ISSN Print: 2394-7500
ISSN Online: 2394-5869
Impact Factor: 5.2
IJAR 2015; 1(9): 758-765
www.allresearchjournal.com
Received: 08-06-2015
Accepted: 11-07-2015

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Evaluation of radioactivity in soils of Chidambaram taluk of Cuddalore district (Tamil Nadu, India)

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Abstract
The main objective of the present study is to establish a data base on distribution of primordial radionuclides activity in the virgin and cultivated soil samples of Chidambaram taluk were carried out employing gamma ray spectrometer and Solid State Nuclear Track Detector (SSNTD) technique. The results indicated that, the mean activity concentration of ^{238}U in cultivated soil (7.1 Bq kg^{-1}) was 25 % higher than that of virgin soil (5.7 Bq kg^{-1}), while the mean ^{232}Th and ^{40}K concentration in cultivated soils were not elevated. The mean Radium equivalent (Ra_{eq}) value for virgin and cultivated soil samples were found to be 91.2 Bq $kg^{-1}$and 93.5 Bq kg^{-1}respectively. Application of phosphate fertilizers may the responsible for the elevated level of ^{238}U in cultivated soil. However, the mean absorbed gamma dose rate for soil samples are (42.2 nGy h^{-1} and 42.3 nGy h^{-1}) well below the permissible limit of 55 370 nGy h^{-1}. All other radiological parameters were also calculated and the study concludes that the study area falls under Normal Background Radiation Area (NBRA).

Keywords: Soil radioactivity, gamma ray spectrometry, Chidambaram, Dose assessment

1. Introduction
Natural radioactivity in soils and rocks comes from primordial radionuclides such as ^{238}U and ^{232}Th series and singly occurring natural ^{40}K. Hence, Human beings are continuously exposed to ionizing radiation and external gamma radiations in their homes and places of work (Damla *et al.* 2011) [3]. Therefore, assessment of gamma radiation dose from natural sources is of particular importance as natural radiation is the largest contributor to the external dose of the world population (UNSCEAR 2000) [21]. Terrestrial radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions and appear at different levels in the soils of each region in the world (UNSCEAR 2000) [21]. The radioactivity levels in soils are related to the types of rocks from which the soils originate (Kumar *et al*. 2003; Ramasamy *et al*. 2004)[11, 16]. Such investigations can be useful for both the assessment of public dose rates and the performance of epidemiological studies, as well as to keep reference–data records, in order to ascertain possible changes in the environment radioactivity due to nuclear, industrial, and other human activities. Environmental radioactivity measurement has taken out in many countries of the world (UNSCEAR 2000) [21]. The reports on natural radioactivity levels of soils and rocks in India are limited (Hameed *et al*. 2014; Jeevarenuka *et al*. 2014; Manigandan and Shekar 2014; Singh *et al*. 2005; Mehra *et al*. 2010; Selvasekarapandian *et al*. 2002) [7, 9, 12, 18, 13, 17]. Recently radioactivity in building materials of Tiruchirappalli district and related radiation exposure rate were reported by Pillai *et al*. (2014) [15]. The main objective of the present work is to estimate activity concentration of the primordial radionuclides and associated radiation dose received from soil samples collected from Chidambaram taluk of Cuddalore district. This study will provide a baseline data of natural background radiation level of the region, which is essential for understanding the future changes in natural background radiation of the region, as there are no earlier results. The results obtained in the present study are compared with the findings of other studies.

2. Materials and Methods
2.1. Study area
Chidambaram taluk (Lat: 11.39^0 and Long: 79.69^0) is a taluk of Cuddalore district of the Indian state of Tamil Nadu.

According to the 2011 census, the taluk of Chidambaram had a population of 4, 69, 416 with 2, 36, 170 males and 233,246 females. This taluk is located close to the shores of Bay of Bengal. The topography is almost plain with forests around the town, with no major geological formation. There are no notable mineral resources available in and around the town. The soil types are alluvial and red that are conducive for crops like paddy, pulses and chili peppers.

2.2. Sample collection

Fifteen samples of soils each from virgin fields (V1-V15) and cultivated (C1-C15) were collected from the chosen sampling stations of Chidambaram taluk. About 2 kg of the sample was collected from each sampling station. The solid matrix of the sample was powdered and sieved through 500 µm mesh. The sample was air-dried for several days and then oven dried at 110 ºC until they reached a constant weight. Each sample was placed in 250 ml plastic container having diameter of ~ 6 cm and height of ~ 9.5 cm. The container was sealed hermetically and externally using adhesive tapand kept aside for about a month to ensure secular equilibrium between radium and its daughter products before being taken for gamma ray spectrometric analysis.

2.3. Primordial radionuclides activity measurement

Gamma radiation measurements were carried outwith a ORTEC make (USA) digibase (Model:905-4) γ-ray spectrometer coupled with a 14-pin photo Multiplier Tube, 8K ADC stem, digital Multi Channel Analyzer (MCA),3" × 3"NaI detector, Pre amplifier, HV power supply and MAESTRO-32 software and USB connection (Figure 1). The detector was hermitically sealed and mounted in a massive lead shielding of 10cm thick to reduce to background radiation. In order to reduce the contribution of X-ray fluorescence, the inner surfaces of the lead shield is lined with copper sheet (0.8 mm thick) and cadmium sheet (1.5 mm thick). The standardreference materials RGU-1 (Uranium ore), RGTh-1 (Thorium ore) and RGK-1 (Potassium sulphate) having the activity of 1065 Bq, 1608 Bq and 4810 Bq respectively procured from International Atomic Energy Agency (IAEA) were used for checking the calibration of the system. These standards were obtained from Radiological Safety Division, Indira Gandhi Centre for Atomic Research, Kalpakkam. The sample container was placed on the top of the detector for counting period of 20,000 sec. The same geometry was used to determine peak area of samples and references. The background and samples were counted for a period of 20,000 seconds to minimize the counting error. The background measurement was subtracted in order to get net count for the sample. The concentration of ^{40}K was measured directly by its own gamma ray peak at 1461 keV, whereas ^{238}U and ^{232}Th were estimated with the help of their gamma emitting daughter products such as ^{214}Bi (1764 keV) and ^{208}Tl (2614.6 keV) respectively. The Below Detectable Limits (BDL) was 2.03 Bq $kg^{-1}$for ^{214}Bi, 4.7 Bq kg^{-1} for$^{208}Tl$and 18.9 Bq kg^{-1} for ^{40}K. The activity concentrations were calculated using the following equation prescribed by Ibrahim (1999) [8]

$$A\ (Bq\ kg^{-1}) = \frac{N_p}{\eta * e * m} \qquad (1)$$

Where N_p = net count rate (cps), measured count rate minus background count rate, *e* is the abundance of the γ-line in a radionuclide, *η* is the measured efficiency for each gamma-line observed for the same number of channels either for the sample or the calibration source, and m the mass of the sample in kilograms. The Below Detectable Limit (BDL) for each radionuclides were calculated using the formula Currie (1968) [4]

$$BDL\ (Bq\ kg^{-1}) = \frac{\sigma \sqrt{B}}{\eta * T} \qquad (2)$$

Where σ is the statistical coverage factor (σ = 4.65) with 95% confidence level, B is the background count for specific radionuclide, T is the counting time (sec).The Below Detectable Limits (BDL) for ^{214}Bi was 2.03 Bq kg^{-1}, for ^{208}Tl was 4.7 Bq kg^{-1} and for ^{40}K was 18.9 Bq kg^{-1}.

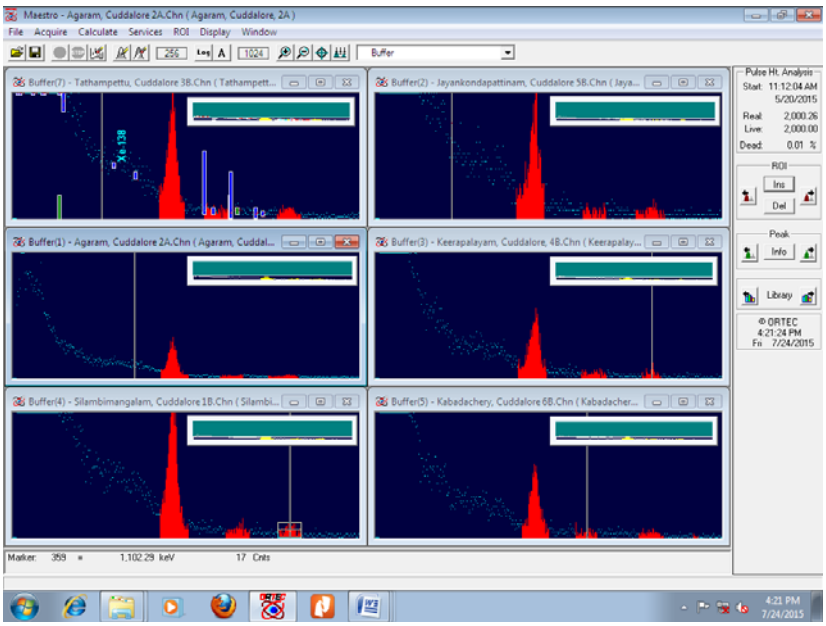

Fig 1: Spectrometric measurement of primordial radionuclides

3. Results and discussion

3.1. Primordial radionuclides

The activity concentrations of ^{238}U, ^{232}Th and ^{40}K in virgin soils of Chidambaram taluk were presented in Table 1 and in cultivated soil in Table 2. The results were graphically given in Figure 3 and 4 respectively.The activity concentration of ^{238}U in virgin soil varied from BDL (<2.03 Bq/kg) to 15.6 Bq kg^{-1} (V4) with the mean value of 5.7 Bq kg^{-1}, ^{232}Th from BDL (< 4.7 Bq kg^{-1}) (V10) to 299.2 Bq kg^{-1} (V13) with the mean value of 45.6 Bq/kg and ^{40}K from 69.2 Bq kg^{-1} (V5) to 532.6 Bq/kg (V10) with mean value of 290.6 Bq/kg.On the other hand, the activity concentration of ^{238}U in cultivated soils ranged from BDL (<2.0 Bq kg^{-1}) to 16.6 Bq/kg (C15) with the mean activity of 7.1 Bq kg^{-1}, ^{232}Th from 10.5 Bq kg^{-1} (C10) to77.5 Bq/kg (C1) with the mean value of 46.5 Bq $kg^{-1}$and ^{40}K from 87.3 Bq kg^{-1} (C7) to 388. Bq kg^{-1}in (C2) with the mean activity of 290.6 Bq kg^{-1}.

Table 1: Activity concentration of primordial radionuclides in virgin soils of Chidambaram taluk

Station code	Name of the station	Activity concentration (Bq kg^{-1})			Ra_{eq} (Bq kg^{-1})
		^{238}U	^{232}Th	^{40}K	
V1	Silambimangalam	BDL	40.9±7.0	393.4±31.1	84.6
V2	Agaram	BDL	31.0±7.4	445.4±333.6	64.9
V3	Tathampettu	BDL	31.3±8.4	239.4±35.0	66.4
V4	Keerapalayam	15.5±3.6	36.8±7.6	154.2±29.5	80.0
V5	Jayamkondapattinam	7.6±3.2	22.8±8.2	69.2±35.5	45.7
V6	Kabadacherry	12.3±3.0	63.1±8.3	275±32.0	123.8
V7	Periyapattu	BDL	17.3±6	292.8±28	49.5
V8	Sivapuri	BDL	48.7±8.3	353.9±35.0	96.9
V9	Kabarapattu	4.1±3.2	63.5±8.5	253.5±34.9	114.5
V10	Periyakumatti	BDL	BDL	532.6±36	41.9
V11	Vadabarirapuram	BDL	46.7±8.6	173.5±35.7	80.1
V12	Parangipettai	5.3±3.9	57.7±8.7	381.6±36.8	117.2
V13	Chinnakumatti	3.7±3.2	122.2±8.2	266.9±30.6	199.0
V14	Varakur	13.1±3.6	64.4±7.9	171.1±33.6	118.4
V15	Keezakundalapaddi	10.0±3.6	33.6±7.7	357.7±33.6	85.8
	Range	BDL-15.5	BDL-122.2	69.2-532.6	41.9-199
	Mean ± Standard Deviation	5.7±4.7	45.6±27.5	290.6±122.5	91.2±40.1

The existence of wide-range of variation in the activities of the three natural radionuclides was evident from the generated data. The mean ratio between ^{232}Th to ^{238}U is always greater than one (>1) indicating that the activity of ^{232}Th is always higher than that of ^{238}U. The enrichment of monazite in igneous rock which was the source of thorium was responsible for the elevated radioactivity level in igneous rock as also reported by Cuney *et al* 1987 [3]. The observation in the present study confirms the report of Slunga *et al* (2008) [19] that the radioactivity fluctuated widely even within a short distance. It was observed from the results that there was a significant difference between the primordial radionuclides.

Table 2: Activity concentrations of ^{238}U, ^{232}Th and ^{40}K in cultivated soils of Chidambaram taluk

Station code	Name of the station	Activity concentration (Bq kg^{-1})			Ra_{eq} (Bq kg^{-1})
		^{238}U	^{232}Th	^{40}K	
C1	Silambimangalam	7.4±3.4	77.5±7.6	286.6±30.6	140.2
C2	Agaram	BDL	40.9±7.3	388.5±31.6	88.5
C3	Tathampettu	4.8±3.9	35.5±8.5	157.8±36.0	67.8
C4	Keerapalayam	5.5±3.6	38.7±8.0	202.4±34	76.7
C5	Jayamkondapattinam	10.2±3.7	67.3±8.2	159.3±33.3	118.8
C6	Kabadacherry	11.3±3.0	47.2±8.1	153.5±34.0	90.6
C7	Periyapettu	8.6±3.0	27.0±6.4	87.3±27	53.9
C8	Sivapuri	13.4±4.0	54.9±8.7	364.2±36.0	120.0
C9	Kabarapattu	BDL	59.1±8.7	320±36	109.1
C10	Periyakumatti	BDL	10.5±6.6	271.7±29.7	36.0
C11	Vadabarirapuram	13.5±3.8	15.1±7.0	210.8±35.1	51.4
C12	Parangipettai	5.3±3.9	57.7±8.7	381.6±36.8	117.2
C13	Chinnakumatti	BDL	69.2±7.9	330.9±32.5	127.1
C14	Varakur	BDL	57.2±8.4	243.1± 35	103.9
C15	Keezakundalapaddi	16.6±3	40.2±8	365.6±35	102.3
Range		BDL-16.6	10.5-77.5	87.3-388.5	36-140.2
Mean ± SD		7.1±4.9	46.5±19.4	261.5±96.5	93.5±30.7

Elevated level of U-238 and Th-232 in cultivated soil could be attributed to application of phosphate fertilizers in agricultural practices which are enriched sources of Uranium and Thorium (Figure 5). The increased concentration of ^{238}U in phosphate fertilizers is linked to the high concentration of uranium in phosphate rock and the chemical process involved in fertilizer productions (Khalifaand El- Arabi, 2005; El-Arabi 2007) [10, 6].

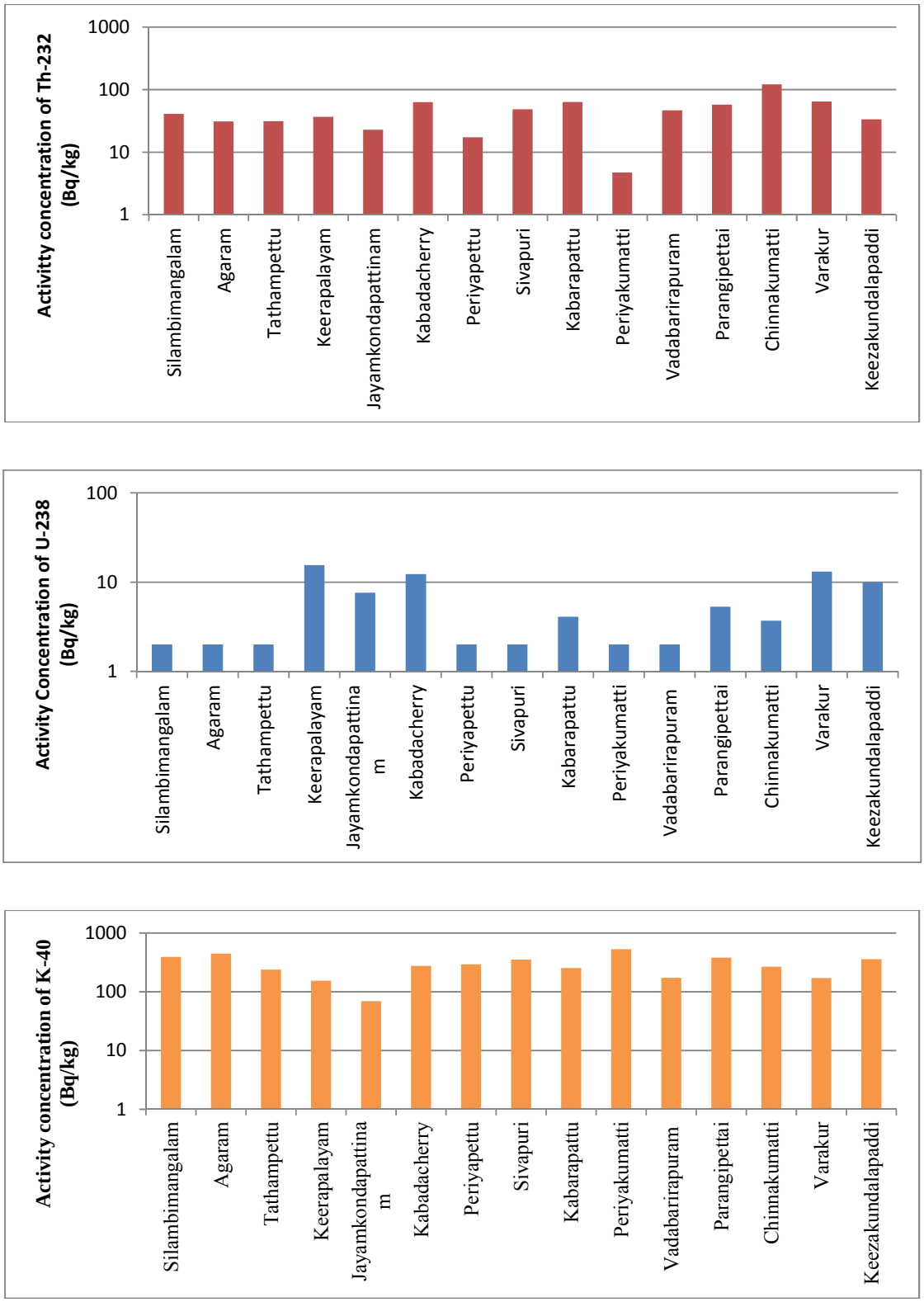

Fig 3: Activity concentration of primordial radionuclides in virgin soils of Chidambaram taluk

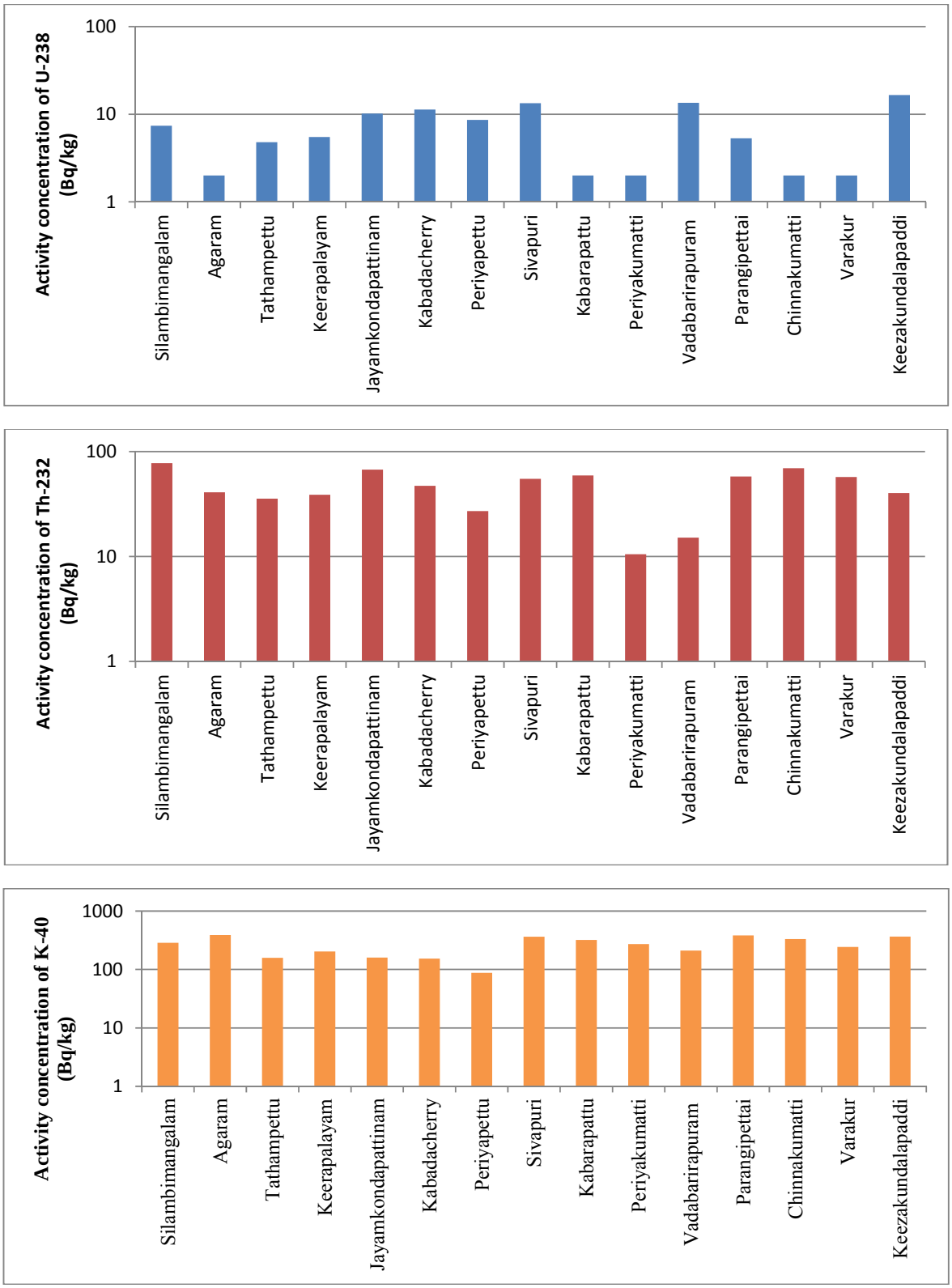

Fig 4: Activity concentration of primordial radionuclides in cultivated soils of Chidambaram taluk

According to (UNSCEAR, 2000) [21] report, the world average value of activity concentration for ^{238}U is 35 Bq kg^{-1}, for ^{232}Th 35 Bq kg^{-1} and for ^{40}K 370 Bq kg^{-1}. The measured value of activity concentrations of U-238 and K-40for the area under investigation for both categories of soil, virgin and cultivated was within the world average range. But, the activity concentration of ^{232}Th for both the soil exceeded the world average.

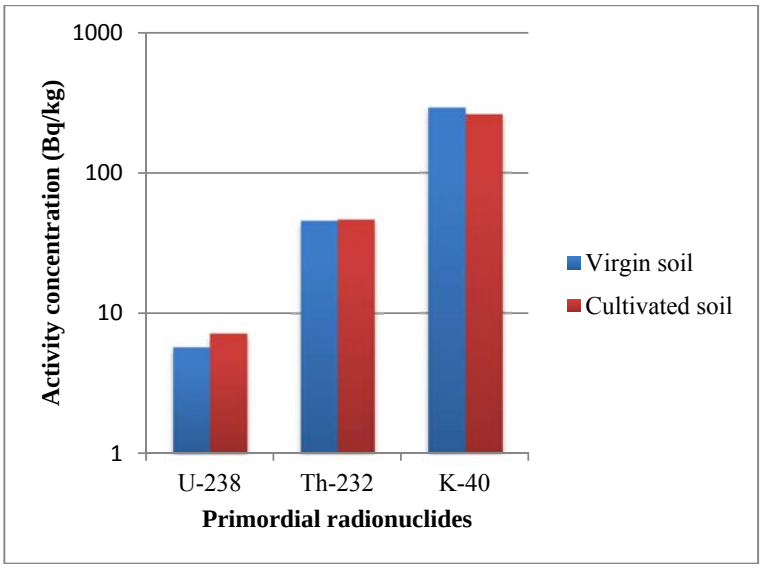

Fig 5: Comparison of activity concentration of primordial radionuclides in virgin and cultivated soil

It is observed from Figure 6, of the total gamma activity, 85 % comes from K-40, 13.7 % from Th-232 and 1.3 % from ^{238}U.

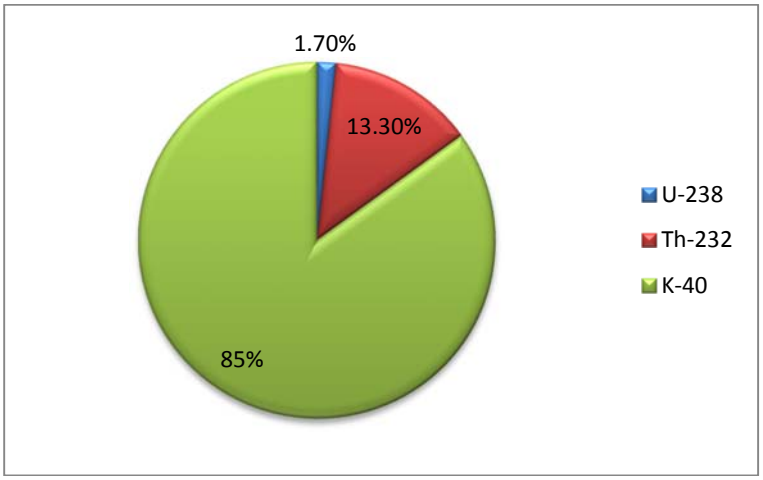

Fig 6: Overall Percentage Concentration of U-238, Th-232 and K-40 in the soils of Chidambaram

3.2. Radium Equivalent Activity (Ra_{eq})

The distribution of primordials in rocks and soils is not uniform. Hence the total exposure to radiation from the primordials has been defined in terms of radium equivalent (Ra_{eq}) in Bq kg^{-1} in order to compare the specific activity of materials containing variable amounts of ^{226}Ra, ^{232}Th and ^{40}K (Beretka & Mathew, 1985) [2]. Therefore, Ra_{eq} of soil samples were calculated using the following formula prescribed by (Sroor *et al*., 2002) [20].

$$Ra_{eq}\ (Bq\ kg^{-1}) = A_U + (A_{Th} \times 1.43) + (A_K \times 0.077) \quad (3)$$

Where, A_U, A_{Th} and A_K are the specific activity of ^{238}U, ^{232}Th and ^{40}K. The safe value of Ra_{eq} for any naturally occurring radioactive materials is reported to be less than 370 Bq kg^{-1} in order to limit the annual effective dose to 1 mSv for the general public (UNSCEAR, 2008) [22].However, in the present study the mean Ra_{eq} value for the both the soil samples were found to be 91.2 Bq kg^{-1} and 93.5 Bq kg^{-1} respectively, which are well below the recommended limit of 370 Bq kg^{-1}.

3.3. The Absorbed Dose Rate in Air from Soil samples

The absorbed dose rate(D) due to gamma radiations in air at 1 m above the ground surface for the uniform distribution of the naturally occurring radio nuclides (^{238}U, ^{232}Th and ^{40}K)were calculated based on guidelines provided by (Beck *et al*., 1972) [1]. It is assumed that the contributions from other radionuclides were insignificant. Therefore, Absorbed Dose (D) can be calculated using the following formula

$$D\ (nGy\ h^{-1}) = 0.462\ A_U + 0.601 A_{Th} + 0.0417\ A_K \quad (4)$$

Where A_U, A_{Th} and A_K are the specific activity of ^{238}U, ^{232}Th and ^{40}K. The results are given in Table 3 and 4. The dose absorbed from both the soil samples of the study area are lower(42.3 nGy h^{-1} and 42.2 nGy h^{-1})than the prescribed limit of55nGy h^{-1}.This is because of the presence of low U-238 and K-40 activity in all the sampling stations. A good correlation exists between the activity of ^{232}Th and the total absorbed dose, whereas no such correlation exists for ^{238}U and ^{40}K. This is due to the fact that major portion of the dose is contributed due to thorium content of the soil.

3.4. Estimation of Annual Effective Dose

The absorbed dose rate is converted into annual effective dose by a conversion factor (0.7 SvG y^{-1}) and indoor occupancy factor of 0.8 as proposed by (UNSCEAR, 2008) [22]. The conversion formula for annual effective dose is given below.

E_{in} (Indoor $= D_{\gamma}$ (nGy h^{-1}) x 10^{-6} x 8760 h/y x 0.8 x 0.7 SvGy^{-1} (5)

(Effective dose)

E_{out} (Outdoor $= D_{\gamma}$ (nGy h^{-1}) x 10^{-6} x 8760 h/y x 0.2 x 0.7 SvGy^{-1} (6)

(Effective dose)

The data on indoor and outdoor annual effective dose from virgin and cultivated soil samples analyzed were also presented in Table 3 and 4.

The mean indoor annual effective dose rate for the virgin soil was 0.21 mSv y^{-1} and cultivated soil was 0.20 mSv y^{-1}. The data also indicated that these values are well within the safety limit (1 mSv y^{-1}) as proposed by (UNSCEAR, 2008) [22].

3.5. External Hazard Index (H_{ex})

Consideration of external radiation exposure is usually associated with gamma radiation emitted by radionuclides of concern. The external hazard index is obtained from Ra_{eq} expression that is allowed maximum value of Ra_{eq}(370 Bq kg^{-1}) (OECD, 1979) [14].

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (7)$$

Table 3: Absorbed dose rate (D), annual effective dose rate (E) and health hazard index (H) in virgin soils from Cuddalore taluk.

Station Code	D (nGy h^{-1})	E_{in} (mSv y^{-1})	E_{out} (mSv y^{-1})	H_{ex}
V1	42.03	0.21	0.05	0.25
V2	38.22	0.19	0.05	0.22
V3	29.81	0.15	0.04	0.18
V4	35.82	0.18	0.04	0.22
V5	20.17	0.10	0.02	0.12
V6	55.26	0.27	0.07	0.33
V7	23.58	0.12	0.03	0.13
V8	45.10	0.22	0.06	0.27
V9	50.82	0.25	0.06	0.31
V10	25.97	0.13	0.03	0.13
V11	36.37	0.18	0.04	0.22
V12	53.21	0.26	0.07	0.32
V13	86.65	0.43	0.11	0.54
V14	52.08	0.26	0.06	0.32
V15	39.83	0.20	0.05	0.23
Range	**20.1-86.6**	**0.1-0.43**	**0.02-0.11**	**0.12-0.54**
Mean ± Standard Deviation	**42.3 ±16.4**	**0.21±0.08**	**0.05±0.02**	**0.2±0.1**

The index value must be less than unity in order to keep the radiation hazard well within the permissible limit.

Table 4: Absorbed dose rate (D), annual effective dose rate (E) and health hazard index (H) in cultivated soils from Cuddalore taluk.

Station Code	D (nGy h^{-1})	E_{in} (mSv y^{-1})	E_{out} (mSv y^{-1})	H_{ex}
C1	62.18	0.31	0.08	0.38
C2	41.83	0.21	0.05	0.24
C3	30.24	0.15	0.04	0.18
C4	34.36	0.17	0.04	0.21
C5	52.00	0.26	0.06	0.32
C6	40.13	0.20	0.05	0.24
C7	23.92	0.12	0.03	0.15
C8	54.54	0.27	0.07	0.32
C9	49.96	0.25	0.06	0.30
C10	18.60	0.09	0.02	0.10
C11	24.15	0.12	0.03	0.14
C12	53.21	0.26	0.07	0.32
C13	56.52	0.28	0.07	0.34
C14	45.61	0.22	0.06	0.28
C15	47.20	0.23	0.06	0.28
Range	**18.6-62.1**	**0.09-0.31**	**0.02-0.08**	**0.1-0.38**
Mean ± Standard Deviation	**42.2 ±13.3**	**0.2 ±0.06**	**0.05 ±0.01**	**0.25 ±0.08**

The values of external hazard index for virgin soil have been found to vary from 0.12 to0.54with the geometric mean value of 0.2 and for cultivated soils from 0.1 to0.38 with the geometric mean value of 0.25. The results of the present study indicated that both the soils from this region do not pose any significant radiological threat to human population.

4. Conclusions

The present study generated a data base on gamma radiation levels in the soils of Cuddalore taluk by undertaking spectrometric analysis of primordials in 30 samples. The distribution of ^{238}U, ^{232}Th and ^{40}K in the tested soil samples (both virgin and cultivated) were non-uniform and widely ranged. In terms of activity, the primordial radionuclides formed the descending order $^{40}K > ^{232}Th > ^{238}U$. The mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K in virginsoil samples were 5.7 Bq kg^{-1}, 45.6 Bq kg^{-1} and 290.6 Bq kg^{-1} respectively. The mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K in cultivated soil samples were 7.1 Bq kg^{-1}, 46.5 Bq kg^{-1} and 261.5 Bq kg^{-1} respectively. The elevated level ofU-238 in cultivated soils may be attributed the application of phosphate fertilizers. The mean Ra_{eq} value for both the soils (91.2 and 93.5 Bq $kg^{-1)}$ are well within the recommended safe limit of 370 Bq kg^{-1} and the mean absorbed gamma dose rate (D) was 42.3 n Gyh^{-1} which is well within the safety limit (55 n Gyh^{-1}). All the risk factors calculated were well below the safe limit and hence there is no radiological risk of human exposure.

Acknowledgements

The authors thankfully acknowledge Atomic Energy Regulatory Board, Govt. of India, Mumbai for research funding (Project No: AERB / CSRP / 45 / 05 / 2010) sanctioned to Prof. Dr. P. Shahul Hameed, Prof. K. Ponnusamy, Chairman, J.J College of Engineering and Technology for providing facilities and Dr. S. Mohamed Salique, Principal, Jamal Mohamed College for academic support.

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