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Radiation hazard of Chemical Fertilizers used in Growing Agriculture Crops in Iraq

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Abstract: Sixteen samples of granular chemical fertilizer types (NPK, DAP, and Urea) were collected from Iraqi nurseries and analyzed using NaI(Tl) gamma-ray detector. Three radionuclides K-40, U-238, and Th-232 were detected in the studied samples. All NPK fertilizers have a specific activity of K-40 greater than the standard limits except Some samples (N.1, N.4, and N.5) were less than standard limits, the maximum specific activity of K-40 2768.395 Bq/kg was detected in the Spain sample, and the mean specific activity of K-40 in NPK is much higher than the mean specific activity of other types. For Dap and Urea samples the maximum specific activity of Bi-214 (4.14 Bq/kg) and Tl-208 (0.384982 Bq/kg) were detected in Lebanon and Iranian samples respectively, the Italian and Russian samples respectively for NPK, all were lower than the standard limits. Depending on the measured specific activities, all the mean of the radiation hazard parameters were less than the worldwide average limit except ELCR for some NPK fertilizers, and within the range for other types. This requires that using of NPK fertilizers must be under the radiological control. The study recommends to use the U.A.E fertilizer of NPK fertilizers (N.2), and the Iraqi fertilizer DAP and Urea fertilizers (D.2).
Keywords: Fertilizers, NaI(Tl) detector , radium equivalent ,Radiological parameters, specific activity.

1 Introduction

In the past, the use of fertilizers was very little around the world, at present time it has become an important global event that cannot develop nor be cultivated without the use of these fertilizers [1].

The productivity of the crop is increased with the use of fertilizers, NPK, DAP, and urea for example [2]. Phosphate rock contains the mineral phosphorus, an ingredient used in some fertilizers to help plants grow strong roots [3]. Many tons of fertilizers are used worldwide every year to increase agricultural crop production and land reclamation for cultivation [4, 5].

Human beings are exposed to radioactivity due to the ingestion of plants raised on fertilized soil and inhalation of rock and fertilizer dust. The use of Phosphate containing fertilizers is the main anthropogenic source of the uranium input in the environment (about 73 % of the total input of uranium) [6]. The use of fertilizers has not shown any serious health problems during the previous ages. However, the frequent use of these fertilizers or their incorrect use may lead to very serious health problems for humans and animals [7]

Natural radionuclides, Ra-226, Th-232, and K-40 in fertilized vegetables were measured by [8]. They found that extensive use of fertilizers rich in phosphate may cause an increase in the activity of natural radionuclides [9]. As for the external exposure, it occurs directly by gamma rays as it emits from the radionuclides of the uranium chain (U-238), thorium chain (Th-232) and K-40 that exists inside phosphate rocks, which is the most important element in the fertilizer industry [10]. It has been revealed that soil available nutrients and result components of fertilizers that absorbed by plants may lead to accumulation of dose in various parts of the human body as well as animals when such plants are ingested or contact with fertilizer dust [11]. Due to the lack of micronutrients that are constantly removed from the soil due to agricultural activities; Fertilizers are used. At the same time, the use of such fertilizers is the main anthropogenic source of the uranium input in the environment (about 73 % of the total input of

uranium) [12]. Plants take small part of the radioactivity present in the fertilizers applied to soil [13]. The aim of this study is to investigate the radioactivity of the fertilizers using in fertilizing agricultural crops and calculate the radiation hazard parameters and recognize their

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radioactivity, to recommend use of the safe fertilizers without control, and use the radioactive types under the radiation protection conditions.

2 Experimental part and Calculations

2.1 Collection samples

Sixteen chemical fertilizers samples from different origins of the most common types of fertilizers used in Iraqi farms are collected from different agricultural offices in Iraq. including 9 NPK samples (triple compound contain different values of three elements essential for growth, Nitrogen (N) Phosphorus (P) and Potassium (K)), 4 DAP samples (Di-ammonium phosphate), and 3 samples of Urea.

2.2 NaI(Tl) Detector

NaI(Tl) detector of "3×3" crystal dimension, type (SCIOIX model 51S51), with a digital multichannel analyzer (bright multichannel SPEC model bMCA) of 4096 channels were used with an operating voltage of 695 volts to perform this work. The measuring time used for the samples is 24 h (86,400 s). Table 1 list the type, code, compositions, origin country, and mass of the studied sample.

Three point sources have been used to find the efficiency calibration (Ba-13, Cs-137, and Co-60), These sources are set at 5 cm from the top of the detector for 24 hours and then recording the spectrum to obtain the net peak area under the peak which can be measured from gamma spectrum.

2.3 Calculations

Three natural radionuclides were detected in the studied samples at three gamma energy peak, K-40 radionuclide is detected at 1460.80 keV, Bi-214 radionuclide related to U-238 series at 1764.5 keV, and TI-208 radionuclide related to Th-232 series is detected at 2614.6 keV. The specific activity of the detected radionuclides were analyzed and computed by [14]:

$$S.A = \frac{N}{\varepsilon \times I\gamma \times t \times m} \quad (1)$$

Where N is the net area under the peak, t is the time of measuring (86400s), I is the intensity of the specific energy, ε is the detection efficiency at energy, and m is the mass of sample in kg.

The radiation parameters of the samples; Radium Equivalent Activity (Ra_{eq}), Absorbed Dose Rate (D_γ), out Annual Effective Dose equivalent (AEDE), External and internal Hazard index (H_{in}, H_{ex}), Gamma and alpha indexes (Iγ, Iα), and Excess lifetime cancer risk (E_{LCR}) were calculated from the following relations [15, 16, 17]:

$$Ra_{eq}(Bq.kg^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_K \leq 370 \quad (2)$$

$$D_\gamma(nGy.h^{-1}) = 0.462A_u + 0.621A_{Th} + 0.042A_K \quad (3)$$

$$AEDE(mSv/y) = D_\gamma \times 8760 \times 0.20 \times 0.7 \times 10^{-6} \quad (4)$$

$$H_{ex} = \frac{A_U}{370\,Bq.kg^{-1}} + \frac{A_{Th}}{259\,Bq.kg^{-1}} + \frac{A_K}{4810\,Bq.kg^{-1}} \leq 1 \quad (5)$$

$$H_{in} = \frac{A_U}{185\,Bq.kg^{-1}} + \frac{A_{Th}}{259\,Bq.kg^{-1}} + \frac{A_K}{4810\,Bq.kg^{-1}} \leq 1 \quad (6)$$

$$I\gamma = \frac{A_U}{300\,Bq.kg^{-1}} + \frac{A_{Th}}{200Bq.kg^{-1}} + \frac{A_K}{3000\,Bq.kg^{-1}} \leq 1 \quad (7)$$

$$I\alpha = A_{Ra}/200 \leq 1 \quad (8)$$

$$ELCR = AEDE \times DL \times RF \quad (9)$$

Where Au, Ath and Ak are the specific activity of U-238, Th-232 and K-40 respectively, and DL is the duration of life (70 years), RF is the risk factor (0.05 Sv-1) [18].

3 Results and Discussion

Tables 2 and 3 illustrate the specific activities of the three natural radionuclides detected in NPK fertilizer group, DAP and Urea fertilizer group respectively, results showed that the mean specific activity of K-40 radionuclide is very high relative to Bi-214 and TI-208 radionuclides in NPK fertilizers, while in DAP and Urea fertilizers the mean specific activity of K-40 radionuclide was not greater than Bi-214 and TI-208 radionuclides.

It's also clear that the mean specific activity of K-40 in NPK group (1271.19±340.6 Bq/kg) is much higher than its amount in Dap and Urae group (10.39±2.37 Bq/kg), because potassium is one of the essential element of NPK [19]. For NPK fertilizers the maximum specific activity of K-40 is measured in the samples having the maximum value of K, such as Spain sample N.6 (2768.395 Bq/kg) has 36% of K, the Norwegian sample N.7 (2503.25 Bq/kg) has 18% K Percentage, and in the Jordanian sample N.3 (2180.995 Bq/kg) has 30% K percentage.
However the maximum specific activity of K-40 (20.19139 Bq/kg) in Dap and Urea group is detected in the Jordanian sample D.6; it's much lower than the minimum specific activity of K-40 in NPK (134.2946 Bq/kg) that detected in the U.A.E sample N.2.
For Bi-214 its maximum and minimum specific activity is found in Dap and Urea group samples, the maximum is in Lebanon sample D.1 which is 4.148675 Bq/kg, and the

minimum is in the Iraqi sample D.2 which is 0.52 Bq/kg. For TI-208 the maximum specific activity is found in Russian sample N.1 which is 1.07 Bq/kg, whilst the minimum is found in the Norwegian sample N.7 which is 0.056 Bq/kg. The specific activities of K-40, U-238, and Th-232 for NPK group and Dap and Urea group are shown in figures 2 and 3 respectively.

Table 1: The name, code, origin country, and mass of the fertilizers sample.

No.	type	Code	Compositions	Origin	Mass(gm)
1	NPK	N.1	N 15%, P 15%, K 15%	Russia	1061
2	NPK	N.2	N 17%, P 17%, K 17%	U.A.E	1004
3	NPK	N.3	N 10%, P 20%, K 30% +3mgo	Jordan	1009
4	NPK	N.4	N 15%, P 15%, K 15%	U.A.E	997
5	NPK	N.5	Npk +TE	Jordan	1002
6	NPK	N.6	N 12%, P 12%, K 36%+TE	Spain	899
7	NPK	N.7	N 12%, P 11%, K 18% yara mila	Norway	986
8	NPK	N.8	N 13%, P 25%, K 14% +3.3 Mgo+1.6 Zn	Spain	920
9	NPK	N.9	N 15%, P 15%, K 15%	Italian	1032
10	DAP	D.1	Supper phosphate P_2O_5	Lebanon	1068
11	Urea	D.2	Di-ammonium phosphate.1	Iraq	997
12	Urea	D.3	Urea 1	Iraq	600
13	Urea	D.4	Urea 2	Iran	643
14	Urea	D.5	Urea 3	Qatar	550
15	DAP	D.6	Di-ammonium phosphate.2	Jordan	888
16	DAP	D.7	Di-ammonium phosphate.3	Jordan	970

Table 2: The specific activities of Bi-214, Tl- 208 and K-40 in NPK samples.

No.	Code	specific activity Bq/kg		
		K-40	U-238 series Bi-214	Th-232 series Tl-208
1	N.1	1121.576	3.5862	1.078656
2	N.2	134.2946	3.866913	0.250966
3	N.3	2180.995	1.937463	0.129091
4	N.4	188.159	1.316895	0.302512
5	N.5	170.7865	3.118056	0.060895
6	N.6	2768.395	1.08547	0.360809
7	N.7	2503.25	2.452148	0.056432
8	N.8	1022.757	3.176818	0.133675
9	N.9	1350.548	7.445846	0.291027
Max.		2768.395	7.445846	1.078656
Min.		134.2946	1.08547	0.056432
Mean±S.E		1271.19±340.6	3.109±0.63	0.29±0.104
Recommended limits [20]		400	35	30

S.E = $\sigma/\sqrt{n}$, σ is the standard deviation, n is the sample number, and S.E is Systematic Error [12].

By comparing the specific activity of our results with the specific activity of the previous studies listed in table 4, it is clear that the range of K-40 in the previous studies is (8-4000 Bq/kg), but the current study range of NPK fertilizers for K-40 is (134.2946 -2768.395 Bq/kg) shown in table 2 is within the range of previous studies, and within the range of other fertilizers (2.317- 20.191 Bq/kg) shown in table 3.

The range of Bi-214 in the previous studies (30.5-1600 Bq/kg) is slightly higher than the range determined in the current study for NPK group (1.08-7.44 Bq/kg), DAP and Urea group (0.52-4.14 Bq/kg). The range of Tl-208 in the previous studies (2-327.1Bq/kg) is slightly higher than the range determined in the current study for NPK fertilizers group (0.056432-1.078656 Bq/kg), DAP and Urea fertilizers group (0.072616-0.384982 Bq/kg).

Table 3: The specific activities of Bi-214, Tl- 208 and K-40 in DAP and urea samples

No.	Code	specific activity Bq/kg		
		K-40	U-238 series Bi-214	Th-232 series Tl-208
1	D.1	13.42721	4.148675	0.253984
2	D.2	2.590955	0.520934	0.072616
3	D.3	11.35502	2.207059	0.116975
4	D.4	2.317602	1.52014	0.384982
5	D.5	12.0168	1.328488	0.320171
6	D.6	20.19139	1.607505	0.323624
7	D.7	10.88571	2.076886	0.308977
Max.		20.19139	4.148675	0.38498
Min.		2.317602	0.520934	0.07261
Mean±S.E		10.39±2.37	1.91±0.42	0.25±0.04
Recommended limits [20]		400	35	30

Table 4: the specific activity of the radionuclides detected in fertilizers used in different counties.

Country	Mean specific activity (Bq/kg)			Ref.
	K-40	Ra-226	Th-232	
Nigeria	2301.8	30.5	327.1	(Faweya, et al, 2017, [21])
Pakistan	221	526	50	(Khan et al, 1998, [22])
Germany	720	520	15	(Pfister, et al, 1976, [23])
Finland	3200	54	11	(Mustonen, (1985), [24])
Egypt	182	571	19	(Hassan, et al, 2017, [10])
Japan	1500	325	9	
Saudi Arabia	2453	64	17	(Alharbi, 2013, [25])
India	2624	120	65	(Pooja Chauhan, et al, 2013, [26])
Italy	4000	120	3.5	(Righi, et al, 2005, [27])
Jordan	8	1044	2	(Olszewska-Wasiolek, 1995, [28])
Morocco	10	1600	20	(Guimond, 1990, [29])
USA	200	780	49	(Guimond and Hardin, 1989, [30])

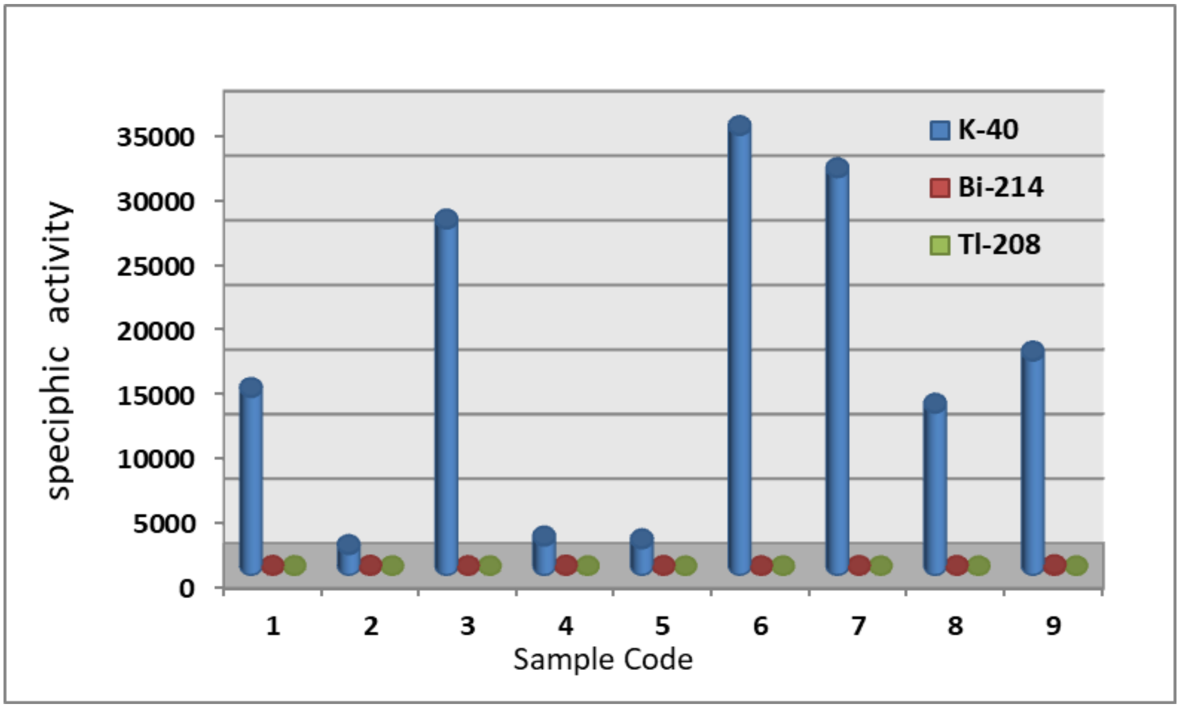

Fig. 1: the specific activities of K-40, U-238, and Th-232 in NPK fertilizer samples.

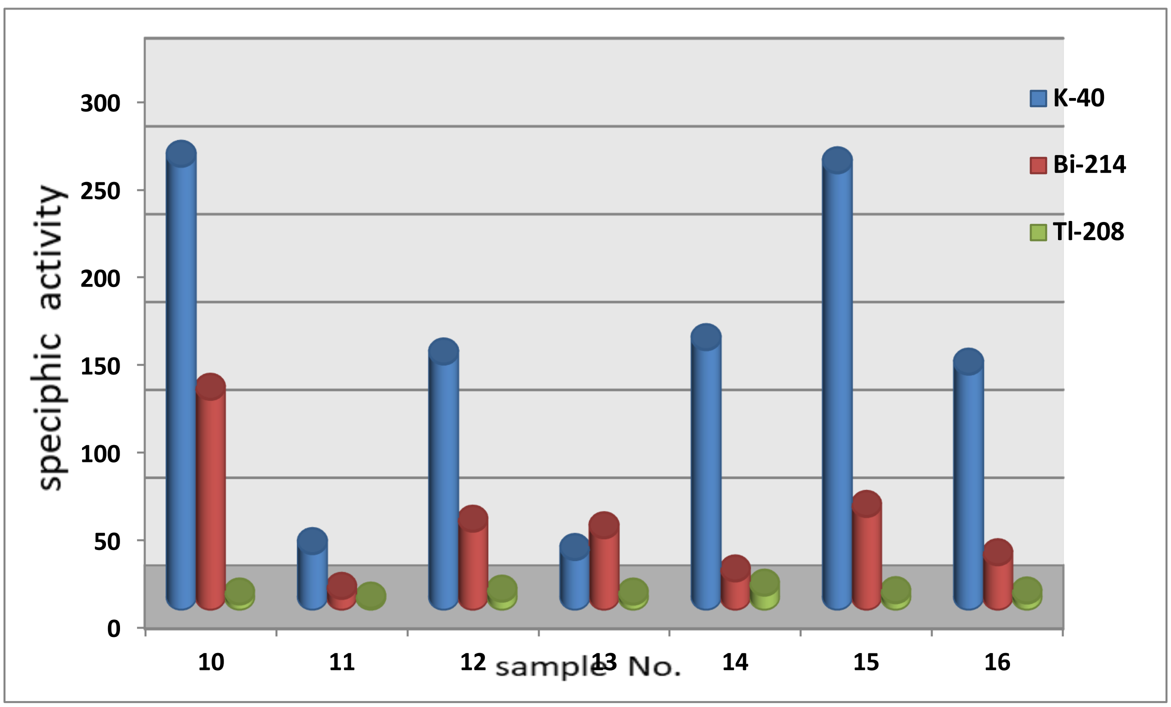

Fig. 2: the specific activities of K-40, U-238, and Th-232 in DAP and Urea fertilizer sample.

Table 5 shown the radiometric parameters of NPK samples, these parameters depends on the specific activity of the detected radionuclides, N.6 has the maximum radiometric parameters because it has the highest specific activity of K-40, except for I_α because it depends on the specific activity of U-238 which has the minimum value. However, the maximum value of I_α is found at N.9 sample because it has the maximum specific activity of U-238. The minimum values of other radiometric parameters (except I_α) are calculated for N.2 which has the lowest specific activity of

K-40. The mean values of the radiometric parameters for NPK type all are less than the permissible limits except I_α and AEDE. Table 6 shows the radiometric parameters of DAP and Urea samples, the maximum values are calculated for D.1 that has the highest specific activity of K-40 and Bi-214. All mean radiation hazards parameters for this type of fertilizers are lower than the permissible limits.

Table 5: the radiometric parameters of the NPK fertilizer samples.

No	Code	Ra_{eq} Bq/kg	H_{in}	Hex	I_γ	I_α	Dγ nGy/h	AEDE mSv/y	AGDE (mSv/y)	ELCR x10^{-3}
1	N.1	91.49	0.256	0.247	0.391	0.017	49.43	0.062	0.367	0.212
2	N.2	14.56	0.049	0.039	0.059	0.019	7.58	0.009	0.055	0.032
3	N.3	170.05	0.464	0.459	0.734	0.009	92.57	0.113	0.691	0.397
4	N.4	16.23	0.047	0.043	0.069	0.006	8.69	0.0107	0.064	0.038
5	N.5	16.35	0.052	0.044	0.068	0.015	8.65	0.0106	0.063	0.037
6	N.6	214.76	0.582	0.579	0.928	0.005	116.99	0.1434	0.874	0.502
7	N.7	195.28	0.533	0.527	0.84	0.012	106.30	0.1303	0.793	0.456
8	N.8	82.12	0.230	0.221	0.35	0.015	44.50	0.0545	0.331	0.191
9	N.9	111.85	0.322	0.302	0.47	0.037	60.34	0.074	0.448	0.259
Max.		214.76	0.58	0.57	0.928	0.037	116.99	0.143	0.87	0.50
Min.		14.56	0.049	0.039	0.059	0.005	7.58	0.009	0.055	0.032
Mean±S.E		101.4±26.08	0.27±0.071	0.26±0.074	0.43±0.11	0.01±0.003	55.01±14.2	0.06 ± 0.017	0.405 ± 0.10	0.23 ± 0.06
Global limit [20]		370	≤1	≤1	≤1	≤1	84	1	0.3	0.29 x10^{-3}

Table 6: the radiometric parameters of the DAP and Urea fertilizer samples.

No	Code	Ra_{eq} Bq/kg	H_{in}	Hex	Iγ	Iα	Dγ nGy/h	AEDEm Sv/y	AGDE (mSv/y)	ELCR x10^{-3}
1	D.1	5.5457	0.026	0.014	0.020	0.020	2.6383	0.0032	0.01809	0.0113
2	D.2	0.8242	0.003	0.002	0.003	0.002	0.3945	0.0004	0.00272	0.0016
3	D.3	3.2486	0.015	0.008	0.012	0.011	1.5692	0.0019	0.01087	0.0067
4	D.4	2.2491	0.010	0.006	0.008	0.007	1.0387	0.0012	0.00703	0.0044
5	D.5	2.7116	0.011	0.007	0.010	0.006	1.3172	0.0016	0.00921	0.0056
6	D.6	3.6250	0.013	0.010	0.014	0.008	1.7916	0.0021	0.01266	0.0076
7	D.7	3.3569	0.014	0.009	0.012	0.010	1.6085	0.0019	0.01112	0.0069
Max.		5.5457	0.026	0.014	0.020	0.020	2.6383	0.0032	0.01809	0.0113
Min.		0.8242	0.003	0.002	0.003	0.002	0.3945	0.0004	0.00272	0.0016
Mean±S.E		3.08 ± 0.54	0.01±0.002	0.008±0.001	0.01±0.001	0.009±0.002	1.47±0.26	0.0017±0.0003	0.0102±0.001	0.006±0.001
Global limit [20]		370	≤1	≤1	≤1	≤1	84	1	0.3	0.29 x10^{-3}

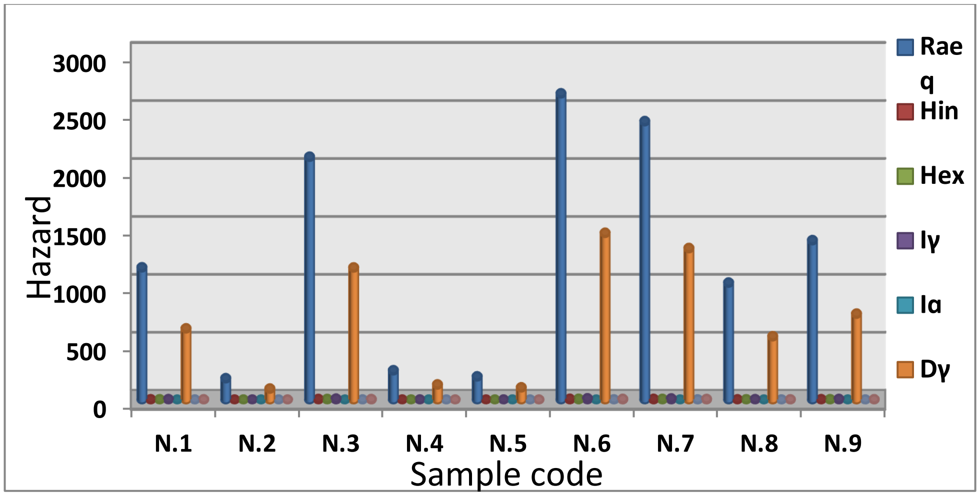

Fig..3: the radiometric parameters of the NPK fertilizer samples.

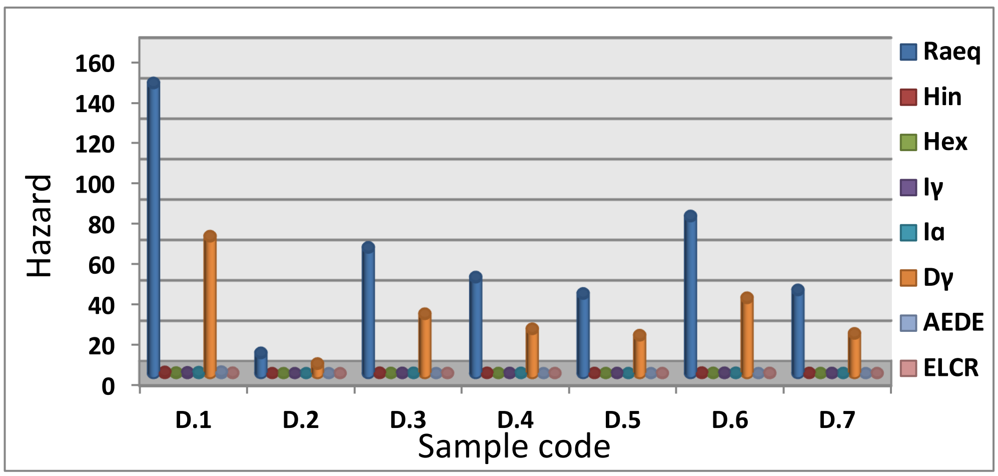

Fig. 4: the radiometric parameters of the DAP and Urea fertilizer samples.

4 Conclusions

1. The mean specific activity of K-40 radionuclide is very high relative to Bi-214 and TI-208 radionuclides in NPK fertilizers.
2. The mean specific activity of K-40 in NPK fertilizers is higher than the mean specific activity of K-40 in other types, because of the difference in their components.
3. The highest specific activity of K-40 in NPK fertilizers are investigated in the samples having the highest value of K (N.6, N.7, and N.3). So, we recommend taking safety when using these types.
4. All the mean of hazard parameters for NPK are less than the worldwide average values except ELCR in samples (N.6, N.7, and N.3). And lower than the word wide average values for other types.
5. Comparing with previous studies, the mean specific activity of K-40 in NPK fertilizers is within the range of its specific activity in other studies. However, for Bi-214 and Tl-208 their mean specific activity is less than the ranges of all fertilizers in previous studies
6. Based on radium equivalent calculations, study recommends using the U.A.E fertilizer (N.2) of NPK fertilizers, and the Iraqi fertilizer (D.2) of other types.

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