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ARTICLE

Indicators of Radioactive Contamination by Radionuclides for Samples of Plant Fertilizers and Pesticides

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Abstract: This study aims to evaluate the radiation levels resulting from plant fertilizers and pesticides in Babylon City, Iraq. The specific activity of the studied radionuclides (e.g. ^{238}U, ^{232}Th, and ^{40}K) was identified using the spectroscopy approach of the sodium-activated iodide thallium (NaI (Tl)) with dimensions of 3" × 3". The resulting data revealed that the specific activities for the aforementioned nuclides were 6.254 Bq/kg, 4.3694 Bq/kg, and 112.751 Bq/kg, respectively. The results indicated that the average radiation hazard metrics for fertilizers (i.e., gamma index, alpha index, absorbed dose rate, ambient dose equivalent rate, annual gonadal dose equivalent, and excess lifetime cancer risk) were lower than the reference levels.

Keywords: Radiation hazard, Fertilizer, Pesticides, Risk parameters, Gamma activity.

1. Introduction

Soil is considered to be one of the key elements of the environment. This is because it is a medium that provides humans with food resources. However, when soil becomes contaminated, it can lead to long-term environmental pollution, affecting food, water, and air. Using phosphate-based fertilizers, which are expected to involve high levels of radiation in the surrounding environment, is the most important human activity that causes radiological exposure [1]. Natural radiological elements are known to be widely spread out in the environment and constitute the main source of human radiation exposure. In this regard, the agricultural activity of human affects the radioactivity in soil [2].

The Earth has harbored radioactive elements since its formation, many of which have decayed over time. Considering what remains to this day of the aforementioned radionuclides of terrestrial origin, they have a lifespan that is greater than the age of the Earth. These are known by the series of ^{238}U, ^{235}U, and ^{232}Th, as well as potassium ^{40}K [3]. Humans and other living organisms are expected to be subjected to radiation on a regular basis from both natural and anthropogenic sources [4].

Soil pollution depends on the movement of radioactive materials and their accumulation or transmission, which are governed by the interaction of the materials and compounds with the hard part of the soil. The soil's physical, chemical, and biological characteristics affect how well it can bury radioactive contaminants. Moreover, the amount and type of radionuclides movement are determined by soil management practices, the types of cultivated plants, rainfall rates, and the amount of irrigation water used [5, 6]. In this context, fertilizers are very important since they enrich the agricultural soil with

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essential elements such as potassium, nitrogen, and phosphorous, which are often depleted due to repeated cultivation. Phosphate fertilizers are valuable because of their phosphorous content, a key element for plant growth.

However, phosphate fertilizers are also a source of concern because they often contain uranium, a naturally occurring radioactive element that is geochemically associated with phosphate wherever it is found. The excessive use of fertilizers has been shown, through many investigations, to increase the levels of nuclides in the soil and even in groundwater. These nuclides can then enter the human body via the intake of contaminated drinking water and food [6].

In order to establish a database on the radioactivity level to be later used as a reference point for radiological accidents, a large number of studies have been undertaken to measure radioactivity. One such study was conducted in the northern regions of Sudan [7]. In this work, the dose ranged from 44 to 53 nGy, while the annually effective dose ranged from 53 to 65 μSv y^{-1}. The average absorbed dose in eastern Sudan ranged from 24 to 48, with an average of 38 nGy h^{-1}. Central Sudan showed air-absorbed dose measurements at a height of 1 m ranging from 31 to 47 nGy h^{-1}, and the effective annual dose was between 6 and 47.8 Sv y^{-1}. In western Sudan, the absorbed dose ranged from 500 to 7000 nGy h^{-1} [8]. Through the latter studies, it was found that agricultural awareness among the farmers in these regions is generally low. However, there is a potential for improvement in agricultural practices by using natural animal fertilizers instead of chemical fertilizers and adopting modern irrigation systems that are more compatible with the requirements of crops [9]. These measures could help preserve agricultural products and reduce the environmental risks posed by radioactive contamination.

The aim of this study is to evaluate the natural radioactivity levels of ^{238}U, ^{232}Th, and ^{40}K in samples of plant fertilizers available in Babylon City, Iraq, using gamma-ray spectroscopy with NaI (Tl). Additionally, the radiological hazard risks are calculated in all samples of the present study.

2. Materials and Methods

2.1 Preparation of the Study Samples

Various types of fertilizers and pesticides utilized in the soils of agricultural land were gathered from local markets in Iraq. Table 1 provides an overview of the different fertilizers considered in this study, including the country of origin and the fundamental elements of each type.

The fertilizer and pesticide samples were crushed in a grain mill to convert them into a fine powder and then sieved using a single-hole sieve with a diameter of 2 mm. These samples were then left to dry for one day at ambient temperature to obtain a steady weight. After drying, each sample weighed 700 grams.

The dried samples were put in cylindrical plastic containers, chosen to fit the detector container. The containers were sealed tightly and stored for 30 days to allow the radium and its decay products to reach equilibrium. After this period, the samples were ready for gamma radiation spectroscopic analysis.

2.2 Measuring System

The technique of gamma-ray spectroscopy based on the activated sodium iodide detector NaI(Tl) whose dimensions are 3"x3" was employed to identify the concentration levels of ^{238}U, ^{232}Th, and ^{40}K in all fertilizer and pesticide samples. The scintillation detector operates at a voltage of 778 V, with an efficiency limit of around 95%, and the detector's energy resolution ranges between 6.5% and 8.5%. The detector base is supported by a lead shield to minimize background radiation. The analysis of the resulting gamma-ray spectra was done using the laboratory's newly developed Maestro-32 software.

2.3 Theoretical equations

The specific activity (in Bq/kg) of the radionuclides investigated in this work was computed as follows [14-16]:

$$A\left(\frac{Bq}{kg}\right) = \frac{N}{t*\varepsilon*I_\gamma*m} \pm \frac{\sqrt{N}}{t*\varepsilon*I_\gamma*m} \quad (1)$$

where N refers to the count under the photopeak, t is the counting time (in seconds), Iγ refers to the probability of absolute transition gamma emission, m refers to the net sample mass (kg), and ε represents the detectors' efficiency for a specific gamma energy.

The absorbed dose rate (AD_γ) for external gamma dose irradiation in the air at a distance of 1 m above the ground surface, due to ^{238}U, ^{232}Th, and ^{40}K, was calculated using the equation [17]:

$$AD_{out}\left(\frac{nGy}{h}\right) = DCF_U\ A_U + DCF_{Th}\ A_{Th} + DCF_K\ A_K \quad (2)$$

where DCF_U, DCF_{Th}, and DCF_K refer to the absorbed dose rate conversion factors for ^{232}Th, ^{238}U, and ^{40}K, respectively, (in nGy/h), while A_{Th}, A_K, and A_U are the activity concentrations of ^{232}Th, ^{40}K, and ^{238}U, respectively. The $DCF_k = 0.0417$ nGy/h/Bq/kg, $DCF_U = 0.462$ nGy/h/Bq/kg, and $DCF_{Th} = 0.604$ nGy/h/Bq/kg. The γ-ray dose (Din) from ^{238}U, ^{232}Th, and ^{40}K indoors was calculated using the equation [18]:

$$AD_{in}\left(\frac{nGy}{h}\right) = 0.92\ A_U + 1.1\ A_{Th} + 0.081\ A_K \quad (3)$$

The annual effective dose comes in two different forms, namely the annual indoor effective dose rate and the annual outdoor effective dose rate (AEDEout) (AEDEin). The following equations were used to determine the AEDEout and AEDEin. [19, 20]:

$$AEDE = \text{Absorbed Dose Rates}\ (AD\gamma) * 0.7 * 8760 * 0.2 \quad (4)$$

$$AEDE_{outdor} = \left[AD_{out}\left(\frac{nGy}{h}\right) * 8760\left(\frac{h}{y}\right) * 0.2 * 0.7\left(\frac{10^3 mSv}{10^9 nGy}\right)\right] \quad (5)$$

$$AEDE_{outdor} = AD_{out} * 1.2264 * 10^{-3}\left(\frac{mSv}{y}\right) \quad (6)$$

$$AEDE_{indoor} = \left[AD_{in}\left(\frac{nGy}{h}\right) * 8760\left(\frac{h}{y}\right) * 0.8 * 0.7\left(\frac{10^3 mSv}{10^9 nGy}\right)\right] \quad (7)$$

$$AEDE_{indoor} = AD_{in} * 4.9056 * 10^{-3}\left(\frac{mSv}{y}\right) \quad (8)$$

The total dose from internal and external effects was calculated by summing the $AEDE_{indoor}$ and $AEDE_{outdoor}$.

It is used to compute the sum of the activities of each of ^{232}Th, ^{238}U, and ^{40}K in (Bq/kg) and then to assess the associated hazards with sample materials that contain ^{232}Th,^{238}U, and ^{40}K in Bq/kg as follows [21]:

$$Ra_{eq.}\left(\frac{Bq}{kg}\right) = A_U + 1.43\ A_{Th} + 0.077\ A_K \quad (9)$$

To calculate the dose rates in air due to the existence of radionuclides in soil via adopting the appropriate conversion factors, the activity utilization index (AUI) can be assessed using the following formula[22,23]:

$$AUI = \frac{A_U}{50\frac{Bq}{Kg}} * f_U + \frac{A_{Th}}{50\frac{Bq}{Kg}} * f_{Th} + \frac{A_k}{500\frac{Bq}{Kg}} * f_K < 2 \quad (10)$$

where f_U = (0.462), f_{Th} = (0.604), and f_K = (0.041) are the partial contributions to the overall dose rate in air resulting from the gamma irradiation by the decay of the radionuclides. The reported standard activities for ^{232}Th, ^{238}U, and ^{40}K in soil (A_{Th}, A_U, and A_K) are 50, 50, and 500 Bq kg^{-1}, respectively.

The annual gonadal dose equivalent (AGDE) refers to the quantity of radiation that is received by the gonads, bone marrow, and bone cells. The following formula can be used to compute AGDE resulting from activity concentrations of 232Th, 238U, and 40K in the tailing enriched soil samples [24]:

$$AGDE\left(\frac{mSv}{y}\right) = 3.09\ A_{(U)} + 4.14\ A_{(Th)} + 0.314\ A_{(K)} \quad (11)$$

where 3.09, 4.14, and 0.314 are the corresponding conversion factors that convert the specific activities of ^{238}U, ^{232}Th, and ^{40}K into the overall organs dose.

Considering the assumption that the highest value of the external hazard index (Hex.) corresponds to the upper limit of Raeq (i.e. 370 Bq kg-1), it can be obtained using the expression for Raeq. It indicates the risk associated with exposure to radiation from ^{238}U, ^{232}Th, and ^{40}K in the analyzed soil samples. The following formula was used to calculate it [25]:

$$H_{ex.} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (12)$$

The internal radiation hazard (Hin.) plays an important role in assessing the internal exposure to ^{222}Rn and gamma rays. This index can be estimated as follows [26]:

$$H_{in.} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (13)$$

The quantity I-alpha, suggested by Kriege and Stoulos, is given by [27]:

$$I_\alpha = \frac{A_{238_U}}{200} \quad (14)$$

The representative level index (Iγ) is a monitoring index that confirms the analogy of environmental samples to established dose standards for soil. Iγ can be estimated via the equation below [28]:

$$I_\gamma = \frac{A_U}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \quad (15)$$

Iγ should be ≤ 1, which refers to the annual effective dose of ≤ 1 mSv, in order to coincide with the given dose criteria [16].

To assess the probability of cancer risk in a population exposed to radiation from soil, the excess lifetime cancer risk (ELCR) can be calculated. This calculation is based on the estimated AEDE using the following equations [29]:

$$ELCR_{outdoor} = AEDE_{outdoor} * LF * RF \quad (16)$$

$$ELCR_{indoor} = AEDE_{indoor} * LF * RF \quad (17)$$

where LE refers to the life expectancy (60 years), and RF (Sv^{-1}) refers to the risk factor per Sievert, which is 0.05. The overall excess lifetime cancer risk can be determined as below [30]:

$$ELCR_{total} = ELCR_{outdoor} + ELCR_{indoor} \quad (18)$$

3. Results and discussion

The average reported activity of ^{238}U, ^{232}Th, and ^{40}K have mean values of 35 Bq/kg, 30 Bq/kg, and 400 Bq/kg, respectively, according to the UNSCEAR report 2008 [31]. The results of the specific activity levels of these natural radioactive elements in the analyzed plant fertilizers and insecticides are presented in Table 1. Among the fertilizer samples, NPK Granules had the lowest specific activity of ^{238}U at 2.12±1.09 Bq/kg, while the Combe Star sample had the greatest specific activity of ^{238}U at 26.91±5.69 Bq/kg, with a mean of 6.254 Bq/kg. According to recent studies, the globally permitted limit for ^{238}U specific activity in fertilizer samples is below 35 Bq/kg (UNSCEAR). The elevated specific activity of ^{238}U in certain samples could be attributed to the higher phosphate content, which often occurs due to the blending of phosphate ores with nitrogen and potassium during fertilizer production. The average specific activity of ^{232}Th was found to be 4.3694 Bq/kg, with values ranging from 0 Bq/kg in NPK Powder to 21.01±2.93 Bq/kg in Norwegian Chemical / Root Fertilizer. The findings demonstrate that the amount of ^{232}Th in the fertilizer samples exceeded the recommended levels. The specific activity levels of ^{40}K varied significantly among the samples, with the lowest value of 40.99±6.11Bq/kg observed in NPK 10 20 10+6S Chemical Magnesium, and the highest value of 495.04±19.31 Bq/kg found in agricultural sulfur, with a mean of 112.751 Bq/kg (see Fig.1). The specific activity levels of ^{40}K were lower than the universally reported limit of 400 Bq/kg, which is considered acceptable according to UNSCEAR 2008.

TABLE 1. The specific activity of ^{40}K, ^{238}U, and ^{232}Th in fertilizer and pesticide samples (Bq/kg)

No.	fertilizer and pesticides	^{40}K Series	^{238}U Series	^{232}Th Series
		^{40}K	^{214}Bi (Bq/kg)	^{208}Tl (Bq/kg)
1	Shoot	218.95±19.28	18.19±5.34	14.28±2.86
2	Man colax	412.15±16.61	10.82±2.59	4.02±0.95
3	Master power	288.50±15.86	4.27±1.85	3.00±0.94
4	Carbaryl Chemical Compound	226.24±13.64	11.40±2.94	1.72±0.69
5	Norwegian Chemical / Root Fertilizer	309.16±19.36	6.39±2.67	21.01±2.93
6	NPK 10-20-10. Granular Compound Fertilizer	336.74±17.32	16.46±3.68	4.18±1.12
7	NPK Powder	416.70±19.43	10.21±2.92	0.00
8	agricultural sulfur	495.04±19.31	4.18±1.71	4.56±1.08
9	NPK 10 20 10+6S (Chemical Magnesium)	165.23±10.47	3.62±1.49	3.41±0.87
10	DAP $(NH_4)_2HPO_4$)	40.99±6.11	4.88±2.03	7.91±1.56
11	NPK Granules	353.29±14.61	2.12±1.09	4.28±0.93
12	Sulphur –dusting fungicide	471.57±26.78	21.93±5.55	10.17±2.29
13	Master flour	274.94±16.86	9.07±2.94	0.00
14	Vector(chania)	162.69±15.30	14.23±4.35	7.98±1.97
15	Di ammonium phosphate	52.44±8.29	5.21±2.51	8.28±1.91
16	ORTs 509 mg	434.04±21.89	4.08±2.04	6.90±1.60
17	ATLAS	396.83±20.38	22.45±4.66	5.48±1.39
18	Qanas super	408.49±25.05	12.06±4.14	15.04±2.79
19	EDTA iron sodium salt(Hunan, China)	374.27±23.53	7.11±3.12	7.90±1.99
20	Master Power Phosphoric Acid	349.60±22.34	7.38±3.12	8.72±2.05
21	NPK 20-20-20(Combe star) China	275.68±18.93	26.91±5.69	8.43±1.92
	Mix	495.04±19.31	26.91±5.69	21.01±2.93
	Min	40.99±6.11	2.12±1.09	
	Average	112.751	6.254	4.3694

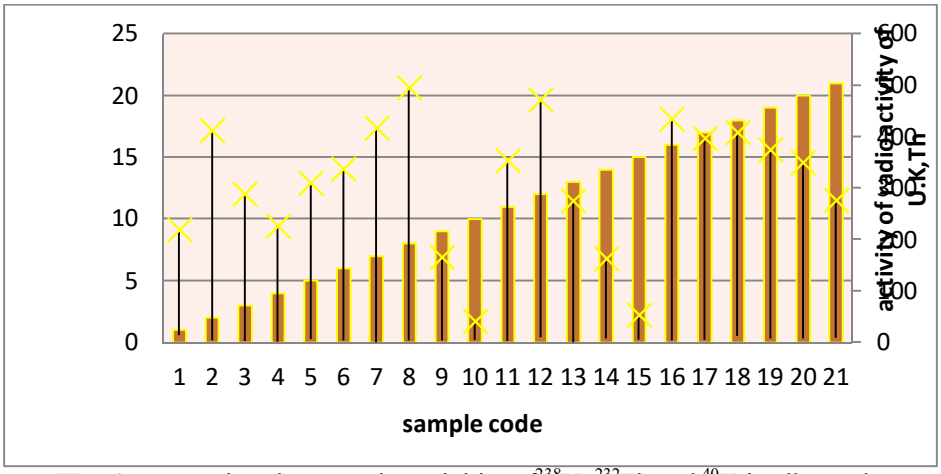

FIG. 1. Comparison between the activities of ^{238}U, ^{232}Th and ^{40}K in all sample.

The radium equivalent activity (Ra_{eq}) was found to be less than the safe levels set by the Organization for Economic Co-operation and Development, ranging from 19.34753 to 72.78399 Bq/kg equivalent in all regions (ECD). For the conversion of the absorbed dose rate into the effective dose equivalent, a conversion factor of 0.7 mSv/y was used. Additionally, an outdoor occupancy factor of 0.2 was employed to estimate the annual effective dose rate equivalent. According to Table 2, the average absorbed dose rates for indoor (D_{in}) and outdoor (D_{out}) exposure were 6 nGy/h and 12.90 nGy/h, respectively. Outdoor exposure values ranged from 16.51 to 69.56 nGy/h. These figures are slightly below the global average absorbed dose rate of 55 nGy/h.

TABLE 2. Absorbed dose rate (AD) indoor and outdoor (nGy/h), annual effective dose (AEDE) indoor, outdoor, and total dose ($mSvy^{-1}$) in the studied types of plant fertilizer and pesticides samples.

No.	Ra_{eq} (Bq/kg)	Absorbed dose rate(AD) (nGy/h)		Annual effective dose equivalent (AEDE) ($mSvy^{-1}$)		$AEDE_{total}$ (mSv/y)
		$AD_{outdoor}$	AD_{indoor}	OUTDOOR	INDOOR	
1	55.46955	25.61478	50.17775	0.031414	0.246152	50.20916
2	48.30415	24.31216	47.76055	0.029816	0.234294	47.79037
3	30.7745	15.68917	30.5969	0.019241	0.150096	30.61614
4	31.28008	15.43489	30.70544	0.018929	0.150629	30.72437
5	60.23962	28.263	54.03176	0.034662	0.265058	54.06642
6	48.36638	23.72244	47.01714	0.029093	0.230647	47.04623
7	42.2959	21.82795	43.1459	0.02677	0.211657	43.17267
8	48.81888	25.19709	48.95984	0.030902	0.240177	48.99074
9	21.21901	10.511	20.46503	0.012891	0.100393	20.47792
10	19.34753	8.575053	16.51079	0.010516	0.080995	16.52131
11	35.44373	18.22023	35.27489	0.022345	0.173045	35.29724
12	72.78399	35.31778	69.55977	0.043314	0.341232	69.60308
13	30.24038	15.41952	30.61454	0.01891	0.150183	30.63345
14	38.16853	17.76847	35.04749	0.021791	0.171929	35.06928
15	21.08828	9.418028	18.14884	0.01155	0.089031	18.16039
16	47.36808	24.01145	46.50084	0.029448	0.228115	46.53029
17	60.84231	29.61853	58.82523	0.036324	0.288573	58.86155
18	65.02093	31.30115	60.72689	0.038388	0.297902	60.76528
19	47.22579	23.43912	45.54707	0.028746	0.223436	45.57582
20	46.7688	23.01928	44.6992	0.028231	0.219276	44.72743
21	60.19226	28.27819	56.36028	0.03468	0.276481	56.39496
Mix	72.78399	35.31778	69.55977	0.043314	0.341232	69.60308
Min	19.34753	8.575053	16.51079	0.010516	0.080995	16.52131
Average	13.5432	6.573181	12.89831	0.008061	0.063274	12.90637
Worldwide [32]	370	55	84	0.07	0.34	≤1

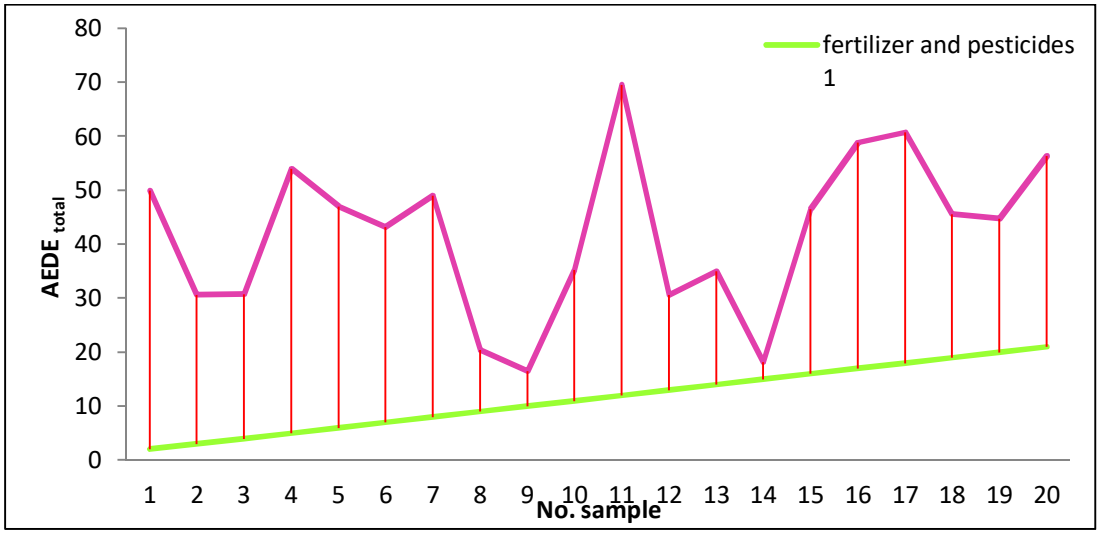

FIG. 2. Relationship between $AEDE_{total}$ and fertilizer samples

The health risks associated with naturally occurring radionuclides in the test samples were investigated and reported in Table 3. For a sample to be considered safe, both H_{ex} and H_{in} must be numerically less than 1. The representative level index was estimated using Eq. (16). It was noticed that all the obtained values exceeded the safety limits advised by UNSCEAR. Table 3 demonstrates the excess lifetime cancer risk (ELCR), which was computed according to Eqs. (17)-(19). The ELCR for the samples was determined to be 1.480503 times greater than the UNSCEAR safety level. Prolonged exposure to these radionuclides, such as in the case of factory workers or phosphate ore miners, may significantly increase the risk of developing cancer.

TABLE 3. Annual gonadal dose equivalent (AGDE), activity utilization index (AUI) in excessive lifetime cancer risk in soil samples, gonadal dose equivalent (AGDE), and activity utilization index (AUI) in excessive lifetime cancer risk in soil samples.

No.	H_{ex}	H_{in}	I_Y	$ELCR_{out}$	$ELCR_{in}$	$ELCR_{total}$ X10^{-3}	AGDE	AUI
1	0.149817	0.198979	0.221922	0.120944	0.947685	1.068629	183.772	0.358094
2	0.130451	0.159694	0.173341	0.114793	0.902032	1.016826	178.0039	0.18151
3	0.083103	0.094643	0.100029	0.074079	0.57787	0.651949	115.1693	0.098775
4	0.084487	0.115298	0.129676	0.072878	0.57992	0.652798	112.55	0.144213
5	0.162664	0.179935	0.187994	0.133448	1.020474	1.153922	203.4065	0.337577
6	0.130634	0.17512	0.195881	0.112009	0.887992	1.000001	172.7232	0.229524
7	0.114227	0.141821	0.154699	0.103064	0.814878	0.917941	160.7259	0.127676
8	0.131822	0.14312	0.148392	0.118972	0.924683	1.043655	185.4394	0.133311
9	0.057301	0.067085	0.071651	0.049629	0.386514	0.436143	76.6609	0.08786
10	0.052252	0.065441	0.071596	0.040488	0.311832	0.35232	60.8499	0.143923
11	0.095704	0.101434	0.104107	0.086029	0.666221	0.752251	133.9611	0.099554
12	0.196576	0.255846	0.283506	0.166758	1.313745	1.480503	256.461	0.363212
13	0.081674	0.106187	0.117627	0.072805	0.578203	0.651009	113.2577	0.105802
14	0.103094	0.141553	0.159501	0.083896	0.661927	0.745823	127.761	0.240899
15	0.056952	0.071034	0.077605	0.044469	0.342769	0.387238	66.9657	0.152358
16	0.127905	0.138932	0.144078	0.113373	0.878241	0.991614	176.0016	0.155774
17	0.164335	0.225011	0.253326	0.139848	1.111006	1.250854	215.2942	0.305383
18	0.175589	0.208184	0.223395	0.147793	1.146922	1.294715	226.7645	0.325797
19	0.127529	0.146745	0.155713	0.110671	0.860227	0.970899	171.0156	0.19107
20	0.126296	0.146242	0.15555	0.108689	0.844214	0.952903	167.6298	0.201497
21	0.162592	0.235322	0.269262	0.133519	1.064452	1.197971	203.8501	0.372537
Mix.	0.196576	0.255846	0.283506	0.166758	1.313745	1.480503	256.461	0.372537
Min.	0.052252	0.065441	0.071596	0.040488	0.311832	0.35232	60.8499	0.08786
Average	0.036576	0.047676	0.054191	0.031036	0.243605	0.274641	47.885	0.088761
Ref. [33]	0353	0.451	0.60	-	-	0.994	-	-

Fig. 3 shows the change in the lifetime cancer risk in fertilizer samples depending on the ^{238}U concentration. It can be noted that the lifetime cancer risk exceeded the internationally permissible limit in sample 12. Fig. 4 illustrates the excess lifetime cancer risk value levels in fertilizer samples.

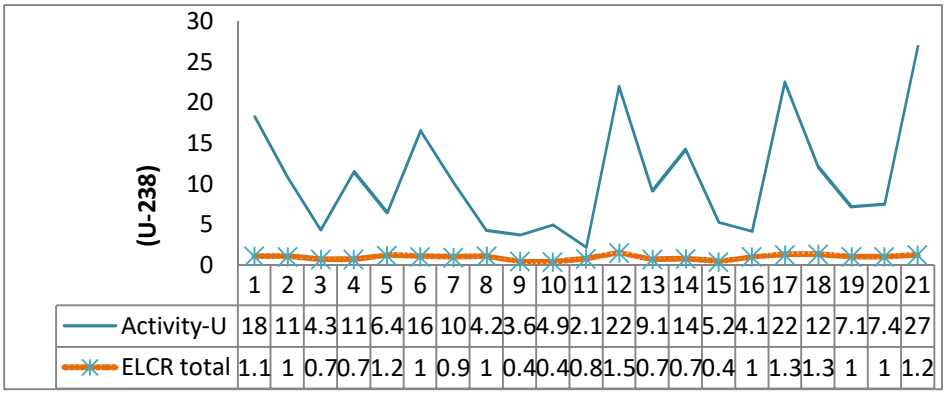

FIG. 3. Relationship between U-238 and $ELCR_{total}$ in all samples of fertilizers and pesticides.

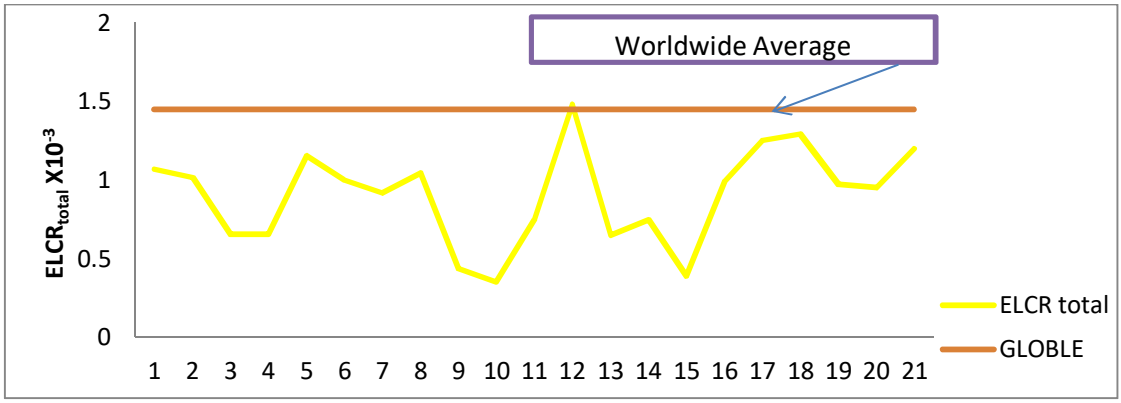

FIG. 4. Excess lifetime cancer risk value levels in fertilizer samples.

Conclusion

The external and internal risks associated with fertilizers are generally less than one. However, caution is advised when using phosphate fertilizers, especially those with high concentrations of phosphates, as they can contain significant amounts of uranium and its decay products. When comparing the results of the current study with those from UNSCEAR, it is clear that all findings are within the internationally permissible limits, although the risk increases with cumulative doses. Based on the results obtained, the following recommendations are made for the use of phosphate fertilizers when planting:

1. The use of chemical or phosphate fertilizers, as well as pesticides, to treat plants should be reduced, because this increases the possibility of contamination with radioactive elements.
2. It is preferable to use environmentally friendly fertilizers that do not cause radioactive or chemical pollution, affecting human and animal health and increasing soil, air, and water pollution.

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