

# Activity Concentrations of Natural Radionuclides in Soils of Rainforest Sites in Western Ghats

\*P.K.Manigandan, and K.K.Natrajan

\*Departemnt of Engineering, Al Musanna College of technology-Oman

\*pkmgs@yahoo.com

**Abstract**– Assessments of naturally occurring radionuclides in soil collected from a tropical rainforest forest of western Ghats, India were conducted. These radionuclides were distributed unevenly in the forest soil. For all soil samples, the terrestrial gamma dose rate and the corresponding outdoor annual effective dose equivalents were evaluated. The activity concentration of  $^{232}\text{Th}$  and average outdoor gamma dose rates were found to be higher than the global average which appears to affects Western Ghats environment in general, the radiological hazard indices were found to be within the International Commission on Radiological Protection recommended limits. Hence, obtained results for natural radionuclides in the forest soils were within the range specified by UNSCEAR (2000) report for virgin soils except  $^{232}\text{Th}$ .

**Key words**– Naturally occurring radionuclides, Western Ghats, Monazite, radiological hazard

## I. INTRODUCTION

We have previously reported that activity concentration of thorium 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. Literature indicates that the deposit of monazite on the coastal areas of Kerala and Tamil Nadu were 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). 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 (Valithan 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 radionuclides in the ecosystem and contributes to the total absorbed dose via ingestion, inhalation and external irradiation. Forest soils in comparison with agriculture soils are more suitable for radionuclide investigations, because they not are usually disturbed by cultivation over long period of time. Characteristics of forest soils may modify radionuclide transfer in the and their bioaccumulation in comparison with other ecosystems (Segovia et al. 2003)[5]. These are 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 (Gasó et al. 1998; Vaca et al. 2001) [6-7]. Therefore, thorough knowledge about the level of exposure to natural radiation from natural gamma-emitting radionuclides is important to the authorities and policy makers for making the right decisions.

## II. MATERIALS AND METHODS

### A. Study Area

The soils analyzed were collected from elevations of between 2000 and 2400 m the Nilgiri Highlands, Tamil Nadu, South India, which are situated between  $11^{\circ} 00'$  and  $11^{\circ} 30' \text{ N}$  and between  $76^{\circ} 00'$  and  $77^{\circ} 30' \text{ E}$ . 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. 1. The vegetation above 2000 m in the highlands is a mosaic of high-elevation evergreen forests, called 'shola' locally, and grasslands with different compositions of flora, including C4 grasses (Sukumar et al. 1995; Rajagopalan et al. 1997) [8-9].

### B. Sample Collection

The study area was divided into a 4-km grid and soil samples were collected from 15 sampling points in the natural, uncultivated, and grass-covered level areas within the grid, conforming to International Atomic Energy Agency recommendations (IAEA 1989)[10]. The 15 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 position and elevation of each sampling point was determined using a global positioning system.

### C. Sample Processing

The soil samples were transported to the laboratory and 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 equilibrium had been established between radium and its short-lived daughters. Detailed gamma-ray spectrometry analysis was performed on the soil samples.

### D. Activity Determination

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



Figure 1 Distribution of Monazite sand along the Kerala Coast

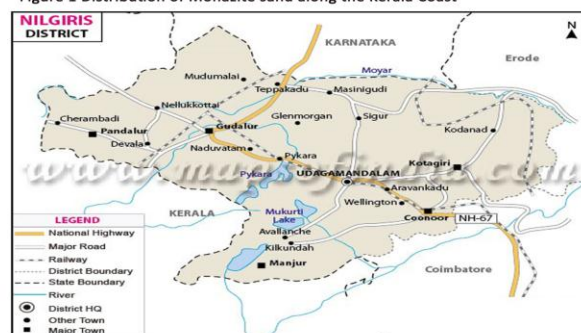


Figure 2 Study area: Nilgiri district

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

In addition to the gamma-ray spectrometric analysis, a low-level survey environmental radiation dosimeter (type ER 705; Nucleonic System PVT Ltd., Hyderabad, India) meter was used to measure the ambient radiation levels in the forest in the study area. The dosimeter had a halogen quenched Geiger–Müller detector (Ind. Inc., U.S.A) powered by a rechargeable battery, and was designed to read the exposure rate at two levels, 0.1  $\mu\text{R/h}$  and 1  $\mu\text{R/h}$ . The dosimeter was calibrated using a standard source before use.

### III. RESULTS AND DISCUSSION

The activity concentration of naturally occurring radionuclides in forest soil of Western Ghats is shown in Table 1. The mean activity concentration ranges for  $^{238}\text{U}$  in soil was 15.12 to 41.21Bq/ kg with an averages of  $26.26 \pm 9.1\text{Bq/kg}$ . 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 NATURALLY OCCURRING RADIONUCLIDES AND RAEQ VALUES IN SOIL SAMPLES

Location	Activity Concentration [Bq/kg]			Radium Equivalent ( $R_{aeq}$ )	Observed Dose(ERD) [nGy/h]
	$^{238}\text{U}$	$^{232}\text{Th}$	$^{40}\text{K}$		
S-1	33.42	61.32	224.56	138.40	115.72
S-2	41.21	70.28	233.71	159.71	118.23
S-3	44.11	76.13	248.12	172.08	123.81
S-4	37.91	64.61	221.5	147.36	100.82
S-5	19.99	46.5	127.54	96.31	90.45
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	15.12	39.17	198.79	86.44	93.98
S-12	21.03	45.89	205.37	102.47	96.59
S-13	19.99	47.76	202.77	103.90	86.36
S-14	21.42	48.91	195.39	106.41	94.32
S-15	20.19	53.55	209.67	112.91	78.41
Range	15.12 - 41.21	39.17- 76.13	127.54 - 248.12	86.44- 172.08	78.41- 123.81
Mean $\pm$ $\sigma$ *	$26.26 \pm 9.1$	$53.61 \pm 10.4$	$204.08 \pm 30.4$	$118.66 \pm 25.3$	$96.96 \pm 12.9$

\*  $\sigma$  is Standard Deviation

On the other hand, the activity concentration of  $^{232}\text{Th}$  was much higher than  $^{238}\text{U}$  at all the locations. The activity of  $^{232}\text{Th}$  in soil ranged from 39.17 to 76.13Bq/kg with a mean of  $53.61 + 10.4\text{Bq/kg}$ . The spectral measurement clearly exposed 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  $^{232}\text{Th}$  series viz,  $^{212}\text{Pb}$ ,  $^{228}\text{Ac}$ ,  $^{208}\text{Tl}$ ,  $^{208}\text{Pb}$ ,  $^{212}\text{Bi}$ ,  $^{228}\text{Ac}$ ,  $^{212}\text{Bi}$  and  $^{208}\text{Tl}$ , respectively. Hence, this observation endorses presence of  $^{232}\text{Th}$  series in soil and 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  $^{40}\text{K}$  in soil ranged from 127.54 to 248.12Bq/ kg with a mean of  $204.08 \pm 30.4\text{Bq/kg}$ . The previous background radiation survey by Selvasekarapandian et al (2000)[2] showed that mean activity of  $^{232}\text{Th}$ -series,  $^{238}\text{U}$ -series and  $^{40}\text{K}$  are 4.4, 1.9 and 0.742 time was higher than the world average values reported by the UNSCEAR 2000 Report (Such as  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  were 35Bq/kg, 30Bq/kg and 400Bq/kg respectively)[11]. The mean activity of  $^{232}\text{Th}$  observed in the present work is 1.5 times higher than the world average value whereas the mean activity of  $^{238}\text{U}$  and  $^{40}\text{K}$  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 ( $^{232}\text{Th}$  and  $^{238}\text{U}$ ), it was observed that both the series are linearly related i.e. concentration of  $^{232}\text{Th}$ -series increases with increase of  $^{238}\text{U}$ -series, but Y- intercept is clearly different from zero. This fact reflects that the  $^{232}\text{Th}/^{238}\text{U}$  activity ratio is not constant across the forest soil.

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

$$x = aC_s^b$$

Where  $X$  is the activity ratio,  $C_s$  is concentration of  $^{238}\text{U}$  radionuclide in the soil and  $a$  and  $b$  parameters to determined. Using the above equation, the following function is obtained.

$$^{232}\text{Th}/^{238}\text{U} = 9.2 (^{238}\text{U})^{-0.456},$$

(With regression coefficients of -0.9)

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

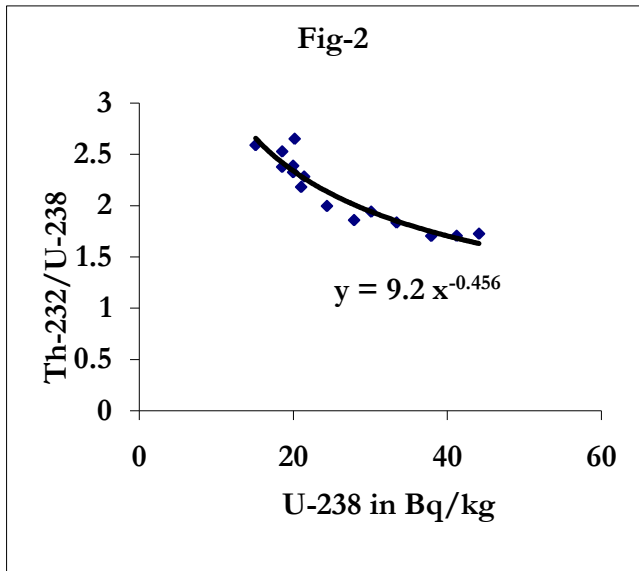


Fig 2. <sup>232</sup>Th/<sup>238</sup>U activity ratio vs concentration of <sup>238</sup>U in soil

**A. Dose Calculation**

1) *Absorbed and observed dose rate:* The mean activity concentrations of <sup>232</sup>Th and <sup>40</sup>K are converted in to dose rate based on the conversion factor given by UNSCEAR (2000) [11] (Table 2).

$$D = (0.462C_U + 0.604C_{Th} + 0.0417C_K) \text{ nGy/h} \dots\dots\dots(1)$$

Where D is calculated the absorbed dose rate (nGy/h) C<sub>U</sub>, C<sub>Th</sub> and C<sub>K</sub> are the activity concentrations (Bq/kg) of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in soil samples respectively. The range of calculated absorbed dose rates is from 38.93 nGy/h to 76.71 nGy/h with an average of 53.03 ± 11.2nGy/h that similar the world average value of 51nGy/h reported in UNSCEAR (2000) [11].

The outdoor gamma dose rates were measured 1 m above the ground by a portable digital ERD at all the sampling sites. A total of five readings were recorded at each spot and the average was taken (Table 1). Other studies indicate an average outdoor gamma dose rate of 60 nGy/h in the world ranging from 10 to 200nGy/h (Taskin et al. 2009)[13] but also similar to our determination within the experimental range.

The present study in Western Ghats shows that in the field, measured average gamma dose rate is 96.96 ±

12.9nGy/h, which is slightly higher than the world average. The level of gamma radiation is directly associated with the activity concentrations of radionuclides in the soil and cosmic rays (Taskin et al. 2009) [13]. The excess dose measured in the field with the portable dosimeter (96.96±12.9 nGy/h) in comparison with the absorbed dose expected on the basis of radionuclide concentrations determined in soil samples (53.03±11.2 nGy/ h) 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.

**B. The Annual Effective Dose Equivalent (AEDE):**

The absorbed dose to effective dose conversion coefficient (0.7 Sv/Gy) and an outdoor occupancy factor (0.2), which have been proposed by UNSCEAR (2000)[11], were used to estimate the annual effective dose rates, as shown in Eq. 2.

$$\text{Effective dose rate (Outdoor)}(\mu\text{Sv/y}) = D (\text{nGy/h}) \times 8760\text{h} \times 0.7\text{Sv/Gy} \times 0.2 \times 10^{-3} \dots\dots\dots (2)$$

The outdoor annual effective dose equivalents obtained for the samples are presented in Table 2 and it was found to be 65.03 ± 13.8μSv which is within the world average value of 70μsv (Orgun et al. 2007) [14].

**C. 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<sub>ex</sub> is defined as follows:

$$H_{ex} = \left[ \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \right] \dots\dots\dots(5)$$

where C<sub>U</sub>, C<sub>Th</sub> and C<sub>K</sub> are mean activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in Bq/kg respectively, Hazard indices of all sites samples were found to be less than unity (permissible level)(Orgun et al. 2007) [14].

TABLE 2

RADIOLOGICAL PARAMETERS FOR THE SOIL SAMPLES

Location	D, Absorbed Dose (nGy/h)				External Hazard Index (H <sub>ext</sub> )	Outdoor Annual effective dose Equivalent <sub>t</sub> (μSv/y)
	<sup>238</sup> U	<sup>232</sup> Th	<sup>40</sup> K	Total		
S-1	15.44	37.04	9.36	61.84	0.37	75.84
S-2	19.04	42.45	9.75	71.23	0.43	87.36
S-3	20.38	45.98	10.35	76.71	0.46	94.07
S-4	17.51	39.02	9.24	65.78	0.40	80.67
S-5	9.24	28.09	5.32	42.64	0.26	52.29
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	6.99	23.66	8.29	38.93	0.23	47.75
S-12	9.72	27.72	8.56	46.00	0.28	56.41
S-13	9.24	28.85	8.46	46.54	0.28	57.07
S-14	9.90	29.54	8.15	47.59	0.29	58.36
S-15	9.33	32.34	8.74	50.42	0.30	61.83
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
Mean±σ	12.13 ± 4.2	32.38 ± 6.3	8.51±1.27	53.02±11.2	0.31±0.07	65.03±13.8

\* σ is SD (Standard Deviation)

**D. Radium Equivalent (Raeq):**

Exposure to radiation can be defined in terms of many parameters. It is well known that, Radium equivalent activity (Raeq) is also a widely used Radiation hazard index. The

indices were defined as below (Beretka and Mathew 1985) [15]

$$Ra_{eq} = (A_U + 1.43 A_{Th} + 0.077 A_K) (Bq/kg)$$

.....(4)

Where A<sub>U</sub>, A<sub>Th</sub> and A<sub>K</sub> are the activity concentration of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K (Bq/kg) 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 Bq/kg of <sup>232</sup>Th, 370 Bq/kg of <sup>226</sup>Ra and 4810 Bq/kg of <sup>40</sup>K produce the same gamma radiation dose rates. The use of materials whose radium equivalent activity concentration exceeds 370 Bq/kg is discouraged to avoid radiation hazards. The annual effective dose for Raeq of 370 Bq/kg 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 ± 25.3Bq/kg which are lower than above said value of 370Bq/kg.

**IV. CONCLUSION**

The average values for <sup>238</sup>U and <sup>40</sup>K in all areas under investigation are within the world wide values reported by UNSCEAR (2000). 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 above average 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 slightly high background radiation, but despite of this, it will not pose much radiological risks regarding harmful effects of ionizing radiation from the naturally occurring radionuclides in soil to the population. Also, the results of measurements will serve as base line data and, as a reference level for soil samples of Western Ghats.

**ACKNOWLEDGEMENT**

The authors are thankful to Dr. A. Natarajan, Head, HASL, IGCAR, Dr. A.R. Lakshmanan. HASL, IGCAR, Dr. A.R.Iyengar, Head, ESL, Kalpakkam for their constant encouragement throughout the period of work.

**REFERENCE**

[1] Manigandan P K (2009). "Activity concentration of radionuclides in plants in the environment of Western Ghats, India". African Journal of Plant Science 3 (9): 205-209,

- [2] Selvasekarapandian S, Manikandan N, Sivakumar R (2000). "Natural radiation distribution of soil at Kotagiritaluk of Nilgiris biosphere in India". Eighth International Conference, October 16-20, Ibaraki, Japan.
- [3] Iyengar M A R, Ganapathy S, Kannan V, Rajan MP, Rajaram S (1990). "Procedure Manual, Workshop on Environmental Radioactivity, Kiga, India".
- [4] M.S Valithan C.C Kartha, C.C. Nair K Shivakumar and T.T Eapan (1994) geochemical basis of tropical endomyocardial fibrosis. *Current Science*. 67(2): 99-104
- [5] Segovia N, Gaso MI, Alvarado E, Pena P, Morton O, Armienta MA, Reyes AV (2003). "Environmental radioactivity studies in the soil of a coniferous forest". *Radiat Meas* 36:525-528
- [6] Gaso MI, Segovia N, Herrera T, Perez-Silva E, Cervantes ML, Quintero E, Palacios J, Acosta E (1998). "Radiocesium accumulation in edible wild mushrooms from coniferous forests around the Nuclear Centre of Mexico". *Sci Total Environ* 223:119-129
- [7] Vaca F, Manjon G, Garcia-Leon M (2001). "The presence of some artificial and natural radionuclides in a Eucalyptus forest in the South of Spain". *J Environ Radioactivity* 56:309-325
- [8] Sukumar R, Suresh HS and Ramesh R (1995). "Climate change and its impact on tropical montane ecosystems in southern India". *Curr. Sci.*, 22 : 533-536.
- [9] Rajagopalan G, Sukumar R, Ramesh R and Pant RK (1997). "Late Quaternary vegetational and climatic changes from tropical peats in southern India - An extended record up to 40,000 years B.P". *Curr. Sci.*, 73: 60-63.
- [10] IAEA (1989), "Measurement of Radionuclides in Food and Environment, IAEA Technical Report Series No: 295", IAEA, Vienna .
- [11] UNSCEAR (2000). "Sources and biological effects of ionizing radiation". Report to general assembly with scientific annexes. United Nations, New York
- [12] Frissel M J, Noordijk KH and van Bergejik K E (1990). "The impact of extreme environmental condition, as occurring in natural ecosystem's on the soil to plant transfer of radionuclides". Elsevier, London and New York: 40-47
- [13] Taskin H, Karavus M, Topuzoglu A, Hindiroglu S and Karahan G, (2009). "Radionuclide concentrations in soil and lifetime cancer risk due to the gamma radioactivity in Kirklareli, Turkey". *Journal of Environmental Radioactivity.*, 100: 49-53.
- [14] Orgun Y, Altinsoy N, Sahin SY, Gungor Y, Gultekin AH, Karaham G and Karaak Z, (2007). "Natural and anthropogenic radionuclides in rocks and beach sands from Ezineregion (canakkale), Western Anatolia, Turkey". *Applied Radiation and Isotopes*, 65: 739-747.
- [15] Beretka J, Mathew P J (1985). "Natural radioactivity of Australian building materials, industrial wastes and by- products". *Health Phys.* 48: 87-95.
- [16] Tahir S N A, Ismail K, Zadi J H (2005). "Measurement of activity concentrations of naturally occurring radionuclides in soils samples from Punjab province of Pakistan and assessment of radiation hazards". *Radiation Protection Dosimetry*. 113 (4): 421-427