

MEASUREMENT OF RADIOACTIVITY IN AN ELEVATED RADIATION BACKGROUND AREA OF WESTERN GHATS

by

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As part of monitoring the exposure of the general public to natural radioactivity, the activity concentration of naturally occurring radionuclides in soil samples in an elevated radiation background area of Western Ghats was determined using gamma-ray spectrometry. Average values of the activity concentration of radionuclides, outdoor terrestrial gamma dose rate, annual effective dose equivalent and radiation hazard indices from soil activity were estimated. The activity concentrations of ²³²Th and average outdoor terrestrial gamma dose rate were found to be higher than the world average, possibly affecting the Western Ghats environment in general. Therefore, radiological risks to the general population from ionizing radiation from the naturally occurring radionuclides in the soil are considered to be significant. However, other radiological hazard indices were found to be within permissible limits.

Key words: naturally occurring radionuclide, Western Ghats, monazite, radiological hazard

INTRODUCTION

Natural radionuclides in soil are responsible for the background radiation exposure of the population. Exposure to gamma radiation is mostly regarded as undesirable at every level, although no harmful effects are known to follow very low levels of exposure. Recently, considerable attention has been given to low-level exposure arising from naturally occurring radionuclides, particularly ²³⁸U, ²³²Th, their decay products, and ⁴⁰K. Natural radioactive concentration depends mainly on geological and geographical conditions and appears at different levels in soils from different geological regions [1] *i. e.*, thorium and uranium may be redistributed during igneous, sedimentary and metamorphic cycles of geological evolution which, under favorable geological processes, might result in small concentrations of deposits. The study of radioactive components in soils is a major link in understanding the behaviour of radioactivity in the ecosystem, because these materials emit radiation by the disintegration of natural radionuclides and contribute to the total absorbed dose via ingestion, inhalation and external irradiation [2]. Also, soil acts as a source of continuous radiation exposure of humans and as a medium of migration for the transfer of radionuclides to biological systems, causing radiological contamination of the

environment. In addition to natural sources, soil radioactivity is also affected by man-made activities. The sources of radioactivity in cultivated soils are mainly due to the extensive use of agricultural fertilizers, rich in phosphates. The concentration of uranium and partial thorium are on the increase in the environment due to these fertilizers. Usually fertilizers are considered as a technologically enhanced source of natural radiation [3]. Hence, soil radioactivity is usually important for the purpose of establishing baseline data for future assessment of radiation impact, radiation protection, and studies.

MATERIALS AND METHODS

Study area

Available information indicates that the deposits of monazite in the coastal areas of Kerala and Tamil Nadu are formed due to the weathering of rocks in Western Ghats. Monazite sands consist of phosphate minerals of elements such as cerium which occur as small brown crystals in the Kerala sands (these monazite sands are mined for both cerium and radioactive thorium oxide). They originate in the granites and gneisses of the Western Ghats and are transported to the coast by more than 47 streams that indent the Kerala coastline [4], as shown in fig. 1.

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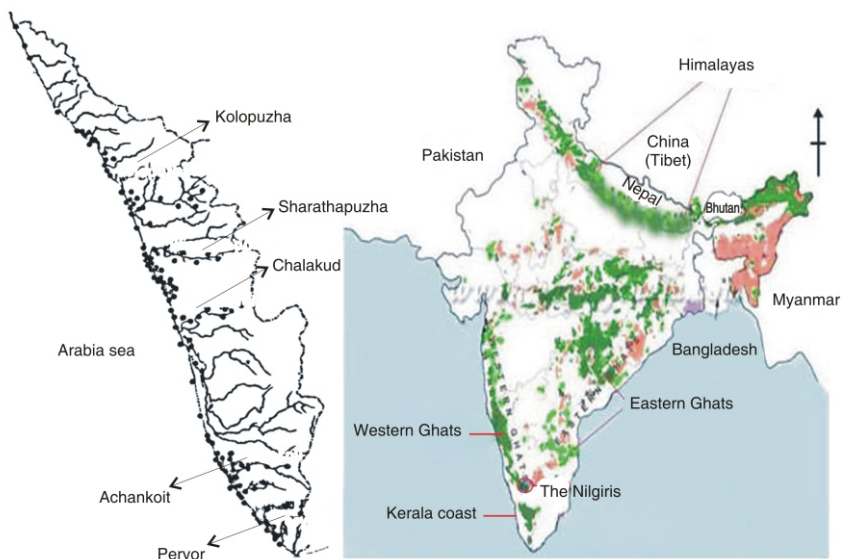


Figure 1. Distribution of Monazite sand along the Kerala coast

The soils analyzed were collected at elevations of between 2000 m and 2400 m: the Nilgiri Highlands, Tamil Nadu, South India, situated between 11°00' and 11°30' N and between 76°00' and 77°30' E. The Nilgiri massif is located at the junction between the Eastern and Western Ghats and is bounded by abrupt slopes. The study area is shown in fig. 2. Vegetation above 2000 m in the highlands is a mosaic of high elevation evergreen forests, locally called “shola”, and grasslands with different compositions of flora, including C4 grasses [5, 6].

Soil sampling and preparation

The study area was divided into a 4 km grid, with soil samples collected from 25 sampling points in the natural, uncultivated, and grass-covered level areas within the grid, conforming to International Atomic Energy Agency recommendations – IAEA 1989 [7]. The 25 sampling points followed a zig-zag pattern. Five 20-cm-deep samples were collected at equal distances along a 1 m circle around the center of each sampling point. This sampling method was used to improve the representativeness of the samples. The posi-



Figure 2. Study area: Nilgiris district

tion and elevation of each sampling point was determined using a global positioning system.

The soil samples were transported to the laboratory where plant roots and other unwanted materials were removed. The samples were then dried in an oven at 105 °C for 12-24 h, ground, and passed through a 2 mm sieve. About 400 g of dry sample was weighed into a plastic container which was capped and sealed. The container was sealed to ensure that none of the daughter products of uranium and thorium that were produced, particularly radon and thoron, could escape. The prepared samples were stored for 1 month before counting, to ensure that the equilibrium between radium and its short-lived daughters had been established. Detailed gamma-ray spectrometry analysis was performed on the soil samples.

Activity determination

The samples were analyzed using a NaI(Tl) spectrometer coupled with a TNIPCAII Ortec model 8K multi-channel analyzer. The ^{232}Th -series, ^{238}U -series, and ^{40}K activities were estimated, as were the amounts of the said radionuclides that would enter the air from the soil. A 3 inch \times 3 inch NaI(Tl) detector was used, with adequate lead shielding, reducing the background by a factor of 95. The energies of interest were found by employing an International Atomic Energy Agency standard source and the appropriate geometry. The system was calibrated in terms of both energy response and counting efficiency. The density of the sample used for the calibration was 1.3 g/cm³ which was the same as the mean density of the soil samples analyzed (1.24 g/cm³). The detector was very well shielded, and the counting time was 20,000 s for each sample. The minimum detectable concentrations, defined as 3σ (standard deviation), were 7 Bq/kg for the ^{232}Th -series, 8.4 Bq/kg for the ^{238}U -series, and 13.2 Bq/kg for ^{40}K .

The concentrations of radionuclides of interest were determined using the counting spectrum for each sample. Peaks corresponding to 1.46 MeV (^{40}K), 1.76 MeV (^{214}Bi), and 2.614 MeV (^{208}Tl) were considered when evaluating the ^{40}K , ^{238}U -series, and ^{232}Th -series activities, respectively. Crystal detector resolution was 6% for ^{40}K , 4.4% for the ^{232}Th -series, and 5.5% for the ^{238}U -series. Gamma-ray spectrum activities for each soil sample were analyzed using dedicated software and references were chosen so as to achieve sufficient discrimination.

In addition to gamma-ray spectrometric analysis, a low-level survey environmental radiation dosimeter-ERD (type ER 705; Nucleonic System PVT Ltd., Hyderabad, India) was used to measure ambient radiation levels in the study areas directly associated with radionuclide activity concentrations in the samples and cosmic rays. The dosimeter had a halo-

gen-quenched Geiger-Müller detector (Ind. Inc., USA) powered by a rechargeable battery and was designed to read the exposure rate at two levels, 0.1 R/h and 1 R/h. Before use, the dosimeter was calibrated using a standard source. Outdoor terrestrial gamma dose rates were measured at 1 m above the ground by a portable digital ERD at all of the sampling sites. A total of five readings were recorded at each spot and the average taken.

Estimation of absorbed dose rate

Radiological risks to the humans and other entities from terrestrial gamma rays and associated with absorbed dose from the natural radionuclides in the soil are considered to be significant. The assessment of the gamma radiation hazard to humans associated with building materials can be done by calculating the different radiation hazard indices [8]. The absorbed dose rate D [nGyh⁻¹] due to terrestrial gamma rays at one meter above the ground level can be estimated by the concentrations of ^{238}U , ^{232}Th , and ^{40}K by applying the factors 0.462, 0.604, and 0.0417 for uranium, thorium, and potassium, respectively [1].

$$D = (0.462C_U + 0.604C_{Th} + 0.0417C_K) \quad (1)$$

where D [nGyh⁻¹] is the absorbed dose rate, C_U [Bqkg⁻¹], C_{Th} [Bqkg⁻¹], and C_K [Bqkg⁻¹] are the activity concentrations of ^{238}U , ^{232}Th , and ^{40}K in soil samples, respectively.

Annual effective dose rates

To estimate the annual effective dose rates, the conversion coefficient from the absorbed dose in air to the effective dose 0.7 Sv/Gy and the indoor occupancy factor of 0.2 proposed by UNSCEAR, 2008, were calculated. Thus, annual effective doses were determined as follows [8]

$$\begin{aligned} \text{Annual effective dose (outdoor)} [\mu\text{Sv}] = \\ = D [\text{nGyh}^{-1}] \times 8760 [\text{h}] \times 0.7 [\text{SvGy}^{-1}] \times 10^{-3} \quad (2) \end{aligned}$$

$$\begin{aligned} \text{Annual effective dose (indoor)} [\mu\text{Sv}] = \\ = D [\text{nGyh}^{-1}] \times 8760 [\text{h}] \times 0.2 [\text{SvGy}^{-1}] \times 10^{-3} \quad (3) \end{aligned}$$

Calculation of radium equivalent

The radium equivalent index, Ra_{eq} is generally introduced as the weighed sum of ^{238}U , ^{232}Th , and ^{40}K activities based on the assumption that 10 Bq/kg of ^{238}U , 7 Bq/kg of ^{232}Th , and 130 Bq/kg of ^{40}K will produce the same dose rates of gamma rays. Values of Ra_{eq} were calculated using eq. [9]

$$Ra_{eq} [Bqkg^{-1}] (C_U \ 1.43C_{Th} \ 0.077C_K) \quad (4)$$

where C_U , C_{Th} , and C_K are defined as in eq. 1.

External hazard index

The consideration of the external radiation exposure is usually associated with gamma radiation emitted by radionuclides of concern. The value of the Hazard index should be below one in order to assure the safe use of building materials corresponding to the upper limit of Ra_{eq} (370 Bq/kg) and so as to keep the radiation hazard insignificant [8]

$$H_{ex} \quad \frac{C_U}{370} \quad \frac{C_{Th}}{259} \quad \frac{C_K}{4810} \quad 1 \quad (5)$$

$$H_{in} \quad \frac{C_U}{185} \quad \frac{C_{Th}}{259} \quad \frac{C_K}{4810} \quad 1 \quad (6)$$

where, C_U , C_{Th} , and C_K are defined as in eq. 1

Radioactivity level index

Gamma radiation hazardous levels associated with natural radionuclides in the building material samples were assessed by means of the radioactivity level index, I_γ . In accordance with the European Commission guidelines, the representative level of I_γ values was estimated according to the equation [10]

$$I_\gamma \quad \frac{1}{150Bq/kg} C_U \quad \frac{1}{100Bq/kg} C_{Th} \quad \frac{1}{1500Bq/kg} C_K \quad (7)$$

where, C_U , C_{Th} , and C_K are defined as in eq. 1

Excess Life time Cancer Risk (ELCR): ELCR is calculated using the equation [11]

$$ELC = AEDE \ DL \ RF \quad (8)$$

where, $AEDE$, DL , and RF are the total annual effective dose equivalent [Sv], duration of life (70 years) and risk factor [Sv^{-1}] (fatal cancer risk per sievert) for stochastic effects. ICRP 60 uses values of 0.05 for the public [11].

RESULTS AND DISCUSSION

Activity concentrations of natural radionuclides (^{238}U , ^{232}Th , and ^{40}K) for all samples were determined, as shown in tab. 1. Mean activity concentrations for ^{238}U , ^{232}Th , and ^{40}K ranges were as follows: 12.36-85.81 Bq/kg with an average of 36.31

17.3 Bq/kg, 30.28-204.11 Bq/kg, with an average of 107.77 50.4 Bq/kg and 83.12-411.56 Bq/kg with an average of 231.93 79.4 Bq/kg, respectively. At all sampling sites, the mean activity concentration is of the order $^{232}Th < ^{238}U < ^{40}K$. At a few of the sites, the activity concentration of ^{238}U and ^{40}K is high, which may be due to the solubility and mobility of $U(VI)O_2^{2+}$, or the presence of loamy and clay sediment and extensive use of fertilizers for agricultural purposes rich in phosphates [12]. The concentrations of ^{238}U for all measured samples were within the world average values, but the average value of ^{40}K in Western Ghats was observed to be lower than the world average (world average value of ^{238}U and ^{40}K is 32 Bq/kg and 412 Bq/kg, respectively). However, according to UNSCEAR (2008), the global ^{232}Th activity concentration range is 7-50 Bq/kg (mean 45 Bq/kg). The measured ^{232}Th activity concentration in our study area was 2.5 times higher than the global mean, indicating the presence of monazite at that sampling site.

The terrestrial gamma-ray dose rate in the study area ranged from 20.67 to 992.67 nGy/h, while the mean was 133.33 183 nGy/h, but the calculated absorbed dose rates ranged from 39.11 nGy/h to 153.40 nGy/h, with an average of 91.54 34 nGy/h that exceeds the world average value of 56 nGy/h [1]. Thus, the ^{232}Th -series contributed with a 71% of the total gamma-ray dose to the environment, whereas the ^{238}U -series and ^{40}K contributed with 19% and 10% of the total gamma-ray dose. World studies indicate an average outdoor terrestrial gamma dose rate of 60 nGy/h, ranging from 10 to 200 nGy/h [12].

The present study shows that the average terrestrial gamma dose rate is 133.33 nGy/h which is higher than the world average. The gamma radiation level is directly associated with radionuclide activity concentrations in the samples and with cosmic rays [12]. The observed gamma-ray dose rate contributed by more than 50% to the dose calculated from soil activities and the difference may be attributed to the cosmic radiation contribution to the total dose of the Western Ghats environment, situated 2400 m above sea level.

The calculated indoor and outdoor AEDE values are quoted in tab. 1. The average, minimum and maximum values for outdoor and indoor were found to be 112.3 42 μ Sv Sv, and Sv, respectively, and 167 μ Sv Sv, and Sv, respectively.

The values of indoor and outdoor AEDE presented are higher than the world average values (70 Sv per year for Indoor, 410 Sv per year for Outdoor), which can be attributed to the higher activity concentration of ^{232}Th . The outcomes of external and internal radiation hazard indices are shown in tab. 2; in soil, the average values of external and internal hazard indices were found to be 0.56 0.2 and 0.66 0.24, respectively.

Table 1. Activity concentration of radionuclides and corresponding dose rates for different soil samples

Locations	Activity concentration [Bqkg ⁻¹]						Absorbed dose rate [nGyh ⁻¹]	Observed dose rate (ERD) [nGyh ⁻¹]	AEDE [μSv]							
	²³⁸ U		²³² Th		⁴⁰ K				Outdoor	Indoor	Total					
L-1	36.34	3.1	57.32	5.5	303.56	22.8	64.07	88.49	78.57	314.30	392.87					
L-2	18.26	3.5	75.91	6.4	276.37	20.9	65.81	66.52	80.71	322.84	403.55					
L-3	22.34	4.2	93.28	7.5	129.89	16.7	72.08	69.93	88.40	353.59	441.99					
L-4	25.45	2.9	72.45	6.2	285.45	18.3	67.42	111.30	82.69	330.74	413.43					
L-5	85.81	6.2	126.08	6.3	299.45	21.8	128.28	108.13	157.33	629.31	786.64					
L-6	55.65	4.4	204.11	9.3	411.56	21.6	166.15	132.98	203.77	815.09	1018.86					
L-7	35.47	2.8	127.35	6.5	229.34	19.1	102.87	176.58	126.16	504.64	630.80					
L-8	45.56	3.7	169.87	7	138.12	12.3	129.41	109.31	158.71	634.83	793.54					
L-9	26.48	2.6	88.28	6.9	319.54	20.5	78.88	137.85	96.74	386.95	483.69					
L-10	32.36	2.6	56.76	5.7	237.56	19.5	59.14	83.49	72.53	290.12	362.64					
L-11	46.83	3.9	73.26	5.2	350.67	19.8	80.51	61.61	98.73	394.94	493.67					
L-12	34.26	2.6	134.14	9.1	246.67	21.7	107.13	71.10	131.39	525.56	656.95					
L-13	50.02	5.6	113.39	8.4	225.89	18.3	101.02	114.04	123.89	495.55	619.43					
L-14	35.34	4.2	30.28	5.1	107.65	13.6	39.11	106.14	47.96	191.83	239.79					
L-15	45.17	4.3	35.88	5.2	187.45	16.4	50.36	39.76	61.76	247.03	308.79					
L-16	30.31	3.8	220.76	11.1	145.21	18.4	153.40	51.10	188.13	752.51	940.63					
L-17	12.65	2.1	139.56	8.1	83.12	11.4	93.60	165.33	114.80	459.19	573.98					
L-18	36.78	4.2	117.56	6.8	339.56	23.5	102.16	101.36	125.29	501.15	626.43					
L-19	25.03	3.2	50.89	5.6	254.67	18.3	52.92	20.67	64.90	259.61	324.51					
L-20	37.34	2.8	60.58	7.8	195.67	14.5	62.00	96.43	76.04	304.15	380.19					
L-21	42.45	3.7	164.67	9.3	257.12	19.6	129.79	64.46	159.18	636.72	795.90					
L-22	73.32	4.6	163.40	9.1	220.78	18.9	141.77	138.19	173.87	695.49	869.36					
L-23	12.36	2.3	97.23	8	164.88	13.3	71.31	148.97	87.46	349.83	437.29					
L-24	20.67	3.6	119.45	9.3	110.78	15.4	86.32	76.73	105.86	423.44	529.30					
L-25	21.51	4.1	101.78	9.5	277.36	17.5	82.98	992.67	101.77	407.06	508.83					
Mean σ	36.3	17.3	107.8	50.4	231.9	84.3	91.54	34	133.3	183	112.3	42	449.1	167	561.8	208

σ = standard deviation

Radium-equivalent activities (Ra_{eq}) and the representative level index values I , were calculated using the formula given in eqs. 3 and 5. Based on the annual external dose of 1.5 mGy, activity limits in terms of (Ra_{eq}) and I are 370 Bq/kg and 1, respectively, for the safe use of soil products. It has been observed that the mean radium equivalent activity and the representative level index values were 208.28 \pm 79.4 Bq/kg and 1.47 \pm 0.55, respectively, as shown in tab. 2. The Ra_{eq} values were much higher in the terrestrial environment, mainly because of the use of fertilizers rich in phosphates at the agricultural sites [3]. Phosphate rocks contain significant concentrations of U, Th, Ra, and their decay products [13]. Hazard indices of all site samples were found to be less than Unity (the permissible level) [14].

Excess life time cancer risk (ELCR) is calculated using eq. 5, as shown in tab. 2. The range of ELCR is 0.17×10^{-3} to 0.71×10^{-3} , with an average of $0.39 \pm 0.15 \times 10^{-3}$. The average ELCR for all samples is marginally higher than the world average (0.29×10^{-3}). It may be noted that ELCR for Western Ghats is far lower than the ICRP [15] prescribed value of 0.05. According to these results, the risk of cancer is found to be negligible.

Comparison of observed activity concentrations with those found in similar studies

The ²³⁸U, ²³²Th, and ⁴⁰K activity concentrations, Ra_{eq} , and I for the terrestrial samples, were compared with the values established in similar studies in other countries and the results summarized in tab. 3.

As can be seen from tab. 3, the radioactivity found in terrestrial soils varies from country to country. It is important to note that the values shown are not representative of the countries mentioned; they pertain to the geological regions in which these samples were collected.

CONCLUSIONS

Mean activity concentrations of soil samples collected from the terrestrial environment of Western Ghats were found to be within the world and Indian average values. However, activity concentration values of ²³²Th were found to belong to the higher end of the world range. The average outdoor terrestrial gamma dose rate was found to be higher than the world

Table 2. Radiological parameters for the soil samples

Locations	Radium equivalent Ra _{eq} ⁻¹ [Bqkg ⁻¹]	Hazard indices		Activity utilization index	ELCR 10 ⁻³
		H _{ex}	H _{in}	I	
L-1	141.68	0.38	0.48	1.02	0.28
L-2	148.09	0.40	0.45	1.07	0.28
L-3	165.73	0.45	0.51	1.17	0.31
L-4	151.03	0.41	0.48	1.08	0.29
L-5	289.16	0.78	1.01	2.03	0.55
L-6	379.22	1.02	1.17	2.69	0.71
L-7	235.24	0.64	0.73	1.66	0.44
L-8	299.11	0.81	0.93	2.09	0.56
L-9	177.32	0.48	0.55	1.27	0.34
L-10	131.82	0.36	0.44	0.94	0.25
L-11	178.59	0.48	0.61	1.28	0.35
L-12	245.07	0.66	0.75	1.73	0.46
L-13	229.56	0.62	0.76	1.62	0.43
L-14	86.93	0.23	0.33	0.61	0.17
L-15	110.91	0.30	0.42	0.78	0.22
L-16	357.18	0.96	1.05	2.51	0.66
L-17	218.62	0.59	0.62	1.54	0.40
L-18	231.04	0.62	0.72	1.65	0.44
L-19	117.41	0.32	0.38	0.85	0.23
L-20	139.04	0.38	0.48	0.99	0.27
L-21	297.73	0.80	0.92	2.10	0.56
L-22	323.98	0.87	1.07	2.27	0.61
L-23	164.09	0.44	0.48	1.16	0.31
L-24	200.01	0.54	0.60	1.41	0.37
L-25	188.41	0.51	0.57	1.35	0.36
Mean σ	208.3 79.4	0.56 0.2	0.66 0.24	1.47 0.55	0.39 0.15

Table 3. Comparison of activity concentrations with those found in similar studies

Country	Activity [Bqkg ⁻¹]			Ra _{eq} ⁻¹ [Bqkg ⁻¹]	I	Reference
	²³⁸ U	²³² Th	⁴⁰ K			
Western Ghats	26.26	53.61	231.93	118.6	1.47	This study
India	64	93	124	206.5	1.4	Singh <i>et al.</i> , [16]
Algeria	47.01	43	329	132	0.95	Wassila <i>et al.</i> , [17]
Brazil	1.69	5.32	34.15	12	0.1	Becegato <i>et al.</i> , [18]
Egypt	13.7	12.3	1233	126.2	1.04	Ahmed <i>et al.</i> , [19]
Pakistan	27.39	31.16	602.77	142.71	1.02	Akhtar <i>et al.</i> , [20]

average and, thus, the Western Ghats region should be classified as an under-elevated background radiation region of the world. It should also be noted that the calculated activity utilization index was also found to exceed the recommended safe limit values. This implies that the inhabitants of the study area are subjected to a radiation exposure significantly higher than the corresponding exposure levels reported in other areas world wide. In spite of all this, other calculated radiological hazard indices were within the acceptable limits (Safety Limit). I should be pointed out that the results of our measurements will also serve as an excellent baseline data and as a reference level for soil samples from Western Ghats.

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AUTHOR CONTRIBUTIONS

Theoretical analysis and experiments were carried out by P. K. Manigandan. The manuscript was written by B. Chandar Shekar.

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МЕРЕЊЕ РАДИОАКТИВНОСТИ У ЗАПАДНОМ ГАТУ –
ОБЛАСТИ СА ПОВИШЕНИМ НИВОМ ПРИРОДНЕ РАДИОАКТИВНОСТИ

Као део мониторинга излагања популације природној радиоактивности, употребом гама спектрометрије одређена је концентрација активности природних радионуклида у узорцима земљишта области Западног Гата која има повишен ниво природног зрачења. Просечна вредност концентрације активности ових радионуклида, спољашња јачина дозе гама зрачења, годишња ефективна доза и индикатори радијационог хазарда процењени су испитивањем узорака земљишта. Концентрација активности ²³²Th и просечна спољашња јачина дозе гама зрачења биле су више од просека у свету, са могућношћу утицаја на животну средину области Западног Гата. Сматра се, стога да је значајан радиолошки ризик за популацију од јонизујућег зрачења од природних радионуклида у земљишту. Ипак, други индикатори радиолошког хазарда били су у дозвољеним границама.

Кључне речи: природни радионуклиди, Западни Гат, монациј, радиолошки хазард