Determination of Natural Radioactivity in Some Commercial Phosphate Fertilizer Samples

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Abstract: Determinations of specific activity concentrations for Uranium-238, Thorium-232 and Potassium-40 in 10 commercial phosphate fertilizers samples from various countries (Iraq, Iran, Jordan, Kingdom of Saudi Arabia, Italy and Turkey) were accomplished by the use of high purity germanium detector device. Radiation hazard indices [(Annual Effective Dose Equivalent) $_{in}$, (Annual Effective Dose Equivalent) $_{out}$, Equivalent Activity of Radium, Dose Rate of Absorbed Gamma, Activity Concentration Index , Internal and External Hazard Indices and (Excess Life Time Cancer Risk) $_{out}$] were also investigated. The acquired outcomes were fewer than the advisable limits identified by (UNSCEAR, 2000).
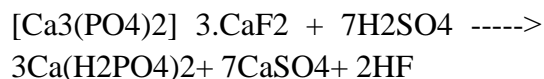
*Keyword*: Radiation hazard indices, fertilizers samples, (HPGe) detector.
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1. Introduction

There are two kinds of phosphate fertilizer [1]:

1.1- Single super phosphate (SSP), which is made according to the following formula:

[Ca3(PO4)2] 3.CaF2 + 7H2SO4 -----> 3Ca(H2PO4)2+ 7CaSO4+ 2HF

This reaction is done by treating the smoothly crushed phosphate rocks with the adequate quantity and concentration of sulfuric acid.

1.2- Triple super phosphate (TSP) (Concentrated), which is made according to the following formula:

[Ca3(PO4)2] 3.CaF2 +14 H3PO4 +10H2O--->10Ca(H2PO4)2. H2O + 2HF
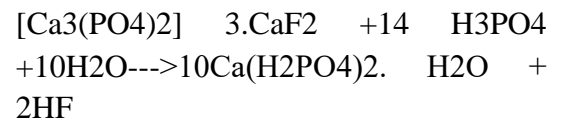
The production of industrial minerals such as phosphates includes production stages that creates residues comprising either the uranium or the thorium radionuclides series or both of them, example of that the phosphogypsum from the phosphate [2].

The present work is concerned with the evaluation of the specific activity concentration of (^{232}Th, ^{238}U and ^{40}K) and their associated hazard indices for some selected commercial phosphate fertilizers samples obtained mostly from

various countries by using a (HPGe) detector device.

2. Materials and Method:

Samples collection and preparation

Ten commercial phosphate fertilizers samples were chosen randomly from the Iraqi markets. These fertilizers samples were made in different factories and countries, three of them were made in Iraq and the other seven samples were made in forging countries. Also these samples were different in kind, i.e. some of them were single super phosphate (SSP) and some of them were triple super phosphate (TSP) while the rest of the samples were a mixed of the two previous kinds.

The samples were bruised to fine squashed by an electrical quern. Samples with 300 μm grain size were secluded by the use of a fine bolter. After that the samples were kept in a 1-kg Marinelli beaker. The beaker stored for 30 days before the test so as to accomplish a temporal balance between ^{238}U and ^{232}Th with their dynasties.

The standardization of energy was gained by using a principle source of Marinelli beaker of ^{152}Eu with energies (964.0, 1408.0, 344.3, 411.1, 444.6, 778.9, 121.8, 1112.0, 1085.8 and 244.7 keV). Efficiency standardization of the (HPGe) detector was gained by the use of the same principle source of ^{152}Eu.

Concentrations of the specific activity for the radionuclides (A) in phosphate fertilizers samples were concluded through the relation [3]:

$$A = \frac{N-BG}{T.I_\gamma(E_\gamma).\varepsilon(E_\gamma).M} \qquad (1)$$

Where:
A: represents the radioactive elements specific activity.
N: area of the photo peak at energy ($E\gamma$).
B.G: net peak area of the background.
$\varepsilon(E\gamma)$: detector efficiency.
$I\gamma (E\gamma)$: abundance.
M: mass of the sample (kg).
T: duration of test procedure (7200 s).

3. Indices of Radiation Hazard:

3.1- Equivalent Activity of Radium (Raeq) [3,4,5]

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

Where A_k, A_U and A_{Th} are concentrations of the specific activity for ^{40}K, ^{238}U and ^{232}Th respectively in (Bq/kg) units.

3.2- Dose Rate of Absorbed Gamma (Dɣ) [5,6]

$$D\gamma = 0.0417 \; A_K + 0.604 \; A_{Th} + 0.462 \; A_U \qquad (3)$$

3.3- The Annual Effective Dose Equivalent (AED$_{in}$, AED$_{out}$) [4,7,8]

$$(AEDE)_{in} = D\gamma \; (nGy/h) \times 0.80 \times (0.7 \; Sv/Gy) \times 8760 \; h/y \times 10^{-6} \qquad (4)$$

$$(AEDE)_{out} = D\gamma \; (nGy/h) \times 0.20 \times (0.7 \; Sv/Gy) \times 8760 \; h/y \times 10^{-6} \qquad (5)$$

3.4- Internal and External Hazard Indices (H$_{in}$, H$_{ex}$) [4,5,7,9]

$$H_{in} = \frac{A_K}{4810} + \frac{A_{Th}}{259} + \frac{A_U}{185} \leq 1 \qquad (6)$$

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

3.5- Activity Concentration Index (I_γ) [5,7,10]

$$I_\gamma = \frac{A_K}{3000} + \frac{A_{Th}}{200} + \frac{A_U}{300} \qquad (8)$$

3.6- Excess Lifetime Cancer Risk (ELCR)

(ELCR) handles with the prospect of having cancer over a lifetime at a given radiation exposure level. We have attempted to determine it using the following relation [11.12,13]:

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

$(AEDE)_{out}$ were adopted for the calculation, (DL) is the average duration of life (70 years) and (RF) is the risk factor (Sv/Y), i.e. fatal cancer risk per Sievert. For stochastic effects, ICRP (International Committee on Radiological Protection) uses (RF) as 0.05 Sv^{-1} for the public exposure [11,12,13].

4. Results and Conclusions

Table (1) includes the outcomes of the present test. It is clear that the specific activity of (^{238}U) was diversified from (13.42 Bq/kg) (Jordan-1origin) to (33.56 Bq/kg) (Turkey-2 origin), with a mean value of (24.936±5.357 Bq/kg), which is fewer than the advisable value (35 Bq/kg) identified by (UNSCEAR, 2000), see Figure (1).

Specific activity of (^{232}Th) was diversified from (18.25 Bq/kg) (Turkey-1 origin) to (33.12 Bq/kg) (Turkey-2 origin), with a mean value of (25.986±4.217 Bq/kg), which is fewer than the advisable value (30 Bq/kg) identified by (UNSCEAR, 2000), see Figure (1).

Specific activity of (^{40}K) was diversified from (135.53Bq/kg) (Turkey-1 origin) to (241.84 Bq/kg) (Iraq-3 origin), with a mean value of (191.146±35.298 Bq/kg), which is fewer than the advisable value (400 Bq/kg) identified by (UNSCEAR, 2000), see Figure (1).

Radium equivalent activity (Raeq) was diversified from (56.343Bq/kg) (Turkey-1 origin) to (98.066 Bq/kg) (Turkey-2 origin), with a mean value of (76.8142±13.237 Bq/kg), which is fewer than the advisable value (370 Bq/kg) identified by (UNSCEAR, 2000), see Figure (2).

Absorbed dose rate (D_γ) was diversified from (25.827 nGy/h) (Turkey-1 origin) to (44.794 nGy/h) (Turkey-2 origin), with a mean value of (35.1868±6.023 nGy/h), which is fewer than the advisable value (55 nGy/h) identified by (UNSCEAR, 2000), see Figure (2).

The (AED)in was diversified from (0.127 mSv/y) (Turkey-1 origin) to (0.22 mSv/y) (Turkey-2 origin), with a mean value of (0.1728±0.02956 mSv/y), which is fewer than the advisable value (1 mSv/y) identified by (UNSCEAR, 2000), see Figure (3).

(AED)out was diversified from (0.032 mSv/y) (Turkey-1 origin) to (0.055 mSv/y) (Turkey-2 origin), with a mean value of (0.0431±0.00752 mSv/y), which is fewer than the advisable value (1 mSv/y)   identified by (UNSCEAR, 2000), see Figure (3).

Activity concentration index (Iγ) was diversified from (0.405) (Turkey-1 origin) to (0.703) (Turkey-2 origin), with a mean value of (0.5536±0.09392), which is fewer than the advisable value of (1) identified by (UNSCEAR, 2000), see Figure (4).

The internal hazard Index (Hin) was diversified from (0.206) (Turkey-1 origin) to (0.356) (Turkey-2 origin), with a mean value of (0.2749±0.05028), which is fewer than the advisable value (1) identified by (UNSCEAR, 2000), see Figure (4).

The external hazard Index (Hex) was diversified from (0.152) (Turkey-1 origin) to (0.265) (Turkey-2 origin), with a mean value of (0.2075±0.0358), which is fewer than the advisable value (1) identified by (UNSCEAR, 2000), see Figure (4).

Values of excess lifetime cancer risk were diversified from (0.113×10^{-3}) (Turkey-1 origin) to (0.193×10^{-3}) (Turkey-2 origin), with a mean value of $(0.152\pm0.0264)\ \times10^{-3}$, which is fewer than the advisable value of $(0.29\ \times10^{-3})$ [11.12.13], see Figure (5).

5. Conclusions

Obtained results of the present work regarding values of the specific activity for $(^{238}U,\ ^{232}Th$ and $^{40}K)$ and indices of radiation hazards [(AED) in ,(AED) out ,Raeq ,D$_{\gamma}$, ELCR, Iγ , Hin and Hex] , were fewer than the allowed limits, and there is no real health risk when using the phosphate fertilizers samples chosen in the present work.
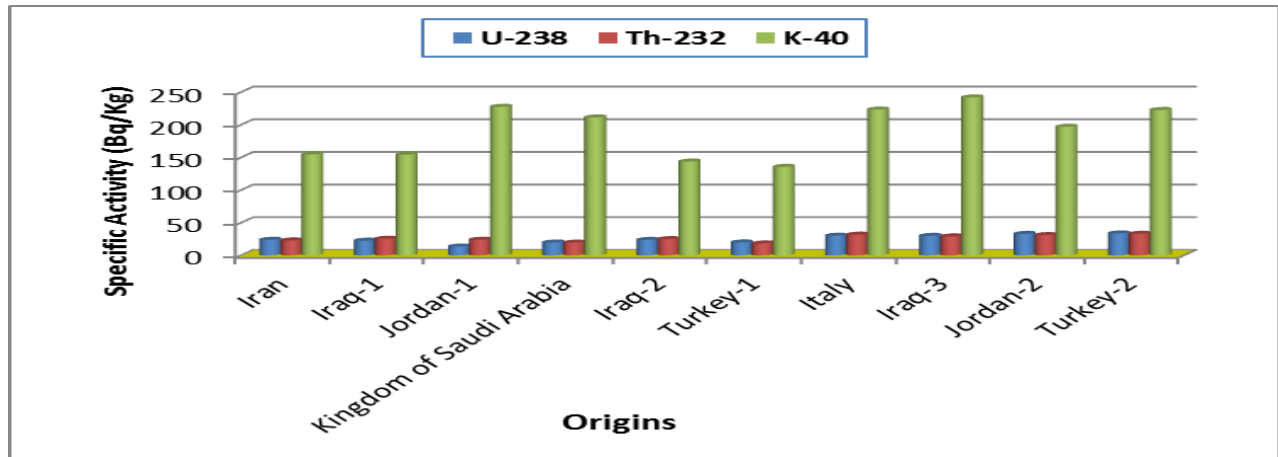
Fig.(1): specific activity of (^{238}U, ^{232}Th and ^{40}K) for all phosphate fertilizers samples.
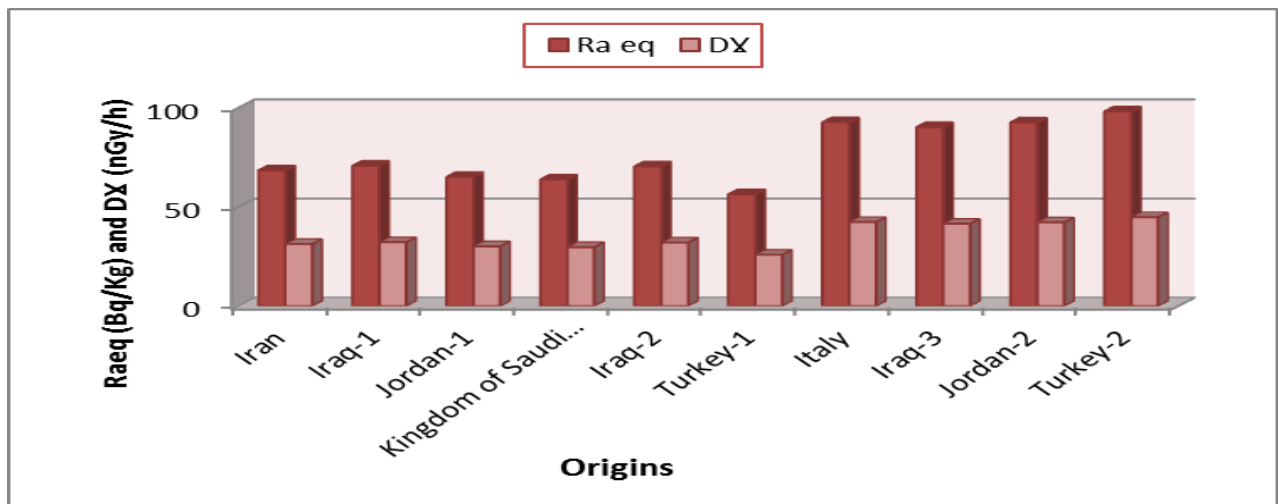

Fig.(2): equivalent activity of radium and dose rate of absorbed gamma for all phosphate fertilizers samples.
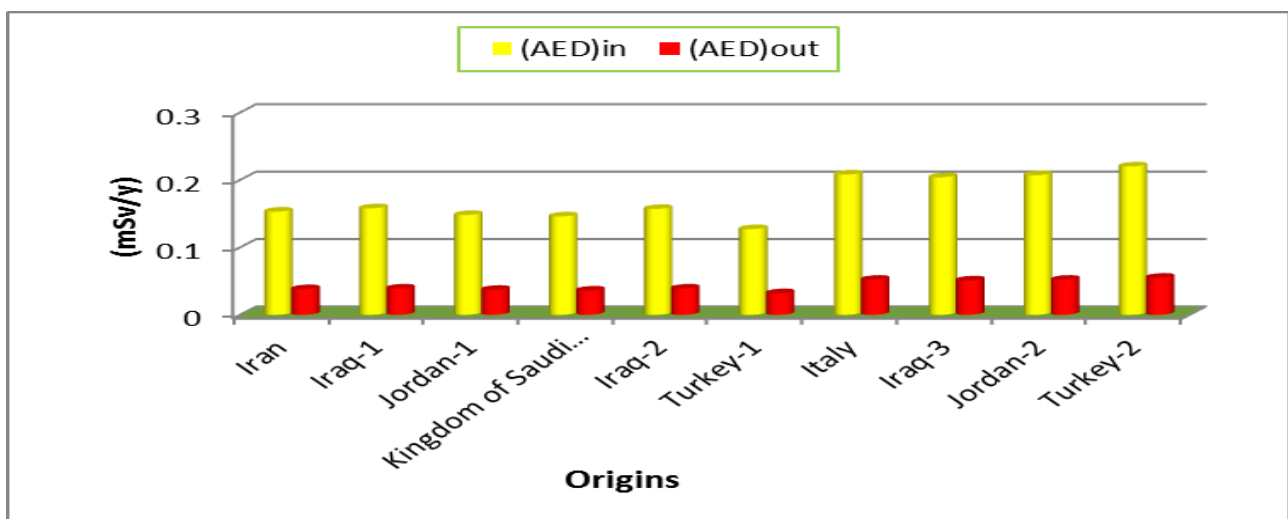

Fig.(3): annual effective dose equivalents (indoor and outdoor) for all phosphate fertilizers samples.
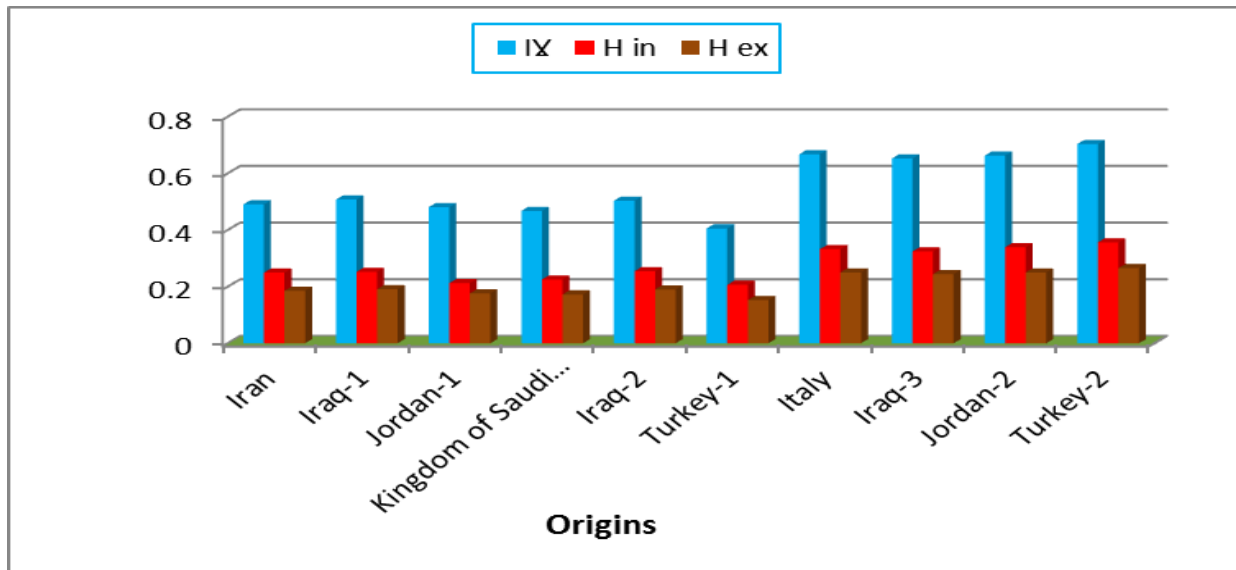
Fig. (4): index of activity gamma, internal and external hazard index for all phosphate fertilizers samples.
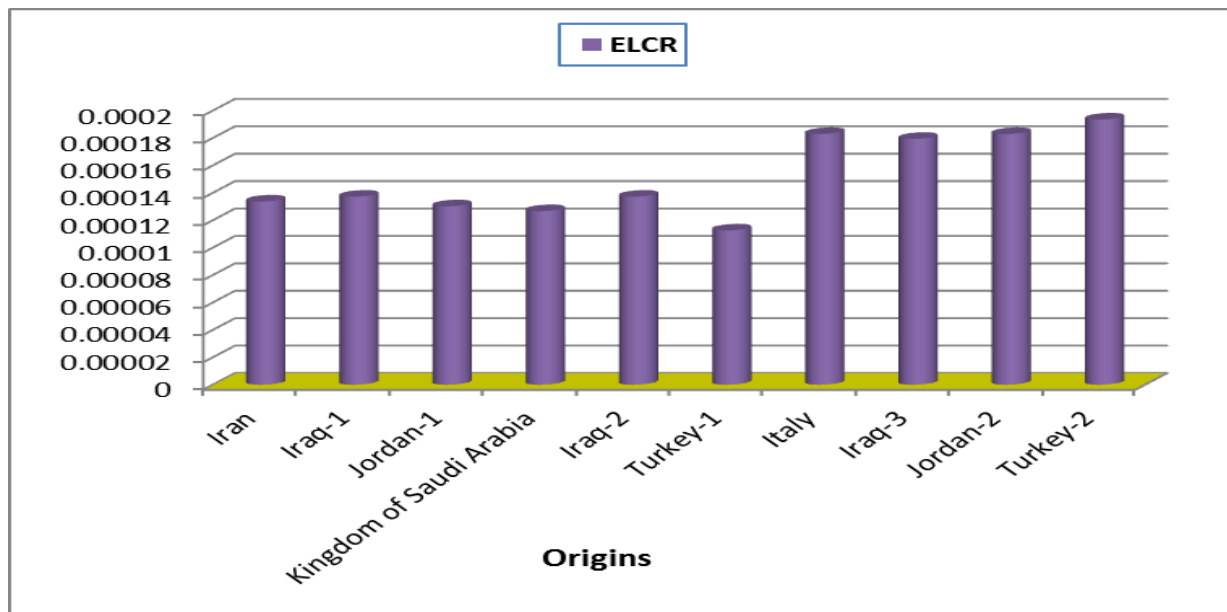

Fig. (5): Excess lifetime cancer risk for all phosphate fertilizers samples.

Table (1): outcomes of the present work for all phosphate fertilizers samples.

| Kind of fertilizer | Origin | ^{238}U (Bq/Kg) | ^{232}Th (Bq/Kg) | ^{40}K (Bq/Kg) | Ra_{eq} (Bq/Kg) | D_γ (nGy/h) | (AED) (mSv/y) | | I_γ | H_{in} | H_{ex} | ELCR $*10^{-3}$ |
							(AED_{in})	(AED_{out})				
M.F	Iran	23.810	22.860	154.540	68.399	31.252	0.153	0.038	0.490	0.249	0.185	0.134
M.F	Iraq-1	22.470	25.430	154.340	70.719	32.177	0.158	0.039	0.507	0.252	0.191	0.137
M.F	Jordan-1	13.420	23.860	227.480	65.056	30.097	0.148	0.037	0.480	0.212	0.176	0.13
T.S.P	Kingdom of Saudi Arabia	19.660	19.490	211.220	63.795	29.663	0.146	0.036	0.467	0.225	0.172	0.127
T.S.P	Iraq-2	23.660	24.940	143.680	70.388	31.986	0.157	0.039	0.503	0.254	0.190	0.137
T.S.P	Turkey-1	19.810	18.250	135.530	56.343	25.827	0.127	0.032	0.405	0.206	0.152	0.113
S.S.P	Italy	30.120	31.710	223.250	92.656	42.378	0.208	0.052	0.667	0.332	0.250	0.183
S.S.P	Iraq-3	29.910	29.120	241.840	90.173	41.492	0.204	0.051	0.652	0.324	0.244	0.179
S.S.P	Jordan-2	32.940	31.080	196.920	92.547	42.202	0.207	0.052	0.662	0.339	0.250	0.183
S.S.P	Turkey-2	33.560	33.120	222.660	98.066	44.794	0.220	0.055	0.703	0.356	0.265	0.193
Ave.		24.936± 5.357	25.986±4 .217	191.146± 35.298	76.8142± 13.237	35.1868± 6.023	0.1728±0 .029	0.0431±0.0 0752	0.5536 ±0.093	0.2749 ±0.050	0.2075 ±0.035	0.152± 0.0264
Min.		13.42	18.25	135.53	56.343	25.827	0.127	0.032	0.405	0.206	0.152	0.113
Max.		33.56	33.12	241.84	98.066	44.794	0.22	0.055	0.703	0.356	0.265	0.193
worldwide average [14]		35	30	400	370	55	1	1	1	1	1	0.29 [11,12, 13]

Where:

M.F: mixed fertilizer.

T.S.P: triple super phosphate fertilizer.

S.S.P: single super phosphate fertilizer.

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