ORIGINAL RESEARCH PAPER

Physics

Measurements of Activity Concentration of Radionuclides in Soil Samples in the Southern Region of Tamil Nadu

KEY WORDS: radioactivity, soil, NaI(Tl) detector, statistical analysis

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ABSTRACT

Human exposure to ionizing radiation from natural sources is continuous throughout life. The present study assesses the gamma radiation levels and the radionuclide distribution in the soil samples collected from the southern region of Tamil Nadu. The activity of 238U vary between 0.389 ± 0.01 and 14.357 ± 1.14 Bq/kg with a mean value of 4.3409 ± 0.30 Bq/kg, 1.550 ± 0.07 to 156.577 ± 12.5 Bq/kg for 232Th with a mean value of 48.8108 ± 3.4 Bq/kg while that of 40K is found to vary from 0.925 ± 0.04 to 954.628 ± 76.3 Bq/kg with a mean value of 356.8706 ± 23.7 Bq/kg. Statistical analysis such as Pearson correlation and ANOVA shows that similar behavior in the environment is assumed for 232Th and 40K radionuclides due to positive correlation that exists between them while the behavior of 238U may differ due to weak correlation with the other two radionuclides.

Introduction

Naturally occurring radioactive materials (NORM) occur naturally in the environment. We are exposed to natural radiation every day of our lives. Humans are continuously irradiated externally and internally. Natural background radiation is of two types: terrestrial and extraterrestrial origin. Terrestrial radiation is from natural sources such as soils, building materials, water, rocks and atmosphere, and extraterrestrial radiation originates from outer space. The radionuclides that enter our bodies in the food and water, people ingest and the air they breathe cause internal radiation [1]. Artificial radioactivity includes medical procedures, commercial products that contain radioactive materials, and fallout from nuclear testing. Radionuclides produced from nuclear activities are dispersed into the environmental ecosystems depending on the physicochemical characteristics of the radionuclides. When rocks are disintegrated, radionuclides are carried to soil by rain and flows [2]. Natural radioactivity arises mainly from primordial radionuclides such as 40K, 232Th and 238U series. These radionuclides emit alpha, beta, and gamma radiation depending on the radioisotope present in the medium. The levels of these radionuclides are relatively distributed in soil based depends on the geological formation, rock composition, chemical composition of the soil, geographical and climatic conditions of the region [3]. The study of distribution of these radionuclides in soil and radiation levels in the environment is important for assess the effects of radiation exposure as it causes health risk to a population. Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions in each region of the world [4]. The radioactivity studies have been previously carried out in soil in other parts of the world [5-10]. The aim of this work is to investigate the activity concentration of ^{238}U, ^{232} Th and 40K from soil samples employing NaI(Tl) gamma spectrometry and to evaluate the radiological hazard indices associated with the corresponding samples. The results can be used as a reference information to assess any change in the radioactivity background level due to the change in the topography of the location, other developments and settlement around it or any artificial influences on the environment.

Materials and Methods

Sampling collection and preparation

Figure 1. Sampling locations of the study area

The present study area includes south east region of Tamil Nadu between Northern Latitude 8o47□ and 78o8□ Eastern Longitude (Fig. 1). About 1 kg of each soil sample at a depth of about 1–5 cm is collected from 10 locations along the southern three districts of Tamil Nadu. The samples are then crushed to fine powder using mortar and pestle and sieved using 0.250 mm size sieve to homogenize soil. The samples are placed in a hot air oven for drying at 110°C for 24 h to ensure that moisture is completely removed. The dried sample is transferred to a 250 ml cylindrical plastic container. The containers are filled fully, sealed to prevent the escape of radon from the samples. The collected samples are stored for a period of 4–5 weeks to attain radioactive equilibrium between ^{238}U and ^{232}Th and their daughter products for the analysis of gamma ray spectrometry.

Experimental methods

Measurement of activity concentrations

In this paper, an NaI(Tl) detector is used to measure the activity concentrations of gamma emitting radionuclides in soil samples. The analyzing spectra from Multichannel analyzer and obtained results of data at this work are done using a winTMCA scint SPEC. Efficiency calibration for the detector is carried out using the three IAEA Reference Standards for Uranium-238, Thorium-232 and Potassium-40 each of 250 ml geometry available for sample counting. In the NaI(Tl) detector spectra, the activity concentration of ^{238}U, ^{232}Th and ^{40}K present in the samples are computed using the gamma energies 1764 keV of 214Bi (daughter product of ^{238}U), 2614 keV of ^{208}Tl (daughter product of ^{232}Th), 1460 keV (^{40}K).

The samples are placed on the detector and each sample is counted for a period of 2000 s. The activity concentration (Bq/kg) of ^{238}U, ^{232}Th and ^{40}K is calculated using the following relation [11]:

$$A_{Ei} = \frac{N_{Ei}}{\varepsilon_E \times t \times \gamma_d \times M_s}$$

where A_{ei} is the specific activity of a nuclide i for a peak at energy E, NEi is the Net Peak Area of a peak at energy E, $\mathcal{E}_E$ is the detection efficiency at energy E, t is the counting live-time, γd is the gamma ray yield per disintegration of the specific nuclide for a transition at energy E, and M_s is the mass in kg of the measured sample.

Radiological Hazard Indices

Radium equivalent activity (Ra_{eq})

The distribution of ^{238}U, ^{232}Th and ^{40}K in soils is not uniform and is defined with respect to radiation exposure which compares the activity of materials containing different elements of these

primordial radionuclides. A common radiation index termed radium equivalent activity (Raeq) measured in becquerel per kilogram, is used. The index is calculated from the following relation [12]:

$$Ra_{eq}(Bqkg^{-1}) = C_U + 1.43\,C_{Th} + 0.077\,C_k$$

where CU, CTh, Ck are the specific activities of ^{238}U, ^{232}Th and ^{40}K, respectively. The Raeq index is defined based on the assumption that 370 Bq/kg of ^{238}U, 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K produce the same gamma-ray dose rate.

Absorbed dose rate (D)

The absorbed dose rates due to gamma radiation in air at 1 m above the ground surface for the uniform distribution of the naturally occurring radionuclides ^{238}U, ^{232}Th and ^{40}K are estimated from the measured activity concentrations of the above said three radionuclides by converting the mean activity concentrations of ^{238}U, ^{232}Th and 40K into doses based on the conversion factor (0.7Sv/Gy) estimated by the following formula [4]:

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

where D is the absorbed dose rate (nGyh-^{1}), CU, CTh, CK are the activity concentrations (Bq/kg) of ^{238}U, ^{232}Th and ^{40}K respectively.

Annual effective dose equivalent (AEDE)

The annual effective dose equivalent received by individuals is calculated by applying the dose conversion factor of conversion factor of 0.7 Sv/Gy and the outdoor occupancy of 20% and 80% for indoors. The annual effective dose is determined using the following equations [13]:

$$AEDE\ (Outdoor)(mSvy^{-1}) = D(nGyh^{-1}) \times 8760\ h \times 0.7SvGy^{-1} \times 0.2 \times 10^{-6}$$

$$AEDE\ (Indoor)(mSvy^{-1}) \quad D(nGyh^{-1}) \times 8760\ h \times 0.7SvGy^{-1} \times 0.8 \times 10^{-6}$$

Hazard indices (H_{ex} and H_{in})

Radiation hazard incurred due to external exposure to gamma rays and internal exposure from radon and its short-lived daughter radionuclides from the studied soil samples are quantified in terms of the external and internal hazard index. This index can be calculated using the following formula relations [12]:

$$H_{ex} = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1$$

$$H_{in} = \frac{C_U}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} < 1$$

where CU, CTh, CK are the activities of ^{238}U, ^{232}Th and ^{40}K, respectively, in Bqkg-^{1}. The values of the indices must not exceed the limit of unity for the radiation hazard to be insignificant.

Representative level index (Iγr)

A radiation hazard index, called representative level index, Iγr used to assess the hazardous level of radionuclides in the human body when exposed to radioactive nuclides in soils. The calculated values of *I* must be less than or equal to 1 to make sure the soil environment is generally safe. It is calculated using the following equation [14]:

$$I_{\gamma r} = \frac{C_U}{150} + \frac{C_{Th}}{100} + \frac{C_K}{1500} \leq 1$$

where C_U is the specific activity of ^{238}U in Bqkg-^{1}, C_{Th} is the specific activity of ^{232}Th in Bqkg-^{1}, CK is the specific activity of ^{40}K in Bqkg-^{1}.

Excess lifetime cancer risk (ELCR)

The excess lifetime cancer risk (ELCR) is computed using the equation [8]:

$$ELCR_{Out} = AEDE_{Out} \times DL \times RF$$

$$ELCR_{In} = AEDE_{In} \times DL \times RF$$

where AEDE is the Annual Equivalent Dose Equivalent, DL is the average duration of life (estimated to 70 years), and RF is the Risk Factor (Sv-^{1}). For stochastic effects, ICRP uses RF as 0.05 for public [15].

Results and Discussion

Analyzing the soil samples in the studied area, the activity concentrations for the measured radio- nuclides of ^{238}U, ^{232}Th and ^{40}K are obtained. It has been observed that the activity concentration of 40K is higher than the ^{238}U and ^{232}Th and it ranges between 0.93±0.04 Bq/kg and 954.63±76.3 Bq/kg with an average of 356.87±23.7 Bq/kg. The 238U concentration varies from 0.39±0.01 Bq/kg to 14.36±1.14 Bq/kg with an average of 4.34±0.30 Bq/kg. The ^{232}Th concentration ranges from 1.55±0.07 Bq/kg to 156.58±12.5 Bq/kg with an average of 48.81±3.4 Bq/kg. The activity concentration of 40K ranges between 0.93±0.04 Bq/kg and 954.63±76.3 Bq/kg with an average of 356.87±23.7 Bq/kg. The 40K activity seems to be higher in the soil sample Ku1. The higher concentration of 40K in the soil can be attributed to geochemical setting of the area [16]. The differences in naturally occurring radionuclide concentration are attributable to the geochemical composition and origin of soil types in a particular area. The main factor influencing the level of terrestrial radionuclides in soil is the corresponding concentration in the soil forming rocks. The world's average activity concentrations in soil are 30, 30 and 400 Bqkg-1 for ^{238}U, ^{232}Th and ^{40}K, respectively [4].

The calculated values of radium equivalent Raeq due to the presence of ^{238}U, ^{232}Th and ^{40}K in soil samples varies from 7.60 Bq/kg to 298.69 Bq/kg with an average of 101.62 Bq/kg in the sub-urban region respectively which are lower than the recommended limit 370 Bq/kg [4]. The calculated values of the absorbed dose rate in soil samples in the present work vary from 3.55 nGyh-^{1} to 134.97 nGyh-^{1} with an average of 46.37 nGyh-^{1} which is lower than the recommended limit (59 nGy/h). The calculated annual effective dose equivalent (outdoor and indoor) ranges between 0.004 mSvy-^{1} and 0.17 mSvy-^{1} with an average value of 0.06 mSvy-^{1} and varies from 0.02 mSvy-^{1} to 0.66 mSvy-1 with an average value of 0.23 mSvy-1 in soil samples. Both are lower than unity that keeps the radiation hazard insignificant in the studied area.

The values of the representative level index are ranged from 0.06 to 2.21 with an average value of 0.75 for the soil samples which are lower than the recommended value unity. The hazard index (external and internal) associated from exposure of these radionuclides varies from 0.02 to 0.81 with an overall average value of 0.27 and varies from 0.03 to 0.81 with an average of 0.29 respectively in soil samples. The mean values are lower than the recommended maximum value of unity in the soil samples [17]. Statistical analysis such as Pearson Correlation and ANOVA (Analysis of Variance) has been carried out in order to find the extent of the existence of these radioactive nuclides in the studied area. Positive relationship between ^{232}Th and 40K indicates that they may have the same origin but their behavior in the environment differs. Weak correlations between the ^{238}U and $_{232}$Th radionuclides explain that ^{238}U radionuclide may not be related to ^{232}Th bearing heavy minerals. A negative correlation (r= -0.50) is observed between the 238U and ^{232}Th radionuclides in the sub-urban region. An ANOVA test is performed to analyze the differences among the radionuclides shows that there is significant statistical difference among the average values in the sub-urban region.

Table 1: Activity concentration (Bqkg-1) of ^{238}U, ^{232}Th and 40 K in the soil samples

Sample	Activity (Bq/kg)			
	^{238}U	^{232}Th	^{40}K	^{Rae}q
Vm1	1.18* ± 0.06	113.13 ± 7.9	911.55 ± 54.6	233.15
Ku1	1.28* ± 0.06	156.58 ± 12.5	954.63 ± 76.3	298.70
Vr1	0.39* ± 0.01	127.71 ± 7.6	5.20* ± 0.25	183.42
Ni1	3.23 ± 0.19	1.79 ± 0.08	23.41 ± 1.63	7.60
Mu1	0.65 ± 0.03	20.57* ± 1.23	258.28 ± 15.4	49.95
Pi1	1.10* ± 0.07	18.87* ± 0.94	779.11 ± 38.9	88.08
Vlu1	14.36 ± 1.14	2.00 ± 0.09	0.93* ± 0.04	17.29
Vam1	9.75 ± 0.67	26.63 ± 2.39	5.70* ± 0.33	48.26
Mlm1	1.12* ± 0.06	19.27* ± 1.34	605.52 ± 48.4	75.30
Ti1	10.36 ± 0.72	1.55* ± 0.07	24.38 ± 1.70	14.45
Average	4.34 ± 0.30	48.8108 ± 3.4	356.87 ± 23.7	101.62

* MDA - Minimum Detectable Activity

Table 2: Radiation hazard parameters in the soil samples

Sample	D (nGyh-^{1})	AEDE (mSvy-^{1})		H_{ex}	H_{in}	Iγr	ELCR	
		Outdoor	Indoor				Outdoor	Indoor
Vm1	106.89	0.13	0.52	0.63	0.63	1.75	0.46	1.84
Ku1	134.97	0.17	0.66	0.81	0.81	2.21	0.58	2.32
Vr1	77.54	0.10	0.38	0.50	0.50	1.28	0.33	1.33
Ni1	3.55	0.00	0.02	0.02	0.03	0.06	0.02	0.06
Mu1	23.50	4	0.12	0.13	0.14	0.38	0.10	0.40
Pi1	44.40	0.03	0.22	0.24	0.24	0.72	0.19	0.76
Vlu1	7.88	0.05	0.04	0.05	0.09	0.15	0.03	0.14
Vam1	20.82	0.01	0.10	0.13	0.16	0.33	0.09	0.36
Mlm1	37.41	0.03	0.18	0.20	0.21	0.60	0.16	0.64
Ti1	6.74	0.05	0.03	0.04	0.06	0.100	0.03	0.12
Average	46.37	0.01 0.06	0.23	0.27	0.29	0.75	0.20	0.80

Table 3: ANOVA result of radionuclides (238U, 232Th and 40 K) for soil samples in the sub-urban region

Source	SS*	df*	MS*	F*	p-value*
Between groups	737184.9	2	368592.4	6.453	0.005
Within groups	1542165	27	57117.21		
Total	2279350	29			

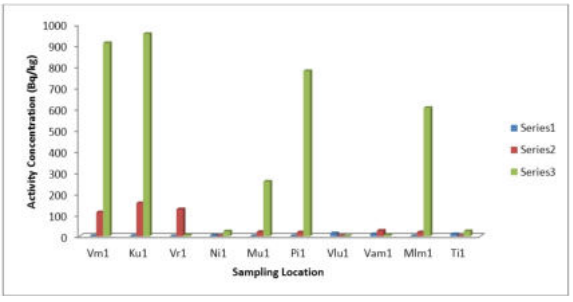

Figure 2: Activity concentration of 238U, 232Th and 40 K at different sampling location in the sub-urban region

Conclusion

The level of naturally occurring radioactivity in soil samples collected from the southern region of Tamil Nadu is evaluated using NaI(Tl) gamma ray spectrometry. The results show that 238U concentration varies from 0.39±0.01 Bq/kg to 14.36±1.14 Bq/kg with an average of 4.34±0.30 Bq/kg. The 232Th concentration ranges from 1.55±0.07 Bq/kg to 156.58±12.5 Bq/kg with an average of 48.81±3.4 Bq/kg whereas 40K ranges between 0.93±0.04 Bq/kg and 954.63±76.3 Bq/kg with an average of 356.87±23.7 Bq/kg. The measured activity concentrations of 40K contributed the most in the soil samples. The present study reveals that the activity values obtained are compared with mean values identified by UNSCEAR (2000). Also, the results can be useful in the assessment of the radiological effects. Statistical analysis such as Pearson Correlation and ANOVA (Analysis of Variance) are performed to determine the relationship between the samples. These results would be useful for establishing a reference data on the gamma background radiation levels in the studied region and represent a basis to assess any further changes in the radioactivity background levels due to various geological formations around the area under considerations. It is observed that the study area is free from radiation hazards. The radiological hazard parameters for all the soil samples are lower than the recommended limit indicates that the investigated areas are safe for human health.

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