

Potential effect of natural radionuclides in riverbed sediments: a statistical approach based on granulometric and magnetic mineral differences

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Abstract South India is one of the regions in the world that has the highest background radiation levels. In this region, river sediments are used in large quantities as building material. Therefore, the knowledge of the radionuclides distribution in such sediments is important for assessing their potential adverse effects on humans residing in buildings made of sediment material. For this goal, we focus on the determination of the natural radioactivity levels and magnetic properties in sediment samples collected from 33 locations along the southwestern Bharathapuzha river originating from the Anamalai hills. The sediment samples were subdivided into two categories according to particle size. It is observed that the average activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K in sediment samples varied greatly with granulometric and geological differences. The average values of ²²⁶Ra, ²³²Th, and ⁴⁰K and its associated radiological hazard parameters for category II samples (particle size between 149 μm and 2 mm) were lower than category I sediment samples (bulk

samples). Moreover, the average radionuclide activity concentrations (except for ⁴⁰K) and the calculated radiation hazard parameters are higher in the lowland region compared to the highland and the midland regions. The mass-specific magnetic susceptibility values ranged widely along the river, as well as between physiographic regions, e.g., average values for category I sediment samples were 950.2×10^{-8} , 351.1×10^{-8} and $131.8 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$ (for high-, mid- and lowland regions, respectively). Differences between physiographic regions and sediment fractions from both radioactivity determinations and magnetic parameters were analyzed with statistical tests and multivariate analysis, which showed the advantages of using both independent techniques.

Keywords Natural radioactivity · Magnetic measurements · Statistical analysis · Bharathapuzha river sediments

Introduction

All human beings and their surrounding environment are greatly affected by natural radionuclides such as ⁴⁰K and the radionuclides from ²³²Th and ²³⁸U series, which are invariably present in soil, sediment, and the atmosphere. The worldwide average concentrations of radium, thorium, and potassium in the Earth's crust are 35, 30, and 400 Bq kg⁻¹, respectively (UNSCEAR 2000). According to the UNSCEAR (1993) report, about half of the annual radiation dose received by humans from external radiation is due to the exposure from natural radionuclides. Since natural radionuclides are not uniformly distributed, it is necessary to understand the exposure of natural background radiation at different locations (Sroor et al. 2001; Chiozzi et al. 2002).

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In buildings, human exposure to radiation is due to both external and internal sources. The external exposure is mostly due to direct gamma radiation, whereas the internal exposure is due to the inhalation of radon (^{222}Rn), thoron (^{220}Rn), and their short-lived decay products. Radon—an inert gas belonging to the radioactive decay series of uranium is present in almost all building materials. It can move rather freely through porous media and can enter into the indoor air from building materials such as bricks, sand, cement. In general, indoor ^{220}Rn concentrations are very low, but, if building materials contain higher concentrations of thorium, then it may become an important parameter for radiation exposure (European Commission 1999). According to Korhonen et al. (2001), up to 19% of radon exposure comes from building materials. Elevated levels of natural radionuclides in building materials like river sediments may release radon gas, which accumulates in indoor air and gets deposited in the human respiratory system leading to severe health hazards (Quindos et al. 1987). The dose rate measurement of natural radionuclides due to gamma rays is needed to implement precautionary measures whenever the dose is found to be above the recommended limits. Therefore, the analysis and assessment of gamma radiation dose from natural sources is of particular importance as natural radiation is the largest contributor to the external dose of the world population (UNSCEAR 1988). External gamma dose estimation due to the terrestrial sources is essential as these doses vary depending upon the concentrations of the natural radionuclides.

Magnetic techniques in environmental magnetism have been successfully developed and improved, becoming a very useful tool in order to investigate and understand the processes occurring in different environments (Petrovsky and Ellwood 1999). Magnetic susceptibility can be measured in an easy and fast way with low cost. This parameter proved to be a good qualitative proxy for environmental changes and contamination. In the present study, both kinds of variables (radioactivity and magnetic) were gathered and studied using statistical analysis, which allows us to assess particle size categories of sediments and their potential adverse influence to natural radiation emission.

It is well known that India is one of the countries in the world which has the highest background radiation levels. The highest background radiation areas in India are the coastal plains of Kerala, Karnataka, Tamil Nadu, and the southwestern coast of India (Radhakrishna et al. 1993; Kannan et al. 2002; Mishra 1993; Sunta 1993). The reason for the elevated background radiation levels is primarily due to the presence of monazite sands (UNSCEAR 2000). Manigandan and Natrajan (2014) reported higher concentrations of thorium in the region of Western Ghats (Nilgiri highlands, Tamil Nadu) due to the presence of monazite

sand. The phosphate-rich monazite sands are derived from crystalline rocks (Valiathan et al. 1994), mostly granites and gneisses (Manigandan and Chandar Shekar 2015) along the Western Ghats and are transported to the rivers flowing in the coastal regions of Kerala. Thus, the knowledge of radionuclides distribution and radiation level in this environment (Bharathapuzha river basin) is important for assessing the effects of radiation exposure. Bharathapuzha river is the broadest and the second longest river in Kerala with a length of 209 km and basin area of 6186 km² (Sreela et al. 2012). The river basin slope varies between 0° and 70° and is mainly controlled by the local geology and erosion cycles (Magesh et al. 2013).

Since India (South India—Kerala State) is one of the regions in the world with the highest background radiation level, the present study focuses in determining the variation of natural radionuclides (^{226}Ra , ^{232}Th , and ^{40}K), its associated hazard parameters and magnetic minerals with physical (particle size) and geological variations (physiographies regions) in bottom sediments of Bharathapuzha river. The understanding of natural radioactivity concentrations and magnetic minerals with varying particle size is very important when sediments are used as building materials. In addition, statistical studies of radionuclides and magnetic parameters are proposed in order to (1) characterize both particle size categories used for construction; (2) identify the potential adverse influence (natural radiation emission) of sediments by means of particle size and magnetic parameters; and (3) classify sites of interest along the river with most potentially dangerous sediments.

Materials and methods

Study area and sample collection methodology

Bharathapuzha river basin, with a large basin area of 6186 km², is the largest among all the 44 river basins of Kerala and the second longest river (209 km) of the State. In the Western Ghats, Bharathapuzha river originates from Anamalai hills, flows toward the west direction and then empties into the Arabian Sea at Ponnani. As estimated by the Centre for Water Resources Development and Management (CWRDM, Basak et al. 1995; Sreela 2009), the physiography of the river basin is divided into three parts: highlands (topographic height >75 m a.s.l.), midlands (topographic height between 8 and 75 m a.s.l.), and lowlands (topographic height <8 m a.s.l.). In the river basin, about 52% of the land is used for cultivation, forest land includes 26, 8% fallow, and 5% of barren and cultivable land (Lakshmi and Zareena Begum 2016).

Sediment samples were collected from 33 locations along the main channel of the Bharathapuzha river starting

from Upper Aliyar (foot of Anamalai Hills) in Coimbatore district, Tamil Nadu State, to Ponnani (mouth of the river) in Malappuram district, Kerala State (Fig. 1). Each sampling location is separated approximately by a distance of about 5 km. The deposited samples were collected manually by hand from the bottom of the river at a water depth less than 30 cm. The collected samples were immediately packed in polyethylene bags to avoid contaminations, sealed, and then labelled. Much care is taken to ensure only sediment samples are used for further analysis by removing plant matter and other contents from the collected samples.

Classification of sediments and its purpose

From previous studies (Narayana and Rajashekara 2010; Breitner et al. 2010), it is clear that the prime factor affecting the activity concentration of ^{238}U and ^{232}Th is the particle size of sand and clay content of the samples. In particular, the activity concentration is high in fine particle size samples (McCubbin et al. 2004). This is due to the fact that the radioactive elements can be more adsorbed in fine particles and gradually decreases as the particle size increases. Blanco Rodriguez et al. (2008) reported that silt and clay fractions contribute greatly to the highest concentration of natural radioactivity. Similar observation was also reported by Suresh et al. (2012) in Ponnaiyar river and Madruga et al. (2014) in Tejo river sediments. Therefore, to evaluate the variation in the natural radionuclide concentration and magnetic minerals with particle size, the collected sediment samples were classified into two categories:

1. Category I sediments: bulk samples containing sediments of all particle sizes up to 2 mm.
2. Category II sediments: sediment samples containing particle sizes ranging between 149 μm and 2 mm (after complete removal of very fine particles $<149\ \mu\text{m}$ from category I samples). For this, sediment samples from alternate locations (1, 3, 5, ..., 33) were used for analysis. The threshold size for category II (149 μm) was chosen with the objective of removing the contribution of the clay and silt fractions ($<63\ \mu\text{m}$) to radioactivity emission; however, for easy and practical sieving, the very fine sand fraction (between 63 and 149 μm) was also removed. Therefore, the fixed threshold permits to keep fine, medium, and coarse sand fractions (149 μm –2 mm) in category II sediments.

Radioactivity measurements

The collected samples were air-dried followed by oven drying at 110 $^{\circ}\text{C}$ and crushed after passing through 2-mm mesh sieve. The homogenized sediment samples of about 0.5 kg were then packed in a 250-ml plastic container,

shielded hermetically and stored for a period of 1 month to enable radioactive equilibrium to take place among the daughter products of ^{222}Rn and ^{220}Rn and their short-lived decay products (Schotzing and Debertin 1983).

The activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K in the Bharathapuzha river sediment samples was measured using a coaxial n-type high-purity Germanium detector (producer: EG&G, ORTEC, Oak Ridge, USA). The relative efficiency of the detector is 20%, and it has a resolution of 2.0 keV at 1332 keV. To reduce the background level of the system, the detector was maintained in a vertical position and shielded using 4 inch lead bricks on all sides of the detector to reduce the background radiation from building materials and cosmic rays. The output of the detector is analyzed using a 4-K multichannel analyzer system connected to a PC and an ADC for data acquisition.

For the efficiency calibration of the system, the International Atomic Energy Agency (IAEA)-standard reference materials RG U-1 (uranium ore), RG Th-1 (thorium ore), and RG K-1 potassium (K_2SO_4) are used in the same geometry. The spectrum is calibrated with known sources of radioactivity such as ^{152}Eu , and the efficiency values are plotted against the energy for particular geometry. The samples were placed symmetrically on top of the detector and measured for a counting period of 20 h (72,000 s). The spectra are analyzed for the peak of radium, thorium daughter products, and potassium by subtracting counts due to Compton scattering of higher peaks and other background sources from the total area of the peaks. Gamma transitions of ^{40}K were determined by measuring the 1461 keV gamma ray emitted during its decay, whereas gamma rays of the following energies were used to measure the activity of other radionuclides: 186 keV for ^{226}Ra ; 295 keV and 352 keV for ^{214}Pb ; 609 keV, 1120 keV, and 1764 keV for ^{214}Bi ; 338 keV, 463 keV, 911 keV, and 968 keV for ^{228}Ac ; 727 keV for ^{212}Bi ; and 238 keV for ^{212}Pb . The spectral data are analyzed using the software “CANDLE” (Collection and Analysis of Nuclear Data using Linux Network) developed locally by the Inter University Accelerator Centre, New Delhi, India (Ajith Kumar et al. 2001, <http://www.iuac.res.in/NIAS/>, <http://www.iuac.res.in/NIAS/Downloads/candle.txt>).

The activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K and their associated radiation hazard parameters were calculated by using the formula as listed in Table 1.

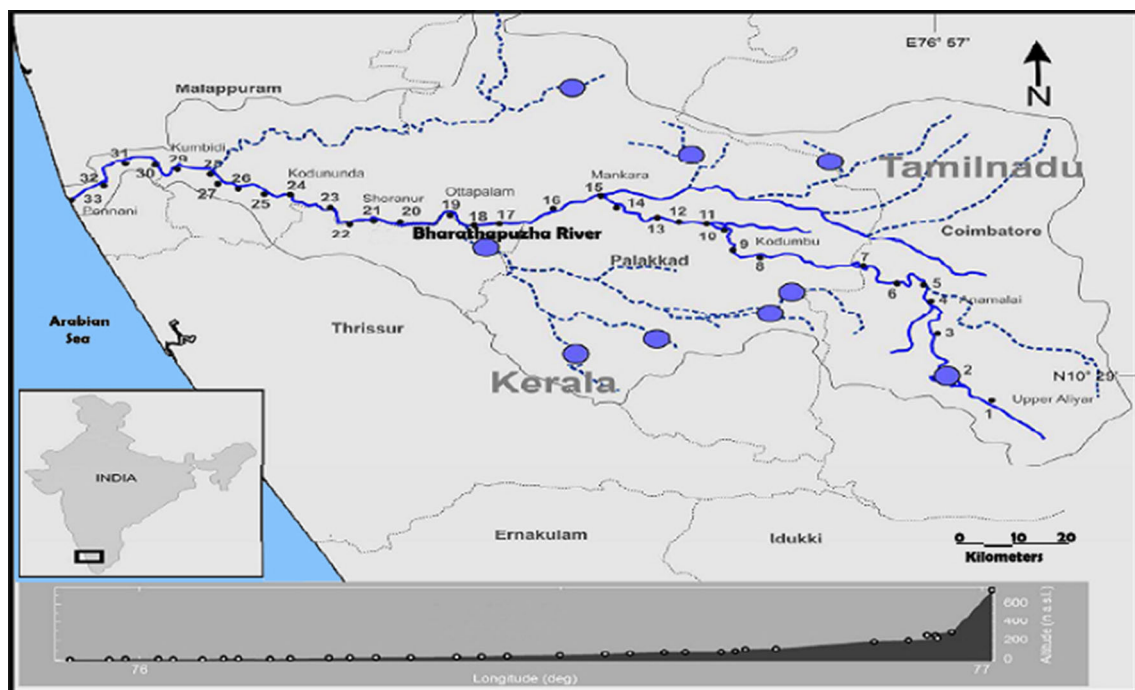
Magnetic measurements

Seventeen sediment samples were subsampled for magnetic studies using plastic containers (2.3 cm^3). Dry samples were sieved (sieve opening of 2 mm) to remove the gravel fraction, separated in the two categories described in “Classification of sediments and its purpose” section, and then

Table 1 Formula used for the estimation of activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K and its associated radiation hazard parameters from activity mass concentrations in bottom sediment samples from different origin along the Bharathapuzha river basin

Parameter	Formula used	Reference
Activity concentration of ^{226}Ra , ^{232}Th and ^{40}K	Activity (Bq kg^{-1}) = $\frac{\text{CPS} \times 100 \times 100}{\text{B.T.} \times \text{Eff}} \pm \frac{\text{CPS}_{\text{error}} \times 100 \times 100}{\text{B.T.} \times 100}$	Mehra et al. (2010)
Air-absorbed dose rate	$D_{\text{AA}} (\text{nGy h}^{-1}) = 0.427 C_{\text{Ra}} + 0.662 C_{\text{Th}} + 0.0432 C_{\text{K}}$	UNSCEAR (1988)
Indoor gamma dose rate	$D_{\text{IN}} (\text{nGy/h}) = 0.92 C_{\text{Ra}} + 1.1 C_{\text{Th}} + 0.080 C_{\text{K}}$	UNSCEAR (1993, 2000) and European Commission (1999)
Radium equivalent activity	$\text{Ra}_{\text{eq}} (\text{Bq kg}^{-1}) = C_{\text{Ra}} + 1.43 C_{\text{Th}} + 0.077 C_{\text{K}}$	Beretka and Mathew (1985)
Annual effective dose equivalent	$\text{AEDE}_{\text{Indoor}} (\mu\text{Sv y}^{-1}) = (D_{\text{AA}}) \text{nGy h}^{-1} \times 8760 \text{ h} \times 0.8 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-3}$ $\text{AEDE}_{\text{Outdoor}} (\mu\text{Sv y}^{-1}) = (D_{\text{AA}}) \text{nGy h}^{-1} \times 8760 \text{ h} \times 0.2 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-3}$	UNSCEAR (2000) UNSCEAR (2000)
Annual gonad dose equivalent	$\text{AGDE} (\mu\text{Sv y}^{-1}) = 3.09 C_{\text{Ra}} + 4.18 C_{\text{Th}} + 0.314 C_{\text{K}}$	Mamont-Ciesla et al. (1982) and Arafa (2004)
Excess life time cancer risk	$\text{ELCR} (\times 10^{-3}) = \text{AEDE} \times \text{DL} \times \text{RF}$	Taskin et al. (2009)

C_{Ra} , C_{Th} , and C_{K} are the activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K , Bq kg^{-1} , respectively

**Fig. 1** Map of sampling locations

packed, weighted, and labelled. Magnetic measurements were carried out at the Institute of Physics IFAS (Tandil, Argentina). Magnetic susceptibility measurements were performed by using a magnetic susceptibility meter MS2, Bartington Instruments Ltd., linked to MS2B dual-frequency sensor (0.47 and 4.7 kHz). The magnetic susceptibility frequency dependence ($\kappa_{\text{FD}} \% = 100 \times [\kappa_{0.47} - \kappa_{4.7\text{kHz}}] / \kappa_{0.47\text{kHz}}$), mass-specific susceptibility (χ), and various other magnetic-dependent parameters (anhysteretic remanent magnetization: ARM, saturation isothermal remanent magnetization: SIRM, remanent coercivity: H_{Cr} , remanence

ratio: S-ratio, SIRM/χ , $\kappa_{\text{ARM}}/\kappa$, SIRM/χ , and ARM/SIRM) are also computed. More details regarding the magnetic measurements are provided in Chaparro et al. (2015).

Statistical methods

Statistical analyses were performed using the R free software (R version 2.15.0 2012). The data set included in the present work is based on magnetic variables, radioactivity variables and its associated hazard parameters for both sediment type samples. The Kruskal–Wallis test (Kruskal

Table 2 Activity concentrations of natural radionuclides (^{226}Ra , ^{232}Th , and ^{40}K) in category I and II sediment samples

Sample number	Activity concentration (Bq kg^{-1})					
	Category I sediment samples			Category II sediment samples		
	^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K
1	30.5	80.59	628.9	40.25	60.36	379.43
2	40.93	52.47	638.88			
3	29.98	38.13	840.08	37.24	58.29	351.77
4	30.25	75.85	899.66			
5	33.96	35.6	400.23	41.21	57.96	285.66
6	32.25	39.48	760.71			
7	45.25	38.06	518.68	39.25	54.32	368.54
8	21.21	39.39	863.83			
9	32.85	58.5	798.14	41.36	56.21	334.49
10	34.25	84.64	749.76			
11	42.58	41.78	690.88	44.88	52.01	380.04
12	32.43	33.49	285.99			
13	36.34	34.74	308.48	32.21	50.02	356.19
14	40.56	34.67	232.25			
15	42.06	51.48	299.62	34.21	48.25	289.68
16	52.83	74.48	271.94			
17	27.28	36.77	308.98	31.12	48.39	329.69
18	45.43	43.42	440.68			
19	50.96	70.92	332.88	40.23	58.36	331.9
20	40.54	41.86	408.56			
21	44.73	46.17	270.73	38.37	54.39	330.07
22	50.7	43.33	331.78			
23	52.57	64.35	428.03	29.26	51.23	306.01
24	32.79	33.56	308.12			
25	66.03	93.1	388.49	44.44	51.78	289.15
26	50.25	66.75	310.19			
27	34.1	45.96	441.4	35.23	54.39	373.03
28	63.7	88.65	475.55			
29	55.91	57.71	386.76	32.09	58.23	338.57
30	40.99	41.95	440.18			
31	62.81	83.44	443.13	50.21	57.28	384.68
32	42.25	93.1	475.55			
33	41.98	45.96	386.76	46.37	59.38	414.44

and Wallis 1952), which is a non-parametric one-way analysis of variance, was used to compare physiographic regions. In order to determine whether there are significant differences between the sediment types, both univariate and multivariate statistical analyses were used. The univariate sign test was applied because the category I and II samples are paired samples and are not normally distributed. The null hypothesis is that the difference median between the continuous distributions is zero. The combination of principal component analysis (PCA) and non-hierarchical clustering (HCA) was implemented to analyze differences between sediment types. This methodology (joint application of PCA and HCA) has two different

perspectives and can reinforce the reliability of our conclusions (Husson et al. 2010).

Results and discussion

Gamma ray analysis

Estimation of radium, thorium, and potassium activity concentration

The distribution of the observed natural radioactivity concentration of ^{226}Ra , ^{232}Th , and ^{40}K in the sediments collected

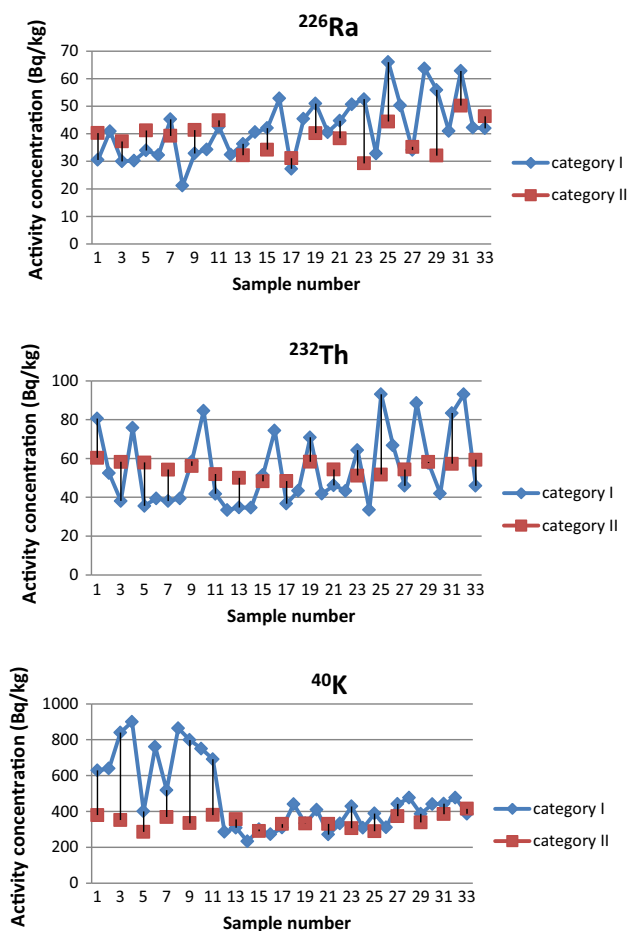


Fig. 2 Activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K in sampling locations in both category I ($N = 33$) and II sediments ($N = 17$) along the Bharathapuzha river basin

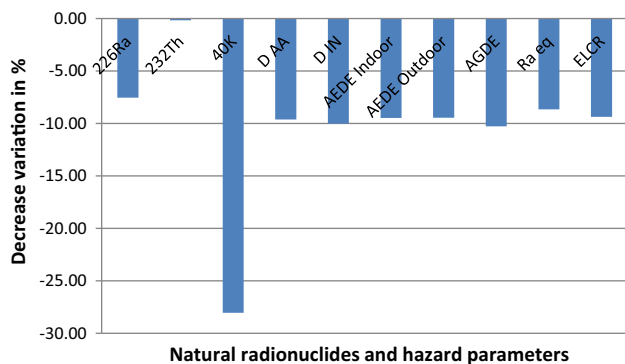


Fig. 3 Decrease percent of average activity concentration and radiological hazard parameters of category II with category I sediment samples

from different locations along the Bharathapuzha river basin for both category I and II samples is shown in Table 2 and Fig. 2. The average activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K in the category I sediment samples are 41.86, 54.86, and 477.75 Bq kg⁻¹, respectively, and in the category II

sediments it is 38.7, 54.76, and 343.73 Bq kg⁻¹, respectively. The higher values of radionuclide concentration in category I sediments are due to the deposition of fine clay particles at the bottom of the river (Gonzalez-Fernandez et al. 2010, 2012), whereas, in category II sediments, such fine particles were completely removed, thus lowering the concentration as indicated in Fig. 3.

Estimation of air-absorbed, indoor gamma dose rate, effective dose, and annual gonad dose equivalent

According to UNSCEAR (2000), the worldwide average value of air-absorbed gamma radiation dose rate from terrestrial sources (D_{AA}) is 57 nGy h⁻¹ and the indoor gamma dose rate (D_{IN}) due to the emissions of gamma rays from natural radionuclides is 54 nGy h⁻¹. The calculated mean values of D_{AA} and D_{IN} for category I sediments are 74.83 and 137.07 nGy h⁻¹, respectively (Table 3), whereas for category II sediments it is 74.83 and 137.07 nGy h⁻¹, respectively (Table 4). For category I and II sediments, the D_{AA} values are about 30 and 20% greater than the world average values, respectively, and the D_{IN} values are, respectively, about 60 and 45% higher. Construction materials like sediments with high values of D_{IN} may cause severe health hazards, which can be prevented to a large extent by using category II sediments instead of category I sediments.

The calculated average values of indoor and outdoor annual effective dose rate ($AEDE_{Indoor}$, $AEDE_{Outdoor}$) for category I sediments are 367.08 and 91.77 $\mu\text{Sv y}^{-1}$, respectively (Table 3). The mean $AEDE_{Indoor}$ and $AEDE_{Outdoor}$ values for category II sediments are 332.3 and 83.08 $\mu\text{Sv y}^{-1}$, respectively (Table 4). The world average values for indoor and outdoor AEDE are 450 and 70 $\mu\text{Sv y}^{-1}$, respectively (Orgun et al. 2007). From Table 4, it should be noted that $AEDE_{Indoor}$ is about 20 and 25% less than the world average value, but $AEDE_{Outdoor}$ is about 30 and 20% higher for category I and II sediments, respectively. Here, for category II sediments, the $AEDE_{Indoor}$ and $AEDE_{Outdoor}$ values are both ~ 10% less than category I sediments (Table 4). Higher values of $AEDE_{Indoor}$ and $AEDE_{Outdoor}$ in category I sediments may be due the presence of high activity concentration of ^{232}Th and ^{40}K (Ramasamy et al. 2011). Therefore, for building construction purposes, category II sediment samples can be extensively used due to the fact that they have lower values of indoor and outdoor AEDE.

The obtained values of annual gonad dose equivalent (AGDE) are between 330 and 716.71 $\mu\text{Sv y}^{-1}$ for category I (Table 3), between 398.35 and 521.63 $\mu\text{Sv y}^{-1}$ for category II sediments, respectively (Table 4). The calculated average value is 70% higher than the world recommended value 300 $\mu\text{Sv y}^{-1}$ for category I sediments and is about 50% for category II sediments. Also, when category II sediments are used as construction material by avoiding

Table 3 Descriptive statistics on natural radionuclides and hazard parameters of Bharathapuzha river sediments based on its geological variations (category I samples)

	Activity concentration (Bq kg ⁻¹)			Dose rate (nGy h ⁻¹)		Annual effective gamma dose rate (μSv y ⁻¹)		Annual gonad dose equivalent (μSv y ⁻¹)	Radium equivalent activity (Bq kg ⁻¹)	Excess life time cancer risk (×10 ⁻³)
	²²⁶ Ra	²³² Th	⁴⁰ K	D _{AA}	D _{IN}	AEDE _{Indoor}	AEDE _{Outdoor}	AGDE	Ra _{eq}	ELCR
Region	All regions: site no. (1–33), n = 33									
Minimum	21.21	33.49	232.25	48.37	89.55	237.28	59.32	330	102.34	0.21
Maximum	66.03	93.1	899.66	106.61	194.24	522.99	130.75	716.71	229.09	0.46
Median	40.99	45.96	428.03	72.77	133.95	356.98	89.25	503.54	147.28	0.31
Average	41.86	54.86	477.75	74.83	137.07	367.08	91.77	508.66	157.09	0.32
S.D	10.90	19.56	195.6	17.81	31.34	87.37	21.84	118.05	37.55	0.08
Region	Highland region: site no. (1–13), n = 13									
Minimum	21.21	33.49	285.99	48.37	89.55	237.28	59.32	330	102.34	0.21
Maximum	45.25	84.64	899.66	103.05	184.59	505.52	126.38	695.05	213.02	0.44
Median	32.85	39.48	690.88	74.33	136.73	364.68	91.17	515.81	149.2	0.32
Average	34.06	50.21	644.94	75.64	138.16	371.08	92.77	517.63	155.52	0.32
S.D	6.24	18.64	207.97	17.64	30.45	86.56	21.64	117.69	35.53	0.08
Region	Midland region: site no. (14–24 and 26–27), n = 13									
Minimum	27.28	33.56	232.25	49.34	90.26	242.04	60.51	335.01	103.65	0.21
Maximum	52.83	74.48	441.4	83.61	153.39	410.16	102.54	565.83	180.28	0.36
Median	44.73	45.96	310.19	64.67	119.29	317.25	79.31	439.23	138.21	0.28
Average	43.45	50.29	337.32	66.41	122.27	325.79	81.44	450.36	141.33	0.29
S.D	8.23	14.11	69.78	12.63	22.6	61.95	15.49	83.23	27.56	0.06
Region	Lowland region: site no. (25 and 28–33), n = 7									
Minimum	40.99	41.95	386.76	64.29	119.07	315.38	78.85	440.23	134.87	0.28
Maximum	66.03	93.1	475.55	106.61	194.24	522.99	130.75	716.71	229.09	0.46
Median	55.91	83.44	440.18	100.22	179.32	491.64	122.91	669.03	212	0.43
Average	53.38	71.99	428.06	88.94	162.54	436.32	109.08	600.26	189.29	0.38
S.D	11.32	22.67	40.54	19.05	33.56	93.43	23.36	124.26	41.52	0.08

S.D indicates standard deviation

Table 4 Variation of natural radionuclides and radiological hazard parameters in category I and II sediments

Parameters	Average (calculated)		Average (world)	Increase/decrease variation of calculated average with world average in %		Decrease average value of category II compared to category I sediments in %
	Category I	Category II		Category I	Category II	
²²⁶ Ra	41.86	38.7	35 (Bq kg ⁻¹)	(+) 19.6	(+) 10.57	(-) 7.55
²³² Th	54.86	54.76	30 (Bq kg ⁻¹)	(+) 82.87	(+) 82.53	(-) 0.18
⁴⁰ K	477.75	343.73	400 (Bq kg ⁻¹)	(+) 19.44	(-) 14.07	(-) 28.05
D _{AA}	74.83	67.62	57 (nGy h ⁻¹)	(+) 31.3	(+) 18.63	(-) 9.64
D _{IN}	137.07	123.33	84 (nGy h ⁻¹)	(+) 63.2	(+) 46.82	(-) 10.02
AEDE _{Indoor}	367.08	332.3	450 (μSv y ⁻¹)	(-) 18.43	(-) 26.16	(-) 9.47
AEDE _{Outdoor}	91.77	83.08	70 (μSv y ⁻¹)	(+) 31.1	(+) 18.69	(-) 9.47
Ra _{eq}	157.09	143.47	370 (Bq kg ⁻¹)	(-) 57.54	(-) 61.22	(-) 8.67
AGDE	508.66	456.4	300 (μSv y ⁻¹)	(+) 70	(+) 52.13	(-) 10.27
ELCR	0.32 × 10 ⁻³	0.29 × 10 ⁻³	0.29 × 10 ⁻³	(+) 10.34	0	(-) 9.38

(+) denoted increase and (-) denotes decrease in value

Table 5 Concentration of magnetic parameters in sediment samples of Bharathapuzha river (category I)

Sample number	χ ($10^{-8} \text{ m}^3 \text{ kg}^{-1}$)	ARM ($10^{-6} \text{ Am}^2 \text{ kg}^{-1}$)	SIRM ($10^{-3} \text{ Am}^2 \text{ kg}^{-1}$)	Z_{ARM} ($10^{-8} \text{ m}^3 \text{ kg}^{-1}$)	K_{FD} (%)	K_{ARM}/K (dimension less)	ARM/SIRM (dimension less)	SIRM/ χ (kA m^{-1})	H_{cr} (mT)	S-ratio (dimension less)
1	1848.7	1131.5	155.4	1546.2	0.31	0.8	0.007	8.4	37.5	0.944
2	752.5	506.1	71.4	677.8	0.97	0.9	0.007	9.5	42.8	0.883
3	1092.6	570.2	74.1	760.8	0.29	0.7	0.008	6.8	35.2	0.92
4	1392.8	591.7	82.1	817.1	0.69	0.6	0.007	5.9	31.8	0.942
5	1231.6	546.1	72.9	703.7	-0.06	0.6	0.007	5.9	31.8	0.934
6	617.0	326.7	40.9	416.2	2.14	0.7	0.008	6.6	36.7	0.932
7	447.0	242.8	29.1	293.8	0.40	0.7	0.008	6.5	36.4	0.929
8	820.7	414.7	56.4	551.8	0.41	0.7	0.007	6.9	33.1	0.944
9	694.8	546.6	67.1	707.6	0.44	1	0.008	9.7	39.9	0.938
10	633.1	321.5	36.3	423.1	0.95	0.7	0.009	5.7	35.9	0.886
11	2160.6	1080.3	149.6	1450.8	1.35	0.7	0.007	6.9	36.2	0.944
12	298.5	205.5	23.1	263.1	1.31	0.9	0.009	7.7	37.2	0.998
13	362.7	227.1	24.3	304.6	0.80	0.8	0.009	6.7	39.1	0.811
14	542.2	336.9	40.1	442.5	0.96	0.8	0.008	7.4	36.8	0.934
15	898.4	547.7	73.3	697.3	0.27	0.8	0.007	8.2	39.1	0.927
16	1043.4	434.2	53.7	598.8	0.54	0.6	0.008	5.1	33.7	0.974
17	442.2	351	40.1	428.8	0.13	1	0.009	9.1	38.5	0.947
18	298.8	166.3	20.2	209.2	0.63	0.7	0.008	6.8	38.5	0.911
19	35.4	67.8	3.5	90.1	10.87	2.5	0.02	9.8	36.4	0.942
20	72.4	62.7	5.3	75.1	5.88	1.0	0.012	7.3	39.8	0.881
21	291.2	234.2	27.4	293.9	2.44	1.0	0.009	9.4	41.7	0.922
22	74.3	61.8	5.5	77.9	4.81	1.0	0.011	7.4	40.3	0.856
23	325.5	138	15.8	169.2	1.64	0.5	0.009	4.8	39.9	0.843
24	92.3	92.3	9.3	109.9	1.58	1.2	0.01	10.1	45.6	0.881
25	290.7	173.3	16.8	210.8	2.74	0.7	0.01	5.8	41.3	0.856
26	114.4	119.9	10.3	155.6	9.11	1.4	0.012	9.0	43.3	0.905
27	333.8	219.7	27.5	280	1.67	0.8	0.008	8.2	39.1	0.885
28	125.0	87.0	9.3	107.5	2.24	0.9	0.009	7.5	41	0.898
29	163.8	104.9	11.9	130.1	2.08	0.8	0.009	7.2	40.1	0.905
30	109.2	87.9	8.2	100.9	2.11	0.9	0.011	7.5	41.2	0.881
31	42.7	50.4	3.9	59.8	1.46	1.4	0.013	9.1	41.7	0.908
32	153.9	119.5	12.0	149.5	0.24	1.0	0.01	7.8	42.6	0.895
33	37.6	54.0	3.5	68.2	1.16	1.8	0.015	9.3	42.6	0.837

Table 6 Concentration of magnetic parameters in sediment samples of Bharathapuzha river (category II)

Sample number	χ ($10^{-8} \text{ m}^3 \text{ kg}^{-1}$)	ARM ($10^{-6} \text{ Am}^2 \text{ kg}^{-1}$)	SIRM ($10^{-3} \text{ Am}^2 \text{ kg}^{-1}$)	χ_{ARM} ($10^{-8} \text{ m}^3 \text{ kg}^{-1}$)	k_{FD} (%)	$k_{\text{ARM}/k}$ (dimension less)	ARM/SIRM (dimension less)	SIRM/ χ (kA m^{-1})	Hcr (mT)	S-ratio (dimension less)
1	1862.7	520.0	59.2	693.9	0.20	0.4	0.009	3.2	37.3	0.893
3	1082.3	292.4	28.4	232.9	2.50	0.2	0.010	2.6	37.3	0.904
5	1082.8	197.5	28.8	252.6	2.10	0.2	0.007	2.7	37.8	0.876
7	351.0	119.5	12.1	141.6	0.00	0.4	0.010	3.4	37.9	0.850
9	386.3	288.1	25.3	368.9	0.00	1.0	0.011	6.5	45.1	0.882
11	2038.3	438.2	60.3	537.4	2.20	0.3	0.007	3.0	37.6	0.825
13	283.6	169.9	13.7	210.5	0.00	0.7	0.012	4.8	40.0	0.903
15	887.6	279.8	33.8	200.7	1.50	0.2	0.008	3.8	36.7	0.913
17	584.5	211.3	21.9	81.9	0.80	0.1	0.010	3.8	37.6	0.843
19	25.5	39.6	2.0	39.5	0.00	1.5	0.019	8.0	38.4	0.879
21	245.8	117.4	10.5	129.9	1.90	0.5	0.011	4.3	46.6	0.838
23	286.9	73.4	10.9	115.9	0.40	0.3	0.009	3.8	45.1	0.702
25	280.9	68.3	10.0	98.9	0.20	0.3	0.009	3.6	46.1	0.699
27	273.9	142.0	15.6	132.8	4.10	0.5	0.009	5.7	47.0	0.801
29	124.3	85.2	5.6	68.5	1.40	0.6	0.015	4.5	44.9	0.719
31	24.4	188.3	5.9	152.3	0.00	6.3	0.032	24.3	40.0	0.984
33	13.2	45.6	1.5	23.8	0.00	1.8	0.029	11.7	52.4	0.845

category I sediments, the risk factor related to the damage of bone marrow and the bone surface cells can be reduced by $\sim 10\%$ (see Table 4).

Estimation of radium equivalent activity and excess life time cancer risk

The radium equivalent activity (Ra_{eq}) is calculated for all the collected samples for category I and II sediments and the average values are presented in Table 4. The average value of Ra_{eq} calculated for the sediment samples and building material is $157.09 \text{ Bq kg}^{-1}$ for category I sediments and $143.47 \text{ Bq kg}^{-1}$ for category II sediments. These values are about 60% less than the worldwide average value for category I and II sediments, respectively (Table 4). It is inferred that for all the sediment samples analyzed, Ra_{eq} value is well within the permissible limit of 370 Bq kg^{-1} (OECD 1979), which implies that the external dose rate exposure to the public is below 1.5 mGy y^{-1} (Krisiuk et al. 1971). Moreover, according to Ramasamy et al. (2011), lower values of Ra_{eq} in sediment samples may be due to the discharge of heavy metals by the constant flow of the river water.

The calculated excess lifetime cancer risk (ELCR) values are between 0.21×10^{-3} and 0.46×10^{-3} and the average value is 0.32×10^{-3} for category I sediments, whereas for category II sediments, the minimum and maximum values are 0.25×10^{-3} and 0.33×10^{-3} , with an average of 0.29×10^{-3} (Table 4). From Table 4, it is clear that, for category I sediments, the average value is about 10% higher than the world average value estimated by UNSCEAR (2000), whereas for category II sediments, the average ELCR is within the permissible hazard level.

The mean values of the radionuclide hazard parameters values for the various physiographic regions are also presented in Table 3. It is noted that the mean values of the hazard parameters are higher in the lowland region than the mean values of highland and midland regions. Such increase may be due to higher concentrations of natural radionuclides, more specifically ^{226}Ra and ^{232}Th , in lowland sampling locations.

Spatial distribution of magnetic susceptibility

Results of magnetic parameters for category I and II samples are displayed in Tables 5 and 6, respectively. The χ values display wide variations in the area, ranging from 35.42×10^{-8} to 2160.62×10^{-8} for category I sediments, and from 13.2×10^{-8} to $2038.3 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$ for category II sediments, respectively. Moreover, the average χ value was 7% higher in category II sediment samples compared with category I sediment samples (Table 7). Hence, assuming the dominance of magnetite-like mineral, the concentration of magnetite ranges between 0.01 and 1% (Thompson and Oldfield 1986). As well-known, the $\kappa_{FD} \%$ values can help to discriminate ultrafine ($<0.03 \mu\text{m}$) superparamagnetic minerals (SP) from single- and multidomain (SD and MD) grains. The $\kappa_{FD} \%$ values for coarser fraction (category II sediment samples) vary from 0 to 4.1% (Table 6), and hence it is indicative of no or scarce presence of SP grains (Dearing et al. 1996) in fraction $>149 \mu\text{m}$ samples. The average $\kappa_{FD} \%$ was about 46% lower in category II sediment samples compared with category I sediment samples (Table 7). The magnetic comparison between bulk (category I) and coarser (category II) sediments shows a decrease in concentration of magnetic minerals as well as of the content of finer magnetic SP grains due to the removal of fine grain materials (fraction $<149 \mu\text{m}$). The graphical representation of the increase/decrease percentage with the calculated mean values of magnetic parameters of category II sediments compared to category I sediments is shown in Fig. 4.

The analysis of magnetic parameters with the altitude was studied using groups of samples regarding three physiographic regions: highland ($n = 13$), midland ($n = 13$) and lowland ($n = 7$) are presented in Table 8. As appreciated in Table 8, the magnetic results reveal distinctive differences between the average values of χ and $\kappa_{FD} \%$. A decreasing pattern is clearly observed for the average values of the magnetic concentration-dependent parameter, in particular, χ shows a regular trend according to the sampling location: It is the highest for samples from highlands ($950.2 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$), it assumes

Table 7 Variation of magnetic concentration, mineralogy and particle-size-dependent parameters of Bharathapuzha river sediments

Parameter	χ	ARM	SIRM	χ_{ARM}	$\kappa_{FD} \%$	κ_{ARM}/κ	ARM/SIRM	SIRM/ χ	H_{cr}	S-ratio
Category I sediments	540.6	309.7	38.8	405.2	1.90	0.93	0.009	7.6	38.7	0.91
Category II sediments	578.5	192.7	20.3	204.8	1.02	0.9	0.013	5.9	41.6	0.84
Variation % of category II sediments compared to category I sediments	(+) 7.01	(-) 37.76	(-) 47.62	(-) 49.45	(-) 46.32	(-) 3.23	(+) 38.60	(-) 22.69	(+) 7.62	(-) 7.70

(+) indicates increase in value and (-) indicates decrease in value

Table 8 Descriptive statistics of magnetic parameters with geological variations in sediments along the Bharathapuzha river basin (category I samples)

Sampling region	Average, median, range and SD	χ ($10^{-8} \text{ m}^3 \text{ kg}^{-1}$)	ARM ($10^{-6} \text{ Am}^2 \text{ kg}^{-1}$)	SIRM ($10^{-3} \text{ Am}^2 \text{ kg}^{-1}$)	Z_{ARM} ($10^{-8} \text{ m}^3 \text{ kg}^{-1}$)	$K_{\text{FD}} \%$ (%)	$K_{\text{ARM}/K}$ (dimension less)	ARM/SIRM (dimension less)	SIRM/ χ (kA m^{-1})	H_{cr} (mT)	S-ratio (dimension less)
All regions (n = 33)	Average	540.6	309.7	38.8	405.2	1.90	0.93	0.009	7.6	38.7	0.910
	Median	333.8	227.1	27.4	293.8	1.20	0.80	0.009	7.4	39.1	0.910
	Range	35.4–2160.6	50.4–1131.5	3.5–155.4	59.8–1546.2	-0.10 to 10.90	0.50–2.50	0.007–0.020	4.8–10.1	31.8–45.6	0.811–0.998
Highland region >75 m AMSL (n = 13)	SD	530.1	269.9	38.2	367.2	2.45	0.39	0.003	1.4	3.4	0.040
	Average	950.2	516.2	67.9	685.9	0.78	0.75	0.008	7.2	36.4	0.920
	Median	752.5	506.1	67.1	677.8	0.70	0.70	0.008	6.8	36.4	0.930
Range	298.5–2160.6	205.5–1131.5	23.1–155.4	263.1–1546.2	-0.10 to 2.10	0.60–1.00	0.007–0.009	5.7–9.7	31.8–42.8	0.811–0.998	
Midland region 8–75 m AMSL (n = 13)	SD	571.8	295.3	42.7	406.9	0.57	0.12	0.001	1.3	3.1	0.0440
	Average	351.1	217.9	25.5	279.1	3.12	1.02	0.010	7.9	39.4	0.910
	Median	298.8	166.3	20.2	209.2	1.60	1.00	0.009	8.2	39.1	0.910
Range	35.4–1043.4	61.8–547.7	3.5–73.3	75.1–697.3	0.10–10.90	0.50–2.50	0.007–0.020	4.8–10.1	33.7–45.6	0.843–0.974	
SD	317.3	156.3	21.3	205.2	3.52	0.51	0.003	1.7	3.0	0.038	
Lowland region <8 m AMSL (n = 7)	Average	131.8	96.7	9.4	118.1	1.71	1.07	0.011	7.7	41.5	0.880
	Median	125.0	87.9	9.3	107.5	2.10	0.90	0.010	7.5	41.3	0.890
	Range	37.6–290.7	50.4–173.3	3.5–16.8	59.8–210.8	0.20–2.70	0.70–1.80	0.009–0.015	5.8–9.3	40.1–42.6	0.837–0.908
SD	85.8	42.0	4.7	51.7	0.82	0.39	0.002	1.2	0.9	0.027	

Table 9 Kruskal–Wallis test for group of samples belonging to different regions of Bharathapuzha River. Analysis for each type of sample. Different letters (a, b and c) indicate statistical differences ($p < 0.05$)

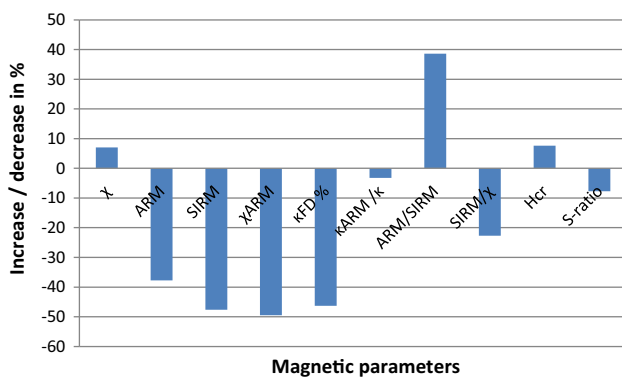
Region	Category I samples							Category II samples						
	χ	κ_{FD} %	^{226}Ra	^{232}Th	^{40}K	D_{AA}	D_{IN}	χ	κ_{FD} %	^{226}Ra	^{232}Th	^{40}K	D_{AA}	D_{IN}
Highland	c	a–b	A	–	b	–	–	II	–	–	–	–	–	–
Midland	b	b–c	B	–	a	–	–	a–b	–	–	–	–	–	–
Lowland	a–b	c	B	–	a–b	–	–	I	–	–	–	–	–	–

I and II indicate sediment category; a, b and c indicate statistical differences

Table 10 Sign test for category I and II sediment samples belonging to Bharathapuzha River. Different letters (a and b) indicate statistical differences ($p < 0.05$)

Category of sediment sample	χ	κ_{FD} %	^{226}Ra	^{232}Th	^{40}K	D_{AA}	D_{IN}
Category I (bulk samples)	a	–	–	–	I	–	–
Category II (>149 μm)	b	–	–	–	II	–	–
p value	0.0023	0.3323	0.999	0.6291	0.049	0.6291	0.6291

I and II indicates sediment category; a and b indicates statistical differences

**Fig. 4** Increase/decrease in percent of the average values of magnetic parameters of category II with category I sediment samples

intermediate values for samples from midlands ($351.1 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$), and the lowest values correspond to samples from χ lowlands ($131.8 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$).

Statistical analysis using radioactivity (^{226}Ra , ^{232}Th , ^{40}K , D_{AA} and D_{IN}) and magnetic variables (χ and κ_{FD} %)

The statistical tests were accomplished in order to investigate: (1) the statistical differences between physiographic regions; (2) the statistical differences between bulk samples (category I) and coarse-grained samples (category II) along the river basin. For category I sediment samples, the average values of magnetic concentration-dependent

parameter (χ) and activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K show differences between highland, midland and lowland regions. Moreover, these average values fit very well with centroids (Groups 1, 2 and 3) from the fuzzy clustering classification (FCC) reported in Krishnamoorthy et al. (2014). It is worth mentioning that this FCC was made without any physiographic information a priori. The Kruskal–Wallis test for this case reveals no statistical differences between both classifications from FCC and physiographic data. This fact reveals that the magnetic/activity data show a distinctive behavior in relation to each physiographic region. On the other hand, for each type of category I samples ($n = 33$) and category II samples ($n = 17$), differences between the three regions were investigated using the Kruskal–Wallis test. As observed in Table 9, there are statistical differences ($p < 0.05$) between the average values of χ , κ_{FD} %, ^{226}Ra and ^{40}K of highland, midland, and lowland regions for category I samples. On the contrary, there are no statistical differences ($p < 0.05$) between the average values for category II samples, except for χ . These results indicate that sediments with different magnetic characteristics and activity concentrations of ^{226}Ra and ^{40}K can be collected along the Bharathapuzha river from different physiographic regions for category I sediment samples. Such differences in magnetic concentration are also possible when the coarse fraction (category II sediments) were collected from different physiographic regions. The differences between the two types of samples were studied using the sign test. Comparison between categories I and II for each region did not reveal statistical

Fig. 5 Principal component analysis (PCA) of samples category I and II. Variables are shown in the plane of the first principal components (PC 1 and PC 2) and (PC 1 and PC 3); variables not reconstructed at the 50% (inner circle) were not represented

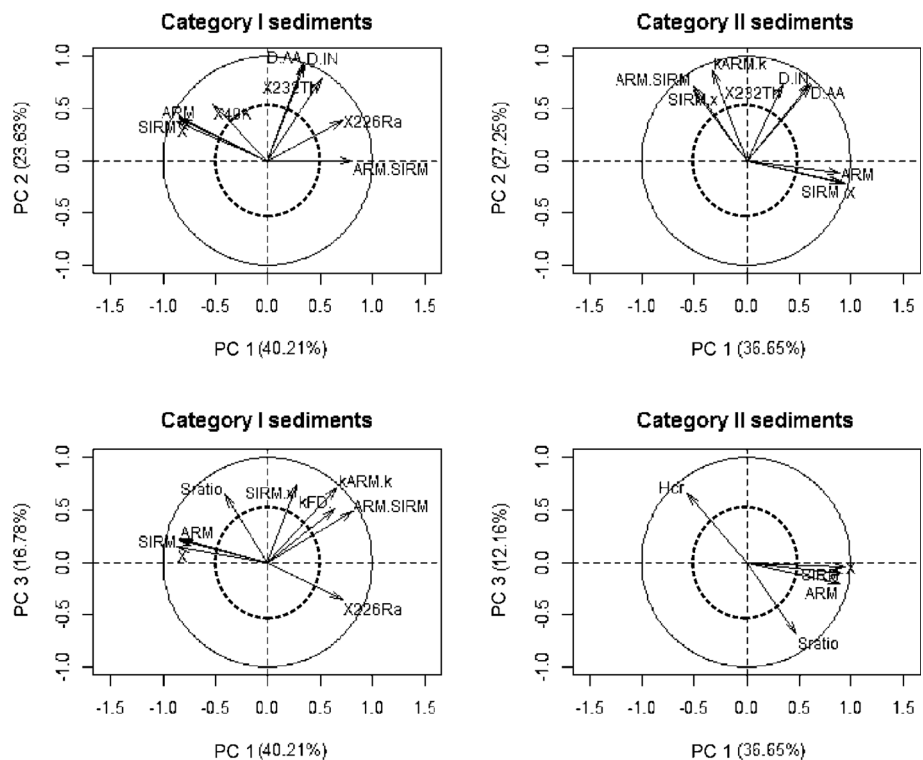


Table 11 Groups for both category I and II sediment samples

Sample no.	Region	Clust I	Clust II
1	H	1	2
11	H	1	2
3	H	1	2
5	H	1	2
9	H	1	3
33	L	2	1
13	H	2	3
15	M	2	3
17	M	2	3
21	M	2	3
27	M	2	3
7	H	2	3
31	L	3	1
19	M	3	3
23	M	3	3
25	L	3	3
29	L	3	3

H highland region, M midland region, and L lowland region

differences between median values. However, there are statistical differences ($p < 0.05$) between category I and II samples for median values of χ and ^{40}K (Table 10) if all sites along the river are taking into account.

These bivariate conclusions were corroborated through multivariate statistics. PCA shows that the relation among

magnetic variables, i.e., magnetic concentration and mineralogical dependent parameters, is similar for both types of samples, as observed from the plots in coordinate planes PC1–PC2 and PC1–PC3 shown in Fig. 5. For example, the relation between variables D_{IN} , D_{AA} , and ^{226}Ra in PC1–PC2 for samples category I is also observed for samples category II in PC1–PC3. Both types of samples show correlation between magnetic concentration-dependent parameters (χ , ARM and SIRM). In addition, for category I samples, these concentration-dependent variables are directly correlated with ^{40}K (PC1–PC2) and inversely correlated with ^{226}Ra (PC1–PC3). Other variables, magnetic mineralogy-dependent parameters: κ_{ARM}/κ , $SIRM/\chi$ and $ARM/SIRM$, are correlated and grouped. The possible influence of extreme values and outliers were analyzed. Although some samples (1, 11, 31 and 33) are potential outliers, the statistical analysis did not change significantly with and without them. Therefore, they are not considered as outliers. As concluded by univariate statistical analysis (sign test, Table 10), it is important mentioning that there are not significant differences between both types of samples.

After the PCA and the analysis of outliers, a cluster analysis was done. The number of clusters was determined using the following criterion,

$$\min_{q_{\min} \leq q \leq q_{\max}} \frac{\Delta(q)}{\Delta(q+1)}$$

Table 12 Descriptive values for each group and for each sample type

Category I sediments					Category II sediments				
Region = H	V. test	Mean C	Mean O	<i>p</i> value	Region = L	V. test	Mean C	Mean O	<i>p</i> value
G1-I					G1-II				
χ	3.29	1405.7	629.4	0.001	ARM/SIRM	3.66	0.031	0.013	0.000
ARM	3.26	774.9	369.7	0.001	SIRM/ χ	3.47	18.0	5.9	0.001
SIRM	3.23	103.8	46.8	0.001	$\kappa_{\text{ARM}}/\kappa$	3.23	4.05	0.90	0.001
^{40}K	3.18	671.65	463.07	0.002	D_{IN}	2.58	140.56	123.33	0.010
^{226}Ra	-2.06	33.97	42.93	0.039	D_{AA}	2.52	76.50	67.62	0.012
H_{cr}	-2.39	36.10	38.60	0.017	^{226}Ra	2.47	48.29	38.70	0.014
Region = Null					Region = H				
G2-I					G2-II				
^{40}K	-2.00	362.10	463.10	0.046	χ	3.48	1516.5	578.5	0.000
^{232}Th	-2.20	42.70	54.30	0.030	SIRM	3.09	44.2	20.3	0.002
D_{AA}	-2.80	60.50	74.30	0.005	ARM	2.85	362.0	192.7	0.004
D_{IN}	-2.80	111.70	136.30	0.004	H_{cr}	-1.99	37.5	41.6	0.047
Region = L					Region = M				
G3-I					G3-II				
^{226}Ra	3.40	57.70	42.90	0.001	ARM	-1.96	145.0	192.7	0.049
^{232}Th	2.80	73.90	54.30	0.005	χ	-2.17	339.1	578.5	0.030
D_{IN}	2.70	166.00	136.30	0.008	^{232}Th	-2.18	53.23	54.76	0.029
D_{AA}	2.60	90.60	74.30	0.009	^{226}Ra	-2.42	36.16	38.70	0.015
$\kappa_{\text{FD}} \%$	2.20	3.80	1.60	0.026	D_{AA}	-2.76	65.01	67.62	0.006
ARM/SIRM	2.00	0.000	0.000	0.043	D_{IN}	-2.77	118.35	123.33	0.006
SIRM	-2.10	10.4	46.8	0.039					
ARM	-2.10	106.9	369.7	0.035					

I and II indicates sediment category; G1, G2 and G3 indicates different groups; V. test indicates whether the mean of the cluster is lower (-) or greater (+) than the mean global; Mean C is the mean of the cluster for each corresponding category; Mean O is the mean global for each corresponding category; *p* value is the probability value corresponding to the test of the following hypothesis: "The mean of the cluster is equal to the mean global"

where $\Delta(q)$ is the between-clusters inertia increase when moving from $q + 1$ to q clusters, q_{min} (and q_{max} , respectively) the minimum (or maximum, respectively) number of clusters chosen by the user.

The obtained groups in each partition for both types of samples are detailed in Table 11. The first groups, G1 and G2, show the same number of individuals for category I (G1-A) and category II (G2-B), and therefore they are not different. These groups show higher values (than the mean global values) for the concentration variables χ , ARM, and SIRM, and lower values for the variable H_{cr} (Table 12). The group G2-A is made of samples from Region M mainly, and it is similar in composition to the G3-B. This group of samples is characterized, in both classifications, by lower values (than the mean global values) for variables ^{232}Th , D_{AA} and D_{IN} . In particular, magnetic variables for G3-B are lower than the mean global values. The group G3-A is made of samples from Region M and L and has similar characteristics to the group G1-B. It is observed

from the variables ARM/SIRM, D_{AA} , D_{IN} , and ^{226}Ra ; which mean values are higher than the mean global values in each group.

Conclusion

The radiological and magnetic measurements performed on sediments sampled from the southwestern Bharathapuzha river basin and the successive statistical analysis have revealed results of relevant impact about the safety of the use of such sediments as building materials. In fact, these sediments belong to one of the region of the world with the highest natural radioactive level. The activity concentration of natural radionuclides (^{226}Ra , ^{232}Th , and ^{40}K) and the associated radiological hazard parameters (except $\text{AEDE}_{\text{indoor}}$ and Ra_{eq}) for category I sediments are slightly higher than the recommended world average values. For category II sediments, the average values of ^{226}Ra and

^{232}Th are slightly higher than the world average values similar to category I sediments, whereas it is about 15% less for ^{40}K , indicating larger accumulation of ^{40}K in coarse-grained sediments. Difference in magnetic susceptibility was also noticed between category I and II sediment samples. Similar variations are noted for the various other magnetic parameters between category I and II sediments. From the statistical analysis, comparison between category I and II sediments for each region did not reveal statistical differences between median values. However, there are statistical differences ($p < 0.05$) between category I and II samples for median values of χ and ^{40}K if all sites along the river are taken into account. This fact indicates lower values of χ and ^{40}K may be obtained if finer particles ($<149\ \mu\text{m}$) are sieved and eliminated from the bulk sediment samples. On the other hand, the statistical test indicates that the sediments with different magnetic characteristics (χ and $\kappa_{\text{FD}}\%$) and activity concentrations of radionuclides (^{226}Ra and ^{40}K) should be chosen from different physiographic regions. For both category I and II, samples from Region H are grouped in a unique group: G1-A and G2-B, respectively. This group has the highest mean values for concentration variables (χ , ARM, SIRM and χ_{ARM}) and the lowest for mineralogy variables ($\kappa_{\text{ARM}}/\kappa$, SIRM/ χ and H_{cr}). Regions M and L for samples category I are clearly characterized by groups G2-A and G3-A. On the other hand, for samples category II, the group G1-B describes the Region L and group G3-B the Region M. In both classifications, samples belonging to Region L have higher mean values (than the mean global values) for variables ARM/SIRM, D_{AA} , D_{IN} , and ^{226}Ra .

Since India is not the only region in the world with high background radiation level that uses sediments for construction purposes, this methodology (the use of at least two particle size sediment fractions) may be applied in other countries/regions and should be considered for assessing sediment and other related materials. In the present study, the average activity concentration of natural radionuclides (^{226}Ra , ^{232}Th , and ^{40}K) were reduced to a considerable percentage and is due to the removal of fine clay, silt and fine sand particles ($<149\ \mu\text{m}$ particles) from category I bulk sediments. As a result, the calculated dose rates (D_{AA} , D_{IN}) and all radiological hazard parameters are lowered in category II sediments making it suitable for the use of building constructions. Thus, it is recommended to local construction industries a sieving procedure of sediments to retain and use coarser sands, hence discarding potentially dangerous finer fractions.

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