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ANALYSIS OF RADIONUCLIDE LEVEL IN SOIL AND ROCK POWDER SAMPLES AND ASSESSMENT OF RADIOLOGICAL RISKS

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Abstract

The activity concentration of natural radionuclides such as ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K in rock and the surrounding soil samples were collected from 20 different locations in the lower hills of western ghats, Kanyakumari district. The measurements were carried out using a 3" × 3" NaI gamma spectrometry. Gamma spectrometric analyses were performed and the mean activity concentrations obtained for each of the radionuclides expressed in Bq/Kg are 19.0, 7.59, 20.35 and 192.52 for ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K for soil samples and 18.51, 6.41, 20.43 and 199.89 for ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K for rock samples respectively. Also the radium equivalent activity, representative level index, external hazard index, Internal hazard index, Annual gonadal dose equivalent, Gamma absorbed dose were found to be within the permissible limits and would not pose any significant radiological threat to the population resident in the studied areas.

Keywords: Distribution, Western Ghats, Radionuclides, Impact, Kanyakumari district.

Introduction

Radionuclides are the sources of radioactivity and emit nuclear radiations which have become a part of our daily lives. The most common forms of ionizing radiation are alpha particles, beta

particles and gamma rays (Lilley *et al* 2001). Radiation can arise not only from natural radio nuclides but it can also be from man-made sources. The properties of radiation have been widely applied to various purposes such as medicine, biology, Industry, agriculture, and electric power generation (Eisenbud *et al* 1997). As a result of the applications of radiation, humans can be exposed to the radiation emitting from different radioactive sources depending upon their activities and surroundings (Klement *et al* 1982). However, not all of the population is subjected to all the various sources of radiation exposure. For instance, patients who are treated with medical irradiation or members of staff who work in the nuclear industries may receive higher radiation exposure levels than members of general public (Watson *et al* 2005). Natural back ground radiation is inevitably present everywhere in the environment and gives the largest contribution to the total radiation dose received by humans. The natural sources of radiation are cosmic rays and naturally occurring radioactive substances existing in the Earth itself and inside the human body. Naturally occurring radionuclides, especially ^{238}U and ^{232}Th series and ^{40}K, in soils are one of the main contributors to outdoor external gamma radiation (UNSCEAR 2000). Naturally occurring radionuclides contribute to a major portion to the effective dose of the world wide population. Natural radionuclides in soil generate a significant component of the background radiation exposure of the population (Merdanoglu *et al* 2006). The study of distribution of these radionuclides in soil and rock samples is of great importance for radiation protection and measurements. The knowledge of radionuclide distribution and radiation levels in the environment is important for assess the effects of radiation exposure due to both terrestrial and extra terrestrial sources. Terrestrial radiation is due to radioactive nuclides present in varying amounts in soils, building materials, water, rocks and atmosphere (Akhtar *et al* 2005). Natural radio nuclides in soil generate a significant component of the back ground radiation exposure of

the population. The ^{226}Ra, ^{232}Th and ^{40}K which pose exposure risks externally due to their gamma ray emissions and internally due to radon and its progeny, are distributed everywhere in the earth's environment with different concentrations. The upper most layer of the earth is covered by soil. The soil is a complex mixture of different compounds and rocks in the natural environment. The natural radionuclides enter into the soil from the earth's crust is the principal source of natural radionuclide in soils and rocks (Tahir *et al* 2005; UNSCEAR 2000). The study of natural radioactivity present in rocks and ornament stones, such as granite, is an important subject in environmental radiological prortection (Anjos *et al* 2005). The concentrations of natural radionuclides in rock have been found to depend on the geological condition and as such they vary from one place to another. Higher concentrations and higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks. There are exceptions, however, as some shale and phosphate rocks have relatively high content of radionuclides (Arafa *et al* 2004). Most of the granites contain radioactive elements such as ^{238}U, ^{232}Th, ^{40}K and ^{226}Ra. In terms of natural radioactivity of granites exhibit an enhanced elemental concentration of U and Th compared with the very low abundance of these elements observed in the mantle and the crust of the Earth. Igneous rocks of granite composition are strongly enriched in U and Th, compared with rocks of basaltic or ultramafic composition. (Faure *et al* 1986, Beretka *et al* 1985). Investigations and measurements of natural back ground radiation and radioactivity are of great importance in health physics, not only for many practical reasons but also for more fundamental scientific reasons. Natural back ground radiation is the main source of human exposure, studies on the dose from this source and its effect on health can improve the understanding of radiation damage. Therefore, it is proposed to get the base line data about the distribution of ^{226}Ra, ^{232}Th and ^{40}K at different places in Kanyakumari district, Tamil Nadu state,

India. The main objective of the study is to find out the present level of natural radionuclide activity concentration and radiological risks of Rock and soil samples around Kanyakumari district.

Materials and Methods

Study area

The present study was carried out in Kanyakumari District, Tamilnadu, India. The district is situated in the south most part of India. The southern region of the district is surrounded by the Indian ocean. It lies between 77^0 15'E and 77^o 36'E long. and 8^o 03' N and 8^o 35' N lat. The sampling stations are shown in Fig.1. The district is located in the southern meridian region of the state. Total geographical area of the district is 1684 sq.kms.

Sample collection and preparation

Twenty rock and soil samples were collected from the different locations of the study region.

The sample location were recorded in terms of degree – minute – decimals (Latitudinal and Longitudinal position) using hand-held Global positioning system (GPS) (Model: GARMIN GPS-12) unit.

Fig.1. Map showing the study area

Experimental technique

The measurement of environmental radioactivity of soil and rock samples were collected at different locations around Kanyakumari district. Gamma spectrometry uses semiconductor detectors like Ge (Li) or Si (Li). In this investigation, a High purity Germanium Detector (HPGe) spectrometer was used to measure the concentrations of gamma emitting radionuclides in soil and rock powder samples. From each site, approximately 2 Kg soil and rock samples were collected and analysed for radioactivity. Finally, the samples were mixed thoroughly and extraneous materials like plants, debris, big pieces of stones, and pebbles were removed (Sannappa *et al* 2006). Composite samples of about 2 kg were taken and sealed in a Polythene bag. The samples were transferred to a porcelain dish and oven dried over night at 110^{o}c. The samples were powdered and sealed in a 500 ml Plastic container, and kept for a month before counting by gamma spectrometry, in order to ensure that radioactive equilibrium was reached between ^{226}Ra, ^{222}Rn and its progeny. Assuming that the two primordial radionuclides ^{238}U and ^{232}Th were in secular equilibrium with their corresponding decay products, the ^{238}U, ^{228}Th activity concentration was calculated through 1764 Kev of ^{214}Bi and 2614.5Kev of ^{208}Tl respectively. ^{40}K activity was calculated through 1460 Kev. The gamma ray energies used to estimate the concentration of ^{226}Ra were ^{214}Bi at 609 Kev and 1120 Kev. The gamma spectrum was recorded using a PC based multichannel analyser and processed using the NETSWIN software. The activity of radionuclides was calculated using the following equation.

$$\gamma = \frac{\text{Net counts}}{T} \times \frac{100}{\text{Efficiency}} \times \frac{1}{\text{Sample weight}} \times 1000 \text{ Bq/kg}$$

Net counts = Sample counts – Back ground counts

W = Weight of the sample in grams

T = Time in seconds

Calculation of radiological effects

Radium equivalent activity (Ra_{eq})

Distribution of ^{226}Ra, ^{232}Th and ^{40}K in environment is not uniform, so that with respect to exposure to radiation, the radioactivity 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 (Beretka *et al* 1985)

$$Ra_{eq} = C_{Ra} + 1.43\ C_{Th} + 0.077\ C_k$$

Where C_{Ra}, C_{Th}, and C_k are the specific activity concentration in $Bq.Kg^{-1}$ of ^{226}Ra, ^{232}Th, and ^{40}K respectively. The index is useful to compare the specific activity of materials containing different concentrations of ^{226}Ra, ^{232}Th and ^{40}K.

Representative level index (I_{yr})

In order to examine whether the samples meets these limits of dose criteria, Another radiation hazard index, the representative level index, I_{yr}, used to examine the level of γ-radiation hazard associated with the natural radionuclides in specific investigated samples, is defined as (NEA-OCED, 1979) from the following equation.

$$I_{yr} = [A_{Ra}/150 + A_{Th}/100 + A_k/1500]$$

Where A_{Ra}, A_{Th}, and A_k are the activities of ^{226}Ra, ^{232}Th, and ^{40}Km respectively, in Bq/Kg.

External hazard index (H_{ex})

The external hazard index is defined as (Beretka and Mathew 1985)

$$H_{ex} = [A_{Ra}/370 + A_{Th}/259 + A_k/4810]$$

Where A_{Ra}, A_{Th}, and A_k are the activities of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/Kg, respectively.

Internal hazard index (H_{in})

The internal exposure to ^{222}Rn and its radioactive progeny is controlled by the internal hazard index (H_{in}) Which is given by (Quindos *et al* 1987; Cottens 1990).

$H_{in} = [A_{Ra}/185 + A_{Th}/259 + A_k/4810]$

Where A_{Ra}, A_{Th}, and A_K are the activities of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/Kg, respectively.

Annual Gonadal Dose Equivalent (AGDE)

The Gonadals, Active bone Marrow and the bone surface cells are considered the organs of interest Therefore, the AGDE (μSvy^{-1}) owing to the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K was calculated using the following formula (Arafa *et al* 2004).

$AGDE = 3.09A_{Ra} + 4.18\ A_{Th} + 0.314\ A_k$

Where A_{Ra}, A_{Th}, and A_K are the activities of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/Kg, respectively.

Absorbed dose rate

The total absorbed dose rate (nGy/h) in air due to the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K (Bq/Kg) in air at 1m above the ground surface was calculated using the following formula :

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

Results and Discussion

The radionuclide activity concentrations of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K in rock and soil samples collected from Kayakumari district. The concentration of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K in rock and soil samples with their Radium equivalent activity (Ra_{eq}), $^{226}Ra/^{232}Th$ activity ratio, Representative level index (I_{yr}), External hazard index (H_{ex}), Annual Gonadal Dose Equivalent (AGDE) with the gamma absorbed dose were calculated and are shown in tables (1) & (2). Correlation between ^{238}U and ^{232}Th activity concentration in soil samples are mentioned in the Fig. 2. In soil samples Radium equivalent activity (Ra_{eq}), varies from 32.70 to 87.55 with a

geometric mean value of 58.21. In soil samples $^{226}Ra/^{228}Th$ activity ratio varies from 0.47 to 1.43 with a geometric mean value of 0.86. In soil samples Representative level index (I_{yr}) varies between 0.24 to 0.53 with a geometric mean value of 0.42. External hazard index (H_{ex}) varies from 0.08 to 0.23 with a geometric mean value of 0.15. In Internal hazard index (H_{in}) varies from 0.11 to 0.26 with a geometric mean value of 0.19. and Annual Gonadal Dose equivalent (AGDE) varies from 107.26 to 283.07 with a geometric mean value of 191.25. Gamma absorbed dose varies from 15.22 to 33.99 with a geometric mean value of 27.06.

Table 1: Activity concentration of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K radionuclides in soil samples and their hazard index

S.No	Locations	Activity (Bq/kg)				Ra_{eq}	$^{226}Ra/^{228}Th$	I_{yr}	H_{ex}	H_{in}	AGDE	Gamma absorbed dose (nGy/h)
		^{226}Ra	^{238}U	^{232}Th	^{40}K							
1.	Chunkankadai	22.36	6.13	25.21	202.32	73.98	0.88	0.53	0.20	0.26	237.99	33.99
2.	Aralvaimozhi	23.64	8.12	16.46	126.17	56.89	1.43	0.40	0.15	0.21	205.37	26.12
3.	Bhudhappandi	14.45	5.23	15.34	118.56	45.51	0.94	0.32	0.12	0.16	145.99	20.88
4.	Kulasekaram	19.07	4.17	17.74	124.11	45.94	1.09	0.38	0.14	0.19	172.05	24.70
5.	Navalkadu	15.75	7.14	22.23	132.13	57.71	0.70	0.41	0.15	0.19	183.07	26.21
6.	Nattalam	23.48	10.28	17.55	237.68	55.10	1.10	0.39	0.14	0.20	176.46	25.29
7.	Thippiramalai	27.63	11.26	28.26	293.42	87.55	0.97	0.63	0.23	0.31	283.07	40.40
8.	Sitharal	9.12	3.24	10.34	114.22	32.70	0.88	0.24	0.08	0.11	107.26	15.22
9.	Erachakuzham	17.70	6.14	18.07	182.45	53.73	0.97	0.38	0.14	0.19	171.81	24.61
10.	pechiparai	11.81	8.02	13.63	138.25	41.94	0.86	0.30	0.11	0.14	136.87	19.45
11.	Mulankanavilai	22.31	10.23	25.89	245.89	60.56	0.47	0.43	0.16	0.19	192.06	27.40
12.	Munchirai	20.87	9.16	23.13	232.67	58.16	0.64	0.41	0.15	0.19	184.29	26.37
13.	Kakanadu	22.45	8.21	24.15	242.45	64.88	0.51	0.47	0.17	0.20	212.40	30.03
14.	Kumaracoil	28.21	14.23	30.12	284.12	54.28	0.81	0.40	0.14	0.18	181.71	25.60
15.	Kandanvilai	14.67	7.14	18.21	204.14	56.42	0.80	0.41	0.15	0.19	185.54	26.28
16.	Marunkoor	15.23	8.24	19.24	186.02	60.91	0.79	0.45	0.16	0.20	201.59	28.49
17.	Kuzhicode	16.43	6.23	22.34	198.13	66.71	0.73	0.49	0.18	0.22	218.92	31.01
18.	Velimalai	17.32	4.24	15.67	193.14	58.21	1.10	0.43	0.15	0.20	194.42	27.48
19.	Kottaram	18.37	5.21	21.15	178.13	65.41	0.86	0.47	0.17	0.22	213.66	30.35
20.	Kariki	19.15	9.23	22.34	216.42	67.76	0.85	0.49	0.18	0.23	220.51	31.36

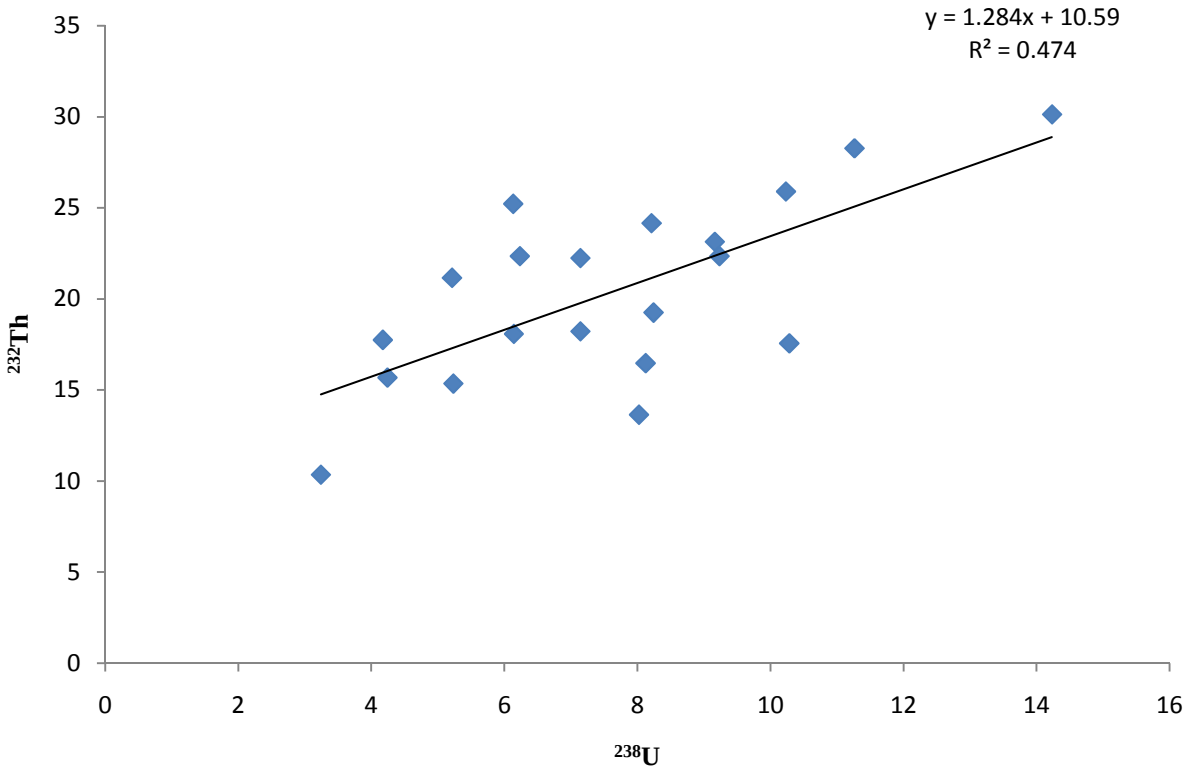

Fig. 2. Correlation between ^{238}U and ^{232}Th activity in Soil samples (Bq/Kg)

In rock samples the geometric mean activity concentrations of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K (Bq Kg^{-1}) were 18.51, 6.41, 20.43 and 199.89 respectively. Correlation between ^{238}U and ^{232}Th activity concentration In rock samples are mentioned in the Fig. 3. Radium equivalent activity (R_{eq}), varies from 8.61 to 27.23 with a geometric mean value of 54.02. In rock samples $^{226}Ra/^{232}Th$ activity ratio varies from 0.47 to 1.53 with a geometric mean value of 0.91. Representative level index (I_{yr}) varies from 0.34 to 0.50 with a geometric mean value of 0.38. External hazard index (H_{ex}) varies from 0.07 to 0.21 with a geometric mean value of 0.14. In internal hazard index (H_{in}) varies from 0.09 to 0.24 with a geometric mean value of 0.18 and the Annual Gonadal Dose Equivalent (AGDE) varies from 93.22 to 256.12 with a geometric mean value of 175.62. Gamma absorbed dose varies from 13.22 to 31.84 with a geometric value of 24.99.

Table 2 : Activity concentrations of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K radionuclides in rock samples and their hazard index

S.no	Locations	Activity (Bq/kg)				Ra_{eq}	$^{226}Ra/^{228}Th$	I_{yr}	H_{ex}	H_{in}	AGDE	Gamma Absorbed dose
		^{226}Ra	^{238}U	^{232}Th	^{40}K							
1.	Chunkankadai	21.23	3.23	23.22	192.22	69.23	0.91	0.50	0.18	0.24	223.01	31.84
2.	Aralvaimozhi	22.14	4.24	14.41	116.27	51.69	1.53	0.36	0.13	0.19	165.15	23.78
3.	Bhudhappandi	13.25	6.12	13.31	121.66	41.65	0.99	0.30	0.11	0.14	134.77	19.23
4.	Kulasekaram	17.06	4.36	15.64	188.11	48.51	1.09	0.34	0.13	0.17	155.17	22.25
5.	Navalkadu	14.65	4.56	20.19	122.13	52.92	0.72	0.38	0.14	0.18	168.01	24.05
6.	Nattalam	23.34	6.27	28.25	226.48	47.88	1.07	0.34	0.12	0.17	153.95	22.03
7.	Thippiramalai	26.43	10.56	31.21	273.32	79.01	1.09	0.57	0.21	0.28	256.12	36.56
8.	Sitharal	8.61	2.34	9.11	104.21	28.23	1.06	0.20	0.07	0.09	93.22	13.22
9.	Erachakuzham	17.70	6.56	16.03	222.45	50.05	1.10	0.35	0.13	0.18	160.14	22.96
10.	pechiparai	10.11	7.83	12.61	138.15	38.77	0.80	0.28	0.10	0.13	127.32	18.04
11.	Mulankanavilai	22.22	8.32	23.59	235.19	55.36	0.47	0.40	0.14	0.17	175.72	25.06
12.	Munchirai	23.57	9.23	27.14	222.17	54.63	0.61	0.39	0.14	0.18	172.83	24.73
13.	Kakanadu	21.41	8.34	23.25	252.45	61.01	0.49	0.45	0.16	0.19	199.15	28.17
14.	Kumaracoil	27.23	12.12	34.09	284.12	49.63	0.86	0.37	0.13	0.16	167.06	23.50
15.	Kandanvilai	12.57	7.68	16.23	194.14	51.72	0.83	0.38	0.13	0.17	170.73	24.16
16.	Marunkoor	14.33	6.42	17.28	216.02	55.67	0.82	0.41	0.15	0.18	184.34	26.06
17.	Kuzhicode	18.42	5.12	21.14	228.13	63.21	0.72	0.46	0.17	0.21	207.64	29.40
18.	Velimalai	16.31	3.56	14.57	230.14	54.86	1.11	0.40	0.14	0.19	183.56	25.93
19.	Kottaram	17.38	4.14	18.25	211.36	59.75	0.95	0.43	0.16	0.20	196.35	27.86
20.	Kariki	22.34	7 .26	29.24	219.11	66.72	1.16	0.48	0.18	0.24	218.25	31.07

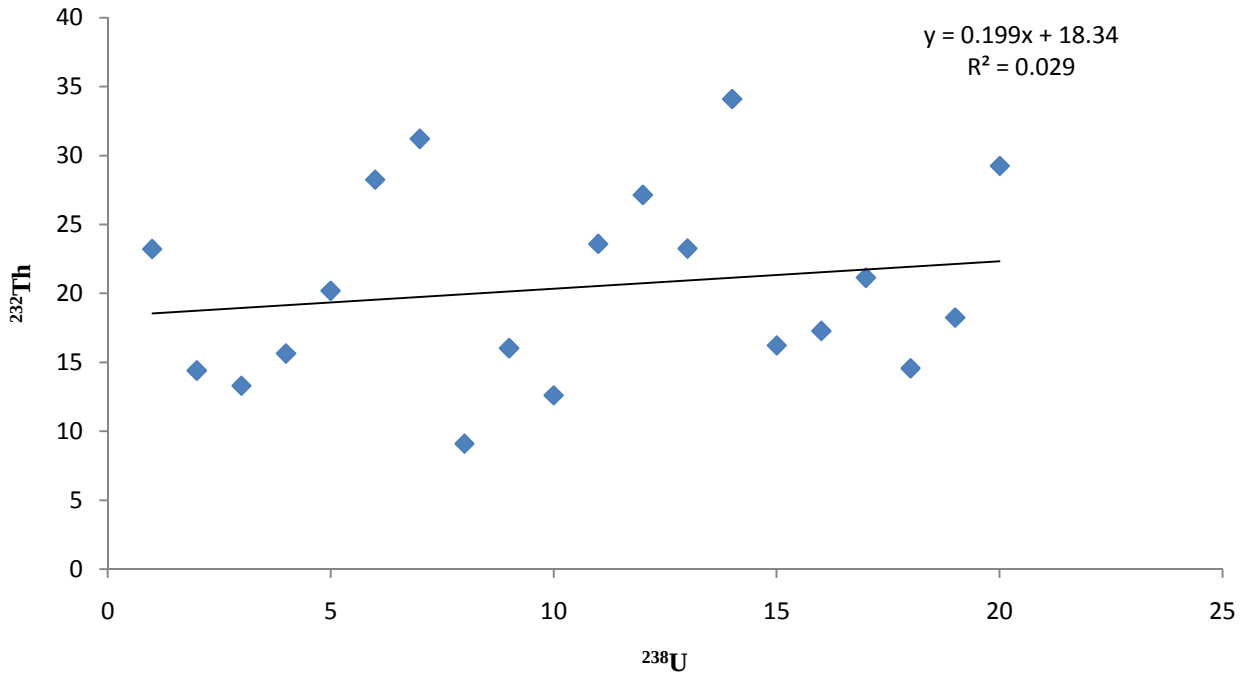

Fig. 3 Correlation between ^{238}U and ^{232}Th activity in rock samples (Bq/Kg)

The concentrations of radionuclides in soil samples activity concentration was found to be higer than in rock samples. The result is good agreement with (Singh et al., 2003; Beretka and Mathew 1985; Arafa et al., 2004).

Conclusion

The concentrations of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K in soil and rock samples collected in Kanyakumari district. Gamma spectrometric analyses were performed and the mean activity concentrations obtained for each of the radionuclides expressed in Bq/Kg are 19.0, 7.59, 20.35 and 192.52 for ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K for soil samples and 18.51 , 6.41, 20.43 and 199.89 for ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K for rock samples respectively. The concentrations of radionuclides in soil samples activity concentration was found to be higer than in rock samples. The Radium equivalent activity (Ra_{eq}), Representative level index (I_{yr}), External hazard index ($H_{ex)}$, Internal hazard index (H_{in}), Annual Gonadal dose equivalent (AGDE), Gamma absorbed dose, for ^{226}Ra,

^{238}U, ^{232}Th and ^{40}K were found to be within the permissible limits and would not pose any significant radiological threat to the population resident in the studied areas.

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