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ORIGINAL ARTICLE

Radiation impact assessment of naturally occurring radionuclides and magnetic mineral studies of Bharathapuzha river sediments, South India

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Abstract The natural radioactivity levels and magnetic measurements in sediment samples of Bharathapuzha river for the first time have been determined. Bottom sediments from 33 locations were collected to determine ^{226}Ra, ^{232}Th and ^{40}K using a HPGe detector based on the high-resolution gamma spectrometry system, and magnetic susceptibility by using Bartington MS2 magnetic susceptibility meter. The calculated activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K have been found to vary from 21.21 to 66.03 Bq kg^{-1}, 33.49 to 93.10 Bq kg^{-1} and 232.25 to 899.66 Bq kg^{-1}, respectively. The results have been compared with worldwide recommended values and also with radioactivity measurements in river sediments of India and other parts of the world. The air-absorbed dose rate, indoor and outdoor annual effective dose rates and radium equivalent activity are calculated with an aim to access the radiation hazards arising due to the use of these materials in the construction of buildings and their mean values obtained are 74.83 nGy h^{-1}, 367.08 μSv y^{-1}, 91.77 μSv y^{-1} and 157.09 Bq kg^{-1}, respectively. The mass-specific magnetic susceptibility values ranged widely from 35.4 to 2,160.5 × 10^{-8} m^3 kg^{-1} and compared with other rivers in South India. Multivariate statistical analyses were performed to describe the magnetic and radioactivity relevance of the different groups of samples. The data obtained in the present study may be useful for radiological and magnetic mapping of the study area in the future.

Keywords Natural radioactivity · Hazard indices · Magnetic susceptibility · Fuzzy c-means clustering · River sediments · Bharathapuzha river

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Introduction

Natural radionuclides are present in each and every place in the world. Humans are exposed to natural radioactivity associated mainly to primordial radionuclides such as ^{40}K and the radionuclides from ^{232}Th and ^{238}U series present in varying amounts in soil, building materials, water, rocks and atmosphere (Tzortzis et al. 2004). Building materials like brick, stone, cement and sediments (sand) contain higher concentrations of radionuclides (Turtiainen et al. 2008). The radiation exposure from these natural radionuclides contributes significantly to the average annual dose to humans from all sources of radiation (Senthilkumar et al. 2010). It is a well-known fact that 7–19 % of radon originates from building materials (Korhonen et al. 2001).

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Higher values of indoor dose rates are due to higher concentrations of naturally occurring radionuclides (Petropoulas et al. 2002). Gamma radiations of ^{226}Ra, ^{232}Th decay series and ^{40}K cause external exposure and inhaling of ^{222}Rn, ^{220}Rn and their short lived progeny caused internal exposure of the respiratory track and acts as agents of lung cancer (Sevc et al. 1976; Savidou et al. 1996) especially in buildings which have low ventilation (Porstendorfer 1994). Activity concentration measurements in sediments are used to access radiological risk from elucidation to Naturally Occurring Radioactive Materials (NORM) (Rao et al. 2009).

In India (mostly in South India), river sediments are used primarily for the construction of buildings. Sediments (sand) are nothing but mineral deposits formed through weathering and erosion of either igneous or metamorphic rocks (Veiga et al. 2006). River sediments can be considered as an environmental host for many chemical, biological and aquatic waste products discharged by humans because water pollution by radionuclide components gets deposited in the sediments. The concentration of radiation levels differs by geological location and by the type of rock, soil or sediments (Mirza et al. 1991; UNSCEAR 2000). Thus the knowledge and detailed understanding of the various types of radioactive materials are very essential to humans in monitoring the changes in natural background radiation (Sroor et al. 2001; Chiozzi et al. 2002; El-Bahi 2004). The study of natural radioactivity in sediments provides information about the nature of radionuclides in aquatic environments and the health hazards due to radionuclides in rivers (Lu et al. 2008). Such data can be used as a baseline from which we can study the artificial release of radioactive nuclides in the future (Abdi et al. 2009; Agusa et al. 2004; Yii et al. 2009).

Magnetic susceptibility is, perhaps, the best parameter for assessing magnetic concentration in environmental samples, assuming uniform mineralogy and consideration of paramagnetic and diamagnetic components (Peters and Dekkers 2003). In environmental magnetism, magnetic techniques have been successfully developed and improved, becoming a very useful tool to investigate and understand the processes occurring in different environments (Chaparro et al. 2006). The magnetic susceptibility method in magnetism has been very well accepted for mapping anthropogenic pollution, becoming useful because it uses only one single parameter (Petrovský and Ellwood 1999). Measurements are easy, prompt and cost-effective to carry out. Thus, it becomes possible to establish dense grids of sites under study (e.g., Blundel et al. 2009), and therefore, to produce 2D or 3D maps of magnetic parameters, which could be used as proxies of certain pollutants. It allows us to assess study areas (e.g., soils, rivers, etc.) for reduced or wide extensions.

The goal of the present study is to measure the activity concentrations of natural radionuclides and to calculate the radiological hazard indices, as well as, to measure the magnetic susceptibility of the bottom sediments of Bharathapuzha river, Tamilnadu/Kerala, South India. It is also of interest for this study to assess the distribution of radionuclides and magnetic minerals along the river using a multivariate statistical analysis with integrated variables.

Materials and methods

Study area

The Bharathapuzha river is the second longest river in Kerala with a length of 209 km. The river originates in the Anamalai hills located in the Western Ghats region in Tamil Nadu. The basin lies approximately between 10°26′ and 11°13′ north latitudes and 75°53′ to 77°13′ east longitudes. For the first 40 km, Bharathapuzha river flows in the northwards till Pollachi in Coimbatore District, Tamilnadu State. The Kannadippuzha and Kalpathippuza, the tributaries of Bharathapuzha meet at Parli and flow in the west direction and end at Ponnani into the Arabian sea draining through Palakkad and Malappuram Districts in Kerala State (Fig. 1). There are several dams constructed across this river among which the Malampuzha dam is the largest. With a drainage basin of 6,186 km^2, Bharathapuzha basin is one of the largest river basins in Kerala, useful to irrigate an area of 773 km^2. Out of the total drainage area two-thirds, which is 4,400 km^2, are in Kerala and the remaining 1,786 km^2 are in Tamil Nadu.

Sample collection

Sediment samples were collected along the Bharathapuzha river region of 33 locations starting from Upper Aliyar (foot of Anamalai Hills) in Tamilnadu State to Ponnani (Arabian Sea) in Kerala State during the dry season period between March 2012 and June 2012. Locations of sampling site are given with their latitude and longitude in terms of degree–minute–second. Each sampling location is separated approximately by a distance of 4–5 km. The deposited samples were collected by hand from the bottom of the river in polyethylene bags and then were air dried on aluminum trays at room temperature in open air.

Radioactivity measurements

Sample preparation

The collected samples were sieved through 2-mm mesh sieve and then were dried in an oven 110 °C till constant dry weight was obtained, crushed and homogenized. The fine quality of the sample is obtained using a scientific

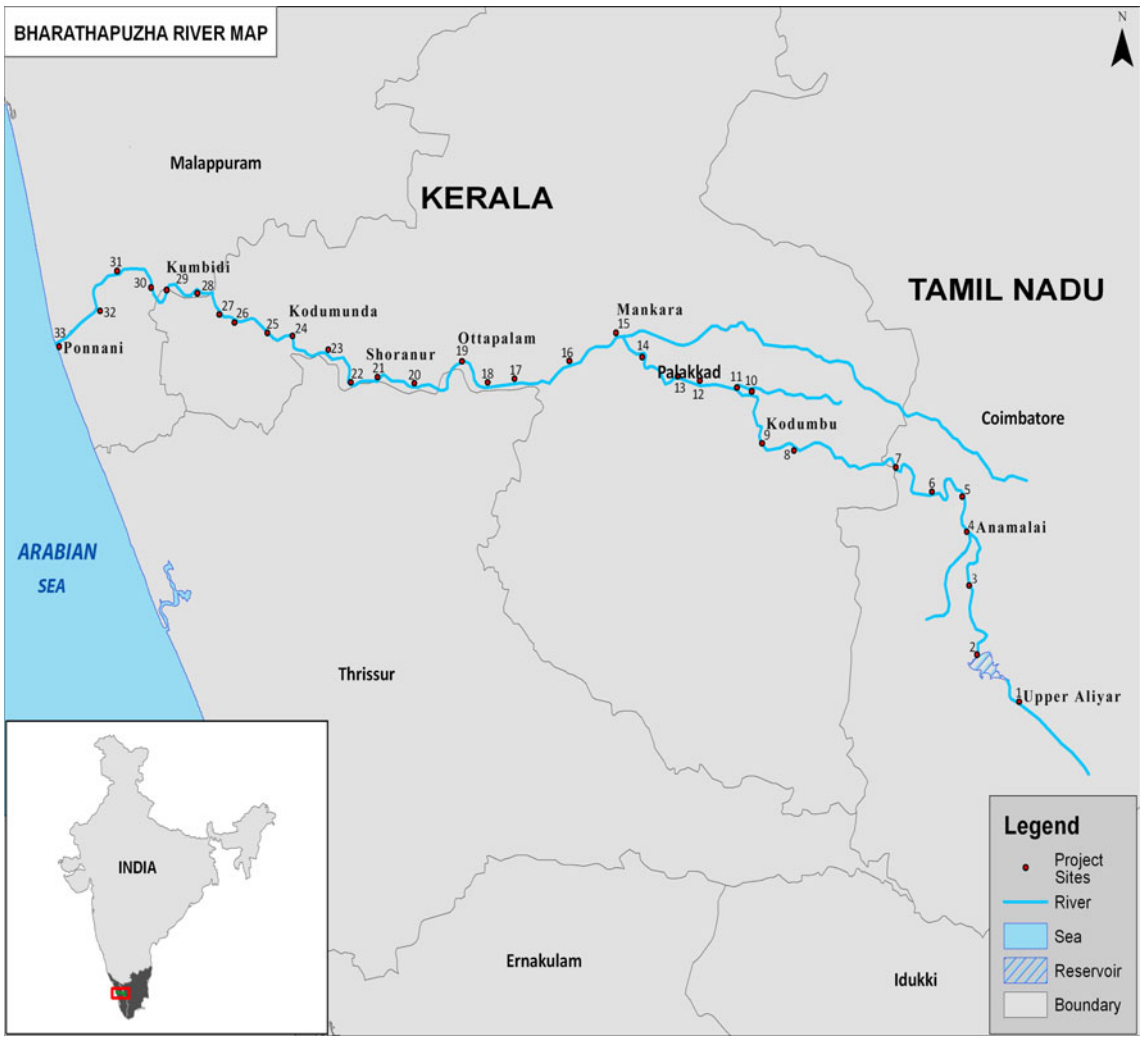

Fig. 1 Map of the study area showing sampling locations along Bharathapuzha river

sieve of 150 microns-mesh size. The homogenized samples were packed in a 250 ml plastic container (9 cm × 6.5 cm: Height × Diameter) to its full volume. These containers are shielded hermetically and also shielded externally to ensure that all daughter products of uranium and thorium, in particular, radon isotope formed, do not escape. The samples were stored for a period of 4 weeks to enable radioactive equilibrium among the daughter products of radon ^{222}Rn, thoron ^{220}Rn and their short lived decay products (Schotzing and Debertin 1983). An average of about 0.50–0.60 kg of sediment is used per sample.

Instrument used and procedure

Using a coaxial n-type HPGe detector based on high-resolution gamma spectrometry system (EG&G, ORTEC, Oak Ridge, USA), the activity concentration of the primordial radionuclides ^{226}Ra, ^{232}Th and ^{40}K in sediment samples is counted. The detector having a resolution of 2.0 keV at 1,332 keV gamma energy of Co-60 and a relative efficiency of 20 % was placed in 4″ shield of lead brick on all sides 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.

A secondary standard which was calibrated with the primary standard (RGU-1) obtained from the International Atomic Energy Agency (IAEA) was used to calibrate the low background counting system. 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. Gamma transitions of 1,461 keV for ^{40}K; 186 keV of ^{226}Ra and 609 keV of ^{214}Bi for ^{226}Ra; 338, 463, 911, 968 keV of ^{228}Ac, 727 keV of ^{212}Bi, 238 keV of ^{212}Pb and 583 keV of ^{208}Tl for ^{232}Th, were used for the laboratory measurement of activity concentration of potassium, radium and thorium. The samples were counted for a period of 72,000 s and the spectra

are analyzed for the photo peak of radium, thorium daughter products and potassium (Mehra et al. 2007). The concentrations of radionuclides are calculated using the equation:

$$\text{Activity}\left(\text{Bq kg}^{-1}\right) = \frac{\text{CPS} \times 100 \times 100}{\text{B.I.} \times \text{Eff}} \pm \frac{\text{CPS}_{\text{error}} \times 100 \times 100}{\text{B.I.} \times 100} \quad (1)$$

where, CPS, net count rate per second; B. I., branching Intensity; and Eff, efficiency of the detector.

Magnetic measurements

Sample preparation and instrument used

Thirty-three sediment samples were sub sampled for magnetic studies using plastic containers (2.3 cm^3). Dry samples were sieved (2 mm) to remove gravel fraction, and then they were packed, weighted, and labeled. Magnetic measurements were carried out in the Institute of Physics IFAS (Tandil, Argentina). Magnetic susceptibility

Table 1 Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1} (dry weight) and magnetic susceptibility measurements in sediments of Bharathapuzha river

Sample number	Sample location	Latitude	Longitude	Activity concentration (Bq kg^{-1})			Magnetic parameters	
				^{226}Ra	^{232}Th	^{40}K	χ (10^{-8} m^3 kg^{-1})	κ_{FD}% (%)
1	Upper Aliyar	10°26′15.65″N	77°0′43.68″E	30.5	80.59	628.9	1,848.7	0.3
2	Aliyar	10°29′1.42″N	76°57′51.87″E	40.93	52.47	638.88	752.5	1
3	Vepparai	10°32′50.88″N	76°57′2.28″E	29.98	38.13	840.08	1,092.6	0.3
4	Anamalai	10°34′47.13″N	76°56′19.38″E	30.25	75.85	899.66	1,392.8	0.7
5	Ambarampalayam	10°38′13.91″N	76°56′50.35″E	33.96	35.6	400.23	1,231.6	−0.1
6	KG Pudur	10°38′30.62″N	76°54′46.09″E	32.25	39.48	760.71	617	2.1
7	Meenakshipuram	10°39′56.65″N	76°52′18.71″E	45.25	38.06	518.68	447	0.4
8	Vilayodi	10°40′55.61″N	76°45′19.98″E	21.21	39.39	863.83	820.7	0.4
9	Ambattampalayam	10°41′21.12″N	76°43′8.47″E	32.85	58.5	798.14	694.8	0.4
10	Dharmanagar	10°44′22.39″N	76°42′25.93″E	34.25	84.64	749.76	633.1	1
11	Kodumbu	10°44′36.43″N	76°41′24.98″E	42.58	41.78	690.88	2,160.6	1.4
12	Kannadi	10°44′59.84″N	76°38′51.78″E	32.43	33.49	285.99	298.5	1.3
13	Thirunellai	10°45′15.51″N	76°37′22.11″E	36.34	34.74	308.48	362.7	0.8
14	Anikode	10°46′23.07″N	76°34′56.27″E	40.56	34.67	232.25	542.2	1
15	Parali	10°47′54.45″N	76°33′10.67″E	42.06	51.48	299.62	898.4	0.3
16	Mankara	10°46′14.55″N	76°29′56.59″E	52.83	74.48	271.94	1,043.4	0.5
17	Lakkidi	10°45′7.28″N	76°26′11.26″E	27.28	36.77	308.98	442.2	0.1
18	Pallapuram	10°44′56.77″N	76°24′21.72″E	45.43	43.42	440.68	298.8	0.6
19	Ottapalam	10°46′7.61″N	76°22′35.46″E	50.96	70.92	332.88	35.4	10.9
20	Mannanur	10°44′51.45″N	76°19′20.03″E	40.54	41.86	408.56	72.4	5.9
21	Shoranur	10°45′13.07″N	76°16′49.33″E	44.73	46.17	270.73	291.2	2.4
22	Mundaya	10°44′53.98″N	76°14′58.54″E	50.7	43.33	331.78	74.3	4.8
23	Karakkad	10°46′52.42″N	76°13′28.97″E	52.57	64.35	428.03	325.5	1.6
24	Pattambi	10°47′56.92″N	76°40′43.88″E	32.79	33.56	308.12	92.3	1.6
25	Kannanur	10°47′48.70″N	76°9′17.70″E	66.03	93.1	388.49	290.7	2.7
26	Thrithala	10°48′23.12″N	76°7′1.81″E	50.25	66.75	310.19	114.4	9.1
27	Pattithara	10°48′51.04″N	76°5′59.00″E	34.1	45.96	441.4	333.8	1.7
28	Koodalur	10°50′6.91″N	76°4′28.47″E	63.7	88.65	475.55	125	2.2
29	Kumbidi	10°50′17.67″N	76°2′22.61″E	55.91	57.71	386.76	163.8	2.1
30	Kuttipuram	10°50′26.88″N	76°1′19.85″E	40.99	41.95	440.18	109.2	2.1
31	Thavanur	10°51′23.81″N	75°58′59.07″E	62.81	83.44	443.13	42.7	1.5
32	Chamravattom	10°49′4.02″N	75°57′49.99″E	42.25	93.1	475.55	153.9	0.2
33	Ponnani	10°46′59.54″N	75°55′2.30″E	41.98	45.96	386.76	37.6	1.2

measurements were performed using a magnetic susceptibility meter MS2, Bartington Instruments Ltd., linked to MS2B dual frequency sensor (0.47 and 4.7 kHz). The volumetric susceptibility (κ), magnetic susceptibility frequency-dependence [κ_{FD} % = 100 × ($\kappa_{0.47} - \kappa_{4.7}$)/$\kappa_{0.47}$] and mass-specific susceptibility ($\chi = \kappa/\rho$, where ρ is the density) were computed.

Statistical methods

Multivariate statistical analyses were performed using the R free software: R version 2.15.0 (2012-03-30) (2012). The descriptive statistics and correlation Pearson's coefficients for all variables and data were calculated as a first step. Then, the fuzzy *c*-means clustering (FCC) was studied. Thirty-three ($n = 33$) samples were used for this investigation. The data set included one magnetic variable (χ) and three radioactivity variables (^{226}Ra, ^{232}Th and ^{40}K).

Results and discussions

^{226}Ra, ^{232}Th and ^{40}K activity concentrations

Activity concentrations of the radionuclides ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1} for the river bottom sediments of Bharathapuzha river are summarized in Table 1 and shown in Fig. 2. The activity concentrations are between 21.21 and 66.03 Bq kg^{-1} for ^{226}Ra, 33.49 and 93.10 Bq kg^{-1} for ^{232}Th and 232.25 and 899.66 Bq kg^{-1} for ^{40}K. The average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides are 41.86, 54.86 and 477.75 Bq kg^{-1}, respectively.

In our present study, the activity concentration of sediment samples collected from different sampling sites varies greatly and this is due to the large variation in their physical, chemical, mineralogical and geological structures (Krmar et al. 2009). ^{226}Ra is considered as the most highly radio-toxic natural radionuclide (Akhtar et al. 2005). From the data presented in Table 1, the concentration of ^{226}Ra is more towards the mouth of the river. It must be noted that Ra is the daughter nucleus of U. The higher concentration of ^{226}Ra towards the mouth may be due to the solubility and mobility of U (VI) O_2^{2+} (Powell et al. 2007) and from the extensive exploitation of phosphate fertilizers in the surrounding agricultural area (Bikit et al. 2005). This is also due to the fact that the sampling locations are located in the downstream of the river (War et al. 2012).

The activity concentration of ^{232}Th was found to be higher than ^{226}Ra in almost all the sampling locations. This may be due to the fact that ^{232}Th is insoluble in water and also due to its low geochemical mobility (Suresh et al. 2011). The increasing value of ^{232}Th may be due to the high content of monazite in the study area (Orgun et al. 2007). Also the concentration of ^{232}Th depends upon the presence of metamorphic rocks like shale and the nature of geological sites (Agbalagba and Onoja 2011).

The higher concentration levels of ^{40}K are observed at sites near to the origin of the river than towards its mouth. This may be due to the fact that, potassium fertilizers are used excessively in agricultural lands in these areas (Akhtar et al. 2005). Higher concentration of ^{40}K may be also due to the presence of clay sediments. In sampling locations towards the mouth of the river, lower values of ^{40}K are observed, which may be due to the decrease in loamy and clay sediments (El-Gamel et al. 2007).

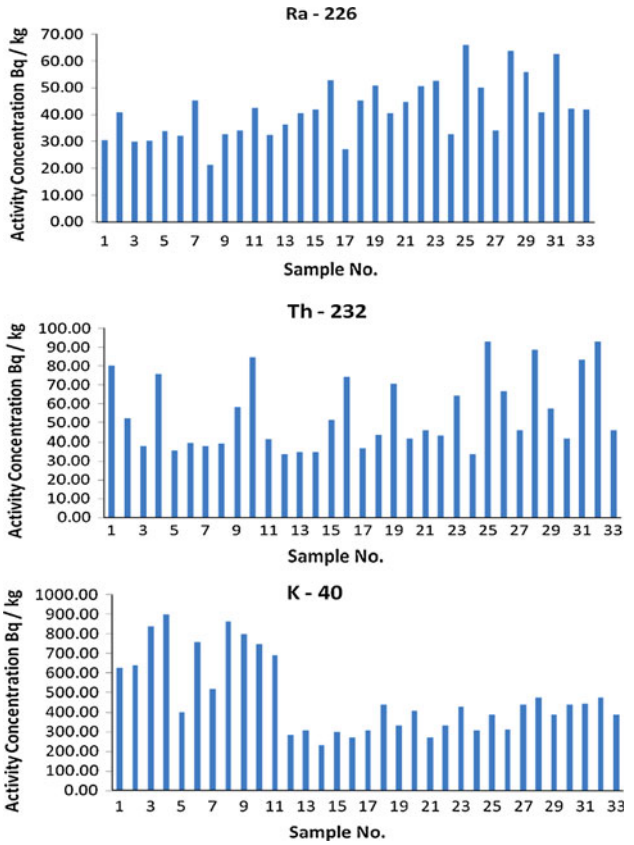

Fig. 2 Activity concentration for ^{226}Ra, ^{232}Th and ^{40}K in river sediment samples

Table 2 Average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1}) and its International and Indian recommended values (UNSCEAR 2000) in sediment samples (dry weight)

Radionuclide	Average activity concentration (Bq kg^{-1})	UNSCEAR (International) (Bq kg^{-1})	UNSCEAR (Indian) (Bq kg^{-1})
^{226}Ra	41.86	35	28.67
^{232}Th	54.86	30	63.83
^{40}K	477.75	400	400

Table 3 Summary of average activity concentration measurements of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in river sediments worldwide including India

Name of the river	Country	Activity concentration (Bq kg^{-1})				Reference
		^{238}U	^{226}Ra	^{232}Th	^{40}K	
Dudvah river	Slovak Republic	–	33.4	43.6	587	Frantisek et al. (2008)
Ogun river	Nigeria	–	12.65	11.78	499.48	Jibiri and Okeyode (2012)
Reedy river	USA	37.8	21.4	45.3	609.3	Powell et al. (2007)
Shango river	Bangladesh	25.4	–	57.5	255	Chowdhury et al. (1999)
Chao Phraya river	Thailand	60.2	–	64.9	431.8	Santawamaitre et al. (2011)
Bakircay river	Turkey	–	ND-160.57	ND-131.49	ND-839.19	Sac et al. (2012)
Gediz river	Turkey	–	17–257	1.22–69.58	2.96–1,038.96	Bakac and Kumru (2001)
Firtina river	Turkey	–	15–116	17–87	51–1,605	Kurnaz et al. (2007)
Wei river	China	–	10.4–39.9	15.3–54.8	514.8–1,175.5	Lu et al. (2008)
Ponnaiyar river	India	7.31	–	46.85	384.03	Ramasamy et al. (2011)
Bharathapuzha river	India	–	41.86	54.86	477.75	Present study

ND not detected

In almost all the sampling locations, the activity concentration is of the order $^{226}Ra < {}^{232}Th < {}^{40}K$. According to the report by UNSCEAR (2000), the worldwide average concentrations of the radionuclides ^{226}Ra, ^{232}Th and ^{40}K are 35, 30 and 400 Bq kg^{-1}, respectively. The average activity concentrations obtained from the results show that ^{226}Ra and ^{40}K are 1.2 times higher than the International recommended average value whereas ^{232}Th is 1.8 times greater. It is to be noted that the ^{226}Ra is 1.46 and ^{40}K is 1.2 times greater than the Indian average value of 28.67 and 400 Bq kg^{-1}, respectively, whereas ^{232}Th is within the Indian recommended value (63.83 Bq kg^{-1}) (Ramasamy et al. 2011) as shown in Table 2. The measured concentrations of radionuclides are also compared to rivers of other countries and in India (Table 3). From the comparison, it is found that the mean value of ^{226}Ra is higher than the values obtained in other river sediments across the world. According to Gonzales-Fernandez et al. (2012), accumulation of radionuclides is due to the restricted water circulation and deposition of fine particles. These fine particles settled at the bottom of the river increase the activity concentration of natural radionuclides (Gonzales-Fernandez et al. 2010).

Evaluation of radiological hazard parameters

Air-absorbed dose rate (D)

The measured activities of ^{226}Ra, ^{232}Th and ^{40}K were converted into doses (nGy h^{-1}/Bq kg^{-1}) by applying the conversion factors 0.427, 0.662 and 0.0432 for uranium, thorium and potassium, respectively (UNSCEAR 1988). It is assumed that ^{137}Cs, ^{90}Sr and the ^{235}U decay series can be neglected as they contribute very little to the total dose from an environmental background (Kocher and Sjoreen 1985). These factors were used to calculate the total adsorbed gamma dose rate in air at 1 m above the ground level using the following equation:

$$D(\text{nGy h}^{-1}) = 0.427C_{\text{Ra}} + 0.662C_{\text{Th}} + 0.0432C_{\text{K}} \quad (2)$$

where, C_{U}, C_{Th} and C_{K} are the activity concentrations (Bq kg^{-1}) of radium, thorium and potassium in the samples. The air-absorbed dose rates for the locations under study varied from 48.37 to 106.61 nGy h^{-1} and are listed in Table 4. The international average value for gamma radiation dose level from terrestrial sources is 57 nGy h^{-1} (UNSCEAR 2000) and the Indian average value is 69 nGy h^{-1} (Kamath et al. 1996). The calculated average air-absorbed dose rate for the Bharathapuzha river sediments is 74.83 nGy h^{-1}. This value is 1.3 times greater than the world average value and 1.08 times greater than the Indian average value. It is also calculated that the relative contribution of ^{226}Ra, ^{232}Th and ^{40}K to the air-absorbed dose rate is 23 % from ^{226}Ra, 49 % from ^{232}Th and 28 % from ^{40}K, which means that the absorbed dose rate in air in the present sampling locations is primarily due to ^{232}Th.

Annual effective dose equivalent (AEDE)

To estimate annual effective doses, account must be taken of (a) the conversion coefficient from absorbed dose in air to effective dose and (b) the indoor occupancy factor. Annual estimated average effective dose equivalent received by a member is calculated using a conversion factor of 0.7 Sv Gy^{-1}, which is used to convert the

Table 4 Air-absorbed dose rate (D), indoor and outdoor annual effective dose equivalent (AEDE) and radium equivalent activity (Ra_{eq}) for the sediment samples

Sample number	Air-absorbed dose rate (D) (nGy h^{-1})				Annual effective dose equivalent (AEDE) (μSv y^{-1})		Radium equivalent activity (Ra_{eq}) (Bq kg^{-1})
	^{226}Ra	^{232}Th	^{40}K	Total (D)	Indoor	Outdoor	
1	13.02	53.35	27.17	93.54	458.89	114.72	194.17
2	17.48	34.74	27.6	79.81	391.52	97.88	165.15
3	12.8	25.24	36.29	74.34	364.67	91.17	149.2
4	12.92	50.21	38.87	102	500.35	125.09	207.99
5	14.5	23.57	17.29	55.36	271.56	67.89	115.69
6	13.77	26.14	32.86	72.77	356.97	89.24	147.28
7	19.32	25.2	22.41	66.92	328.30	82.07	139.61
8	9.06	26.08	37.32	72.45	355.42	88.86	144.06
9	14.03	38.73	34.48	87.23	427.92	106.98	177.96
10	14.62	56.03	32.39	103.05	505.50	126.38	213.02
11	18.18	27.66	29.85	75.69	371.29	92.82	155.53
12	13.85	22.17	12.35	48.37	237.30	59.32	102.34
13	15.52	23	13.33	51.84	254.31	63.58	109.77
14	17.32	22.95	10.03	50.3	246.76	61.69	108.01
15	17.96	34.08	12.94	64.98	318.76	79.69	138.74
16	22.56	49.31	11.75	83.61	410.17	102.54	180.28
17	11.65	24.34	13.35	49.34	242.03	60.51	103.65
18	19.4	28.74	19.04	67.18	329.56	82.39	141.46
19	21.76	46.95	14.38	83.09	407.61	101.90	178.01
20	17.31	27.71	17.65	62.67	307.45	76.86	131.86
21	19.1	30.56	11.7	61.36	301.01	75.25	131.6
22	21.65	28.68	14.33	64.67	317.23	79.31	138.21
23	22.45	42.6	18.49	83.53	409.79	102.45	177.54
24	14	22.22	13.31	49.53	242.97	60.74	104.51
25	28.19	61.63	16.78	106.61	523.00	130.75	229.09
26	21.46	44.19	13.4	79.04	387.76	96.94	169.58
27	14.56	30.43	19.07	64.05	314.22	78.56	133.81
28	27.2	58.69	20.54	106.43	522.12	130.53	227.09
29	23.87	38.2	16.71	78.79	386.51	96.63	168.22
30	17.5	27.77	19.02	64.29	315.37	78.84	134.87
31	26.82	55.24	19.14	101.2	496.44	124.11	216.25
32	18.04	61.63	20.54	100.22	491.63	122.91	212
33	17.93	30.43	16.71	65.06	319.16	79.79	137.48

absorbed rate to human effective dose equivalent with an outdoor occupancy of 20 and 80 % for indoors (UNSCEAR 2000; Singh et al. 2009).

The annual effective doses are determined as follows:

$$\text{AEDE indoor } (\mu\text{Sv y}^{-1}) = (\text{Absorbed dose}) \text{ nGy h}^{-1} \times 8760\, h \times 0.8 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-3} \quad (3)$$

$$\text{AEDE outdoor}\left(\mu\text{Sv y}^{-1}\right) = (\text{Absorbed dose}) \text{ nGy h}^{-1} \times 8760\, h \times 0.2 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-3} \quad (4)$$

The calculated values of annual effective indoor dose rate are between 237.3 and 523.00 μSv y^{-1} with an average value of 367.08 μSv y^{-1} and annual effective outdoor dose rate ranges between 59.32 and 130.75 μSv y^{-1} with an average value of 91.77 μSv y^{-1} (Table 4). The world average values for indoor and outdoor AEDE are 450 and

70 μSv y^{-1}, respectively (Orgun et al. 2007). It should be noted that the average value of indoor AEDE is within the world average value whereas the outdoor AEDE exceeds the maximum limit by 1.31 times. This may be due the presence of high activity concentration of ^{232}Th and ^{40}K (Ramasamy et al. 2011). Sediment samples which have lower values of indoor and outdoor AEDE can be used as construction material in terms of radiation safety.

Radium equivalent activity index (Ra_{eq})

The natural radioactivity in building materials is not uniform and is usually determined from ^{226}Ra, ^{232}Th and ^{40}K contents. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq kg^{-1} to compare the specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity. It is a most commonly used hazard index and is calculated through the following relation (Beretka and Mathew 1985; Orgun et al. 2007):

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (5)$$

where, C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. While defining Ra_{eq} activity, it has been assumed that 370 Bq kg^{-1} ^{226}Ra or 259 Bq kg^{-1} ^{232}Th or 4,810 Bq kg^{-1} ^{40}K produces the same gamma dose rate (Krisiuk et al. 1971). Ra_{eq} is estimated for all the collected samples and summarized in Table 4. These values varied from 102.34 to 229.08 Bq kg^{-1} with an average value 157.09 Bq kg^{-1}. The estimated average values are lower than the maximum permissible value of 370 Bq kg^{-1} suggested for building materials from the point of radiation hazard (OECD 1979; Iqbal et al. 2000). Lower values of Ra_{eq} may be due to the fact that the heavy metal gets discharged by the constant flow of water in the river (Ramasamy et al. 2011). It should be noted that if Ra_{eq} <370 Bq kg^{-1}, then the external dose rate will be below 1.5 mGy $year^{-1}$ (Krisiuk et al. 1971).

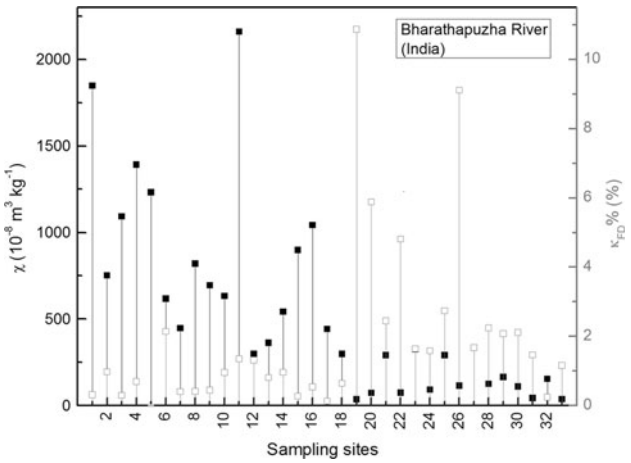

Fig. 3 Magnetic susceptibility measurements of sites along the Bharathapuzha river

Magnetic parameters

Results of magnetic concentration (χ), and grain size (κ_{FD}%) dependent parameters are displayed in Table 1 and Fig. 3. The concentration-related parameters display wide variations in the area, ranging from 35.4 to 2,160.5 × 10^{-8} m^3 kg^{-1}, values of χ are higher at sites 1–16 as can be appreciated in Fig. 3. Specific susceptibility is a magnetic concentration-dependent parameter, and its values vary according to the contribution of materials, i.e.: diamagnetic (∼−6 × 10^{-9} m^3 kg^{-1}), paramagnetic (∼10^{-6} m^3 kg^{-1}), antiferromagnetic (∼6–7 × 10^{-7} m^3 kg^{-1}), and ferrimagnetic materials (∼0.5–5.6 × 10^{-3} m^3 kg^{-1}, in detail in Maher et al. 1999). The magnetic concentration is dominated by ferromagnetic minerals, among them, magnetite-like and hematite minerals are expected.

Table 5 Magnetic data reported on river sediments from South India

Study area	Magnetic parameter	Minimum	Maximum	Mean	Standard deviation	Reference
Palaru river	χ (10^{-8} m^3 kg^{-1})	9.1	279.7	46.6	59.8	Chaparro et al. (2008)
	κ_{FD}% (%)	−0.6	4.4	1.1	1.1	
Cauvery River	χ (10^{-8} m^3 kg^{-1})	29.5	988.6	194.6	232.1	Chaparro et al. (2008)
	κ_{FD}% (%)	−0.5	2.7	0.7	0.8	
Vellar river	χ (10^{-8} m^3 kg^{-1})	14.3	518.0	141.0	143.8	Chaparro et al. (2011)
	κ_{FD}% (%)	−1.8	1.9	0.4	1.0	
Ponnaiyar river	χ (10^{-8} m^3 kg^{-1})	11.0	151.1	54.0	54.2	Chaparro et al. (2013)
	κ_{FD}% (%)	−2.6	5.7	0.2	2.3	
Bharathapuzha river	χ (10^{-8} m^3 kg^{-1})	35.4	2,160.6	540.6	530.1	Present study
	κ_{FD}% (%)	−0.1	10.9	1.9	2.5	

Table 6 Statistical results from the fuzzy *c*-means cluster analysis and members of each cluster

Group	χ	^{226}Ra	^{232}Th	^{40}K
Group 1: sites 1, 2, 3, 4, 5, 6, 8, 9, 10, 11, 15, 16 ($n = 12$)				
Mean	1,098.8	35.3	56.0	653.5
CV %	44.6	23.1	32.7	33.2
Min	617.0	21.2	35.6	271.9
Max	2,160.6	52.8	84.6	899.7
Group 2: sites 7, 12, 13, 14, 17, 18, 21, 23, 25, 27 ($n = 10$)				
Mean	363.3	42.5	47.1	362.4
CV %	23.6	26.3	39.5	25.8
Min	290.7	27.3	33.5	232.2
Max	542.2	66.0	93.1	518.7
Group 3: sites 19, 20, 22, 24, 26, 28, 29, 30, 31, 32, 33 ($n = 11$)				
Mean	92.8	48.4	60.7	390.9
CV %	48.4	20.1	34.8	16.2
Min	35.4	32.8	33.6	308.1
Max	163.8	63.7	93.1	475.5

Values correspond to the centroids of each variable

CV coefficient of variation

Although the presence of this kind of minerals will be confirmed by additional studies that will be published elsewhere, they were reported in rivers of the area of influence (e.g., Chaparro et. al. 2013; Ramasamy et al. 2011).

Assuming the dominance of magnetite-like mineral, the concentration can be estimated from the Thompson plot (Thompson and Oldfield 1986), giving values between 0.01 and 1 %. The Bharathapuzha river showed mean values of χ (540.6×10^{-8} m^3 kg^{-1}) higher than other rivers of southern India (Chaparro et al. 2011, 2013). As can be noted in Table 5, the mean values are about 3–11 times higher than Palaru, Ponnaiyar, Vellar and Cauvery rivers. This difference can come from minerals that have originated by disintegration of parent rocks during the pedogenesis, by lithogenic process and by anthropogenic activities. However, the magnetic data of this study are comparable to data from rivers Cauvery and Vellar, which were reported with higher pollutant loads.

The κ_{FD}% values can help to discriminate ultrafine (<0.03 μm) superparamagnetic minerals (SP) from single and multi-domain (SD and MD) grains. There seems to be a maximum observational limit on κ_{FD}% of 15 % (Dearing et al. 1996). In Table 1 and Fig. 3, it is possible to note higher values of κ_{FD}% (between 0 and 10.9 %), which are indicative of the presence and dominance (the high values) of SP grains (Bartington Instruments Ltd 1994; Dearing et al. 1996). In addition, the mean values are higher than other rivers of South India (Table 5).

Multivariate statistical analysis using radioactivity and magnetic variables

The statistical studies were accomplished to get two aims: (a) the relationship between variables; (b) a description of the activity/magnetic concentration along the river.

The Pearson's coefficients showed statistically significant correlation values between most concentration variables. The positive correlation values are observed for χ-^{40}K ($R = 0.526$, $p < 0.01$) and ^{232}Th-^{226}Ra ($R = 0.522$, $p < 0.01$), and negative values for χ-^{226}Ra ($R = -0.406$, $p < 0.05$) and ^{40}K-^{226}Ra ($R = -0.442$, $p = 0.01$).

The FCC method allows us to get a fuzzy clustering using relevant variable for this work. The classification

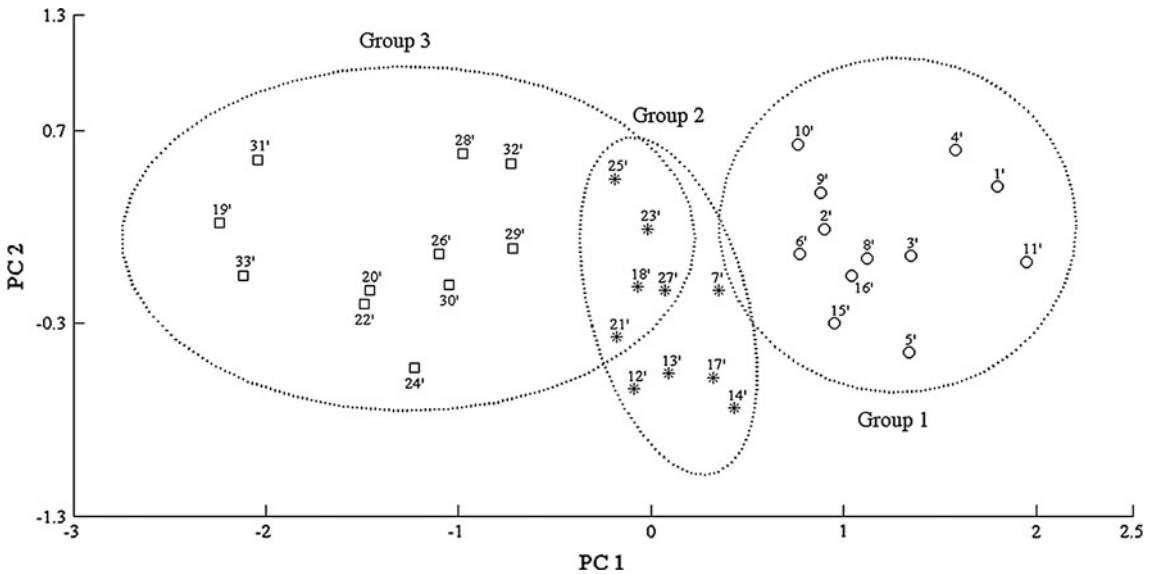

Fig. 4 FCC classification, the obtained groups are represented in the coordinate plane (PC 1 and PC 2)

obtained by this method allows us to describe the magnetic and the radioactivity relevance of the different group of samples. As a first step, the variables were transformed using a logarithmic function (Ln) and then several numbers of groups, from $N = 3$ to $N = 6$, were studied for this dataset. The optimum number of clusters is three ($N = 3$), which was chosen according to the high value (0.5066) of the normalized Dunn's index (Dunn 1973). Each group is characterized by three centroids that are summarized in Table 6. In addition, each sample is classified from its membership value (from 0 to 1) of each group. It is worth mentioning that each sample is classified according to the maximum value of membership, e.g.: sample 2 has membership values of 0.86, 0.12 and 0.02 for Group 1, 2 and 3, respectively, hence this sample is classified as Group 1.

These groups can be observed in the graphic representation of PC1 and PC2 (Fig. 4). As can be distinguished, the group 1 (comprising the river head, sites 1–6, 8–11, 15–16) has samples with the highest magnetic susceptibility and ^{40}K concentration and the lowest values of ^{226}Ra concentration; and the contrary is observed for group 3 (comprising the river mouth, sites 19–20, 22, 24, 26, 28–33).

Conclusion

The natural radioactivity levels of ^{226}Ra, ^{232}Th and ^{40}K have been measured in bottom sediments of Bharathapuzha river using gamma-ray spectroscopy. The average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were 41.86, 54.86 and 477.75 Bq kg^{-1}, respectively. The mean concentrations of ^{226}Ra, ^{232}Th and ^{40}K are higher than the worldwide average value. The air adsorbed dose rates (D) have been calculated because of humans residing along the bank of the river and their average value is 74.83 Bq kg^{-1} which is 1.3 times greater than the recommended value (UNSCEAR 2000). This may sometimes cause additional radiological health risks to the humans residing in this area, and in future radiological parameters must be carefully monitored. The annual effective dose rates (indoor and outdoor) and radium equivalent activity are calculated to evaluate the radiological hazard of sediments since it is widely used as construction material in this region. The overall mean value of outdoor AEDE is 1.31 times greater than the recommended value whereas indoor AEDE and Ra_{eq} are lower than the recommended level. Bharathapuzha river sediments which have lower values of activity concentration and radiological hazard parameters can be safely used for construction purposes. The concentration related parameters display wide variations in the area. The Bharathapuzha river showed mean values of χ (540.6×10^{-8} m^3 kg^{-1}) higher (up to 11 times higher) than others rivers of southern India (e.g., Palaru, Ponnaiyar, Vellar and Cauvery rivers). The magnetic concentration is dominated by ferromagnetic minerals. Magnetite concentration varied between 0.01 and 1 %, showing high contents of this magnetic mineral and a magnetic enhancement in various sites, which was more evident for sites 1–11 and 15–16. Such magnetic enhancement may arise from the presence of pollution. The FCC method allows us to get a fuzzy clustering using relevant variable for this work. The classification obtained (three groups) by this method allows us to describe the magnetic and the radioactivity relevance of the different groups, e.g., the group 1 has sampled with the highest magnetic susceptibility and ^{40}K concentration and the group 3 has samples with the lowest magnetic susceptibility and the highest ^{226}Ra, ^{232}Th concentration.

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