.60	0.151
17	34.59	60	42.42	169.69	0.197	0.211	0.59	0.148
18	37.45	60	45.93	183.74	0.215	0.229	0.63	0.161
19	40.18	60	49.28	197.11	0.232	0.247	0.67	0.172
20	42.68	60	52.34	209.35	0.248	0.261	0.73	0.183
21	42.48	55	52.10	208.40	0.247	0.261	0.72	0.182
22	45.58	85	55.90	223.61	0.266	0.280	0.78	0.196
23	49.42	60	60.60	242.42	0.291	0.304	0.86	0.212
24	55.39	55	67.93	271.70	0.329	0.341	1.00	0.238
25	60.10	60	73.71	294.83	0.359	0.371	1.12	0.258
26	56.16	70	68.88	275.51	0.333	0.347	1.02	0.241
27	55.43	60	67.98	271.91	0.329	0.342	1.01	0.238
28	61.49	60	75.41	301.66	0.368	0.381	1.15	0.264
29	63.40	70	77.76	311.03	0.379	0.392	1.20	0.272
30	66.30	70	81.31	325.25	0.397	0.411	1.25	0.285
31	68.46	65	83.95	335.82	0.411	0.426	1.31	0.294
32	73.45	60	90.08	360.31	0.443	0.457	1.46	0.315
33	65.72	60	80.60	322.42	0.394	0.406	1.26	0.282
34	75.36	100	92.42	369.67	0.455	0.467	1.50	0.323
35	78.58	110	96.37	385.46	0.476	0.488	1.60	0.337
36	79.63	90	97.65	390.61	0.482	0.494	1.62	0.342
37	79.21	120	97.14	388.57	0.478	0.490	1.59	0.340
38	80.18	140	98.34	393.35	0.485	0.497	1.63	0.344
39	80.82	130	99.12	396.48	0.488	0.500	1.62	0.347
40	86.17	150	105.68	422.73	0.522	0.535	1.79	0.370
Average	47.07	73.37	57.73	230.92	0.277	0.29	0.90	0.202
Maximum	86.17	150	105.68	422.73	0.522	0.53	1.79	0.370
Minimum	16.48	45	20.21	80.82	0.091	0.12	0.28	0.071

The calculated indoor and outdoor AEDE values are quoted in table 2. The average, minimum and maximum value for outdoor and indoor is found to $57.73\mu Sv/y$, $20.21\mu Sv/y$ and $105.66\mu Sv/y$, respectively and $230.92 \mu Sv/y$, $80.82 \mu Sv/y$ and $422.73 \mu Sv/y$, respectively.

Radiological Hazard Indices: The Gamma ray radiation hazards due to the specified radionuclides in river sediments are assessed by calculating different indices. Even though total activity concentration of radionuclides is calculated, it does not provide the exact indication about the total radiation hazards. Also these hazard indices are used to select the right materials.

Hazard Indices (H_{ex} and H_{in}): The two indices are that represent the external and internal radiation hazards. These indices are calculated (table 2) by following relation (Orgun *et al.*, 2007).

$$H_{ex} = (C_U/370 + C_{Th}/259 + C_K/4810) \le 1$$
$$H_{in} = (C_U/185 + C_{Th}/259 + C_K/4810) < 1$$

Where C_U , C_{Th} and C_K are the mean activity concentrations of ^{238}U , ^{232}Th and ^{40}K in Bq/Kg respectively.

Activity Utilization Index (I): The samples are also examined whether it facilitate the dose criteria when it used as building material. For that reason, the Activity utilization Index (I) is calculated using the equation given by Tzortzis and Haralabos (2003) and El-Gamal *et al.*, (2007).

The calculated I values for all the samples are presented in table2. The values range from 0.28 to1.79 with an average of 0.90, exhibit that I<2, which corresponds to an annual effective dose < 0.3 mSv/y (El-Gamal *et al.*,2007).

Excess Lifetime Cancer Risk (ELCR): Excess Lifetime Cancer Risk (ELCR) is calculated using below equation and shown in Table 2.

$$ELCR = AEDE \times DL \times RF$$

Where AEDE, DL and RF is the annual effective dose equivalent, duration of life (70 years) and risk factor (Sv⁻¹), fatal cancer risk per sievert. For stochastic effects, ICRP 60 uses values of 0.05 for the public (Taskin *et al.*, 2009). The range of ELCR is 0.071 x 10^{-3} to 0.37 x 10^{-3} with an average of 0.202 x 10^{-3} .

DISCUSSION

The activity concentrations vary from site to site, because river bottoms can exhibit large variation in chemical and mineralogical properties (Krmar *et al.*, 2009). In all sampling sites, mean activity concentration is of the order 238 U < 232 Th < 40 K. In particular S₂, the activity concentration of ²³⁸U is high, which may be due to the solubility and mobility of U (VI)O₂²⁺(Powell *et al.*, 2007). However, the S_{40} is having high activity concentrations. Increasing concentration of ²³²Th and ⁴⁰Kmay be due to the high content of monazite (Orgun et al., 2007). The increasing trend of ⁴⁰K is due to presence of loamy and clay sediments (El-Gamal et al., 2007). Ramasamy et al., (2004 and 2006) reported the values of Palar and Cauvery rivers, which are higher than the present values. The concentration of ²³⁸U, ²³²Th and ⁴⁰K for all measured samples are below the world and Indian average values (World average value of ²³⁸U, ²³²Th and ⁴⁰K is 50Bq/kg, 50 Bq/kg and 500 Bq/kg, respectively. Indian average value of ²³⁸U, ²³²Th and ⁴⁰K is 28.67 Bq/kg, 63.83 Bq/kg and 327.6 Bq/kg respectively). However in some sampling sites, concentration of ²³²Th is higher than world average value, indicating that monazite may exist at that sampling site. Average absorbed dose rate for all samples are lower than the world average value (51nGy/h) (UNSCEAR, 2000). Studies indicate an average outdoor terrestrial gamma dose rate of 60 nGy/h in the world ranging from 10 to 200 nGy/h (Taskin et al., 2009). The present study shows that the average terrestrial gamma dose rate is 73 nGy/h and higher than the world average. The level of gamma radiation is directly associated with the activity concentrations of radionuclides in the sediment samples and cosmic rays (Taskin et al., 2009). The present values of indoor and outdoor AEDE is lower than the world average values (70 μ Sv/y for outdoor, 450 µSv/y for indoor) (Orgun et al., 2007). Hazard indices of all site samples are less than Unity (permissible level) (Orgun et al., 2007). Average ELCR for all samples is less than the world average (0.29 x 10⁻³) (Taskin et al., 2009). The last eight sampling sites have higher ELCR value (S_{33} to S_{40}).

CONCLUSION

The average activity concentrations of Ponnaiyar river sediments were within the world and Indian average value, although some extreme values had been determined. The average outdoor terrestrial gamma dose rate is higher than world average. The other calculated radiological hazard indices are below the acceptable limit (Safety Limit). The calculated activity utilization index is less than two; this indicates that the Ponnaiyar river sediments can be used for safety construction of buildings. This information is an important information for local peoples to utilize the Ponnaiyar river sediments.

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