Retraction for *Environmental Science: Processes & Impacts*:

Radioactivity Concentration and Dose Assessment for soil samples of Western Ghats

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I, the Editor, Harpal Minhas, hereby wholly retract this article as it contains significant overlap including the reuse of data with the article "Distribution of radionuclide in the forest soils (Western Ghats-India)", P. K. Manigandan, S. Selvasekarapandian and N. Manikandan, *Iranian Journal of Radiation Research*, 2007, **5**, 17-22 without sufficient attribution to this earlier work being given.

The principal author declined to sign the retraction notice and did not agree to the retraction.

Signed: Harpal Minhas, Editor, *Environmental Science: Processes & Impacts*. Retraction published 26 February 2014

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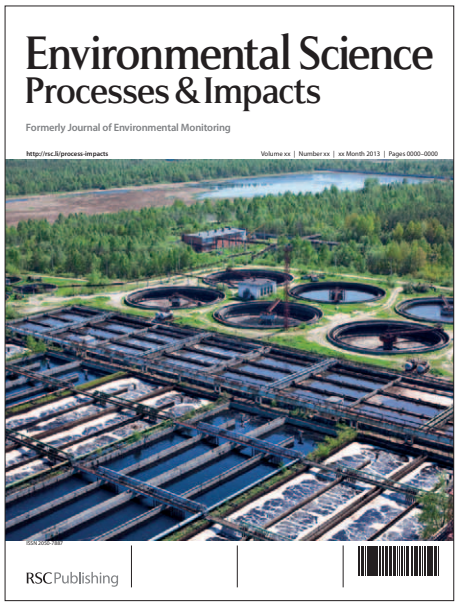

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Radioactivity Concentration and Dose Assessment for soil samples of Western Ghats

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Abstract

Surveys of naturally occurring radionuclides in soils from the tropical forest of western Ghats, India were conducted. The activity concentrations of these radionuclides were varied marginally in the soil. For all soil samples, the absorbed gamma dose rate and the corresponding effective dose rate, due to external exposure in outdoor were evaluated. Although the activity concentration of ^{232}Th and average outdoor gamma dose rates were found to be higher than the world average causing an alarming situation in the Western Ghats environment, the radiological hazard indices were found to be within the ICRP (International Commission on Radiological Protection) recommended limits. Also, the levels of the studied natural radionuclides in the forest soils were within the range specified by the United Nations scientific Committee on the effect of Atomic Radiation report for regular soils except ^{232}Th.

Key words: Primordial radionuclides, Western Ghats, igneous rock, radiological hazard

Introduction

We have previously reported that activity concentration ofthorium was high in the region of Western Ghats especially around the Nilgiri hill station due to the presence of monazite sand (Manigandan. 2009; Selvasekarapandian. 2000; Iyengar et al. 1990) [1-3]. The external radiation levels from monazite sands in India are higher than that of radiation level reported from Brazil. High content of thorium and traces of uranium are also reported from these areas. These thorium and uranium may be redistributed during igneous, sedimentary and metamorphic cycles of geological evolution, which might have resulted in small concentrations of deposits under favorable geological processes. Available information indicates that the monazite deposits on the coastal areas of Kerala and Tamil Nadu are formed due to the weathering of rocks in Western Ghats. Monazite sands comprise 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). The sands 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 (Valiathan et al 1994)[4] and it is shown in the Figure 1.

The study of the radioactive components in soil is a fundamental link in understanding the

behavior of radioactivity in the ecosystem as these materials emits radiation by the disintegration of natural radionuclides and contributes tothe total absorbed dose via ingestion, inhalation andexternal irradiation. Forest soils are suitable for radionuclide investigations, because they not are usually disturbed by cultivation over long periods of time. The presence of radionuclides in the forest can affect the evolution of ecosystems, since the soil properties of these ecosystems produce dramatic differences in biotic concentrations of radionuclides from similar levels of deposition (Segovia et al. 2003)[5]. These are very important factors that might result in additional population exposure due to external irradiation or intake of radioactivity by the people. This might have economic consequences due to possible recreational or industrial use of the forest or its products (Gaso et al. 1998: Vaca et al. 2001) [6-7]. Therefore, thorough knowledge about the level of exposure to natural radiation from these natural gamma-emitting radionuclides is important to the authorities and policy makers for making the right decisions.

Materials and Methods

The soils presented here were sampled in the Nilgiri highlands (Tamil Nadu, South India) which are situated between 11°00'-11°30' N and 76°00'-77°30' E, at elevations ranging between 2000 and 2400 m. The Nilgiri massif is located at the junction of the Eastern and Western Ghats and is bound by abrupt slopes. The study area is shown in Fig.2. The vegetation of the highlands (above 2000 m) is a mosaic of high-elevation evergreen forests, locally named 'shola', and grasslands of different floristic compositions containing C4 grass species (Sukumar et al. 1995, Rajagopalan et al. 1997)[8-9].

The sampling sites were selected to randomly cover the forests with a 4 km grid. The soil samples were collected from 15 sampling points in natural, uncultivated, grass covered level areas in conformity with the IAEA recommendations [10]. Sampling was done in a zig – zag pattern comprising of 15 sampling point for a depth of 20cm. From each point, five representative samples were collected at equal distance along the 1m circumference around the point. This will improve the representative homogeneity of the samples from each sampling point. The position and elevation of each sampling point was determined using a global positioning system (GPS). Soil samples were taken to the lab and cleaned from plant roots and other foreign materials. Samples were dried in an oven at 105 °C for 12–24 hours and were grounded and sieved using 2 mm sieve to get homogeneous samples. About 400g of dry sample was weighed into a plastic container, capped, sealed and then labeled. This sealing was to ensure that all the daughter products of uranium and thorium and in particular radon and thoron daughters that would be formed thereafter would not escape. These prepared samples were stored for one month before counting to ensure equilibrium between radium and its short-lived daughters. All the soil samples were subjected to detailed gamma ray spectrometry analysis.

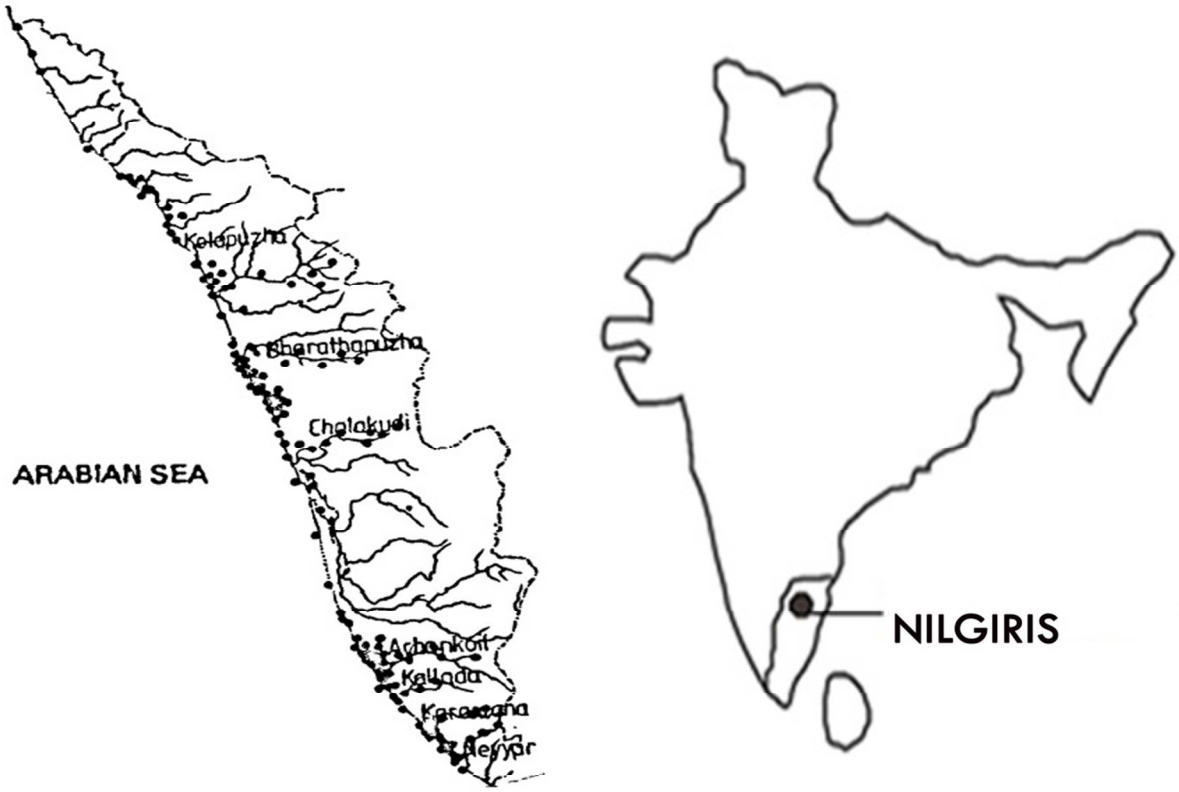

Figure 1 Distribution of Monazite sand along the Kerala Coast

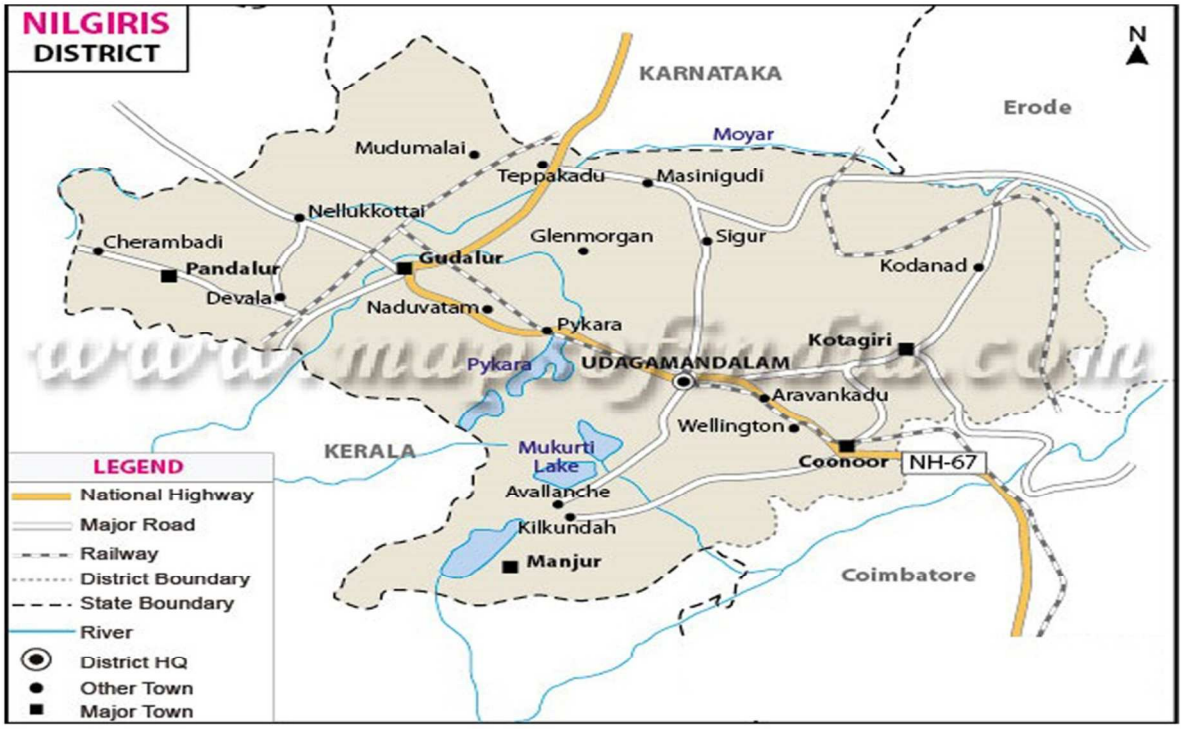

Figure 2 Study area: Nigiri district

To estimate the activity levels of ^{232}Th-series, ^{238}U-series and ^{40}K and to evaluate the absorbed dose rate in air from these radionuclides in soil, the samples were analyzed by NaI(Tl) spectrometer which was coupled with TNIPCAII Ortec model 8K multi-channel analyzer. A 3"×3" NaI(Tl) detector was employed with adequate lead shielding which reduced the background by a factor of 95. The efficiency of various energies was arrived at using IAEA standard source and the required geometry. The system was calibrated both in terms of energy response and also for counting efficiency. The density of the sample used for the calibration was 1.3gm/cm^3 which was the same as average of soil samples analyzed (1.24gm/cm^3) with a counting time of 20,000sec for each sample and with very good shielding to the detector, the minimum detectable concentration was 7Bqkg^{-1} for ^{232}Th-series, 8.4Bqkg^{-1} for ^{238}U- series and13.2Bqkg^{-1} for ^{40}K at 3σ confident levels.

The concentrations of various radionuclides of interest were determined using the counting spectrum of each sample. The peaks corresponding to 1.46MeV (^{40}K), 1.76MeV (^{214}Bi) and 2.614MeV(^{208}Tl) were considered in evaluating the activity levels of ^{40}K, ^{238}U-series and ^{232}Th, respectively. The resolution of the crystal detector was 6% for ^{40}K, 4.4% for ^{232}Th-series and 5.5% for the ^{238}U-series. The activity analysis of gamma spectrum obtained for each soil sample was performed with dedicated software and the choice of reference was made, so that they were sufficiently discriminated.

In addition to the gamma ray spectrometric analysis, Environmental Radiation Dosimeter was used to measure the ambient radiation level around forest in Western Ghats region. In the present study the ambient gamma radiation level survey was conducted using an environmental radiation dosimeter. ERD type ER 705, supplied by Nucleonic System PVT Ltd., Hyderbad, India, a low-level survey meter. It consist of a halogen quenched G.M. Detector (Ind.lnc.U.S.A) powered by a rechargeable battery. The Survey meter is designed to read exposure rate in two ranges of 0.1μR/h and 1μR/h. The survey meter is calibrated regularly using standard source, before starting survey work.

Results and Discussion

The activity concentration of radionuclides in soil from the forest ecosystem of Western Ghats region is shown in Table 1. The range activity of ^{238}U in soil is from 15.12 to 41.21Bq kg^{-1} with a mean of 26.26 $\pm$ **2.4**Bq kg^{-1}. This shows that, similar activity concentration was found throughout the forestland with less variation. At the same time, samples that were collected from interior parts of the forest showed

high concentration of thorium, since the samples collected from these areas were covered with bushes and trees of various species where soils were generally undisturbed much by weathering.

Table 1: The activity concentration of Radionuclides and Ra_{eq} values for soil samples

Location	Activity Concentration [$Bqkg^{-1}$]			Radium Equivalent(Ra_{eq})	Observed Dose [$nGyh^{-1}$]
	^{238}U	^{232}Th	^{40}K		
S-1	15.12	39.17	198.79	86.44	93.98
S-2	21.03	45.89	205.37	102.47	96.59
S-3	19.99	47.76	202.77	103.90	86.36
S-4	21.42	48.91	195.39	106.41	94.32
S-5	20.19	53.55	209.67	112.91	78.41
S-6	27.9	51.86	218.06	118.85	82.95
S-7	18.57	46.96	201.14	101.21	89.77
S-8	24.38	48.67	148.89	105.44	93.18
S-9	18.56	44.14	211.19	97.94	90.91
S-10	30.12	58.46	214.56	130.24	98.9
S-11	33.42	61.32	224.56	138.40	115.72
S-12	41.21	70.28	233.71	159.71	118.23
S-13	44.11	76.13	248.12	172.08	123.81
S-14	37.91	64.61	221.5	147.36	100.82
S-15	19.99	46.5	127.54	96.31	90.45
Range	15.12 - 41.21	39.17- 76.13	127.54 - 248.12	86.44-172.08	78.41-123.81
AM $\pm$ SE*	26.26 $\pm$ 2.4	53.61 $\pm$ 2.7	204.08 $\pm$ 7.9	118.66 $\pm$ 6.5	96.96 $\pm$ 3.4

*SE(Standard Error = $\sigma/\sqrt{N}$, where σ is SD(Standard Deviation and N is the no of observation

On the other hand, the activity concentration of ^{232}Th was much higher than ^{238}U at all the locations. The activity of ^{232}Th in soil ranged from 39.17 to 76.13Bq kg-1 with a mean of 53.61 + 2.7Bq kg-1. The spectral measurement clearly revealed the spectral photo peaks at 238.3, 373.3, 510.7, 727.3, 911.2, 916, 1587 and 2614KeV which were due to the daughter products of 232Th series viz, ^{212}Pb, ^{228}Ac, ^{208}Tl, ^{208}Tl, ^{212}Bi, ^{228}Ac, ^{212}Bi and ^{208}Tl, respectively. This confirms the abundance radionuclides from ^{232}Th series in soil and the same was observed by Selvasekarapandian et al 2000 [2]. Also the deposits of monazite on the coastal areas of Kerala and Tamil Nadu were formed due to the weathering of rocks in Western Ghats.

The activity of ^{40}K in soil ranged from 127.54 to 248.12Bq kg^{-1} with a mean of 204.08 + 7.9Bq kg^{-1}. The previous background radiation survey by Selvasekarapandian et al (2000)[2] showed that mean activity of ^{232}Th-series, ^{238}U-series and ^{40}K are 4.4, 1.9 and 0.742 time was higher than the world average values reported by the UNSCEAR 2000 Report (Such as ^{238}U, ^{232}Th and ^{40}K were 35Bqkg^{-1}, 30Bqkg^{-1} and 400Bqkg^{-1} respectively)[11] . The mean activity of ^{232}Th observed in the present work is 1.5 times higher than the world average value whereas the mean activity of ^{238}U and ^{40}K was observed to be lower than the world average. These variations in the activity concentration may be explained by the difference in natural ecosystems and the terrestrial ecosystems. There are several important features, the main one being that, in terrestrial ecosystems, soils are periodically ploughed and fertilized, while in natural systems they exhibit a more or less clear subdivision in the upper, mainly organic horizon and the lower, mineral horizon. They differ in several important characteristics such as pH, moisture, nutrient status, biological activity etc.(Frissel et al. 1990) [12].

While comparing radionuclides from different decay chains (^{232}Th and ^{238}U), it was observed that both the series are linearly related i.e. concentration of ^{232}Th-series increases with increase of ^{238}U-series, but Y- intercept is clearly different from zero. This fact reflects that the ^{232}Th/^{238}U activity ratio is not constant across the forest soil.

A graph is plotted between **^{232}Th/^{238}U** activity ratios with the ^{238}U concentration. The curves reflect the variation of activity ratio and expressed mathematically a hyperbolic function:

$$R = {}_a C_s^b$$

Where **R** is the activity ratio, ***Cs*** is concentration of radionuclide ^{238}U in the soil and *a* and*b* parameters to determined. Using the above equation, the following function is obtained.

^{232}Th/^{238}U= 9.1697(^{238}U)$^{-0.4561}$, (With regression coefficients of –0.9058)

This relationship reflects that the activity ratio remains constant only for high concentration of ^{238}U in the soil. For low activity concentration, contamination of radionuclides from ^{232}Th decay chain seems to be undistinguished.

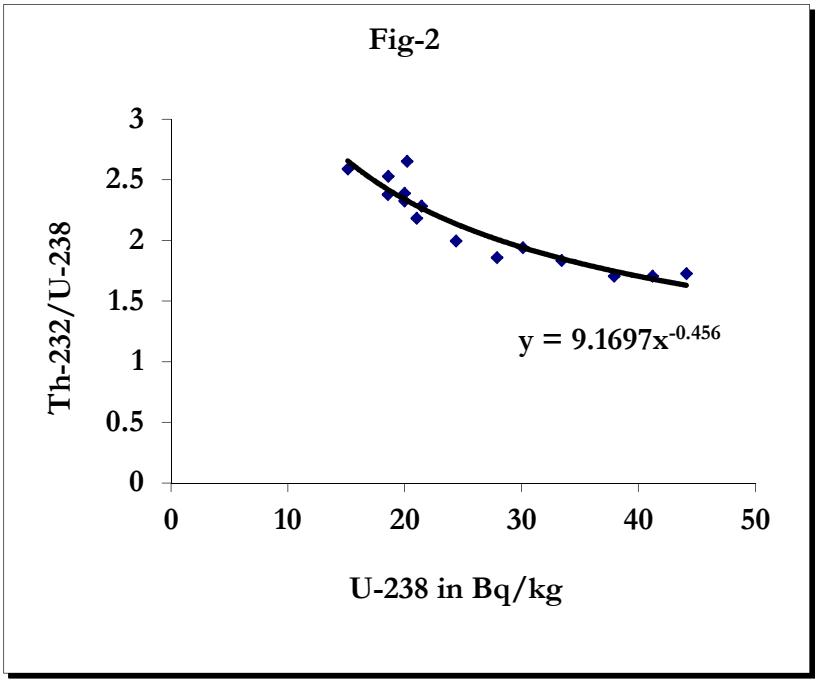

Fig – 3. $^{232}Th/^{238}U$ activity ratio vs concentration of ^{238}U in soil

Dose calculation

Absorbed and observed dose rate: The mean activity concentrations of Th and K are converted in to dose rate based on the conversion factor given by UNSCEAR (2000) [11] (Table 2).

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

Where D is the absorbed dose rate ($nGyh^{-1}$), C_U , U_{Th} and C_K are the activity concentrations ($Bqkg^{-1}$) of ^{238}U,^{232}Th and ^{40}K in soil samples respectively. The range of absorbed dose rates is from 38.93 $nGyh^{-1}$ to76.71 $nGyh^{-1}$ with an average of 53.03 $\pm$ 2.9nGy/h that exceeds the world average value of 51$nGyh^{-1}$ UNSCEAR (2000) [11].

The outdoor gamma dose rates were measured 1 m above the ground by a portable digital ERDM at all the sampling sites. A total of five readings were recorded at each spot and the average was taken (Table 1). Studies indicate an average outdoor gamma dose rate of 60 $nGyh^{-1}$ in the world ranging from 10 to 200$nGyh^{-1}$(Taskin et al. 2009) [13].

Table 2 Radiological parameters for the soil samples

Location	Absorbed Dose ($nGyh^{-1}$)				External Hazard Index (H_{ext})	Outdoor Annual effective dose Equivalent (μSvy^{-1})
	^{238}U	^{232}Th	^{40}K	Total		
S-1	6.99	23.66	8.29	38.93	0.23	47.75
S-2	9.72	27.72	8.56	46.00	0.28	56.41
S-3	9.24	28.85	8.46	46.54	0.28	57.07
S-4	9.90	29.54	8.15	47.59	0.29	58.36
S-5	9.33	32.34	8.74	50.42	0.30	61.83
S-6	12.89	31.32	9.09	53.31	0.32	65.37
S-7	8.58	28.36	8.39	45.33	0.27	55.59
S-8	11.26	29.40	6.21	46.87	0.28	57.48
S-9	8.57	26.66	8.81	44.04	0.26	54.01
S-10	13.92	35.31	8.95	58.17	0.35	71.34
S-11	15.44	37.04	9.36	61.84	0.37	75.84
S-12	19.04	42.45	9.75	71.23	0.43	87.36
S-13	20.38	45.98	10.35	76.71	0.46	94.07
S-14	17.51	39.02	9.24	65.78	0.40	80.67
S-15	9.24	28.09	5.32	42.64	0.26	52.29
Range	6.99-20.38	23.66-45.98	5.32 -10.35	38.93-76.71	0.23 - 0.46	47.75 - 94.07
AM $\pm$ SE*	12.13 $\pm$1.1	32.38 $\pm$1.6	8.51$\pm$0.3	53.02$\pm$2.9	0.31$\pm$0.02	65.03$\pm$3.6

*SE(Standard Error = $\sigma/\sqrt{N}$, where σ is SD(Standard Deviation and N is the no of observation

The present study in Western Ghats shows that the average gamma dose rate is 96.96 ± 3.4nGyh^{-1}, which is higher than the world average. The level of gamma radiation is directly associated with the activity concentrations of radionuclides in the samples and cosmic rays (Taskin et al. 2009) [13]. The excess dose measured by the environmental dosimeter is due to the significant contribution from the cosmic radiation in the present study area, located at 2400m above the sea level, where the contribution of cosmic ray is much higher than the normal one.

The Annual Effective Dose Equivalent (AEDE):

To estimate the annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose (0.7SvGy^{-1}) and outdoor occupancy factor (0.2) proposed by UNSCEAR (2000) [11] are used. Therefore, the annual effective dose rate was calculated by the following equation:

$$\textbf{Effective dose rate (μSv/y) = D(nGy/h) × 8760h × 0.7 Sv/Gy× × 0.2 × } 10^{-3}$$

The outdoor annual effective dose equivalents obtained for the samples are presented in Table 2 and it was found to be 65.03 ± 3.6μSv which is within the world average value of 70μsv (Orgun et al. 2007) [14].

Radiological Hazard Indices:

The Gamma ray radiation hazards caused by the specified radionuclides in samples were assessed by calculating the different indices. Even though total activity concentration of radionuclides is calculated, it does not provide the exact indication of total radiation hazards. Also, these hazard indices are used to select the right materials, because soil potentially contaminated is used for making earthen huts, bricks and pottery items.

The gamma–ray radiation hazards due to the specified radionuclides were assessed by two different indices (Radium-equivalent activity and external radiation hazard). A widely used hazard index (reflecting the external exposure) called the externalhazard index H_{ex} is defined as follows:

$$\textbf{Hex= } (C_U/370) + (C_{Th}/259) + (C_K/4810)$$

where C_U, C_{Th} and C_K are mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K in Bqkg^{-1} respectively,Hazard indices of all sites samples were found to be less than unity (permissible level)(Orgun et al. 2007) [14].

Radium Equivalent (Raeq):

Exposure to radiation can be defined in terms of many parameters. It is well known that, Radium equivalent activity (Ra_{eq}) is also a widely used Radiation hazard indices. The indices were defined as below (Beretka and Mathew 1985)[15]

$$\mathbf{Ra_{eq} = (A_U + 1.43\ A_{Th} + 0.077\ A_K)\ (Bqkg^{-1})}$$

Where A_U, A_{Th} and A_K are the activity concentration of ^{238}U, ^{232}Th and ^{40}K ($Bqkg^{-1}$) in the soil samples respectively. "Radium equivalent activity index (Raeq) represents a weighted sum of activities of the above-mentioned natural radionuclides and is based on the assumption that 259 $Bqkg^{-1}$ of ^{232}Th, 370 $Bqkg^{-1}$ of ^{226}Ra and 4810 $Bqkg^{-1}$ of ^{40}K produce the same gamma radiation dose rates. The use of materials whose radium equivalent activity concentration exceeds 370 $Bqkg^{-1}$ is discouraged to avoid radiation hazards. The annual effective dose for Raeq of 370 $Bqkg^{-1}$ corresponds to the dose limit of 1.0 mSv for the general population" (Tahir et al. 2005) [16]. The calculated average radium equivalent activity value in the present study is 118.66 $\pm$ 6.5$Bqkg^{-1}$ which is lower than above said value of 370$Bqkg^{-1}$.

Conclusion

The average values for ^{238}U and ^{40}K in all areas under investigation are lower than the world wide values reported by UNSCEAR (2000) [11]. The thorium concentration in the Western Ghats region is on the higher side of the world wide range which could be due to the existence of monazite sand in the area of study. The average outdoor gamma dose rate is higher than the world average, and thus Western Ghats region comes under an elevated background radiation in the world. In spite of all these, the other calculated radiological hazard indices are within the acceptable limits, (Safety Limit) and thus we can conclude that forest environment of Western Ghats has high background radiation, but it will not pose much radiological risks from harmful effects of ionizing radiation from the naturally occurring radionuclides in soil to the population. Also, the results of measurements will serve as an excellent base line data and, especially as a reference level for soil samples of Western Ghats.

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The recent study in the Western Ghats region shows that the presence of high activity concentration of primordial radionuclides in soil. It was therefore felt worthwhile to study the radioactivity in some tropical forest in the region of Western Ghats. The present work aims to assess and try to understand the behavior of primordial radionuclides present in forest soils and to measure the radiation in the local environment of long wood forest of west Nilgiris system. Soil radioactivity is usually important for the purpose of establishing baseline data for future assessment of radiation impact, radiation protection, and exploration. Not only that, a comparative analysis are rare nowadays in which an article that gives an assessment and level of exposure to natural radiation of the entire Western Ghats region is important to improve the foundation for decisions of the authorities and policy makers.