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Activities of Primordial Radionuclides in the tobacco cultivated fields of Dindigul and Erode districts (Tamil Nadu, India)

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KEYWORDS

Primordial tobacco field Uranium Thorium Potassium

ABSTRACT:

Present study is a novel attempt to estimate the activities of primordial radionuclides such as ²³⁸U (Uranium-238), ²³²Th (Thorium-232) and ⁴⁰K (Potassium-40) in the soil samples collected from tobacco plantations in Dindigul and Erode Districts. Radioactivity concentrations of the collected soil samples were determined with the aid of a NaI gamma-ray detector, which ranged from BDL (2 Bq/kg) to 20.3 Bq/kg for ²³⁸U, 5.1 to 156.5 Bq/kg for ²³²Th and 87.1 to 712.5 Bq/kg for ⁴⁰K respectively with mean value of 6.41, 54.34 and 296.96 respectively. Frequency distribution of the radionuclides showed the leptokurtic, platykurtic and mesokurtic for ²³⁸U, ²³²Th and ⁴⁰K respectively. The average activity concentrations of primordial radionuclides in the tobacco field soils of Dindigul and Erode Districts were compared to the published global average values. The estimated primordial activity concentration from the current work is lesser than that of the average global values.

1. Background

Primordial radionuclides are released from the terrestrial radiation sources such as earth's crust, rocks, soil, air and water [1]. Radiations are the spontaneous emission of energy from unstable atoms [2]. Natural and anthropogenic radionuclides are a crucial requirement for the evaluation and control of public exposures [3]. Cosmogenic radionuclides arise as a result of the interaction of cosmogenic radiation with elemental substances in the atmosphere [4]. Anthropogenic activity can also be considered as a ubiquitous source of radioactivity which contributes to the background radiation level [5-6]. Natural gamma radiation that occurs in the environment is created by primordial radionuclides from the sky and on Earth [7]. Naturally occurring radionuclides in the soil significantly increase the population's background radiation exposure [8]. The primary external source of irradiation in humans is gamma radiation, which is emitted by naturally occurring radionuclides that are deposited on the ground.

Distribution of natural radiation level depends upon multiple factors such as the place, elevation, latitude as well as the higher abundance of radioactive minerals [9]. The primary sources of naturally occurring radioactivity in soil are the ²³⁸U, ²³²Th decay series and natural ⁴⁰K respectively [10-11]. The uranium and thorium series, which are respectively descended from the ²³⁸U and ²³²Th series [12] have average continental crust levels of 2.7ppm and 9.6ppm, respectively [13]. ²³⁸U and ²³²Th are fission products produce 12 and 11 daughter products and ⁴⁰K is non fission product [2]. The presence of Uranium and Thorium due to fertilizers enhances the natural radiations [14]. U, Th, and K are released into the soil by the progeny of U and Th as well as by the

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weathering of bedrock, which is the main repository of the primordial radionuclides. Three isotopes of potassium exist in nature (³⁹K, ⁴⁰K and ⁴¹K). Only ⁴⁰K, exhibits gamma radioactivity, and it makes up 0.012% of all potassium in nature. The daughter isotopes ⁴⁰Ca and ⁴⁰Ar are produced during the decay of ⁴⁰K, along with beta and gamma radiations. One of the reasons for the presence of high activity in soil is the presence of potassium-containing fertilisers, which have a significant impact on radionuclide concentrations [15]. The present study focused on the distribution of natural radioactivity levels in the surface soils of tobacco cultivated fields which absorbed external gamma radiations from the soil sample of selected stations.

2. Methods

2.1. Location of Study

Location of the study includes Dindigul (11°30.870'N, 077°57.722'E) and Erode (10°32.256'N, 077°57.039'E) districts, Tamil Nadu (India) (Fig. 1). 23 different sites of tobacco cultivated fields comprised of 5000 hec are selected for this study (Table 1). The distance between each site is kept as 3-6 km.

Table 1. GPS Cordinates of the sampling locations

No.	Location	GPS Cordinates			
		Latitude	Longitude		
S1	Vedachandur	077°57.039'E	10°32.256'N		
S2	Lashmanampatt i	077°57.582'E	10°32.740 'N		
S3	Kalanampatti	077°57.988'E	10°32.730'N		
S4	Idayakottai	077° 53.523'E	10°31.436,N		
S5	Puliyurnatham	077°50.025'E	10°32.002'N		
S6	Javathupatti	077° 51.053'E	10° 31.350'N		
S7	Odaipatti	077°47.095'E	10°35.232 'N		
S8	Thangachiamm	077°42.025'E	10°29.232 'N		
	apatti				
S9	Mylambaddi	077°40 .722'E	11° 30.870'N		
S10	Kuruchi	077°41.564'E	11°34.031'N		
S11	Poonachi	077°39.397'E	11°36.294'N		
S12	Olagadam	077°41.633'E	11°33.740 'N		
S13	Kalpavi	077°38.471'E	11°34.151'N		
S14	Alukuzhi	077°21.379'E	11°26.751'N		
S15	Karattupalayam	077°21.353'E	11°26.906'N		
S16	Kurumanthur	077°20.874'E	11°24.815'N		
S17	Nambiyur	077°19.313'E	11°21.679'N		
S18	Elathur	077°18.430'E	11°23.293'N		

S19	Varapalayam	077°13.958'E	11°22.344'N
S20	Kavillipalayam	077°13.885'E	11°23.186'N
S21	Karapaddi	077°12.057'E	11°22.741'N
S22	Nallur	077°08.377'E	11° 30.870'N
S23	Puliyampatti	077°57.039°E	10°32.256'N

2.2. Soil collection and processing

Soil samples (1000-2000 g) of tobacco cultivated fields was collected from the surface regions (0-5 cm depth) during the tobacco cultivated seasons (December to April). Collected soils dried at room temperature for 48 hours and then stored in black polythene bags. Before processing, the samples are again dried at 110°C using hot air oven, to destroy inorganic and organic compounds and moisture. Then the samples were stored in plastic containers (9×6.5cm). After four weeks, samples are attained secular equilibrium between R-226 and its shortlived daughter products. The net weights of the samples are determined before counting.

2.3. Gamma ray spectrometry

The concentration of primordial radionuclides (²³²Th, ²³⁸U and ⁴⁰K) in soil samples is measured using a gamma ray spectrometer with a NaI (TI) detector. The energy resolution (2.0 Kev) with 33% relative efficiency at 1.33 Mev and the counting time 20000 seconds were always ensured prior to the analysis following the calibration with AERB approved sources. A computer programme is used to calculate the activity concentration ²³²Th, ²³⁸U and ⁴⁰K from the counting spectra. The peak corresponds to 1400Kev (K-40) for ⁴⁰K, 1764.54 Kev (Bi-214) for ²³⁸U and 2614.5Kev (Ti-208) for ²³²Th are considered in arriving at the activity levels (Bq/kg).

2.4. Radium Equivalent Activity

Since the activity concentrations of the primordial radionuclides such as 238 U, 232 Th and 40 K in the soil samples, are not uniform and range vary widely, there is some practical difficulty for comparing the gamma radiations emitted from primordial radionuclides from soil samples of various locations. Therefore, the concept of Radium Equivalent (Req) was introduced [16-17]. The total exposure to gamma radiation from the primordial radionuclides has been defined in terms of Raeq activity in Bq/kg. It is calculated on the assumption that 370 Bq/kg 226 Ra or 259 Bq/kg 232 Th or 4810 Bq/kg of 40 K produce same gamma dose rate. Therefore, Raeq of any

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sample can be calculated using the following formula [18].

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

Where A_{Ra} , A_{Th} and A_{K} are the specific activities of ^{226}Ra , ^{232}Th and ^{40}K respectively. The safe value of Ra_{eq} in any environmental matrix is reported to be less than 370 Bq/kg in order to limit the annual effective dose to 1 mSv for the general public [19].

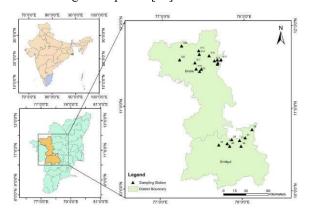


Fig. 1 showing the sampling locations (tobacco cultivated fields)

2.5. Determination of Absorbed Dose Rate

The absorbed gamma dose rate (D), is defined as the amount of energy deposited in a unit mass in human tissue or other media. It is expressed by the SI unit as Gray/kg (Gy/kg) and 1 Gray= 1 Joule. The absorbed dose rate (D) in nGy/h due to terrestrial gamma radiation at 1 m above the ground surface has been computed from the specific activities of ²²⁶Ra, ²³²Th and ⁴⁰K respectively [20].

$$D (nGy/h) = 0.462A_U + 0.604 A_{Th} + 0.0417A_K$$

Where 0.462, 0.604 and 0.0417 nGy/h per Bq/kg are the dose conversion factor for 238 U, 232 Th and 40 K, respectively and A_U , A_{Th} and A_K are the activity concentration of 226 Ra, 232 Th and 40 K respectively.

2.6. Determination of Annual Effective Dose Rate

Effective dose is defined as dose equivalent weight to express the equivalent sensitivity of different human organs to radiation exposure. The unit for effective dose is Sievert (Sv).

Effective dose rate (mSv/y) = D (nGy/h)
$$\times$$
 8760 (h/y) \times 0.2 \times 0.7 (Sv/Gy) \times 10⁻⁶

To estimate the annual effective dose rate, the conversion coefficient from absorbed dose in air to effective dose 0.7 Sv/Gy and the indoor occupancy factor of 0.8 and the outdoor occupancy factor of 0.2 were used [20].

2.7. Radiological Hazard Indices (Hex and Hin)

Beretka and Mathew [19] described two types of hazard index namely external hazards index and internal hazard index. The external hazards index indicates whether the absorbed gamma radiation exceed the permissible limit of unity (<1). Similarly, the internal hazard index is useful in assessing whether alpha radiation from ²³⁸U and its progenies exceeds the maximum permissible limit of unity (<1). The external hazards index is obtained from Raeq expression.

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

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

2.8. Statistical analysis and spatial mapping

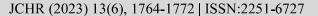
Statistical analysis is performed for the obtained results using the SPSS software (Version 17.0). Skewness, kurtosis, frequency distribution and histogram are used to analyse the results. Spatial analysis of the obtained results was conducted by means of IDW (Inverse distance weighted) interpolation technique and Arc GIS Desktop software (Version 10.5) is used for spatial mapping.

3. Results

3.1. Primordial Radionuclides

The concentrations of primordial radionuclides such as ²³⁸U, ²³²Th and ⁴⁰K in the tobacco cultivated soils of Dindigul and Erode districts varied very widely. The ²³⁸U concentration ranged from Below Detection Limit (BDL) to 20.3 Bg/kg, ²³²Th from 5.1 Bg/kg to 156.6 Bq/kg and ⁴⁰K from 87.1 Bq/kg to 712.5 Bq/kg and mean values of ²³⁸U, ²³²Th and ⁴⁰K were 6.4 Bq/kg, 54.3 Bq/kg, and 296.9 Bq/kg respectively. Puliyurnatham (S5) registered maximum level of three primordial radionuclides ²³⁸U (20.3 Bq/kg), ²³²Th (156.3 Bq/kg) and ⁴⁰K (712.5 Bg/kg) whereas the minimum levels of ²³⁸U (BDL Bq/kg) and 232 Th (5.1 Bq/kg) were recorded in Nambiyur (S17) and Kurumanthur (S16) registered minimum level of ⁴⁰K (87.1 Bq/kg). Spatial distribution of primordial radionuclides confirmed non-uniform distribution (Figure 2-4).

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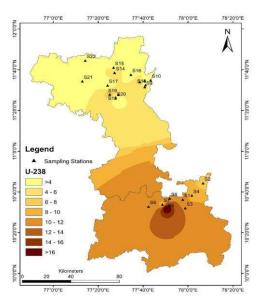


Fig. 2: ²³⁸ U in tobacco cultivated fields soil samples

In Figure 5, histogram with positively skewed (1.164) kurtosis with leptokurtic curve showed for the concentrations of ²³⁸U whereas for ²³²Th, histogram with positively skewed (0.845) kurtosis with platykurtic curve were observed and histogram with normally skewed (0.639) kurtosis with mesokurtic curve for ⁴⁰K were observed.

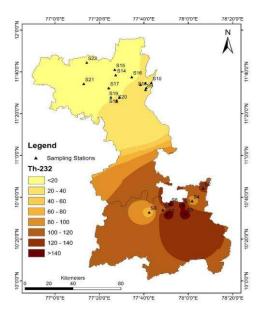


Fig. 3: ²³²Th in tobacco cultivated fields soil samples

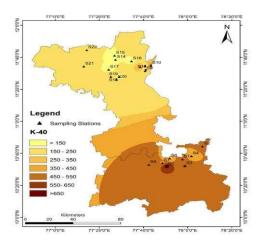


Fig. 4: ⁴⁰K in tobacco cultivated fields soil samples

3.2. Radium Equivalent Activity

The data on the radium equivalent activities in 23 soil samples are presented in Table 2. The maximum Ra_{eq} activity was recorded (298.9 Bq/kg) in Puliyurnatham (S5) in Dindigul district and minimum of 13.6 Bq/kg was found in Nambiyur (S17). It was observed that the mean Raeq activity in Dindigul district (222.1 Bq/kg) was about 5 times higher than the mean Ra_{eq} activity in Erode district (45.6 Bq/kg).

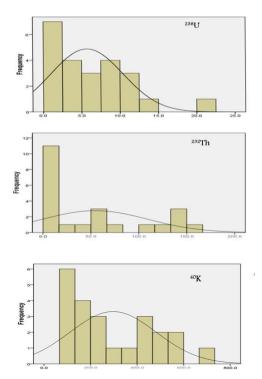


Fig. 5: Frequency distributions of the radionuclides

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3.3. Absorbed Dose Rate, Radiological Hazard Indices

The data on absorbed dose rate, annual effective dose rate and hazard indices (External and Internal Hazard) are presented in Table 2. The absorbed dose rate calculated for 23 sampling stations ranged from 8.9 nGy/h to 133.8 nGy/h with mean values of 48.5 nGy/h. The maximum absorbed dose rate was recorded in the sampling station, S5 (Puliyurnatham) in Dindigul district. Conversely, and the minimum absorbed dose rate in the sampling station S17 (Nambiyur) in Erode district. The same trend was observed with reference to annual effective dose rate. The maximum annual effective dose rate of 0.1 mSv/year was registered in several sampling stations of Dindigul district. However, most of sampling stations in Erode district registered annual effective dose rate of about 0.01 mSv/year. The mean annual effective dose rate for all the sampling stations was 0.04 mSv/year. Both the external hazard and internal hazard indices ranged from 0.05 to

0.86 with mean value of 0.3. It was also observed that the hazard index did not exceed unity in any of the sampling station

4. Discussion

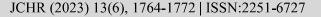
The activity concentrations of primordial radionuclides such as ²³⁸U, ²³²Th and ⁴⁰K were measured in soil samples from Dindigul and Erode districts of Tamil Nadu as the tobacco plants are largely cultivated in these areas. The measurements of radioactive substances in the tobacco cultivated soil assume importance to evaluate the accumulation of radionuclides in tobacco leaves which go into the manufacturing of chewing tobacco and bidis for smoking. Further the measurements are important in the assessment of population exposure to gamma radiation from the soils. Since radiation from natural sources constitutes 96% of total radiation dose to the world population [11, 21], knowledge of concentration of primordial radionuclides becomes essential for the assessment of possible radiological risk to human health.

Table 2. Activity Concentrations of ²³⁸ U, ²³² Th and ⁴⁰ K and Radium Equivalent Activity (Ra_{eq}), Absorbed Dose Rate, Annual Effective Dose Rate, External Hazard (H_{ex}) and Internal Hazard(H_{in}) in Soil Samples from Tobacco Field

Sampling Station	Concentra	ntion of Radion	nuclides (Bq/kg)	Ra _{eq} (Bq/kg)	(nGy h ⁻¹)	Annual Effective Dose Rate (mSv y-1)	Hazard Indi	ces
Station	²³⁸ U	²³² Th	⁴⁰ K				External Hazard (H _{ex})	Internal Hazard (H _{in})
Dindigul D	istrict	· · · · · · · · · · · · · · · · · · ·	•			<u> </u>		
S1	11.7±3.8	146.1±8.9	541.8±33.7	262.3	116.4	0.1	0.709	0.739
S2	6.9±3.7	122.6±8.6	491.2±33.4	220.0	97.86	0.1	0.594	0.613
S3	12.0±3.7	147.0 ±8.6	555.1±32.5	265.5	117.63	0.1	0.716	0.751
S4	8.2±3.5	82.3±7.9	363.9±31.7	153.8	68.76	0.08	0.416	0.151
S5	20.3±4.0	156.5±9.2	712.5±35.8	298.8	133.81	0.1	0.807	0.862
S6	11.3±3.9	143.8±9.1	316.7±34.0	241.2	105.37	0.1	0.658	0.682
S7	8.4±3.6	107.8±8.3	433.6±32.0	195.8	87.2	0.1	0.529	0.558
S8	10.5±3.5	65.8±7.8	460. ±32.6	139.9	69.91	0.08	0.378	0.406
Range	8.2-20.3	65.8-156.5	316.7-712.5	139.9-298.8	68.76-133.81	0.08-0.1	0.378-0.807	0.151-0.862
Mean	11.2	121.4	484.3	222.16	99.62	0.095	0.601	0.623
SD±	4.15	33.3	123.1	55.8	23.30	0.009	0.151	0.306

Erode District								
S9	4.9±3.5	25.9±7.6	217.7±33.0	57.3	27.04	0.03	0.159	0.172
S10	3.9±3.2	36.2±7.1	521.6±31.4	95.7	45.56	0.05	0.259	0.459
S11	BDL	10.4±7.7	102.9±33.0	24.2	11.33	0.01	0.066	0.071

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S12	8.0±3.6	51.7±7.9	435.5±33.8	115.4	53.2	0.06	0.212	0.332
S13	BDL	8.3±6.9	101.1±30.2	20.2	10.0	0.01	0.058	0.062
S14	5.3±3.6	16.6±7.8	132.6±33.8	39.2	18.02	0.02	0.106	0.121
S15	3.7±2.9	11.7±6.9	133.7±27.5	30.6	14.37	0.01	0.082	0.089
S16	BDL	8.2±7.2	87.1±31.1	19.9	9.2	0.01	0.053	0.058
S17	BDL	5.0±4.1	119.8±27.1	13.6	8.92	0.01	0.050	0.055
S18	8.7±3.9	6.7±4.9	234.0±36.7	36.2	17.87	0.02	0.098	0.122
S19	4.3±3.2	13.2±6.9	117.2±29.8	33.02	14.87	0.01	0.087	0.099
S20	BDL	54.9±7.38	194.9±29.8	95.1	42.11	0.05	0.258	0.263
S21	BDL	9.7±7.7	153.6±32.9	27.5	12.94	0.01	0.073	0.078
S22	BDL	10.6±7.9	192.7±34.7	31.9	15.36	0.01	0.086	0.090
S23	7.0±3.5	9.0±7.6	211.5±33.7	36.08	17.54	0.02	0.098	0.117
Range	2-87	5-54.9	87.1-521.6	13.6-115.6	8.92-53.2	0.01-0.06	0.05-0.259	0.055-0.459
Mean	3.84	18.54	197.6	45.6	21.222	0.022	0.116	0.146
SD±	2.49	16.25	124.4	31.47	14.218	0.017	0.0714	0.117
Over all	BDL-20.3	5.1-156.5	87.1-712.5	13.6-298.8	8.92-133.81	0.01-0.1	0.050-0.807	0.055-0.862
Range	DDL-20.3	3.1-130.3	07.1-712.5	13.0-290.0	0.72-133.01	0.01-0.1	0.030-0.807	0.033-0.802
Mean±SD	6.41±4.73	54.34±55.10	296.96±185.7	106.7±95.1	48.49	0.039	0.285	0.303

Gamma radiation levels are inversely correlated with cosmic ray activity and radionuclide concentrations in soil samples from tobacco cultivation [22]. The primordial radionuclides are long lived radionuclides are such as ²³⁸Uranium, ²³²Thorium and ⁴⁰Potassium present in the environment factor such as soil, rocks, sediment [10]. The average primordial radionuclides levels in the present study were found as 87.0±4.0, 98.0±4.0 and 1254.00 ± 62.0 Bq/kg for ²³⁸U, ²³²Th and ⁴⁰K respectively. Due to the heterogeneous soil properties, present study has recorded differed ⁴⁰K activity and is varied greatly between 200 and 854 Bq/kg. Activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in soil samples from the different parts India and world over were reported by several investigators. The data generated in the present study are compared with results of other investigators (Table 3). The generated data on the analysis of primordial radionuclides indicated that the distribution of ²³⁸U, ²³²Th and ⁴⁰K was not uniform and ranged widely. In general, the concentration of 40K was distinctly higher than that of both ²³⁸U and ²³²Th. But the concentration of ²³²Th was always higher than that of ²³⁸U, in all soil samples. When the activity concentrations of primordial radionuclides are converted in to Radium Equivalent (Req) value, a distinct variation in the distribution of the primordials in the soil of Dindigul and Erode districts was observed. It was evident from radium equivalent data that the mean radium equivalent activities in Dindigul district

(222.Bq/kg), was about 5 times higher than the mean radium equivalent activity in Erode district (46 Bq/kg).

Table 3. Comparison of Concentration Radionuclides²³⁸U, ²³²Th and ⁴⁰K in Soil Samples from Different Parts of the World and India

Nation/ State	Conce	ntration	Reference	
	Radionuclides (Bq/kg)			
	²³⁸ U	²³² Th	⁴⁰ K	
Dindigul and	6.41	54.34	296.96	Present
Erode districts				study
(Tamil Nadu)				
Ireland	26.0	37.0	350.0	[24]
Turkey	21.0	37.0	342.0	[25]
(Istanbul)				
South Africa	26.4	32.2	115.0	[26]
Turkey	16.52	17.38	209.0	[27]
China	40 .3	59.6	751.2	[28]
Jordan	284.5	16.5	146.5	[29]
Hong Kong	21.33	21.16	290.0	[30]
Nigeria	39.70	46.81	603.62	[31]
World average	35.0	45.0	420.0	[20]
Indian studies				
Tiruchirappalli	6.50	61.7	380	[32]
(Tamil Nadu)				
Perambular	13.2	66.0	340	[33]
(Tamil Nadu)				
Western Ghats	36.3	107.8	231	[34]

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(Tamil Nadu)				
Punjab and	56.7	87.4	143	[35]
Himachal				
Pradesh				
Sirsa	27.9	72.5	286	[36]
(Haryana)				
Kotagiri	48.8	102	229	[37]
(Tamil Nadu)				
Kalpakkam	16.0	119	40 6	[38]
Coast (Tamil				
Nadu)				
Ramanagara	33.7	77.4	791	[39]
and Tamkur				
(Karnataka)				
Visakhapatnam	38.0	230	520	[40]
(Andhra				
Pradesh)				

The level of natural background radiation in a soil sample is influenced by geological and geographic factors, uranium mineralization and leaching within the earth crust, and chemical and biochemical patterns of uranium, thorium, and their decay products [19]. Another significant factor is the contribution of rocks with high thorium and uranium concentrations [20].

It was observed that higher concentrations of ²³⁸U and ²³²Th were found in the Oddanchittram taluk of Dindigul district. Similarly, in Gobhichettipalaym taluk of Erode district registered higher concentration of ²³⁸U and ²³²Th as compared to other taluks. As regards to ⁴⁰K, the distribution was found to be uniformly higher than ²³⁸U and ²³²Th in both districts. However, Dindigul district registered an elevated level of primordial radionuclides than the Erode district. The higher level of the primordial radionuclides in Dindigul district may be attributed to the rocky environment of the district. Rocks are primary sources of radioactive substances and soil was formed by the weathering of rocks. The geomorphology and lithology characters of Dindigul district may have a bearing on the elevated levels of primordial radionuclides. Dindigul district is naturally enriched with granite rocks (igneous rocks) having higher activity of primordial radionuclides. The enriched of monazite in igneous rocks, which was the source of ²³²Th, was responsible for the higher radioactivity level [23].

The data on activity concentration 238 U, 232 Th and 40 K in the soil samples of Dindigul and Erode districts were

used for the calculation of gamma radiation dose of hazard indices. The mean absorbed gamma dose rate for the soil samples analyzed was found to 48.5 nG/h which is comfortably less than world average of 54 nGy/h. The mean annual effective dose calculated for the soil was found to be 0.04 mSv/year which is well within the safety limit 0.07mSv/year [20]. Similarly, the mean external hazard index (0.29) and internal hazard index (0.30) registered for the soil samples are very well below permissible level unity (< 1). From the analysis of data, it is evident that the gamma activity levels from the soil of Dindigul and Erode districts did not provide any radiological risk to human population of two districts and these results supports our earlier findings [41-42].

4. Conclusion

As a novel attempt, the activity concentration of ²³²Th, ²³⁸U and ⁴⁰K in Dindigul and Erode districts have been evaluated from the soil samples collected at various tobacco cultivated fields. The activity concentration of ²³²Th in soil has found to be 6 times lower than the average world concentration (30 Bq/kg) and the activity concentration of ⁴⁰K has found to be 0.7 times lesser than the average world concentration (400 Bq/kg) and for ²³⁸U is found to be 6 times lesser than the world average (35 Bq/kg). Present study concluded that the concentrations of ²³²Th, ²³⁸U and ⁴⁰K in all sampling stations are low in level and are found below the World's and India's average values.

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