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Risk Determination of Radionuclide Derived from Agriculture Fertilizers in Iraqi Markets by Gamma Spectrometry

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Abstract. A fertilizer has increase production and becomes a necessary tool that uses in agriculture. Different amount of uranium and thorium are including in the fertilizers that mean natural nuclide will be concentrated in high values. In this work, seventeen samples of commonly used fertilizer in Iraqi markets were collected and determine the specific activities of radiation ^{238}U, ^{232}Th, and ^{40}K using a technique of gamma-ray spectroscopy NaI(Tl) detector. The concentration of these nuclides has average values (79.66±0.41Bq/kg, 30.52±0.31Bq/kg, and 181.27±2.91 Bq/kg) comparing with worldwide limitation (35Bq/kg, 35Bq/kg, and 350 Bq/kg), respectively. So the concentration of ^{238}U nuclei greater than its global limit. The radiological hazards of all samples as radium equivalent activity (Raeq), external (H$_{ex}$) and internal (H$_{in}$), alpha and gamma indices, and annual effective dose, due to the presence of these radionuclides, were calculated to assess it hazards and by the statistical study that using SPSS program one can mark the relations between all hazards indices.

Keywords: Radionuclide, Fertilizer, NaI(TI) detector, hazard indices, Cluster statistic.

1. Introduction.

Residents were exposed to naturally occurring radionuclides which are found in various sources, among them fertilizers that are used in agriculture. The use of fertilizers in agriculture gives good information to hand out the specific activities of ^{238}U, ^{232}Th, and ^{40}K in the environment. All corps need to grow up quickly, easily, and healthy. With some steroids as fertilizers in reasonably priced application depending on the soil type and its fertility where some crops do not require fertilizer as legumes, one can get what is desirable. Fertilizers are solid and liquid, the solid fertilizer is mostly inorganic as urea, di-ammonium, phosphate, and potassium chloride. The solid shape either granulate or powder that composed mainly of nitrogen (N), potassium (K), and phosphorus (P). However, the extensive use of fertilizers can increase environmental effects. The increasing consumption of it can affect soil, surface, and ground waters because of the dispersion of element using. Many studies have shown that agricultural fertilizers contain varying amounts of natural radioactive material [1-3]. The radioactive elements that imply fertilizers differ in amount depending on their concentration such as Uranium-238. As the concentration increase of the natural radionuclides in fertilizers, the risk of human health from this nuclide increases.

The use of natural fertilizers in wide-ranging Iraqi markets may lead to the distribution of radionuclide elements in the agricultural areas that will become a source of radioactivity decay. This phenomenon may lead to potential radiation hazards due to external exposure to gamma and alpha particles (external and internal) radiation by workers in factories where fertilizer produced, formers, or

residents of forms. In several markets, there are several types of fertilizers used in agriculture as di-ammonium phosphate and nitrogen urea, and so on. Their concentrations must be measured in fertilizer to specify the safe employment of fertilizers. The concentration of radium and uranium contentment in the soil is increasing the radiation dose and absorbed dose in the human body which leads to unwanted health prohibitions.

This evaluation aims to build a level for concentrations of radioactive nuclei in used samples and find a correlation between concentrations and radioactive parameters comparing with word limits. Besides, existing a relationship between activity concentration and hazard index using statistical methods(cluster, Pearsons, and descriptive).

2. Material and Methods:

Sample collection: Seventeen samples of the most available types of fertilizers in local markets in Iraq were collected and prepared to measure the natural activity. The type samples are listed in Table 1 where each plastic bag is labeled by its name.

Table 1. The details of the fertilizer sample under test.

Samples name	Sample code	Type	Origin
Fertilizer 1	F1	O-DAP Micronutrients	Iraqi (Kufa)
Fertilizer 2	F2	K-Humate (N-13, P-22, K-18)	Iraqi (Kufa)
Fertilizer 3	F3	Nitrogen -Urea	Iraqi (Basra)
Fertilizer 4	F4	Nitrogen -Urea	Ukrainian
Fertilizer 5	F5	Nitrogen -Urea	Iranian
Fertilizer 6	F6	Phosphate daimonian	Jordanian
Fertilizer 7	F7	Phosphate daimonian	Jordanian
Fertilizer 8	F8	N-P component	Iraqi (Basra)
Fertilizer 9	F9	Compost	Iraqi (municipal)
Fertilizer 10	F10	Nitrogen -Urea (EFC)	Egyptian
Fertilizer 11	F11	Nitrogen -Urea	Egyptian
Fertilizer 12	F12	Potassium Nitrate (NP)	Jordanian
Fertilizer 13	F13	Potassium Nitrate (NP)	Chinese
Fertilizer 14	F14	Potassium Nitrate (NP)	Saudi Arabia
Fertilizer 15	F15	NPK	Germania
Fertilizer 16	F16	NPK	Tunisian
Fertilizer 17	F17	NPK	United States

2.1 Sample preparation

The collected sample underwent to milling process for 2 hours with a porcelain mill with different sizes of porcelain balls. The result powder sieving by 2mm meshes to get rid of the coarse suspended material with sample and obtain a homogenous sample free from impurities that may affect on measuring process. The samples dried in the controlled oven at 100 ℃ to remove the moisture and have a constant weight then cooled in desiccators. The sample was left in preservative bags (polyethylene plastic sealed bag) for about 28 days to reach the secular equilibrium condition between ^{238}U and ^{232}Th long-lived radionuclides and its parent daughters. To measure the background radiation of the environment, the homogeneous samples were transported to a new sealed bag and left for about 30 days.

2.2 Measurements

Measurements of natural radioactivity have been done using the gamma-ray spectrometer that includes a multichannel analyzer connected with NaI(Tl) detector. The led cylinder was used to protect the detector and reduced the background level by a factor of about 95%, while the energy calibration was made for the detector by two source ^{137}Cs (0.662 MeV) and ^{60}Co (1.333 MeV).

Spectroscopy analysis: The activity concentrations of radionuclide were determined in Bq/Kg. The activity of ^{40}k was determined from peak energy of gamma at 1460 KeV, while for ^{238}U and ^{232}Th determine from the secular equilibrium and their progenies (^{214}Pb and ^{214}Bi) at energies 295.2 KeV and 315.9 KeV, respectively. The counting time that converting count/s to activity unit (Bq/kg) is19000s.

3. Result and Discussion

3.1 Results

Activity Concentration: The amount of radioactivity in the sample can be determined by the activity concentration as a specific activity following the relation from [4]

$$A_i \left(Bq/kg \right) = \frac{C_i - B.G.}{\varepsilon M_s P_\gamma T} \qquad (1)$$

Where C_i is counting net area under the photo peaks, i represents the fertilizer sample, $(B.G.)$ is background count below, ε denotes absolute energy efficiency for the detector at a specific energy of gamma, $(P\gamma)$ is an absolute γ- transition or emission probability of the specific gamma-ray,(T) is the measurement time of counting inside the detector in(s) and (M_s) is dry of each sample weighted mass in the container in (kg). The specific activities of ^{238}U, ^{232}Th, and ^{40}K and their corresponding uncertainties for samples are represented in Table 2. ^{40}K concentration has value dominated for several samples. The variation in values of concentrations of the radionuclide may due to the chemical formula of fertilizers. The values and average values of each nuclide are shown in fig. (1). The Uranium concentration average value (79.66 Bq/kg) above the global limit, while for ^{232}Th and ^{40}K (30.52 and 181.27 Bq/kg) are less than the worldwide recommended limits reported by UNSCEAR (2008) [5] (35, 35, and 370 Bq/kg, respectively). The variation coefficient shows some data with relatively high and lower values. The high values represent a heterogeneous distribution for data while the lower relatively values indicated homogeneous distribution.

Table 2. Radionuclide specific activity in fertilizer samples.

Sample code	Specific Activity in (Bq/kg)		
	^{238}U	^{232}Th	^{40}K
F1	131.18 ± 1.26	72.17 ± 0.64	83.233 ± 2.91
F2	108 ± 0.83	64.41 ± 0.38	480.16 ± 3.28
F3	173.98± 1.43	62.16 ± 0.45	362.77 ± 2.15
F4	14.9±0.38	33.45±1.02	282.49±1.94
F5	6.9 ± 0.1	8.6 ± 0.21	21.3 ± 1.16
F6	16.18 ± 0.19	4.02 ± 0.09	57.69 ± 1.34
F7	12.27 ± 0.37	6.34 ± 0.32	33.18 ± 2.38
F8	165.81 ± 1.27	11.21 ± 0.51	496 ± 0.87
F9	7.82 ± 0.20	11.93 ± 0.18	33.27 ± 0.42
F10	208.61 ± 2.31	50.14 ± 0.41	22.90 ± 1.24
F11	181.97 ± 1.01	18.73 ± 0.23	17.65 ± 0.43
F12	20.22 ± 0.19	17.91 ± 0.31	47.81 ± 1.13
F13	79.19 ± 0.34	31.82 ± 0.46	1024 ± 8.12
F14	84.10 ± 0.56	22.53 ± 0.23	402.6 ± 12.18
F15	72.43 ± 0.56	121.18 ± 0.58	418..17 ± 9.66

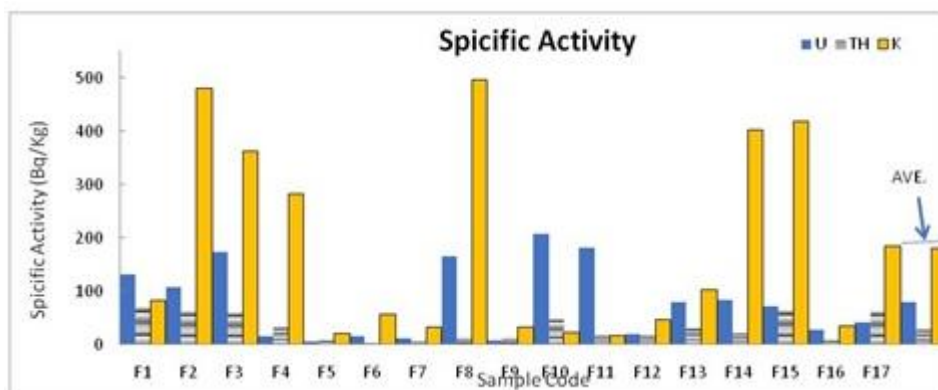|			
F16	28.57 ± 1.13	7.82 ± 0.13	35.16 ± 0.17
F17	42.16 ± 0.76	63.37 ± 1.01	184.76 ± 2.84
ave.	79.66±0.41	30.52±0.31	181.27±2.91

Fig. 1. Represent the concentration of specific activity of the samples with average values for each nuclide.

Table 3. The partial correlations between the specific activities.

Control Variables		^{238}U	^{232}Th	^{40}K
^{238}U	Correlation	1	0.427	0.278
^{232}Th	Correlation	0.427	1	0.398
^{40}K	Correlation	0.278	0.398	1

From Table 3, it is clear that the correlation between three radionuclides is linear and in Table 4 the differences in the activity values were clear from the arithmetic mean (AM) where the highest value for

Table 4. Descriptive Statistics of the main radionuclides underestimation.

Radionuclide	N	Range	Min. value	Max. value	Sum	AM	S.D.	Variance	Skewness	Kurtosis
U	17	202	7	209	1354	79.66	69.693	4857.04	0.600	-1.116
Th	17	68	4	72	552	32.46	24.823	616.197	0.451	-1.559
K	17	478	18	496	3082	181.27	181.704	33016.37	0.724	-1.265

^{40}K (181.27 Bq/kg) and lowest one for ^{232}Th (32.46 Bq/kg). To measure the degree of asymmetry tail distribution.

of the radionuclide, the skewness has positive values and near zero, while the kurtosis has negative values, i.e. the distribution follows the normal one.

Radiological Hazards Effects Evaluation**:** Different radiological indices were estimated to determine the hazard of natural radioactivity in samples. The calculated values were compared with the universal values that represent safety limits.

3.1.1 Radium Equivalent Activity Appreciation

Since the distribution of radionuclide is not uniform with real activity level in the samples, the radium equivalent (Ra_{eq}) is a single radiological hazard index need to determine because of its ability to give a good explaining of radiation protection for human comparing the limits of fertilizer that must be maximum less than 370 Bq/kg to keep the dose of gamma-ray below 1.5 mSv y^{-1} (UNSCEAR, 2000) [6]. The main formula to compute this parameter is [7]:

$$Ra_{eq} = A_U + (1.43A_{Th}) + (0.077A_K) \qquad (2)$$

where A_U, $A_{Th,}$ and A_K are uranium, thorium, and potassium concentrations, respectively. The above equation is based on the assumption that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq/kg of ^{232}Th, and 4810 Bq kg^{-1} of ^{40}K produce the same gamma-ray dose rate. The radium equivalent is related to both the external γ-dose and the internal α-dose from radon and its progeny. Fig. (2) represented the frequency distribution where analysis has been done for all samples and showed a normal (bell-shaped) distribution.
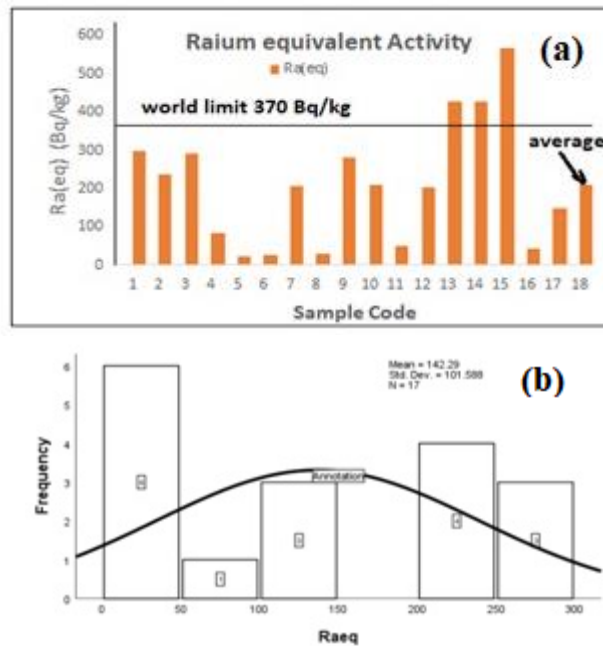

Fig. 2. (a) bar chart (b) normal distribution shapes of Ra(eq) for all samples.

3.1.2 Absorbed Dose and Effective Dose Appreciation

The use of fertilizers in agriculture is a possible source of exposure for the workers in sites and that may increase radionuclide exposure. By applying the following relation one can convert the measured activities of U, Th, and K into doses according to UNSCEAR, 2000 [7] using the same factors to give the absorbed gamma rate in the air at 1m above the ground level[9]:

$$D_{abs}(nGyh^{-1}) = (0.462A_U) + (0.621A_{Th}) + (0.042A_K) \qquad (3)$$

The results were shown in Table 2 and figure (3). the limit recommended by UNSCEAR (2000) [7.]

The estimation of annual effective dose (AEDE) according to UNSCER(2008) [5]it must be taken into account by converting the coefficients from (D_{abs} in nGy.h^{-1})) in the air using conversion factor (0.7 Sv Gy^{-1}) to (AEDE in mSv.y^{-1}) with indoor occupancy factor (fraction of time spent indoor (0.8) and outdoor (0.2)). The AEDE from gamma radiation from ^{226}Ra, ^{232}Th, and ^{40}k in the fertilize samples was calculated from [5]:

$$AEDE(mSv) = D_{abs}(nGyh^{-1}) \times 8760h \times O \times C \text{ (mSv/ nGy)} \qquad (4)$$

Where O is the occupancy factor and C is the absorbed to the effective dose conversion factor for adults (0.7 x 10^{-6}Sv/Gy). AEDE in all samples for gamma radiation from natural nuclide is varied from

to with mean value of, as shown in table()all studied samples of fertilizers are less than the recommended limiting value of 480 μSv y^{-1} [5].
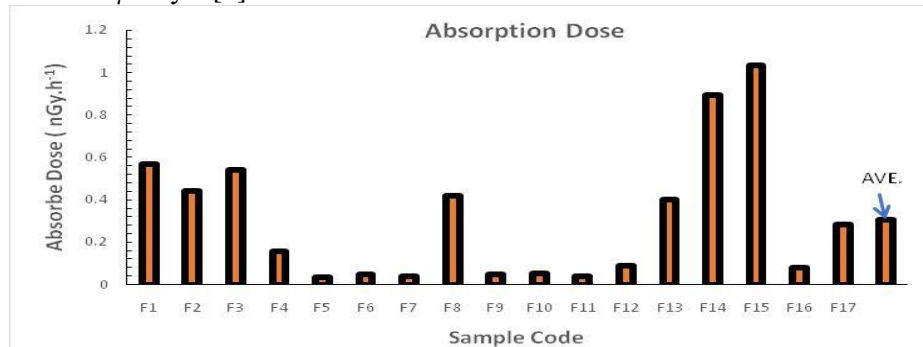

Fig. 3. Absorption Dose values with their average value.

3.1.3. Gamma-ray Index

This index is used to estimate the hazard level of gamma radiation from the fertilizer samples that may harmful by damaging the human body cells. This index limitation according to European Commission Guidelines [8] must be ($2 < I_{\gamma} < 6$) when increasing the annual effective dose to 1*mSvy^{-1}*[9] while $I_{\gamma} \leq 0.5$ corresponding to *0.3mSvy^{-1}*and $I_{\gamma} \geq 0.5$ corresponding to 1 *mSvy^{-1}*. It can be calculated from [10]:

$$I_{\gamma} = \frac{A_U}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \qquad (5)$$

The results and average value are represented in Table 2 and Fig. 4. Four samples from all the measured samples had a radioactivity level index of more than 1, which lead to avoid using because give an exposure effective dose of more than *1mSvy^{-1}* for whom to deal with this production.

3.1.4. Internal Level Radiation Index (Alpha Radioactivity**)**

When radon gas is released from samples there is an alpha particle were emitted causes alpha radiation. The index I_{α} can be calculated from [11]:

$$I_{\alpha} = \frac{A_U}{200} \qquad (6)$$

It should be less than the maximum permissible value of I_{γ} for radium concentration 200 Bq/kg. This index must be less than unity and the results of fertilizes samples show varied values from 0.0345 to 1.043 with a mean value less than 1. The higher sample value is F10 (Nitrogen -Urea (EFC).
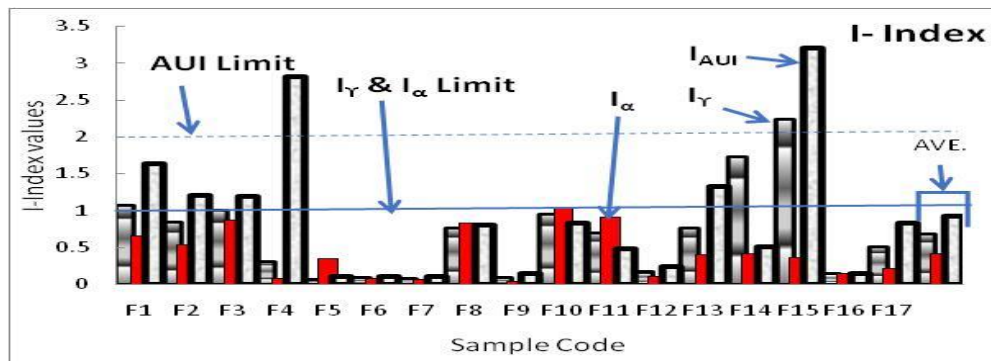
Fig. 4. Radiation hazard indices with limitations and average values.

3.2 Radio hazard indices

Analysis of various known radiological health hazard indicators in radiological studies were used to find a safer conclusion. To assess the radiation risk associated with tested samples, the following indicators were identified.

3.2.1 External and internal hazard indices

To provide information about safe levels of internal exposure to radon and its daughter that have a short-life, the internal index (H_{in}) must be in the value less than one to keep the radiation hazards low. The index can calculate from [12]

$$H_{in} = \left(\frac{A_u}{185}\right) + \left(\frac{A_{Th}}{259}\right) + \frac{A_K}{4810} \qquad (7)$$

The external index (H_{ex}) is an assessment of the natural risks of gamma and is used to evaluate the radioactivity of a substance. H_{ex} represents exposure to external radiation associated with gamma from radionuclide. This index evaluated as following and should also less than one UNSCEAR (2000) [7]:

$$H_{ex} = \left(\frac{A_u}{370}\right) + \left(\frac{A_{Th}}{259}\right) + \frac{A_k}{4810} \qquad (8)$$

3.2.2. Excess Lifetime Cancer Risk (ELCR)

The probability of cancer developing over a lifetime human at a certain exposure level. This indicator gives a value of expected numbers of cancers in limiting people's numbers when exposed to specific carcinogenic at a specific dose. An increase in ELCR led to increase in developing leukemia, breast, and prostate cancer. The ELCR can be evaluated using the equation [13]:

$$ELCR = D_{eff}(AEDE) \times DL \times R \qquad (9)$$

Where DL is the duration of life (approximately 66 years) and RF is a fatal cancer risk factor in (Sv^{-1}) with a value of 0.05 as in ICRP-60 [15].
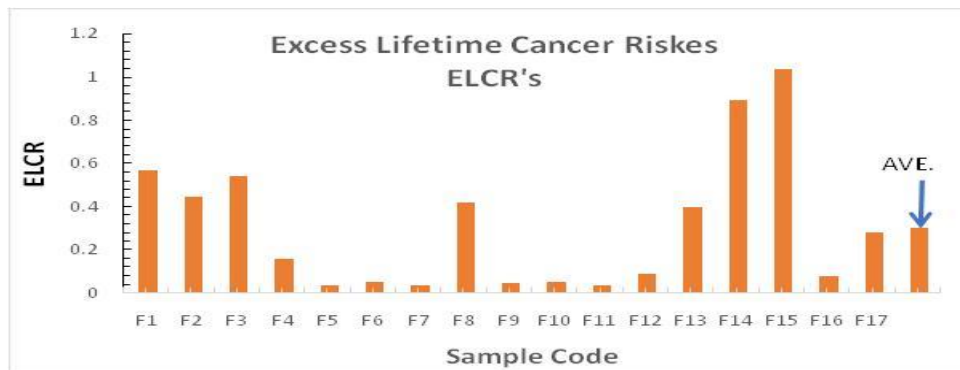
Fig. 5. Excess Lifetime Cancer Risks with the average value.

3.2.3. Activity utilization index (AUI)

The calculation of absorbed dose in the air can be active by using a transfer factor. For this purpose, the activity utilization index building up and calculated using the relation [14]:

$$A_U\ 50(Bq\,/\,kg\,)f_U + A_{Th}\ 50(Bq\,/\,kg\,)\mathrm{f}_{Th} + A_K\ 500(Bq\,/\,kg\,)f_K \qquad (10)$$

Where f_U, f_{Th}, and f_K are the fraction contributions to the total dose rate in the air because of gamma radiation from the studied natural radionuclide f_U = 8.09%, f_{Th} = 47.98%, and f_K = 43.92%. The main limitation of this index is less than 2 for the annual effective dose(AEDE) (<0.3 mSy/y) [15]. The sample F15 recorded the highest value for this index (3.207) while the average value within the limit.

3.2.4. Clark value $^{232}Th/^{238}U$ concentration ratio

This ratio will give an indicator for the samples collected from a markets regions have either higher or lower uranium concentration to be economic for uranium mining and extraction UNSCEAR(2000).
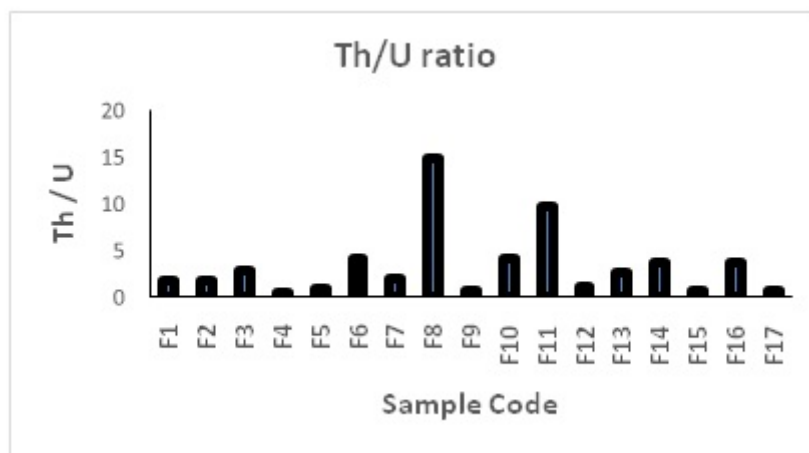

Fig. 6. Th /U ratio.

3.3 Statistical Analysis

To understand the multivariate correlations between radionuclide concentrations and other parameters, Pearsons, descriptive and cluster technique analysis had been carried out through the " Statistical Program of the Social Science (SPSS/PC-IBM) [16].

3.3.1 Pearson's Coefficient Matrix

This method measure the linear relationship between any two continuous parameters. From Table 5, significant correlation is in the bold (") represent the good correlations lies among (0.5) and (1) between any two-parameter and the table show high positive correlations with perfect monotonic relation coefficient (1)**.** The high values correlation of AEDE (in, out) with other parameters was represented while^{238}U has a weak correlation with ^{40}K and AUI. The negative sign in the matrix appears just for Th/U parameter that means the three parameters (Th, ELCR, and AUI) are in the opposite direction**.**

Table 5. Pearson's correlation coefficient matrix for radionuclide and the determined hazard indices of measured parameters.

code	^{238}U	^{232}Th	^{40}K	Ra$_{eq}$	D	AEDE in	AEDE out	I$_\gamma$	I$_\alpha$	H$_{in}$	H$_{ex}$	ELCR	AUI	Th / U
^{238}U	1													
^{232}Th	.441	1												
^{40}K	.289	.407	1											
Ra$_{eq}$	.900**	.709**	.364	1										
D	.900**	.695**	.388	.998**	1									
AEDEin	.567*	.612**	.618**	.613**	.622**	1								
AEDEout	.603*	.736**	.607*	.656**	.665**	.987**	1							
I$_\gamma$	.504*	.606**	.614**	.551*	.561*	.995**	.986**	1						
I$_\alpha$	.695**	.295	.452	.668**	.684**	.467	.487	.422	1					
H$_{in}$	.841**	.569*	.529*	.818**	.825**	.906**	.931**	.873**	.636**	1				
H$_{ex}$	.624**	.664**	.610**	.672**	.680**	.995**	.996**	.988**	.499*	.928**	1			
ELCR	.268	.564*	.739**	.414	.433	.900**	.877**	.912**	.417	.702**	.877**	1		
AUI	.162	.650**	.518*	.342	.351	.575*	.637**	.606**	.148	.456	.596*	.595*	1	
Th / U	.582*	-.315	.218	.303	.331	.112	.104	.065	.572*	.343	.127	-.039	-.216	1

3.3.2. Cluster analysis

Hierarchical cluster analysis is the method used by grouping similar radiometric characters [17] that fall into the same class to have a clear up for the relationships between all parameters and find optimal grouping within each group, and calculate the Euclidean distance for each parameter. The cluster statistics performed based on Pearson's correlation coefficient by the dendrogram shown in Fig. 7 according to Uranium concentration were parameters grouped in the cluster as it present in a population that can be divided into clusters to get the whole population information. It is clear in Fig. 7 there are three clusters, the first represented by samples (1,2,3,4,5 and 6), the second for (7,8,9, and 10), and the third cluster for equal clusters (11,12,13,14,15 and 16). These groups have different areas.
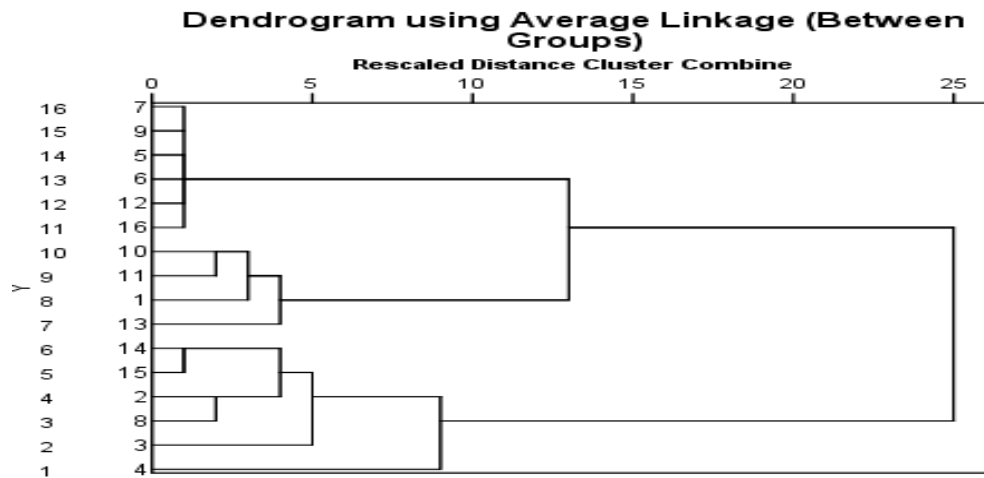
Fig. 7. Dendrogram using Centroid Linkage for classifying parameters with uranium concentration in the fertilizer samples.

3.3.3. Descriptive statistics analysis

From Table 6 the ^{40}K concentration present high average value (496 Bq/kg) than other parameters and twice the uranium average value (209 Bq/kg) with variation coefficient (33.01%), less variation for ^{232}Th (6.16%). This variation coefficient represents the dispersion of the data compared with other parameters variation uranium reach 48.57%.

Table 6. Descriptive statics of the parameters.

Descriptive Statistics								
	N	**Min.**	**Max.**	**Mean**	**S.D.**	**Variance %**	**Skewness**	**Kurtosis**
^{238}U	17	7	209	79.66	69.693	48.57	.600	-1.116
^{232}Th	17	4	72	32.46	24.823	6.16	.451	-1.559
^{40}K	17	18	496	181.27	181.704	33.01	.724	-1.265
Ra_{eq}	17	21	298	142.29	101.588	10.32	.199	-1.478
D	17	9	140	66.65	47.688	22.74	.151	-1.549
AEDE in	17	0	1	.43	.365	.001	.867	.252
AEDE out	16	0	0	.10	.088	.008	.759	.338
I_{gamma}	17	0	2	.68	.615	.378	1.189	1.356
I_{alpha}	17	0	1	.36	.306	.093	.685	-.874
H_{in}	17	0	1	.67	.532	.283	.077	-1.957
H_{ex}	17	0	1	.49	.394	.155	.651	-.191
ELCR	17	0	1	.30	.313	.098	1.146	.538
AUI	17	0	3	.93	.924	.854	1.441	1.647
Th/U	17	0	15	3.24	3.731	1.39	2.328	5.666

4. Conclusion

Natural radioactivity of ^{238}U, ^{232}Th, and ^{40}K in different types of fertilizers was measured. The average specific activities in samples are in the global limits unless the ^{238}U is higher than the recommended limit of UNSCEAR 2008. (35Bq/kg). The radiological hazard indices measurement includes radium equivalent activities, external and internal indexes, gamma index, absorbed radiation,

and annual effective doses of the fertilizers. From all of these results, we deduce that the number of fertilizers that showed high radioactivity should be decreased and used with precautions.

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