

Measurement of Natural Radioactivity and Hazard Indices in Agriculture Soil Using Statistical Approach

V. Prabha

Ph.D. Research Scholar (Reg. No.11105)
Department of Physics and Research Centre,
Women's Christian College, Nagercoil – 629 001.
(Affiliated to Manonmaniam Sundaranar University, Abishekapatti, Tirunelveli – 627 012,
Tamil Nadu, India.)

Dr. G. Shanthi

Assistant Professor (Research Supervisor)
Department of Physics and Research Centre,
Women's Christian College, Nagercoil – 629 001.
(Affiliated to Manonmaniam Sundaranar University, Abishekapatti, Tirunelveli – 627 012,
Tamil Nadu, India.)

Abstract

Naturally occurring radionuclides such as ^{238}U , ^{232}Th and ^{40}K activity concentrations were analyzed using the high purity germanium detector NaI(Tl). The samples were collected randomly from several sites in Kanyakumari district at a penetration of about 0-10, 10-30, 30-50 cm depth. The average activity concentration of ^{238}U , ^{232}Th and ^{40}K were found to be 63.204 ± 4.42 Bq/ Kg, 85.14 ± 5.10 Bq/ Kg and 812.39 ± 48.74 Bq/ Kg. The radiological indices such as absorbed dose rate, annual effective dose rate, radium equivalent activity, life time cancer risk and hazard indices were evaluated. The statistical method was used to investigate the relationship between the calculated radiological parameters and the radionuclides. The result values will also serve as a baseline radiometric data in the area for the future studies.

Keywords: Gamma ray Spectroscopy, Multivariate, Farm soil, radiological hazards, NORMS.

INTRODUCTION

Natural radionuclides such as uranium, thorium and potassium were found everywhere in the environment. The radioactivity concentration in the agriculture soil depends upon the various geological as well as the type. Phosphate rock is the material which contains uranium, thorium and potassium which is also used as a fertilizer for agricultural purposes. The radiation contribution from soil to the humans due to external radiation directly originating from the primordial radionuclides due to inhalation reported by Jibril and Okeyode, 2012. To improve the properties the nutrient deficient lands the usage of fertilizers is increased for the development of crop production as well as to

improve the nutrient deficient lands properties (Bokaetal., 2007). Natural occurring radioactive materials are the contamination of cultivated lands due to the negative effect of the phosphate fertilizers used (Khater Ashraf E.M A.L. Sewaiden H.A., 2008). To determine the safety of the populace the radionuclides concentration on human health have to be calculated. The dose rate differs from sites to sites depending upon the activity concentration of the radionuclides (^{238}U , ^{232}Th and ^{40}K) available in the soil. Egunyinka et al (2009) reported that the high geochemical mobility of these radionuclides in the environment allows them to move easily and to contaminate the environment which human beings come into contact. 87 % of radiation dose received by humans are from natural radiation doses, which comes from the naturally occurring radioactive isotopes of ^{238}U , ^{232}Th and their progeny as well as ^{40}K reported by Isinkaye et al., 2015.

The aim of this study is to overview the distribution of the radionuclides in the selected agriculture farm areas and its radiological parameters to assess human exposure dose rates and health risks.

MATERIALS AND METHODS

Sampling and sample preparation

To measure the natural radioactivity of the agriculture soil samples of different depths were randomly collected from the different locations in the Kanyakumari district. Samples of approximately 2 kg were taken and removed the stones, plants and homogenized by crushing with an agate motor. Before measurement, the samples were dried in an oven at a temperature of 100°C for about 24 hours. The samples were weighed and packed in cylindrical plastic containers for 40 days to provide the radioactive equilibrium and the short lived degradation products between radium and thorium.

Detection technique

Each sample was measured using a gamma ray spectroscopy NAI(Tl) scintillation detector and a multichannel analyzer. The detector is shielding in a chamber of a cylindrical lead (100 mm thick). The shield is used to reduce the radioactivity background. From the 1764 keV gamma peak ^{238}U radionuclide was evaluated and from the 1120 keV gamma peak ^{232}Th radionuclide and from the 1460.7 keV gamma peak ^{40}K radionuclide was measured. Depending on the concentration of the radionuclides the sample were counted for 2000s.

Radioactivity measurement

The specific activity concentration was measured by calculating the area under the curve peak and using the efficiency curve concentration was determined using the formula

$$C = \frac{C_n}{\epsilon P \gamma M_s} \text{Bq/Kg}$$

Where C is the activity concentration of the radionuclide in the sample given in Bq/Kg. C_n is the count rate under the corresponding peak, ϵ is the detector efficiency at

the specific γ ray energy. P_γ is the absolute transition probability of the specific gamma ray and M_s is the mass of the sample.

Table 1: Activity concentrations of ^{40}K , ^{238}U , ^{232}Th in banana farm soil samples

Location code	Depth	Activity concentration of radionuclides (Bq/kg)		
	cm	^{238}U	^{232}Th	^{40}K
CLL	0-10	128.26 $\pm$ 7.69	124.79 $\pm$ 7.48	1106.61 $\pm$ 66.39
	10-30	108.39 $\pm$ 5.41	107.03 $\pm$ 6.42	1071.21 $\pm$ 53.56
	30-50	58.12 $\pm$ 4.06	74.74 $\pm$ 4.48	501.31 $\pm$ 25.06
PKI	0-10	64.43 $\pm$ 4.51	124.74 $\pm$ 8.73	862.88 $\pm$ 43.14
	10-30	71.64 $\pm$ 5.01	115.04 $\pm$ 6.90	731.03 $\pm$ 36.55
	30-50	45.76 $\pm$ 2.28	58.35 $\pm$ 3.50	316.88 $\pm$ 15.84
APM	0-10	91.89 $\pm$ 6.43	115.31 $\pm$ 6.91	1489.03 $\pm$ 89.34
	10-30	52.21 $\pm$ 3.65	59.6 $\pm$ 3.57	741.6 $\pm$ 37.08
	30-50	26.72 $\pm$ 1.33	50.55 $\pm$ 3.53	561.25 $\pm$ 28.06
ENL	0-10	35.74 $\pm$ 2.50	54.61 $\pm$ 3.27	648.05 $\pm$ 38.88
	10-30	19.5 $\pm$ 0.97	28.39 $\pm$ 1.70	385.24 $\pm$ 19.26
	30-50	11.67 $\pm$ 0.58	27.65 $\pm$ 1.65	122.38 $\pm$ 6.11
VAI	0-10	109.44 $\pm$ 6.56	127.62 $\pm$ 8.93	1975.96 $\pm$ 98.79
	10-30	104.38 $\pm$ 6.08	117.74 $\pm$ 5.88	1145.15 $\pm$ 57.25
	30-50	19.91 $\pm$ 0.99	90.99 $\pm$ 4.54	527.35 $\pm$ 26.36
Average		63.204 $\pm$ 4.42	85.14 $\pm$ 5.10	812.39 $\pm$ 48.74

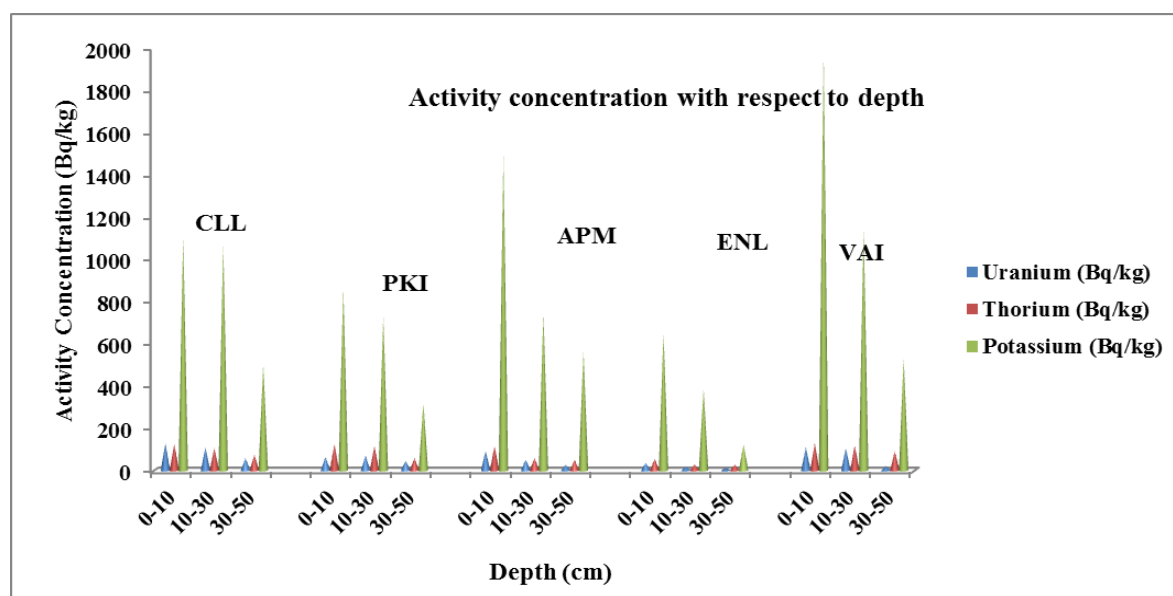


Figure 1: Activity concentration with respect to various depths

RESULTS AND DISCUSSION

Naturally occurring radioactive materials have been analyzed in agriculture farm soil in certain sites in Kanyakumari district about different depths measured using gamma ray spectrometer are shown in table 1. The average values of all radionuclides are higher than the world wide average given by the UNSCEAR 2000. The mean values for ^{40}K , ^{238}U , ^{232}Th are 812.39 ± 48.74 , 63.204 ± 4.42 , 85.14 ± 5.10 Bq/kg respectively. The radiation hazard index is determined due to the natural radioactivity associated with that soil. The observed indices are compared with internationally recommended with safety limits.

Estimation of radiological hazard indices

1. Radium equivalent activity (R_{eq})

Radium equivalent activity is used to predict total activity as well as the gamma radiation hazards.

$$R_{eq} = C_u + 1.43 C_{th} + 0.07 C_k$$

Whereas C_u , C_{th} , C_k are the specific activity concentrations of ^{238}U , ^{232}Th , ^{40}K in Bq/Kg. The average R_{eq} outcome values of the specified sites are examined in column 3 of Table 2 are outlined. Whereas C_u, C_{th}, C_k are the specific activity concentrations of ^{238}U , ^{232}Th , ^{40}K in Bq/Kg. The radium equivalent activity concept allows a number to account for the radiation hazards associated with the mixture of uranium, thorium and potassium in agriculture soil samples in different locations reported by Suresh Gandhi et al., 2013. The mean value of R_{eq} is 247.50 Bq/Kg which is lower than the recommended value of about 370 Bq/kg.

2. Absorbed dose rate (D)

The calculated activity concentrations of ^{238}U , ^{232}Th , ^{40}K are converted into absorbed dose rate by utilizing 0.462 for uranium, 0.604 for thorium and 0.0417 for potassium respectively as the conversion factors. (UNSCEAR 2000)

$$D = 0.461 C_U + 0.623 C_{Th} + 0.0414 C_K$$

C_U, C_{Th}, C_K are the activity concentrations (Bq/Kg) of ^{238}U , ^{232}Th , ^{40}K in agriculture soils respectively. The absorbed dose rate for most of the samples of different depths is higher than the world mean value 60 nGy/h. This may be due to the significant of ^{232}Th in the particular sites. Figure 2 illustrates the radium equivalent activity and absorbed dose rate with various depths of various sites in Kanyakumari district.

3. Annual Effective Dose Equivalent (AEDE)

The annual effective dose equivalent is calculated using a conversion factor of 0.7 SvG/y which is used to convert the absorbed rate to human effective- dose equivalent with an outdoor and indoor occupancy of 20% and 80% respectively (UNSCEAR 2000).

$$AEDE(\text{mSv/y}) = D(\text{nGy/h}) \times 8760 \text{h} \times 0.2 \times 0.7 \text{SvGy}^{-1}$$

The average absorbed dose rate is about 3.47 is higher than the permissible value of 0.48mSv/y.

4.External and internal hazard index (H_{ex} , H_{in}):

Due to the emitted gamma rays of the samples external hazard index were calculated using the formula

$$H_{ex} = \frac{C_u}{370Bq-1} + \frac{C_{th}}{259Bq-1} + \frac{C_k}{4810Bq-1}$$

C_u, C_{th}, C_k are the activity concentrations(Bq/Kg) of $^{238}U, ^{232}Th, ^{40}K$ in agriculture soils respectively.By using the internal hazard index, the internal exposure to radon and its daughter products is qualified using the equation reported by Senthil Kumar et al., 2010.

$$H_{in} = \frac{C_u}{185} + \frac{C_{th}}{259} + \frac{C_k}{4810} \leq 1$$

The mean value of H_{ex} and H_{in} (0.13& 0.56) is less than unity.

5. Excess life time cancer risk (ELCR)

A higher value of ELCR implies higher probability induction of cancer of the individual that was exposed using the below formula was reported by Taskin et al., 2009.

$$ELCR = AEDE \times DL \times RF$$

DL = Duration of Life, RF = Risk factor (S/v)

The average value of ELCR is 0.492×10^{-3} . ELCR value is greater than the world permissible value 0.29×10^{-3} .

Table 2: Radiological hazard parameters of Banana Farm Soil

Location Code	Depth	R_{aeq}	D	AEDE	H_{in}	H_{ex}	I_{yr}	ELCR
	Cm	(Bq/kg)	(nGy/h)	(mSv/y)				($\times 10^{-3}$)
CLL	0-10	391.91	182.68	4.84	0.89	0.22	2.84	0.78
	10-30	343.92	160.99	4.65	0.78	0.19	2.50	0.69
	30-50	203.59	94.11	2.19	0.46	0.11	1.46	0.40
PKI	0-10	309.25	143.13	3.68	0.70	0.17	2.25	0.61
	10-30	292.43	134.96	3.16	0.66	0.16	2.11	0.57
	30-50	153.60	70.56	1.41	0.34	0.08	1.09	0.30
APM	0-10	371.43	175.84	6.30	0.86	0.21	2.75	0.75
	10-30	194.54	91.90	3.15	0.45	0.11	1.43	0.39
	30-50	142.22	67.04	2.35	0.32	0.08	1.05	0.28
ENL	0-10	163.73	77.32	2.73	0.37	0.09	1.21	0.33
	10-30	89.76	42.62	1.62	0.20	0.05	0.67	0.18
	30-50	60.63	27.67	0.52	0.13	0.03	0.43	0.11
	0-10	444.08	211.76	8.33	1.03	0.25	3.32	0.90

VAI	10-30	360.92	168.88	4.94	0.82	0.20	2.63	0.72
	30-50	190.63	87.69	2.19	0.43	0.10	1.39	0.37
Average		247.50	115.81	3.47	0.56	0.13	1.80	0.492

6. Calculation of gamma radiation representative level index(I_γ)

Estimation of the level of gamma radiation associated with different concentrations of some specific radionuclides is known as representative level index (Shanthi et al., 2010) is given by

$$I_\gamma = \frac{C_u}{150Bq-1} + \frac{C_{th}}{100Bq-1} + \frac{C_k}{1500Bq-1}$$

Where C_u, C_{th}, C_k are the activity concentrations of $^{232}U, ^{238}Th, ^{40}K$ in Bq/Kg. Gamma representative index average value is 1.80mSv and in some locations of various depths, the values exceeded unity due to radiological hazardous.

Table 3: Comparison of average activity concentration of $^{238}U, ^{232}Th, ^{40}K$ and R_{eq} in the worldwide agriculture soils.

Location	^{238}U (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	R_{eq} (Bq/kg)	Reference
Northern Jordan	49.90	26.70	291.10	103.10	Alhamameh and Awadallah (2009)
India Amritsar	50.61	78.31	301.80	189.67	Mehra, and singh (2011)
Malaysia	80.63	116.87	200.66	258.36	Ahmad et.al.(2015)
India	63.204	85.14	812.39	247.50	Present work
Worldwide	32	30	400	370	UNSCEAR(2000)

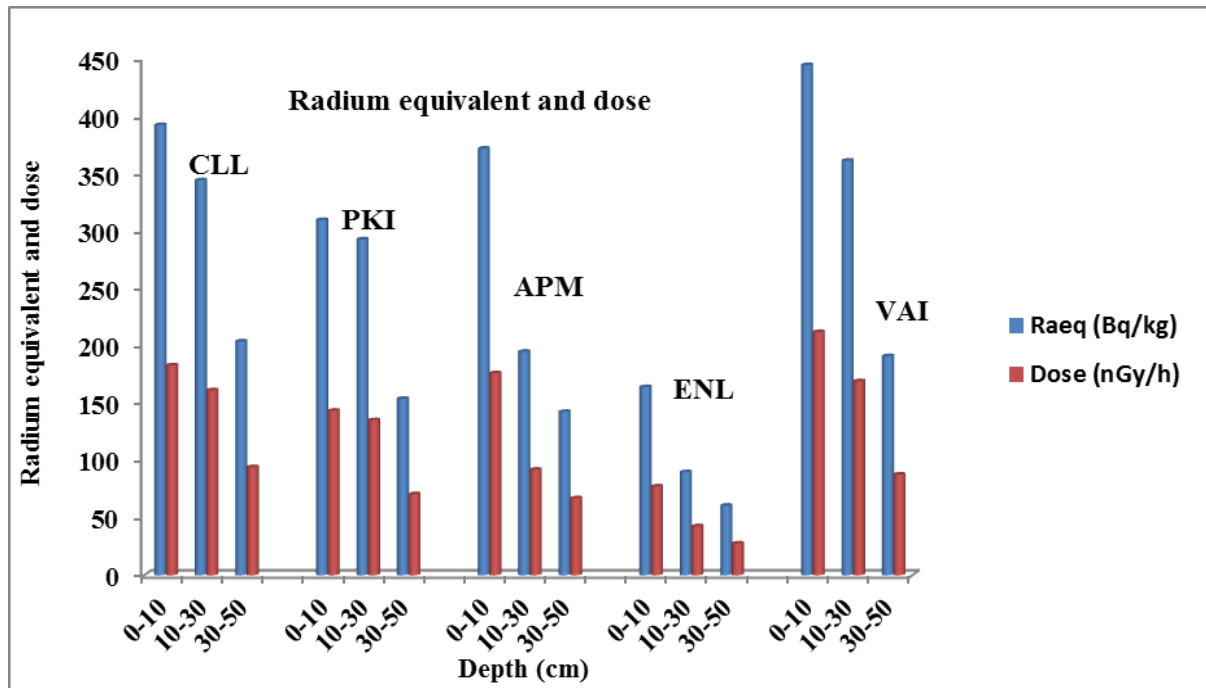


Figure 2: Radium equivalent activity and absorbed dose with respect to depths.

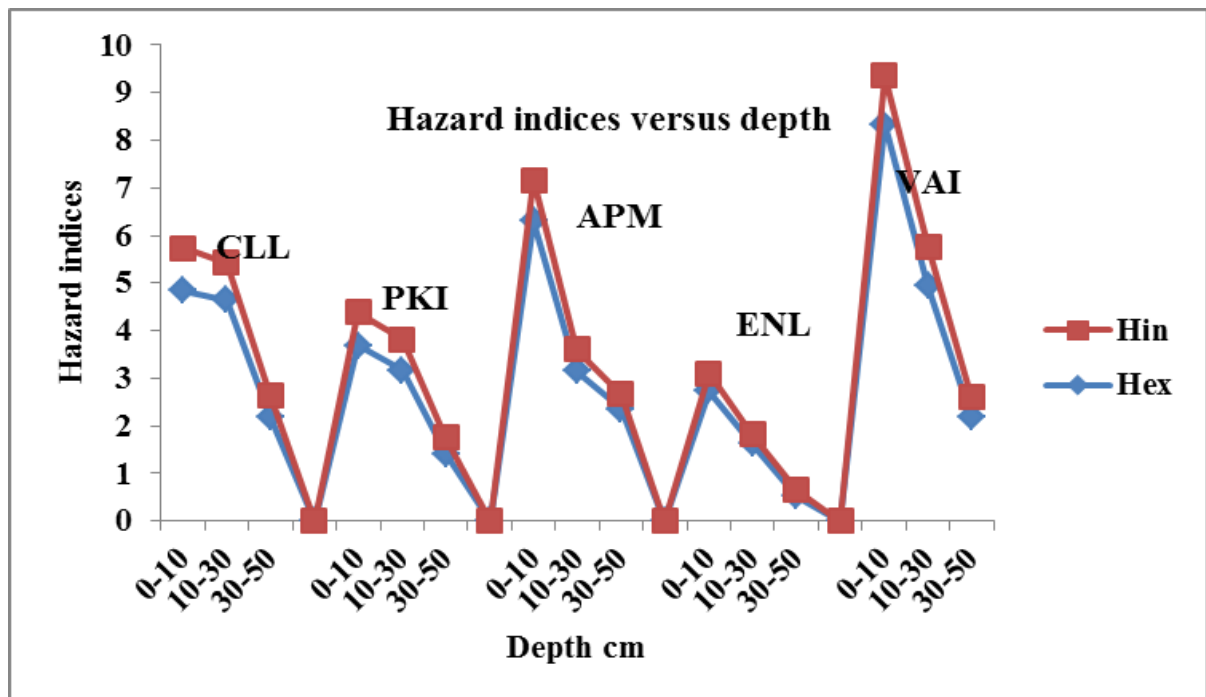


Figure 3: Hazard indices with respect to depth

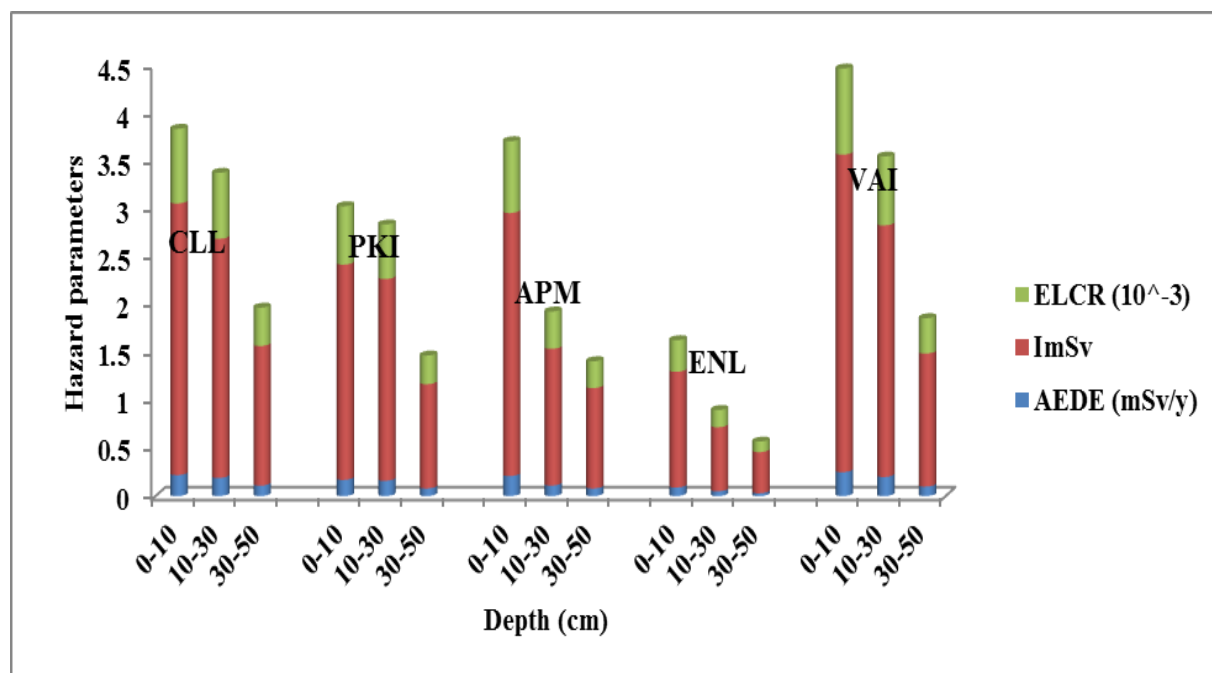


Figure 4: Radiological parameters with respect to various depths

Multivariate Statistical Analysis:

Multivariate statistical procedures for data treatment and histograms were performed by means of the software package SPSS version 16.0 for windows. Pearson correlation and cluster analysis were carried out to clarify the correlation among the variables, the impact of soil indices on the distribution of natural radionuclides. Multivariate statistical methods are effectively used to interpret the relationships between variables in the environmental studies reported by Liu et al., 2003. Laaksoharju et al., 1999 reported that this method can also help to simplify and organize large data sets to provide meaningful insight and also to indicate natural associations between variables.

Basic Statistics:

The statistical behavior of the measured radionuclide is shown in table 4. "Skewness is a measure of the asymmetry of the probability distribution of areal valued random variables" (Adam and Eltayeb 2012). Positive skewness denotes the distribution with an asymmetric tail extending towards values that are positive. Kurtosis is a measure whether the data are peaked or flat relative to a normal distribution. Positive kurtosis indicates flat distribution and positive denotes peak distribution.

Table 4: Basic statistics of the measured data

Variables	²³⁸ U(Bq/kg)	²³² Th(Bq/kg)	⁴⁰ K(Bq/kg)
Mean	12.84	30.25	143.04
SD	58.42	125.21	1881.48
Variance	31.32	59.23	782.61
Kurtosis	13.67	28.479	458.42

Skewness	186.89	811.08	210152.9
Maximum	-0.60	1.433	1.191
Minimum	0.40	1.471	1.012
Frequency distribution	Normal	Normal	Normal

Factor Analysis:

Factor analysis is to determine the interrelationships among radioactive variables and to describe these variables in terms of their common dimensions reported by J.C.Davis et al., 1986. Varimax rotation is performed for factor loading (H.F.Kaiser 1958). Component 1 is loaded with ^{238}U and other parameter explains 50.348 % and component 2 is loaded with ^{40}K which explains 49.610% of total variance. Component 1 explains more than the component 2. The two factors extraction in this study explains 99.958% of the total variance of the data set.

Table 5: Rotated factor loading of variables:

Variables	Component 1	Component 2
^{238}U (Bq/kg)	.937	-.139
^{232}Th (Bq/kg)	.931	-.287
^{40}K (Bq/kg)	.937	.348
R_{eq} (Bq/kg)	.998	-.061
D(nGy/h)	.999	-.035
AEDE (mSv/y)	.945	.327
Hex	.999	-.040
Hin	.999	-.035
I _y	.999	-.031
ELCR($\times 10^{-3}$)	.999	-.036
Percentage of variance explained	50.348	49.610

Pearson Correlation Coefficient Analysis:

Variables	^{238}U	^{232}Th	^{40}K	R_{eq}	D	AEDE	H _{in}	H _{ex}	I _{yr}	ELCR
^{238}U	1									
^{232}Th	0.84	1								
^{40}K	0.81	0.78	1							
R_{eq}	0.94	0.94	0.91	1						
D	0.93	0.94	0.92	0.99	1					
AEDE	0.93	0.94	0.92	0.99	0.99	1				
H _{in}	0.96	0.93	0.90	0.99	0.99	0.99	1			
H _{ex}	0.93	0.94	0.92	0.99	0.99	1	0.99	1		
I _{yr}	0.93	0.94	0.92	0.99	0.99	1	0.99	1	1	

ELCR	0.93	0.94	0.92	0.99	0.99	1	0.99	1	1	1
------	------	------	------	------	------	---	------	---	---	---

Pearson Correlation Analysis is analyzed so as to conclude the related correlations between the radionuclide and radiological indices. Positive correlation is detected amongst the three radionuclides and all the radiation hazard factors imply very strong relationship with agriculture soil and radiological indices. It also indicates that all radionuclides will donate the emission of gamma radiation. The frequency distribution curves of ^{238}U , ^{232}Th and ^{40}K as shown in Figure 5.

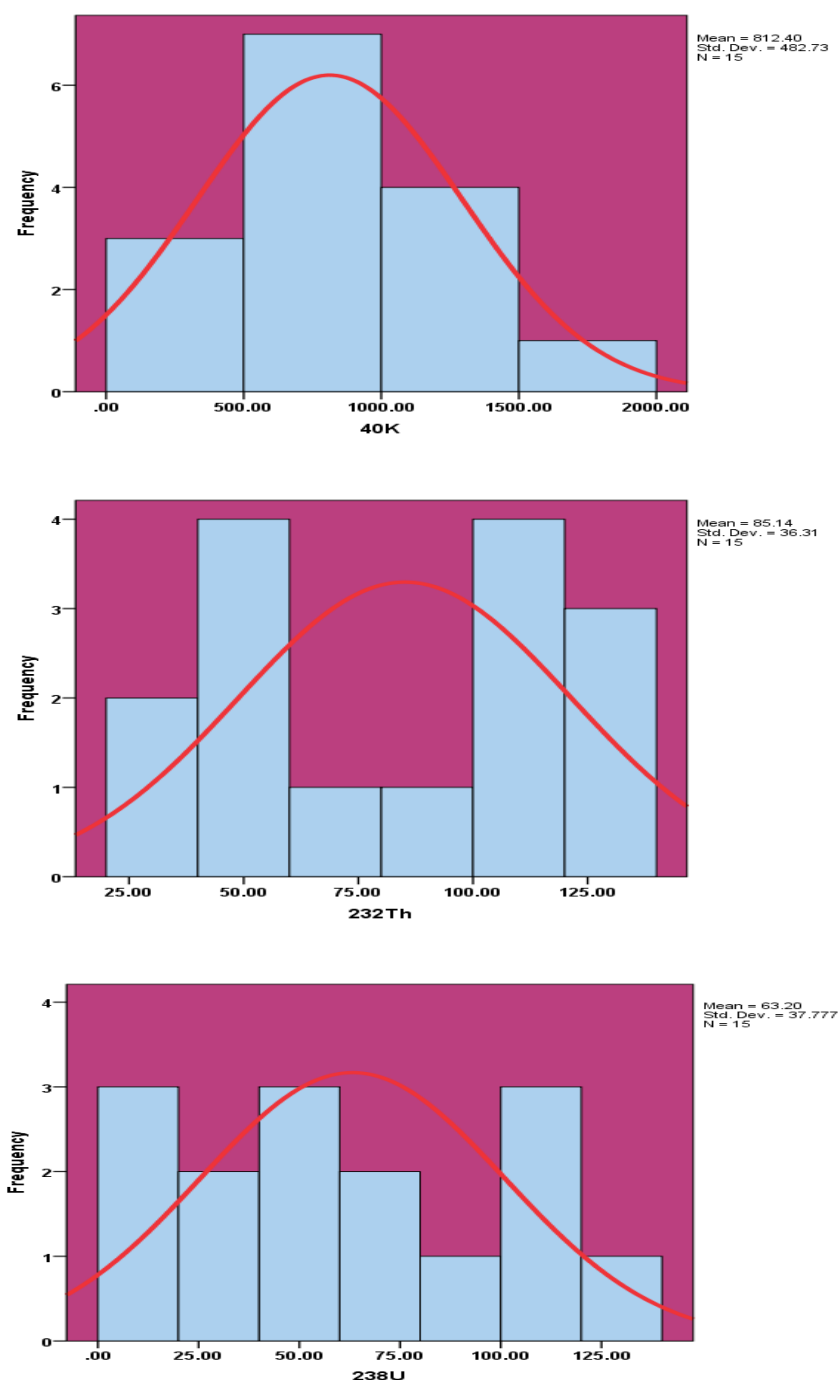


Figure 5: Frequency distributions of ^{238}U , ^{232}Th and ^{40}K in agricultural soils

Cluster Analysis:

Cluster analysis through axes is used to classify analogous characteristics among natural radio isotopes and radiological parameters in the soil. It is also useful to define the linkages among soil samples commencing various localities to investigate the data. Cluster I consists of and ^{40}K and their radiological parameters. Cluster II consists of ^{238}U and Cluster III consists of ^{232}Th . Dendrogram of variables are illustrated in Figure 6.

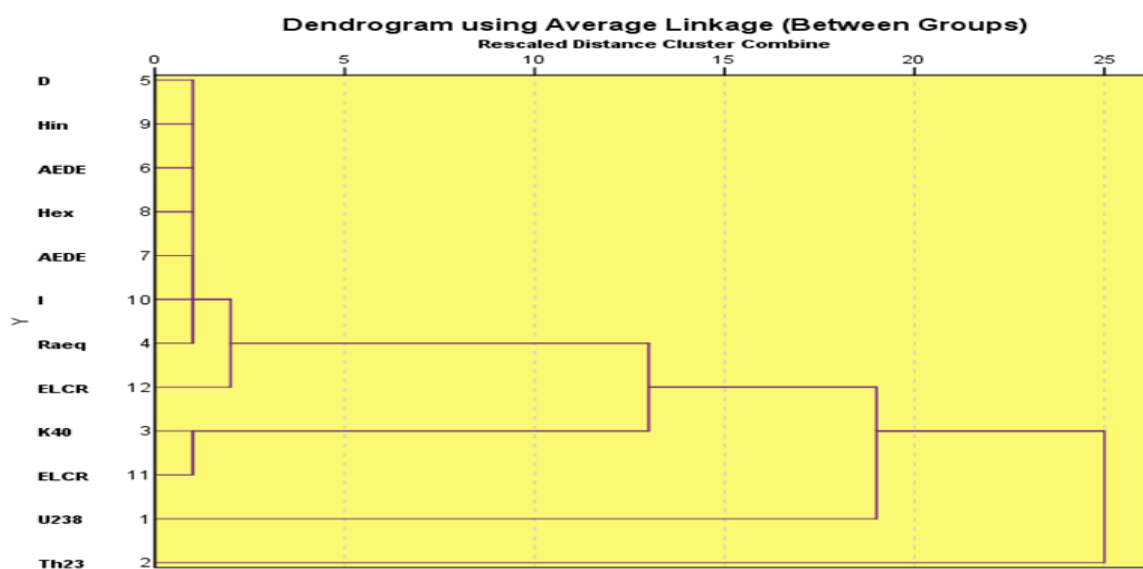


Figure 6: Dendrogram of radioactive variables

Conclusion

The calculated average values of ^{40}K , ^{238}U , ^{232}Th are higher than the world wide range given by UNCEAR (2000). R_{eq} average value is less than the value identified by UNSCEAR (2000). Due to low price and further necessities phosphate fertilizers are used. The existence of high activity of ^{40}K is due to the potassium containing manures. ^{232}Th have the maximum value this may be due to the accumulation of radioactivity in the upper surface of the soil. Due to continuous fertilization the ^{238}U value is less at maximum depth. From multivariate statistical analysis as a result of same origin the positive correlation occurs between ^{40}K , ^{238}U , ^{232}Th . The observed result serves as a reference for the future studies.

References

- [1] Adam A.M, Eltayeb M.A. (2012) Multivariate statistical analysis of radioactive variables in two phosphate ores from Sudan. Journal of Environmental Radioactivity, 107:23-43.

- [2] Bolca M, Sac M, Cokuysal B, Karali T, Ekdal, E. (2007). Radioactivity in soils and various foodstuffs from the Gediz River Basin of Turkey. *Radiation Measurement*. 42:263-270.
- [3] J.C. Davis, *Statistics and Data Analysis in Geology*, Wiley, New York, 1986, p.17.
- [4] Jibiri, N.N., Okeyode I.C. (2012). Evaluation of radiological hazards in these sediments on Ogun River, South Western Nigeria. *Radiation physics and chemistry*, 81, 103-112.
- [5] H.F. Kaiser. Thevarimax criteria for analytical rotation in factor analysis. *Psychometrika* 1958; 23, 187-200.
- [6] Khater Ashraf E.M., AL- Sewaidan H.A., (2008), Radiation exposure due to agricultural uses of phosphate fertilizers, *Radiation measurements* volume 43, Issue 8,1402-1407.
- [7] Liu,W., X.Li, Z.Shen, D.Wang,O,Wai and Y.Li, (2003), Multivariate Statistical Study of heavy metal enrichment in sediments of the Pearl River Estuary, *Environmental Pollution*,121,377-388.
- [8] Laaksoharju M, Skarman C, Skarman E. Multivariate mixing and mass balance (M3) calculations, a new tool for decoding hydrogeochemicalin formation. *Applied Geochemistry* ,1999;14:861-71.
- [9] Taskin. H., Karavus M., Ay, P., Topuzoghi, A., Hindiroglums, S. and Karahan, G.Radionuclide concentration in soil lifetime cancer risk due to gamma radioactivity in Kirklareli, Turkey,*Journal of environmental Radioactivity*,2009;100,49-53.
- [10] UNSCEAR, (2000). Exposure of natural radiation sources. Annex B. Sources and effects of ionizing radiation, United Nations, Newyork. United Nations scientific Committee on the effects Atomic Radiation.
- [11] Egunyinka, O.A., C.J. Olowookere, I.A. Babalo and R.I. Obed. Evaluation of ^{238}U , ^{232}Th and ^{40}K concentrations in the top soil of the University of Ibadam South-western Nigeria. *Pacif. J. Sci.Technol.*, (2009) 10(2):742-750.
- [12] SureshGandhi M, Ravishankar R, Rajalakshmi A, Sivakumar S, Chandrasekaran A, PreamAnand D. 2014.Measurements ofnatural gamma radiation in beach sediments of north east coast of Tamilnadu, India by gamma ray spectroscopy withmultivariate statistical approach. *Journal of Radiation Research and Applied Sciences*. 7: 7-17.
- [13] Senthilkumar B, Dhavamani V, Ramkumar S, Philominathan, P.2010.Measurement of gamma radiation levels from Thanjayur,using gamma ray spectroscopy and estimation of population exposure. *J. Med.Phys.*35:48-53.
- [14] Shanthi G., Thampi J. T. K. Allen G. G. R.andManiyan C. G. Measurement of activity concentration of natural Radionuclides for the Assessment of Radiological indices. *Radiation Protection Dosimetry*.2015; 141(1) 90-96.
- [15] Isinkaye M.O. and Emelue. Natural radioactivity measurements and evaluation of radiological hazards in sediment of Oguta lake, South East Nigeria. *Journal of Radiation Research and Applied Sciences*,2015;8: 459-469.