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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 ^{238}U (Uranium-238), ^{232}Th (Thorium-232) and ^{40}K (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 ^{238}U, 5.1 to 156.5 Bq/kg for ^{232}Th and 87.1 to 712.5 Bq/kg for ^{40}K 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 ^{238}U, ^{232}Th and ^{40}K 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 ^{238}U, ^{232}Th decay series and natural ^{40}K respectively [10-11]. The uranium and thorium series, which are respectively descended from the ^{238}U and ^{232}Th series [12] have average continental crust levels of 2.7ppm and 9.6ppm, respectively [13]. ^{238}U and ^{232}Th are fission products produce 12 and 11 daughter products and ^{40}K 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 (^{39}K, ^{40}K and ^{41}K). Only ^{40}K, exhibits gamma radioactivity, and it makes up 0.012% of all potassium in nature. The daughter isotopes ^{40}Ca and ^{40}Ar are produced during the decay of ^{40}K, 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 (11º30.870'N, 077º57.722'E) and Erode (10º32.256'N, 077º57.039'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	077º57.039'E	10º32.256'N
S2	Lashmanampatti	077º57.582'E	10º32.740 'N
S3	Kalanampatti	077 º 57.988'E	10º32.730'N
S4	Idayakottai	077º 53.523'E	10º31.436,N
S5	Puliyurnatham	077º50.025'E	10º32.002'N
S6	Javathupatti	077º 51.053'E	10º 31.350'N
S7	Odaipatti	077º47.095'E	10º35.232 'N
S8	Thangachiammapatti	077º42.025'E	10º29.232 'N
S9	Mylambaddi	077º40 .722'E	11º 30.870'N
S10	Kuruchi	077º41.564'E	11º34.031'N
S11	Poonachi	077º39.397'E	11º36.294'N
S12	Olagadam	077º41.633'E	11º33.740 'N
S13	Kalpavi	077º38.471'E	11º34.151'N
S14	Alukuzhi	077º21.379'E	11º26.751'N
S15	Karattupalayam	077º21.353'E	11º 26.906'N
S16	Kurumanthur	077º20.874'E	11º24.815'N
S17	Nambiyur	077º19.313'E	11º21.679'N
S18	Elathur	077º18.430'E	11º23.293'N
S19	Varapalayam	077º13.958'E	11º22.344'N
S20	Kavillipalayam	077º13.885'E	11º 23.186'N
S21	Karapaddi	077º12.057'E	11º22.741'N
S22	Nallur	077º08.377'E	11º 30.870'N
S23	Puliyampatti	077º57.039'E	10 º 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 short-lived daughter products. The net weights of the samples are determined before counting.

2.3. Gamma ray spectrometry

The concentration of primordial radionuclides (^{232}Th, ^{238}U and ^{40}K) 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 ^{232}Th, ^{238}U and ^{40}K from the counting spectra. The peak corresponds to 1400Kev (K-40) for ^{40}K, 1764.54 Kev (Bi-214) for ^{238}U and 2614.5Kev (Ti-208) for ^{232}Th 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 (R_{eq}) was introduced [16-17]. The total exposure to gamma radiation from the primordial radionuclides has been defined in terms of Ra_{eq} 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, Ra_{eq} 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].

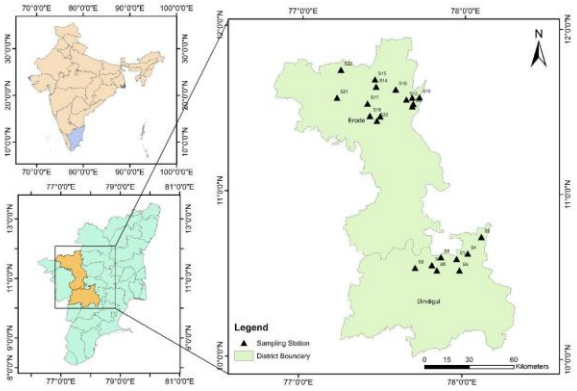

Fig. 1 showing the sampling locations (tobacco cultivated fields)

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 ^{226}Ra, ^{232}Th and ^{40}K 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).

$$\text{Effective dose rate (mSv/y)} = D\ (nGy/h) \times 8760\ (h/y) \times 0.2\times 0.7\ (Sv/Gy) \times 10^{-6}$$

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 (H_{ex} and H_{in})

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 ^{238}U and its progenies exceeds the maximum permissible limit of unity (<1). The external hazards index is obtained from Raeq expression.

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

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

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

Journal of Chemical Health Risks
www.jchr.org
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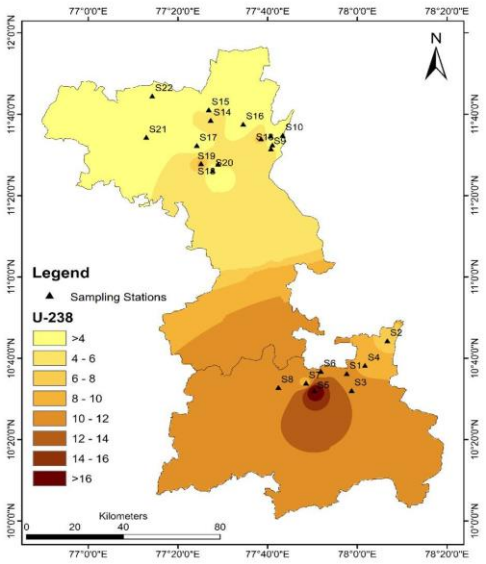

Fig. 2: ^{238}U in tobacco cultivated fields soil samples

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

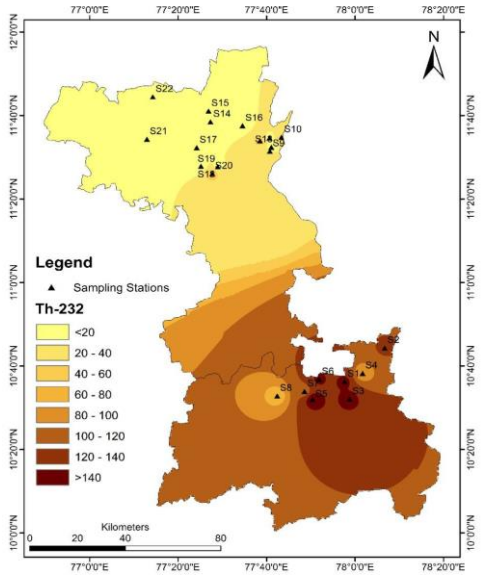

Fig. 3: ^{232}Th in tobacco cultivated fields soil samples

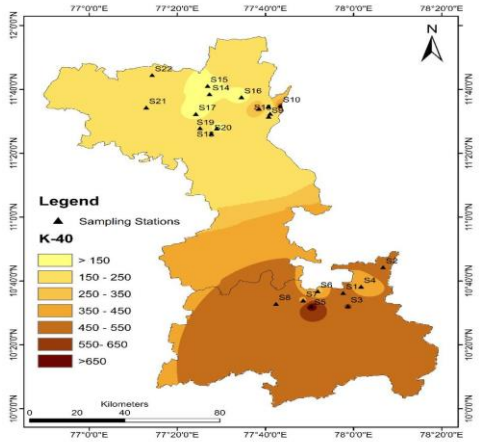

Fig. 4: ^{40}K in tobacco cultivated fields soil samples

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).

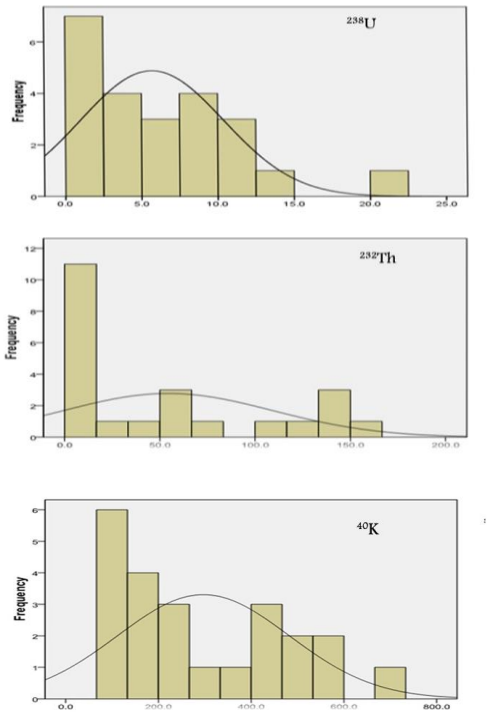

Fig. 5: Frequency distributions of the radionuclides

Journal of Chemical Health Risks

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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 ^{238}U, ^{232}Th and ^{40}K 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	Concentration of Radionuclides (Bq/kg)			Ra_{eq} (Bq/kg)	Absorbed Dose Rate (nGy h^{-1})	Annual Effective Dose Rate (mSv y^{-1})	Hazard Indices	
	^{238}U	^{232}Th	^{40}K				External Hazard (H_{ex})	Internal Hazard (H_{in})
Dindigul District								
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

Journal of Chemical Health Risks

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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 Range	**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**
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 ^{238}Uranium, ^{232}Thorium and ^{40}Potassium 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 ± 62.0Bq/kg for ^{238}U, ^{232}Th and ^{40}K respectively. Due to the heterogeneous soil properties, present study has recorded differed ^{40}K activity and is varied greatly between 200 and 854 Bq/kg. Activity concentrations of ^{238}U, ^{232}Th and ^{40}K 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 ^{238}U, ^{232}Th and ^{40}K was not uniform and ranged widely. In general, the concentration of ^{40}K was distinctly higher than that of both ^{238}U and ^{232}Th. But the concentration of ^{232}Th was always higher than that of ^{238}U, in all soil samples. When the activity concentrations of primordial radionuclides are converted in to Radium Equivalent (R_{eq}) 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 Radionuclides^{238}U, ^{232} Th and ^{40} K in Soil Samples from Different Parts of the World and India

Nation/ State	**Concentration of Radionuclides (Bq/kg)**			**Reference**
	^{238}U	**^{232}Th**	**^{40}K**	
Dindigul and Erode districts (Tamil Nadu)	**6.41**	**54.34**	**296.96**	**Present study**
Ireland	26.0	37.0	350.0	[24]
Turkey (Istanbul)	21.0	37.0	342.0	[25]
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 (Tamil Nadu)	6.50	61.7	380	[32]
Perambular (Tamil Nadu)	13.2	66.0	340	[33]
Western Ghats	36.3	107.8	231	[34]

Journal of Chemical Health Risks

www.jchr.org

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

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 ^{238}U and ^{232}Th were found in the Oddanchittram taluk of Dindigul district. Similarly, in Gobhichettipalaym taluk of Erode district registered higher concentration of ^{238}U and ^{232}Th as compared to other taluks. As regards to ^{40}K, the distribution was found to be uniformly higher than ^{238}U and ^{232}Th 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 ^{232}Th, 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 ^{232}Th, ^{238}U and ^{40}K in Dindigul and Erode districts have been evaluated from the soil samples collected at various tobacco cultivated fields. The activity concentration of ^{232}Th in soil has found to be 6 times lower than the average world concentration (30 Bq/kg) and the activity concentration of ^{40}K has found to be 0.7 times lesser than the average world concentration (400 Bq/kg) and for ^{238}U is found to be 6 times lesser than the world average (35 Bq/kg). Present study concluded that the concentrations of ^{232}Th, ^{238}U and ^{40}K in all sampling stations are low in level and are found below the World's and India's average values.

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