

DETERMINATION OF LEVEL OF RADIOACTIVITY AND EVALUATION OF RADIATION HAZARDOUS NATURE OF THE RECENTLY EXCAVATED RIVER SEDIMENTS

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Abstract: Gamma radiation measurements were performed using high resolution HpGe detector for sediment (sand) samples collected from various sites of Vellar River, Tamilnadu. The concentration and distribution of ²³⁸U, ²³²Th and ⁴⁰K were investigated with an aim of an evaluating radiation hazards to mankind, when it is used for building construction. The mean activity concentration values for ²³⁸U, ²³²Th and ⁴⁰K were found to be 15.14 ± 1.75 Bqkg⁻¹, 29.76 ± 4.49 Bqkg⁻¹ and 135.05 ± 16.03 Bqkg⁻¹ respectively. These values are fall within range of typical world values. To assess the radiological hazards of river sediments, the absorbed and observed dose rate, annual effective dose (outdoor and indoor) equivalence, radium equivalence (Ra_{eq}), radiation heat production (RHP) rate, hazard indices (H_{ex} and H_{in}) and gamma index are calculated. The radiological hazard indices are below the internationally recommended values. Thus, sediments of Vellar River do not pose a significant radiological threat to the population when it is used for construction of buildings.

Keywords: Radioactivity, River sediments, Radiological hazard indices, HpGe detector, radionuclides

1. INTRODUCTION

Human population is always exposed to ionizing radiation from natural 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 and also traces level in construction materials, which may contribute significantly towards an increased radiation dose received by human beings (Faheem et al., 2008 and Sonkawade et al., 2008). Natural radioactive concentrations mainly depend on geological and geographical condition and appear at different level in soils of each different geological region (UNSCEAR, 2000). Building materials contain some amount of natural radionuclides that cause exposure of people to ionizing radiation (Krstic et al., 2007). Ionizing radiation have sufficient energy to cause the ionization in matter, is

incident on the human body, the free radicals are formed as the result the chemical and structural damages of DNA, lipids, and protein and also chromosome aberration, gene mutation and cell death may possible. Building materials are the main source of indoor gamma radiation, besides terrestrial and cosmic radiation (Sonkawade, 2008).

Radioactivity of various building materials was measured by many authors, including brick, cement, ceramics, gypsum, sand, soil, mosaic tiles, marbles, granites, river sediments, etc., in different part of the world (Tsabaris et al., 2007; Krstic et al., 2007; Papaefthymiou et al, 2008; Sonkawade, et al., 2008 and Fatheem et al., 2008).

Among the various building materials, river sediment(sand) is one of the most important and major mixing materials for building construction in India, especially 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 sediment as well (Kumar et al., 2009). Since considerable amount of natural radionuclides can be found in river sediments as the end result of fertilizer washing, industrial activities and also human activities (Kumar et al., 2009). Natural radioactive metals leached from wastes materials also contaminate surrounding ground water, soils and biota (Petrescu & Bilal, 2006 & 2007).

Based on the above discussion, the Ionizing radiation from river sediments are evident due to the natural and artificial radionuclides. Rivers has valuable deposits of sediments, which is used for building construction, and also river is act as a source of drinking water, domestic water, radioactive elements and irrigation water. Nature has gifted lot of precious gifts to mankind; river sediment is one of them. In Tamilnadu, India, building construction mostly depends on the various building materials, among this river sediment is one. 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 present study is focused on the natural radiation level of the Vellar River sediments; this River is one of major rivers in Tamilnadu state, India. The sand (sediments) samples from this river were used. Since, the main goal of the present work is to study the naturally occurring radioactivity level of the Vellar River sediments and its relevant dose and radiological hazards while it is used as a building construction material.

2. MATERIALS AND METHODS

2.1. Study Area

The Vellar River covers four districts (Salem, Perambalur, Villupuram and Cuddalore) in Tamilnadu, India. Location of sampling sites is shown in figure 1.

2.2. Sample Collection and Preparation

The present study area (Vellar River) covers a total length of 200 Km, from which 27 locations were selected. The sample locations were recorded by GARMIN GPS-12 unit. Each location is separated by a distance of 7-8 Km approximately. All sediment samples were collected during the summer season (April-May 2009). Each sample has a weight of 3-4 kg. The collected samples were dried at room temperature in open air for two days and

stored in black polythene bags. Surface layer samples were collected at 0-10 cm depth.

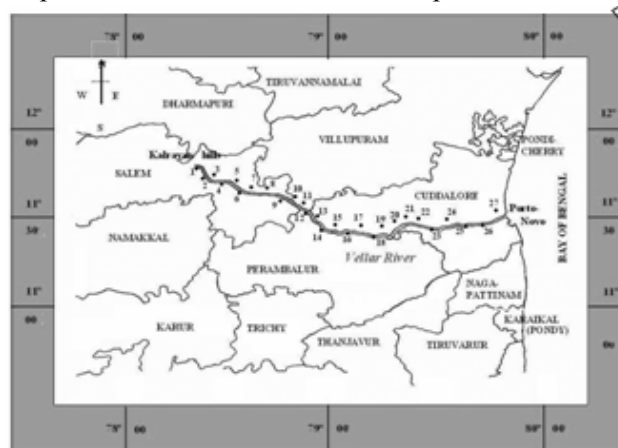


Figure 1. Location of the sampling sites

Sediment 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.

2.3. Radioactivity measurements

The gamma ray spectrometer with HpGe detector (model & efficiency) was used to determine the concentration of primordial radionuclides (^{238}U , ^{232}Th and ^{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 2.0Kev 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 are used for calibration. It was calibrated using standard solution of ^{226}Ra in equilibrium with daughters (obtained from NBS, USA), mixed with simulated soil matrix and counted in the same geometry as that of the soil samples. A standard soil of known radioactivity was taken. From the counting spectra, the activity concentrations of ^{238}U , ^{232}Th and ^{40}K was determined using computer program. The peak corresponds to 1460Kev (K-40) for ^{40}K , 1764.5Kev (Bi-214) for ^{238}U and 2614.5Kev (Ti-208) for ^{232}Th were considered in arriving at the activity levels (Bq/kg).

3. RESULTS AND DISCUSSION

3.1. Activity Concentration of ^{238}U , ^{232}Th and ^{40}K

The activity concentration of natural radionuclides (^{238}U , ^{232}Th and ^{40}K) for all 27 site samples is given in table 1. The activity concentrations are ranged from 4.75 ± 1.02 to $42.64 \pm 3.95 \text{ Bqkg}^{-1}$ with a mean of $15.14 \pm 1.75 \text{ Bqkg}^{-1}$, 10.39 ± 4.85 to $65.29 \pm 1.34 \text{ Bqkg}^{-1}$ with a mean value of $29.76 \pm 4.49 \text{ Bqkg}^{-1}$ and 78.08 ± 17.46 to $267.01 \pm 22.86 \text{ Bqkg}^{-1}$ with a mean value of $135.05 \pm 16.03 \text{ Bqkg}^{-1}$ for ^{238}U , ^{232}Th and ^{40}K respectively. The average activity concentrations of all radionuclides are lower than the world average values (50 Bqkg^{-1} and 500 Bqkg^{-1} for ^{238}U and ^{40}K) (Ramasamy et al., 2010).

The activity concentration ^{238}U is lower than the ^{232}Th and ^{40}K , because ^{238}U is more soluble in water and leached before final deposition in sediments. The organic matter content may also significantly affect ^{238}U activities. Decreasing contents of organic matter are related to higher ^{238}U activities, because all soil profiles have decreasing percentage of organic matter as observed by Navas et al. (2002). In view of this, the minimum values of ^{238}U in this study may indicate that the presence of higher amount of organic matter in that area. The ^{40}K activity concentration dominates over ^{238}U and ^{232}Th elemental activities like what normally happens in soil. In contrasting with very low mobility in the other two profiles (^{238}U and ^{232}Th), ^{40}K seems to be involved in some suspended transport and sorption to colloidal material.

3.2. Dose calculation

3.2.1. Absorbed Dose rate

The mean activity concentrations of ^{238}U , ^{232}Th and ^{40}K were converted in to dose rate based on the conversion factor given by UNSCEAR (2000) and Ramasamy et al., (2010).

$$D = (0.462C_U + 0.604 C_{Th} + 0.0417 C_K) \text{ nGyh}^{-1}$$

Where D is the absorbed dose rate (nGyh^{-1}), C_U , C_{Th} and C_K are the activity concentrations (Bq/kg) of ^{238}U , ^{232}Th and ^{40}K in river sediment respectively.

Absorbed dose rate for all sampling sites are calculated using the above equation. The values are presented in table 1. It is ranged from $16.85 \pm 2.43 \text{ nGy/h}$ to $52.08 \pm 5.04 \text{ nGy/h}$ with an average of $32.90 \pm 4.35 \text{ nGy/h}$. This average is lower than the world average value (51 nGy/h) (Ramasamy et al., 2010). The values are changed according to fractional contribution of the materials, this could be

due to the spatial variation of minerals holding sediments (El-Gamal et al., 2007).

3.2.2. Observed Dose rate

The contribution of natural radionuclides to the absorbed dose rate depends on the concentration of the radionuclides in sediments, but observed dose rate mainly depends on both outdoor terrestrial gamma radiation and cosmic rays (Degerlier et al., 2008). Gamma dose rates were measured at one meter above the ground by a portable digital ERDM (Environmental Radiation Dosimeter) at all the sampling sites. A total five readings were recorded at each spot and average is taken. The measured values are presented in table 1. From the table 1, maximum dose rate of 130 nGyh^{-1} is obtained in the site no.27 and the minimum value of 40 nGyh^{-1} in the site no.15 with the mean of 68.70 nGy h^{-1} . Average observed dose rate is two times of absorbed dose rate, this difference due to the contribution from cosmic rays (Degerlier et al., 2008).

3.2.3. The Annual Effective Dose Equivalent (AEDE)

Since the Vellar River sediments are the main mixing material with cement and cement products for building construction in Tamilnadu, India, the determination of the AEDE of each site sample assumes importance. For that, the living style of the people or indoor or outdoor occupancy factor of a location is considered. AEDE received by a member was 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 was determined using the following equations

$$\text{AEDE (Outdoor)} (\mu\text{Sv/y}) = (\text{Absorbed dose}) \text{ nGy/h} \times 8760 \text{ h} \times 0.7 \text{ Sv/Gy} \times 0.2 \times 10^{-3}$$

$$\text{AEDE (Indoor)} (\mu\text{Sv/y}) = (\text{Absorbed dose}) \text{ nGy/h} \times 8760 \text{ h} \times 0.7 \text{ Sv/Gy} \times 0.8 \times 10^{-3}$$

The calculated indoor and outdoor AEDE values are quoted in table 1. The outdoor AEDE varies between 20.71 ± 6.14 (site no.15) and $66.07 \pm 2.77 \mu\text{Svy}^{-1}$ (site no.27) with a mean of $38.15 \pm 5.24 \mu\text{Svy}^{-1}$. The mean value of annual effective dose rate of outdoor is 0.55 times that of the world average value $70 \mu\text{Svy}^{-1}$. For indoor, it varies between 82.86 ± 24.58 (site no.15) and $264.26 \pm 11.09 \mu\text{Sv y}^{-1}$ (site no.27) with a mean of $152.61 \pm 20.94 \mu\text{Sv y}^{-1}$. The present values of indoor AEDE are lower than the world average values ($450 \mu\text{Sv/y}$) (UNSCEAR 2000).

3.2.4. Radiological Hazard Indices

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

3.2.4.1. Radium Equivalent Activities (Ra_{eq})

Radium equivalent activity is an index that has been introduced to represent the specified activities of ^{238}U , ^{232}Th and ^{40}K by a single quantity, which takes in to account the radiation hazards associated with them. Ra_{eq} was calculated by below equation (Ramasamy et al., 2006 and Orgun et al., 2007).

$$Ra_{eq} = (C_U + 1.43 C_{Th} + 0.077 C_K) \text{ Bq/Kg}$$

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. Ra_{eq} is calculated for all site samples, the calculated values are presented in table 2. The maximum value ($118.67 \pm 4.70 \text{ Bqkg}^{-1}$) at site no. 27 and the minimum value ($35.45 \pm 11.01 \text{ Bqkg}^{-1}$) at site no.15 are observed with a mean of $67.59 \pm 9.42 \text{ Bqkg}^{-1}$. The maximum values of Ra_{eq} in building materials

must be less than the 370 Bq/Kg for safe use (Ramasamy et al., 2006 and Orgun et al., 2007). The present study values show lower value.

3.2.4.2. Radioactive heat production (RHP) rate

During the last few decades, the assessment of the amount of radioactive elements, the major internal heat source of the earth is the subject of several studies due to its importance in modeling the thermal evaluation of the lithosphere. The radioactive isotopes ^{238}U , ^{232}Th and ^{40}K contribute most of the terrestrial heat flow. These elements are fundamental for understanding the nature of the mantle, crust of the earth and their heat generating potential.

In this study, an attempt has also been made to find out the radioactive heat produced by the Vellar River sediments at different sites using the relation given by Rybach (1988) and Adel et al., (2006).

$$A = 10^{-5} \rho (9.52 C_U + 2.56 C_{Th} + 3.48 C_K) \mu\text{W m}^{-3}$$

Where: A is radioactive heat production rate expressed in $\mu\text{W m}^{-3}$, ρ is the sample density in kg m^{-3} , C_U and C_{Th} are the uranium and thorium concentration in ppm and C_K is the total potassium concentration in %.

Table 1. The activity concentrations of ^{238}U , ^{232}Th and ^{40}K , absorbed dose rate and AEDE of all sites

Site no.	Activity concentrations (Bq/kg)			Absorbed dose rate (nGyh ⁻¹)	Observed dose rate (nGyh ⁻¹)	Annual effective dose rate (μSvy ⁻¹)	
	²³⁸ U	²³² Th	⁴⁰ K			Indoor	Outdoor
1	42.64±3.95	11.48±5.05	84.90±18.03	30.37±5.71	60	148.98±28.01	37.25±7.00
2	24.95±2.79	13.07±5.74	93.63±18.01	23.55±5.60	50	115.53±27.47	28.88±6.87
3	27.43±2.36	11.94±4.68	93.91±18.31	24.00±4.76	55	117.73±23.35	29.43±5.84
4	18.80±2.69	12.70±5.21	112.20±17.54	21.25±5.21	60	104.24±25.56	26.06±6.39
5	32.94±2.96	33.91±5.86	136.57±18.05	41.97±5.76	90	205.89±28.26	51.47±7.06
6	24.80±1.31	11.20±4.86	131.80±18.19	23.91±4.38	50	117.29±21.49	29.32±5.37
7	25.11±1.19	10.82±4.72	138.41±18.26	24.09±4.24	55	118.18±20.80	29.54±5.20
8	20.89±1.23	14.43±5.97	155.07±18.56	25.08±5.05	60	123.03±24.77	30.76±6.19
9	17.90±1.37	19.96±5.62	177.43±17.46	28.06±4.85	65	137.65±23.79	34.41±5.95
10	14.87±1.16	53.50±5.52	145.23±16.59	46.15±4.66	100	226.39±22.86	56.60±5.72
11	18.64±1.08	30.84±5.34	140.33±16.29	33.62±4.49	75	164.93±22.03	41.23±5.51
12	15.45±1.17	32.56±5.40	178.06±17.58	34.78±4.63	80	170.62±22.71	42.65±5.68
13	13.78±1.36	26.40±5.18	78.08±17.46	26.02±4.57	55	127.64±22.42	31.91±5.60
14	12.62±1.28	28.14±5.61	197.91±19.56	31.56±4.89	70	154.82±23.99	38.71±6.00
15	9.01±2.64	10.39±4.85	150.47±18.65	16.89±5.01	40	82.86±24.58	20.71±6.14
16	13.83±2.44	11.00±5.30	177.14±17.10	20.61±5.13	50	101.10±25.17	25.28±6.29
17	7.39±1.52	29.71±3.44	125.04±9.62	27.08±3.24	45	132.84±15.89	33.21±3.97
18	8.65±2.61	34.96±3.84	143.55±18.46	31.69±4.36	65	155.46±21.39	38.86±5.35
19	12.81±1.21	39.82±3.96	111.96±10.23	35.32±3.44	75	173.27±16.88	43.32±4.22
20	7.44±1.06	41.22±3.82	121.38±10.82	34.10±3.31	80	167.28±16.24	41.82±4.06
21	6.76±1.54	46.89±2.44	107.42±10.47	36.72±2.66	80	180.13±13.05	45.03±3.26
22	6.14±1.23	49.11±2.40	83±12.02	36.79±2.56	75	180.48±12.56	45.12±3.14
23	4.88±1.11	51.08±4.24	133.20±10.78	39.53±3.60	90	193.92±17.66	48.48±4.42
24	5.23±2.40	22.49±2.21	115.84±11.12	21.21±2.94	50	104.05±14.42	26.01±3.61
25	4.82±1.41	47.45±2.49	99.7±11.45	35.85±2.68	75	175.87±13.15	43.97±3.29
26	6.33±1.22	43.24±6.30	147.09±19.27	35.91±5.28	75	176.16±25.90	44.04±6.48
27	4.75±1.02	65.29±1.34	267.01±22.86	53.87±2.26	130	264.26±11.09	66.07±2.77

Table 2. The Radium equivalent, Hazard indices and Radioactive heat production rate for all sediment samples

Site number	Ra _{eq} (Bq/kg)	Hazard indices			RHP rate A (μWm^{-3})
		H _{ex}	H _{in}	I _{yr} (Bq/kg)	
1	65.59±12.56	0.1773±0.0340	0.2925±0.0446	0.4557±0.0889	0.8342±0.1311
2	50.85±12.38	0.1374±0.0334	0.2048±0.0410	0.3595±0.0880	0.5836±0.1223
3	51.74±10.46	0.1398±0.0282	0.2139±0.0346	0.3649±0.0747	0.5978±0.1004
4	45.60±11.49	0.1232±0.0310	0.1740±0.0383	0.3271±0.0817	0.4759±0.1110
5	91.95±12.73	0.2485±0.0344	0.3374±0.0424	0.6497±0.0904	0.9963±0.1276
6	50.96±9.66	0.1377±0.0261	0.2047±0.0296	0.3652±0.0694	0.5679±0.0880
7	51.24±9.35	0.1385±0.0252	0.2063±0.0285	0.3679±0.0673	0.5612±0.0831
8	53.47±11.20	0.1445±0.0302	0.2009±0.0336	0.3869±0.0803	0.5383±0.0980
9	60.10±10.75	0.1624±0.0290	0.1991±0.0327	0.4372±0.0770	0.6089±0.1024
10	102.56±10.33	0.2772±0.0279	0.3171±0.0310	0.7310±0.0740	1.0592±0.1033
11	73.55±9.97	0.1988±0.0269	0.2490±0.0298	0.5262±0.0715	0.7264±0.0900
12	75.72±10.25	0.2046±0.0277	0.2462±0.0308	0.5473±0.0735	0.7306±0.0953
13	57.54±10.11	0.1555±0.0273	0.1926±0.0310	0.4079±0.0725	0.5828±0.0937
14	54.24±10.81	0.1466±0.0292	0.2180±0.0326	0.4975±0.0777	0.5689±0.0904
15	35.45±11.01	0.0958±0.0298	0.1201±0.0369	0.2643±0.0785	0.3046±0.1047
16	43.20±11.34	0.1168±0.0306	0.1541±0.0372	0.3203±0.0807	0.3688±0.1006
17	59.50±7.18	0.1608±0.0194	0.1807±0.0235	0.4297±0.0509	0.5471±0.0726
18	69.70±9.52	0.1884±0.0257	0.2116±0.0328	0.5030±0.0681	0.6066±0.0925
19	78.37±7.66	0.2118±0.0207	0.2463±0.0240	0.5582±0.0545	0.7081±0.0692
20	75.73±7.36	0.2047±0.0199	0.2246±0.0227	0.5427±0.0525	0.6275±0.0634
21	82.08±5.84	0.2218±0.0158	0.2399±0.0199	0.5856±0.0416	0.6838±0.0540
22	82.76±5.59	0.2237±0.0151	0.2401±0.0184	0.5874±0.0402	0.7186±0.0512
23	88.18±8.00	0.2383±0.0216	0.2513±0.0246	0.6321±0.0570	0.6952±0.0676
24	46.31±6.42	0.1252±0.0173	0.1392±0.0238	0.3370±0.0455	0.3801±0.0651
25	80.35±5.85	0.2172±0.0158	0.2300±0.0196	0.5731±0.0419	0.6532±0.0524
26	79.49±11.71	0.2148±0.0316	0.2317±0.0349	0.5727±0.0840	0.6572±0.0996
27	118.67±4.70	0.3207±0.0127	0.3327±0.0154	0.8626±0.0354	0.7884±0.0316

Using conversion factors quoted by Mohanty et al., (2004), the concentration of radionuclides in Bq kg⁻¹ is converted in terms of ppm and % and substituted in the above relation. As shown in table 2, the heat production rate ranges from 0.3046±0.1047 μWm^{-3} (site no.15) to 1.0592±0.1033 μWm^{-3} (site no. 10) with a mean value of 0.6360±0.0874 μWm^{-3} . The values show low RHP rate (below 1 μWm^{-3}) in the samples of all sites except in site no. 10.

3.2.4.3. Hazard Indices (H_{ex} and H_{in})

The external and internal hazard indices is obtained from Ra_{eq} expression through the supposition that its maximum value allowed (equal to Unity) corresponding to the upper limit of Ra_{eq} (370Bq/Kg). These indices were calculated by following relation (Orgun et al., 2007).

$$H_{ex} = (C_U/370 + C_{Th}/259 + C_K/4810) \leq 1$$

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

where C_U, C_{Th} and C_K are the mean activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in Bq/Kg respectively. Hazard indices (H_{ex} and H_{in}) were calculated for all site samples which are presented in table 2. The maximum values of H_{ex} and H_{in} are 0.3207±0.0127, 0.3374±0.0424 in site no.27. Hazard indices of 27 site samples were less than Unity (permissible level) (Orgun et al., 2007).

3.2.4.4. Gamma Index (I_{yr})

The gamma index takes in to account typical ways and quantities in which material is used in buildings. It is derived to identify, whether dose criterion is met. Activity concentration index is the form to represent the practical monitor of present investigation levels. The Activity concentration index (I_{yr}) was calculated according to below equation suggested by the European Commission (EC 1999)

$$I_r = (C_U/300 + C_{Th}/200 + C_K/3000)$$

Where C_U, C_{Th} and C_K are the mean activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in Bq/Kg respectively. The minimum value of 0.2643±0.0785Bq kg⁻¹ at the site no. 15 and the maximum of 0.8626±0.0354Bq kg⁻¹ are observed at the site no. 27 with a mean of 0.4886±0.0673Bqkg⁻¹. The world average value of I_{yr} is 1Bq kg⁻¹ (Alam et al., 1999). If these (H_{ex}, H_{in} and I_{yr}) hazard indices are higher than unity, which may cause harm to people living in this region. However, in this study, these parameters are within the limit.

4. CONCLUSION

In this study, natural radionuclides ²³⁸U, ²³²Th and ⁴⁰K concentration in Vellar river sediments were detected. The mean activity concentration of all

measured radio nuclides are less than the world average value. The mean value of absorbed dose, observed dose, Annual effective dose equivalent, Radium equivalent activity, Radioactive heat production (RHP) Rate, Hazard indices (H_{ex} and H_{in}) and Gamma index are below the acceptable limit (Recommended Limit). Thus, the Vellar River sediments have acceptable radioactivity level and insignificant radiological hazards. Sediments of the River do not pose a significant radiological threat to the population when it is used for construction of buildings. Hence, human should be aware of their natural environment with regard to the radiation effects due to the naturally occurring radioactive elements.

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