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Evaluation of the activity levels and the radiological hazards associated with the imported chemical fertilizers used in Libya

Asma Mohammed A. AL-abrdi [1] Areej Hazawi [1] Mbrouka A.Abd-Alhamid[2] and Rasha Yousif M. Elfallah [2]

[1]*Physics Department, Faculty of Science, Omar Al-Mukhtar University*

[2] *Physics Department, Faculty of Science,Darna University.*

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A B S T R A C T

This study provides a preliminary assessment of the levels of natural radioactivity in ten types of local chemical fertilizers collected from markets in Libya. The primary objective was to determine the activity concentrations of the naturally occurring radionuclides ^{226}Ra, ^{232}Th and ^{40}K and subsequently evaluate the associated radiological hazard indices. Measurements were performed using gamma spectrometry with NaI (Tl) 3″ × 3″ detector. The determined activity concentration ranges were: ^{226}Ra, ^{232}Th and ^{40}K were 65.27 ± 3.9–97.46 ± 5.09, 70.26 ± 4–86.3 ± 2.36, and 76.77 ± 7.43–131.79 ± 8.8 Bq kg^{-1}, respectively Furthermore, the calculated radiological parameters showed the following ranges: Radium Equivalent Activity from 174.57 to 228.75 Bq kg^{-1} Cancer Risk Factor from 0.09 to 0.12 and Absorbed Dose Rate from 77.05 to 101.45 nGy h^{-1} The obtained concentration and hazard values were systematically compared with established international permissible levels by [UNSEAR 2016]. This research offers crucial baseline data for the radiation safety assessment of chemical fertilizers in Libya .

1 Introduction

The Sources of Terrestrial Radioactivity in the Agricultural Environment Human and environmental exposure to terrestrial natural radioactivity is predominantly governed by geological conditions and local soil characteristics. Chemical fertilizers play a vital role in modern agriculture, serving to replenish nutrient deficiencies and enhance soil properties, which directly contributes to elevated crop efficiency and increased yields [Hallenburg, Jamesk. 2020]. These essential chemical compounds are industrially manufactured, relying primarily on potassium ores, phosphate rocks, and nitrogenous compounds as key raw materials. Radiological Implications of Fertilizer Application Despite the recognized economic and nutritional benefits, the global annual consumption of over 30 million tons of fertilizers [Bayrak Yılmaz, Atik, & Sivri, 2017] poses a potential environmental concern. The primary negative consequence is the potential contamination of cultivated lands by Naturally Occurring Radioactive Materials (NORMs). The natural radioactive content within fertilizers consists mainly of radioisotopes from the Uranium and Thorium decay series, alongside natural Potassium ^{40}K [Bayrak, G. et al., 2018]. Fertilizer Impact on Radionuclide Uptake Variations in fertilizer composition lead to differential effects on plant physiology and metabolism, consequently altering the uptake mechanism for various radionuclides and elements. Therefore, fertilizer application has the potential to elevate radionuclide concentrations within the soil, which subsequently results in enhanced plant uptake. It is important to note that natural radioactivity is an inherent phenomenon throughout the terrestrial environment, detectable in

Asma.alabrdi@omu.edu.ly

phosphate rocks, soil, air, and biological systems (plants and animals). [Hallenburg, Jamesk.2020]

2 Experimental technique

2.1. Sample description and preparation

Ten chemical fertilizers typeslwere collectedlfrom Al gabal al akhder region, markets (Libya).. The investigatedlsamples types are Urealimproved, AmmoniumlNitrate, Single superlphosphate, PotassiumlFertilizers, Magnesium, and Zinc Fertilizers. Table (1) shows the details of the collected samples. They were cleaned, grind as shown in Fig. (1), andldried in the electric oven inlthe laboratory at 110 ℃ for one hour per sample to get rid of moisture in samples as shown in Fig. (2) Sampleslwere placedlin polyethylene beaker, of 350 Cm3 volumes andlweighted for at least 4 weeks; to allow radioactive equilibrium to be reached as shown in Fig. (3). This step is necessary to ensure thatlradon gas is confinedlwithin the volumeland that theldaughters will alsolremain in the sample. [Abbady et al., 2005].

Table (1): Description of the fertilizer samples.

Samples No	Description
Fc1	Magnesium(Italy)
Fc2	NPK0.0.50(Tunisia)
Fc3	NPK20.20.20(Italy)
Fc4	Egyptian high potassium
Fc5	NPK17.17.17(Jordan)
Fc6	NPK20.20.20(Egypt)
Fc7	Zinc(Jordan)
Fc8	NPK18.18.18(Belgian)
Fc9	NPK16.8.24(Egypt)
Fc10	Urea(Italian)

Fig. (1): Grind some samples

Fig. (2): The electric oven device

Fig. (3): The shape of the fertilizer samples after drying and processing

2.2. Instrumentation and calibration

Gamma Spectrometry System and Calibration Activity concentrations were determined using NaI (TI) detector- based gamma spectrometric system. The setup employed 3 × 3 inch. Scintillation detector, whose sealed assembly incorporates the NaI (Tl) crystal, a photomultiplier tube, internal magnetic shielding, and an aluminum housing, coupled to a Canberra Accuspec PC-MCA (Multi-Channel Analyzer). The detector exhibited a specified energy resolution of 6.5\% at the 661.9 keV phitopeak of ^{137}Cs, Key specifications included an aluminum window, an oxide reflector, a magnetic/light shield, and operation at a positive bias voltage of 900 V (dc). To effectively suppress background gamma radiation, the detector was enclosed within a lead shield assembly featuring a fixed base and a movable cover. This 100 mm cylindrical lead shield significantly reduced the contribution of cosmic ray components, such as electrons and photons, to negligible levels [Baqir, Y. et al., 2020]. The system underwent energy calibration utilizing standard sources: ^{60}Co (1173.2 and 1332.5 keV) and ^{137}Cs (661.9 keV). Subsequently, an efficiency calibration curve was generated using various energy peaks spanning the range up to approximately 2000 KeV [Uosif, 2007].

2.3. Activity concentrations

Radioactivity is the volume of radiation produced in a given amount of time. The radioactivity concentration of the different radionuclides was calculated by gamma ray spectrometry with the following relation [Tsoulfanidies, 1983]:

$$(1)\quad A = \frac{\text{Net area (CPS)}}{I_{\gamma} \times \xi \,.M}$$

Where;

A= Activity concentration of the gamma spectral line in Bq/kg.
Net area (cps) = the net detected counts per second corresponding to the energy
ξ = Counting system efficiency of the energy.
M = Mass of sample in kg.
Iγ= Intensity of the gamma spectral.

3. The radiation hazard indices in Chemical fertilizers.

3.1. Radium Equivalent Activity (Bq/kg).

To represent the activity levels of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, a common radiological index has been introduced. This index is called radium equivalent (Ra_{eq}) activity. [Camacho et al., 2010]. It can be calculated from the following relation. [Geremew, H, 2023], [Mwalongo et al., 2023].

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

Where
A_{Ra}, A_{Th} and A_{K} are the activity concentrations **(Bq/Kg)** for **^{238}U,^{232}Th** and **^{40}K** respectively. The maximum value of Ra_{eq} must be less than 370 Bq.kg^{-1}.

3.2. The external and internal hazard index (Hex and Hin)

The hazard indices which reflect exposure are called internal hazard and external hazard respectively. The internal hazard exposure to radon-222 and its daughter products is controlled by an internal hazard index (**H_{in}**) [UNSCEAR, 2000], [Najam et al 2022]

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \qquad (3)$$

The external hazard index (**H_{ex}**) determines for samples gamma-ray emissions and radiological danger was computed by the relation:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \le 1 \qquad (4)$$

Where
A_{Ra}, A_{Th} and A_{K} are the activity concentrations for ^{226}Ra,^{232}Th and ^{40}K in Bq/Kg**[Asma.,2023].**

3.3. Outdoor Absorbed dose rate in Air (D_{out})

The radiological hazard of gamma-ray radiation for the uniform distribution of naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K). The following equation was used to calculate the absorbed dose rate [IAEA,1989].

$$.D_{out} = 0.427A_{U} + 0.662A_{Th} + 0.043A_{K} \quad (5)$$

Where A_{U}, A_{Th} and A_{K} the activities for **^{226}R, ^{232}Th** and **^{40}K** in **Bq /Kg** [I. U. Khan, et al 2020]. 0.427, 0.662 and o. o43 nGy.h^{-1}/BqKg^{-1} are the conversion factors of ^{226}Ra, ^{232}Th and ^{40}K, respectively [Azeez et al,2018]

3.4. Outdoor Annual effective dose (AED_{out})

The amount of outdoor annual effective dose received by human beings is determined using the following equation
.

$$AED_{out}\ (\mu sv/y) = D_{out}\ (nGyh^{-1}) \times 8760 h/y \times 0.2 \times 0.7 (\mu svGy^{-1}) \times 10^{-3} \qquad (6)$$

Where D_{out} is the calculated outdoor dose rate (in nGy/h), 8760 h/y is the number of hours in a year, 0.2 is the outdoor occupancy factor (people stay about 20 % of their time outdoors) and 0.7 is the conversion coefficient from the absorbed dose rate.**[**Asma M., 2023].

3.5. Outdoor excess lifetime cancer risk ($ELCR_{out}$)

This indicator predicts the of developing cancer is the increased of acquiring cancer as result of exposure to a radiation during a person's lifetime. It is determined using the following equation:

$$ELCR\ out = AED_{out} \times D_{L} \times R_{F} \qquad (7)$$

Where AED_{out} is the outdoor annual effective dose equivalent; D_L average life expectancy (70 years) and D_F is the risk factor (0.05) per Sievert, as recommended by ICRP) [Asaduzzaman K., et al., 2015].

4 Results and Discussion.

The values of activitylconcentrations for chemical fertilizer samples the recorded values of radionuclides were varied from (65.27 ± 3.9 to 97.46 ± 5.09), (70.26 ± 4 to 86.3 ± 2.36), and (76.77 ± 7.43 to 131.79 ± 8.8) Bq kg−1 , for ^{226}Ra, ^{232}Th and ^{40}K respectively, Table

(2), The outcomes of radioactive activity concentrations for organic fertilizer samples, including ^{238}U,^{226}Ra, ^{232}Th, and ^{40}K The samples analyzed in this investigation had activity concentrations less the allowable level for uranium (1500 Bq kg-1) [UNSCEAR 2016] ,the activity concentrationslare below the radium (1550 Bq kg-1), the activity concentrations are below the thorium (750 Bq kg-1). the activity concentration is below the potassium suggested ranges (100–700). The following Figure (4) shows the activity concentrations of radium, uranium, thorium, and potassium in organic fertilizers.

Table (2): The average activity concentrations (Bq kg^{-1}) of the radioactive elements (^{238}U,^{226}Ra, ^{232}Th and ^{40}K) of the investigated samples.

samples	^{238}U	Error	^{226}Ra	Error	^{232}Th	Error	^{40}K	Error
Fc1	79.59	±4.80	90.50	±5.87	79.88	±4.20	120.27	±9.53
Fc2	81.88	±5.16	77.88	±5.65	71.96	±6.13	102.36	±9.51
Fc3	66.31	±1.29	65.27	±3.90	72.30	±4.92	76.77	±14.10
Fc4	85.33	±1.69	87.02	±4.09	80.86	±3.17	99.79	±15.81
Fc5	73.53	±1.60	78.32	±4.89	72.72	±2.37	89.56	±11.45
Fc6	76.94	±1.42	77.45	±4.38	70.91	±3.41	83.17	±8.78
Fc7	74.76	±6.76	74.84	±4.28	70.26	±4.00	76.77	±7.43
Fc8	88.68	±2.99	87.89	±5.60	75.78	±5.70	131.79**	±8.80
Fc9	99.02**	±1.50	97.46**	±5.09	86.30**	±2.36	102.36	±11.03
Fc10	70.71	±1.72	73.98	±5.30	73.60	±3.46	76.77	±10.82
Average	79.68	±2.89	81.06	±4.90	75.46	±3.97	95.96	±11.87
P.L	1500		1550		750		100-700	

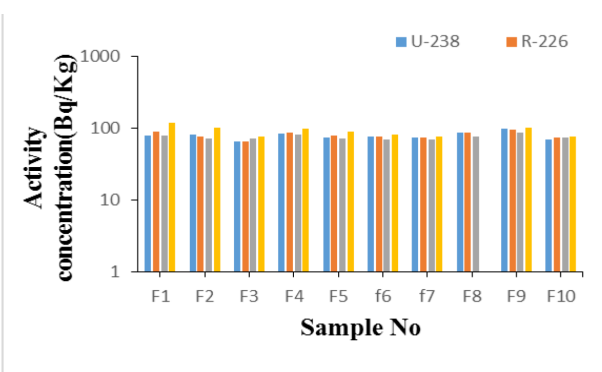

Fig. (4): The activity concentration of Chemical fertilizers.

The Radium equivalent activity (**Ra_{eq}**) is a single quantity that compares the activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in chemical fertilizer samples to obtain a total activity concentration. The results of the calculated (Ra_{eq}) values for chemical fertilizer samples the results range from (174.57 to 228.75 Bq kg^{-1}), with an average value of 196.37 Bq kg^{-1} as shown in Fig. (5), In Table (3), the (Ra_{eq}) values for all fertilizer samples in the present study are lower than the world recommended value 370 Bq kg^{-1} [UNSCEAR, 2016].

Table (3): The value of radium equivalent

Samples code	$Ra_{eq}\ Bq/Kg$
Fc1	213.99
Fc2	188.67
Fc3	174.57
Fc4	210.33
Fc5	189.20
Fc6	185.25
Fc7	181.22
Fc8	206.41
Fc9	228.75
Fc10	185.31
Average	196.37
P.L	**370**

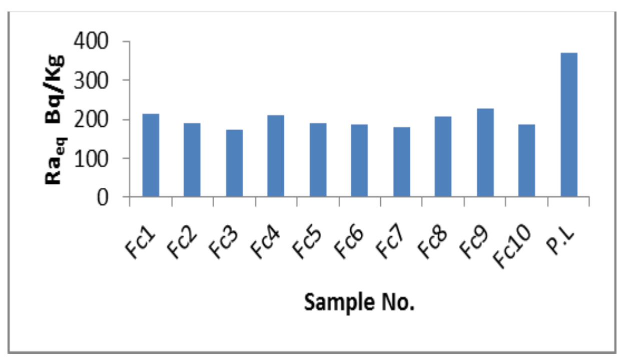

Fig. (5): Radium Equivalent of chemical fertilizer samples.

The absorbed dose rate values for the investigated chemical fertilizer samples range from (77.05 to 101.45 nGy h^{-1}), but the average value of the absorbed dose rate for the chemical fertilizer samples was 86.26 nGy h^{-1}, Fig. (6) shows these results. The values of the absorbed dose rate for all chemical fertilizer samplesbstudied were within the recommended limits [UNSCEAR, 2016], except for the following samples (Fc1, Fc4, Fc9), which were higher thanwthe recommendedmvalues forwthe absorbededose rate of 84 $nGyh^{-1}$

Table (4): Outdoor Absorbed dose rate in Air (D_{out})

Samples code	Dose rate D_{OUT}(nGyh-1)
Fc1	95.11
Fc2	83.75
Fc3	77.05
Fc4	93.23
Fc5	83.87
Fc6	82.10
Fc7	80.23
Fc8	83.91
Fc9	101.45
Fc10	81.85
Average	86.26
P.L	84

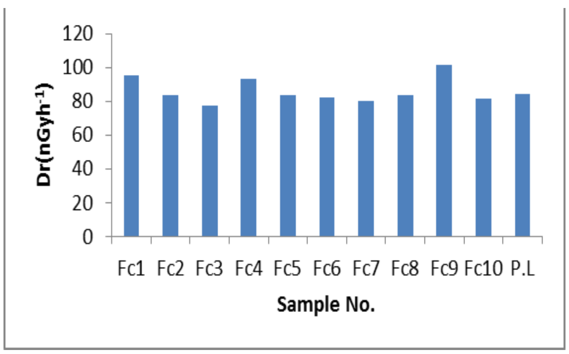

Fig. (6): The absorbedgdose rate of chemical fertilizer samples

The internal and external hazard results were obtained and shown in Table (5). The results value of internal hazard for chemical fertilizer samples ranged from (0.65 to 0.88), with an average value 0.77, as shown in Fig. (7), As a result, all internal hazard values for the fertilizer samples under study were lower than the [UNSCEAR 2016] recommended values. The external hazard for chemical fertilizer samples the results ranged from (0.41 to 0.67), with an average of 0.53, as shown in Fig. (8).

Table (5): The value of internal and external hazards

Samples code	Hin	Hex
Fc1	0.82	0.59
Fc2	0.72	0.51
Fc3	0.65	0.47
Fc4	0.80	0.67
Fc5	0.72	0.51
Fc6	0.71	0.50
Fc7	0.69	0.49
Fc8	0.79	0.56
Fc9	0.88	0.62
Fc10	0.69	0.41
Average	0.77	0.533
P.L	1	1

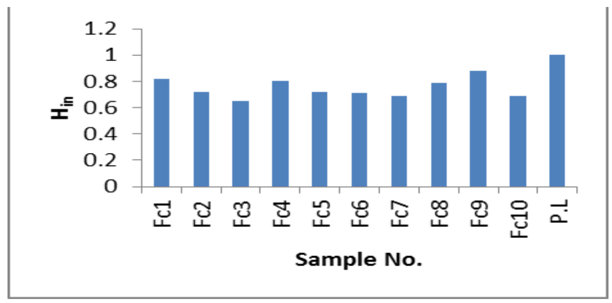

Fig. (7): Internal hazard index of organic fertilizer samples.

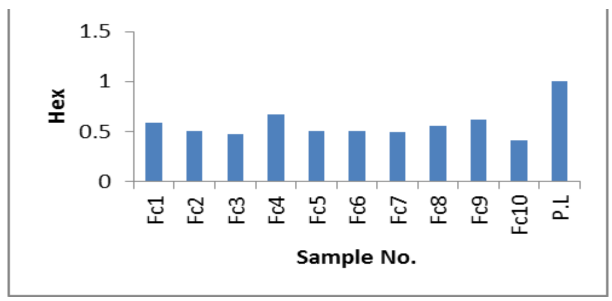

Fig. (8): External hazard index of organic fertilizer samples.

From equation (7) has been calculated of ELCR where the equation dependent of risk factor (0.05 sv^{-1}) and life expectancy (70 years). Table (6) shows the excess lifetime cancer risk factor ELCR for samples, the recorded values of excess lifetime cancer risk ELCR of chemical fertilizer samples the recorded values of ELCR range from (0.19 to 0.41) with an averagegvalue 0.29. The results showed that the ELCR values for all the chemical fertilizer samples studied were within the recommended limits of 0.29×10^{-3} [UNSCEAR, 2016], except for the following sample, Fc9, which were higher than the recommended limits as shown in Fig. (9).

Table (6): The value of Excess Lifetime Cancer Risk (ELCR)

Samples code	Cancer risk factor $ELCR\times10^{-3}$
Fc1	0.20
Fc2	0.29
Fc3	0.27
Fc4	0.25
Fc5	0.24
Fc6	0.19
Fc7	0.20
Fc8	0.27
Fc9	0.41
FC10	0.23
Average	0.26
P.L	0.29

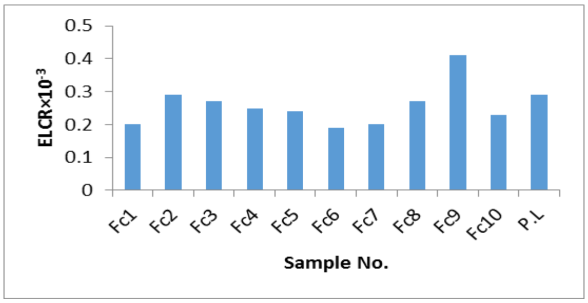

Fig. (9): The cancer risk factor of the chemical fertilizer samples.

5 Conclusions

The study on natural radioactivity levels in chemical fertilizers in Libya, produced the following conclusions:

•Use of simple NaI gamma spectrometer showed potential in the assessment of radioactivity concentration.

•For all chemical fertilizer samples in this study, the activity concentrations of radionuclides ^{226}Ra, ^{232}Th, and ^{40}K were found to be below the world-recommended value.

• All of the samples in this investigation have radium equivalent levels below the global recommended value of 370 Bq kg^{-1}.

•The studied chemical fertilizer samples had absorbedgdose rategvalues below the world recommended limits. except for the following samples (Fc1, Fc4, Fc9), which were higher than the recommended values for the absorbe dose rate of 84 nGyh

•The obtained values of internal hazard index H_{in}, and external hazard index Hex for all the tested sample were lower than the required value.The Excess lifetime cancergrisk (ELCR) values are found to be much lower thangthe permissible level (0.29) for chemical samples except for the following sample, Fc9, which were higher than the recommended limits.

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