Scientific Research Publishing

World Journal of Nuclear Science and Technology, 2025, 15(3), 80-96
https://www.scirp.org/journal/wjnst
ISSN Online: 2161-6809
ISSN Print: 2161-6795

Analysis of Natural Radioactivity in Phosphogypsum by Gamma Spectrometry and Evaluation of Radiological Risk Parameters Associated with Phosphogypsum Storage in Senegal

Moussa Hamady Sy[1*], Ndèye Fatou Fall[1], Djicknack Dione[1], Papa Macoumba Faye[1], Nogaye Ndiaye[1], Oumar Ndiaye[1], Alassane Traoré[1,2], Jean Paul Latyr Faye[1,2], Ndèye Arame Boye Faye[1,2], Ababacar Sadikhe Ndao[1,2]

[1]Institute of Applied Nuclear Technology, University of Cheikh Anta Diop, Dakar, Senegal
[2]Department of Physics, Faculty of Sciences and Technology, University of Cheikh Anta Diop, Dakar, Senegal
Email: *moussahamady.sy@ucad.edu.sn

How to cite this paper: Sy, M.H., Fall, N.F., Dione, D., Faye, P.M., Ndiaye, N., Ndiaye, O., Traoré, A., Faye, J.P.L., Faye, N.A.B. and Ndao, A.S. (2025) Analysis of Natural Radioactivity in Phosphogypsum by Gamma Spectrometry and Evaluation of Radiological Risk Parameters Associated with Phosphogypsum Storage in Senegal. *World Journal of Nuclear Science and Technology*, **15**, 80-96.
https://doi.org/10.4236/wjnst.2025.153007

Received: April 22, 2025
Accepted: June 13, 2025
Published: June 16, 2025

Abstract

The HPGe gamma spectrometer was used to measure the concentrations of the natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in eight samples of phosphogypsum, in order to assess the health risk indices associated with radiation and the excess lifetime cancer risk (ELCR). The ^{226}Ra, ^{232}Th, and ^{40}K were discovered with activity concentrations of 586.26 ± 129.91, 4.05 ± 1.47, and 12.22 ± 6.75, respectively. The average values of the estimated radiological risk parameters were 592.99 ± 141.06 Bq·kg^{-1} for radium equivalent (Raeq), 273.81 nGy·h^{-1} for the external absorbed dose rate (D_{ext}), and 544.81 nGy·h^{-1} for the internal absorbed dose rate (D_{int}). The average annual effective dose (AEDE) was 335799.36 nSv·y^{-1} for the external effective dose (AEDE$_{ex}$t) and 2672601.09 nSv·y^{-1} for the internal effective dose (AEDE$_{in}$t). The annual dose equivalent to the gonads (AGED) was 1832.32 μSv. The average cancer risk rate (ELCR) was 10.22 × 10^{-3}. With the External Risk Index (Hext), the Internal Risk Index (Hint), the Representative Gamma Index (I_γ), and the Alpha Index I_α, the average risk indices were 1.60; 3.19; 1.98; and 2.93. According to this study, the average values of radiological risk parameters for all examined samples exceed the internationally recommended thresholds.

Keywords

Natural Radioactivity, Gamma Spectrometry, Phosphogypsum, Activity

Concentrations, Dose Parameters, Radiological Risks

1. Introduction

Several so-called primordial radioactive materials that were created during the planet's formation are still present on Earth and continue to create others through nuclear disintegration processes. Potassium-40 and the radioisotopes from the natural series of uranium, actinium, and thorium, with the parent nuclei ^{235}U, ^{238}U, and ^{232}Th, as well as the decay products resulting from successive alpha or beta decays, are the most abundant primordial radionuclides [1]. Phosphate rock, a sedimentary mineral mainly composed of apatite ($Ca_5(PO_4)_3(OH, F, Cl)$), contains radionuclides originating from the natural decay series of uranium and thorium, as demonstrated by research [1]-[3]. The phosphate rock is widely exploited to produce phosphoric acid and various brands of chemical fertilizers around the world. When phosphate rock is extracted and processed during mining activities, the radionuclides it contains are released into the environment and end up in finished products and by-products.

Phosphate rocks are treated with concentrated sulfuric acid (H_2SO_4) to produce phosphoric acid in a wet process (see "Equation (1)"), which accounts for about 90% of the world's phosphoric acid production [4].

$$Ca_5F(PO_4)_3 + 5H_2SO_4 + 10H_2O \rightarrow 3H_3PO_4 + 5CaSO_4 \cdot 2H_2O + HF \qquad (1)$$

The by-products, referred to as Phosphogypsum, are the result of this wet chemical treatment of phosphate, a technique known for its importance in enhancing natural radioactivity. It is primarily composed of calcium sulfate dihydrate and contains impurities such as radionuclides and heavy metals (Cd, Pb, As, Cr, etc.). Due to its radioactivity and potential environmental pollution, phosphogypsum is typically stored in large man-made piles.

In Senegal, phosphoric acid production began in 1960 and reached 0.65 million tons in 2010 [5]. The phosphogypsum (PG), produced during this chemical reaction is stored in extensive piles near the plant. Furthermore, the fact that arable land remains occupied by the vast piles of phosphogypsum, natural radioactivity, the presence of heavy metals, and the impurities found in this material can have a toxic and radioactive impact on the local environment.

Soil contamination by heavy metals and radionuclides, following their dispersion or leaching by rain, can alter its structure and fertility. Water, whether groundwater or surface water, can be polluted by toxic leachate from this waste, with consequences for drinking water quality and aquatic ecosystems. The air can be laden with fine dust and radon, a radioactive gas naturally emitted by these residues, which represents a health hazard if inhaled. Living beings, fauna, and flora, can suffer the effects of the bioaccumulation of these toxic substances, causing ecological disruption. Finally, humans are indirectly exposed through contaminated water, food, or

air, which can increase the risk of respiratory, kidney, or cancerous diseases.

Today, the search for a limited impact on the environment, driven by the identification of issues related to inadequate management, pressure from the population, and a better scientific understanding of the risks associated with radiological elements, has led governments and industries to take a series of regulatory measures to reduce the impact of this type of management. As part of a sustainable approach to this by-product, its enhancement has been encouraged worldwide. Various uses of phosphogypsum have been studied, particularly in the civil engineering sector to produce plaster [6]. In agriculture, it is used to fertilize and improve the soil [7] as well as in construction materials to replace natural gypsum in the production of Portland cement in order to regulate the hydration reaction rate of the cement and aggregates [8] [9]. Natural radioactivity must therefore be assessed not only in phosphate rock but also in the by-products. High radioactivity can lead to significant exposure for miners, manufacturers, and end users. The objective of this study is to analyze the levels of natural radionuclides such as ^{226}Ra, ^{232}Th, and ^{40}K in phosphogypsum samples using high-resolution gamma spectrometry. Furthermore, this study will also examine the radiological risk factors associated with the outdoor storage of phosphogypsum.

2. Materials and Methods

2.1. Sample Collection and Preparation

The sampling was carried out at the storage piles of the industrial facilities for phosphoric acid production, in ICS (Chemical Industries of Senegal). The map below of the area, which is the focal point of the study, shows the sampling sites as well as the main communes located in this region (Figure 1).

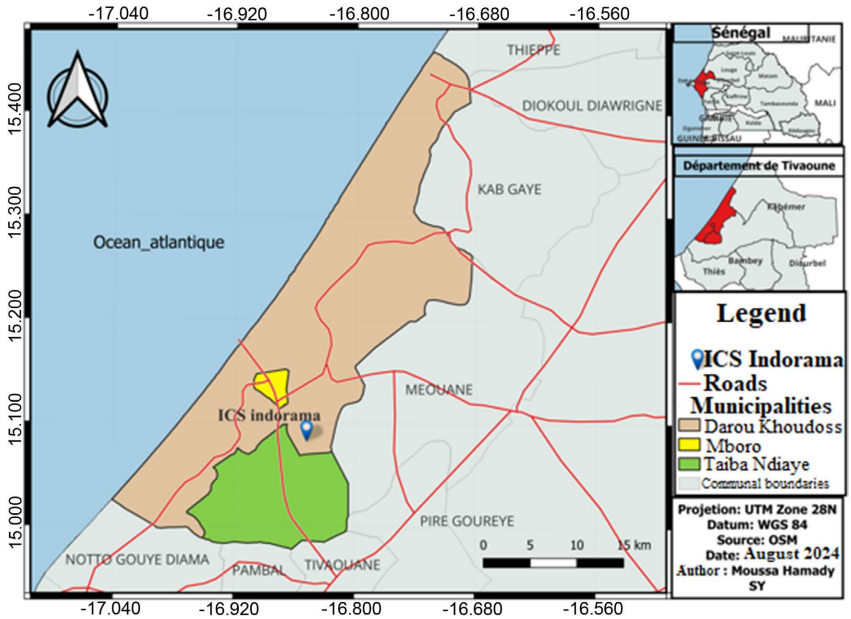

Figure 1. Location of the main cities and study area: Darou Khoudoss, Mboro, and Taiba Ndiaye, sample collection points.

Eight samples of phosphogypsum were collected from stockpiles and stored in polyethylene bags. After being collected in polyethylene bags, the samples were placed in appropriate measuring containers and sealed to prevent any cross-contamination. The samples were prepared according to standard packaging norms to ensure their suitability for spectrometric analysis. In the laboratory, the samples were first dried in an oven at 105˚C for 24 hours to remove moisture until a constant weight was reached. Then, they were crushed and passed through a series of sieves to check their homogeneity [10]. A very fine powder was obtained from the pulverized samples, but to ensure homogeneity, the fractions passing through the 250-micrometer sieves or smaller were used, and a mass of the sieved sample was weighed. The homogeneous samples were transferred into Marinelli Beaker-type containers up to the gauge mark, and the mass that will be used for the measurement was recorded. The sealed samples were left to rest for 4 weeks to allow for the establishment of secular equilibrium between radon-222 (^{222}Rn) and its descendants, which is essential for determining the activity of radium-226 (^{226}Ra). To monitor this balance, additional measurements of 86,400 seconds were taken for each sample, with a one-week interval between each series of measurements, repeated four times.

2.2. Determination of Phosphogypsum Activity Concentration by Gamma Spectrometry

The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were measured on various samples using a high-resolution gamma spectrometry system. A high-purity germanium (HPGe) N-type detector from the brand CAMBERRA is used in this spectrometry system, with a relative efficiency of 15%, for an energy range of 10 keV to 10 MeV. A multi-nuclide reference standard was used to calibrate the energy and relative efficiency of the detector, emitting gamma radiation in an energy range from 5.9 to 1332 keV. The typical energy resolutions of the detector used in this study range from 0.45 keV to 5.9 keV and from 1.9 keV to 1332.5 keV, respectively, with 55Fe and 60Co. The adjustment and verification of the geometry were carried out using the mathematical software Labsocs. All measurements were taken for at least 86,000 seconds to ensure accurate detection of radionuclides and minimize statistical errors. The peak total energy of its descendant ^{228}Ac at 911.6 keV was used to assess the activity concentration of thorium ^{232}Th. In gamma spectrometry, the direct quantification of radium ^{226}Ra is challenging due to the overlap of its peak at 186.2 keV with that of uranium ^{235}U at 185.77 keV. The problem is all the more complex because the limited activity of uranium-235 makes its precise measurement difficult [11]. In this study, the activity concentration of radium ^{226}Ra was determined from its short-lived decay products once secular equilibrium was reached. The activity concentrations of radium ^{226}Ra were calculated by averaging the measured activities for two distinct photoelectric peaks of its daughter nuclides: ^{214}Pb at 351.92 keV and ^{214}Bi at 609.31 keV. For potassium ^{40}K, the activity concentration was determined by using the gamma line at 1460.81 keV as illustrated in **Table 1**.

Table 1. Major gamma lines used for natural radionuclides.

Radionuclides	E, keV	P_γ %
^{226}Ra		
^{214}Bi	609.31	46.30
^{214}Pb	351.92	37.20
^{232}Th		
^{228}Ac	911.60	27.70
^{40}K	1460.81	10.67

The following equation was used to calculate the activity concentration of each radionuclide (Bq·kg^{-1}).

$$A = \frac{1000 \cdot N}{m \cdot \varepsilon(E) \cdot P_\gamma(E) \cdot t_{\text{mesured}}} \tag{2}$$

where:

- A represents the activity concentration at the time of sampling (in Bq/kg);
- N is the net number of counts of the peak (cps);
- t_{mesured} is the live measurement time in seconds (s);
- m is the mass of the sample used in grams (g);
- $\varepsilon(E)$ is the detector efficiency for gamma radiation (dimensionless); and
- $P_\gamma(E)$ is the probability of gamma emission by the radionuclide (dimensionless).

2.3. Evaluation of Dose and Radiological Risk Parameters

The following equations have been used to calculate the radiological hazard indices related to exposure to excessive gamma radiation from phosphogypsum resulting from phosphoric acid production.

2.3.1. Radium Equivalent Activity

In general, the concentration and distribution of ^{40}K, ^{226}Ra, and ^{232}Th in the soil vary by country [12]. Thus, in order to represent the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K with a single measure, taking into account the radiological hazard associated with them, it is common to calculate a widely used common radiological hazard index: the radium equivalent activity (R_{aeq}) [13]. The radium equivalent activity can be expressed as follows:

$$R_{\text{aeq}}\left(\text{Bq}\cdot\text{kg}^{-1}\right) = A_{\text{Ra}} + A_{\text{Th}} \times 1.43 + A_{\text{K}} \times 0.077 \tag{3}$$

where A_{Ra}, A_{Th}, and A_{K} representing the specific activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq·kg^{-1}, respectively.

2.3.2. Absorbed Dose Rates

An assessment of the external dose rate of radiation (D_{ext}) at 1 meter above the

ground surface is conducted using the gamma radiation emitted by ^{226}Ra, ^{232}Th, and ^{40}K, which are assumed to be evenly distributed in the soil. The following equations have been used to calculate the doses of external and internal radiation. (D_{ext} et D_{int}) [3]:

$$D_{ext}\left(\text{nGy}\cdot\text{h}^{-1}\right)=0.462A_{Ra}+0.604A_{Th}+0.0417A_{K} \quad (4)$$

$$D_{int}\left(\text{nGy}\cdot\text{h}^{-1}\right)=0.92A_{Ra}+1.1A_{Th}+0.081A_{K} \quad (5)$$

2.3.3. Annual Effective Dose Equivalent

The following relationships were used to calculate the expected annual effective dose equivalent (AEDE) for the general public due to the radioactivity present in phosphogypsum:

$$\text{AEDE}_{ext}\left(\text{nSv}\cdot\text{y}^{-1}\right)=D_{ext}\left(\text{nGy}\cdot\text{h}^{-1}\right)\times 8760\ \text{h}\cdot\text{y}^{-1}\times 0.2\times 0.7\ \text{Sv}\cdot\text{Gy}^{-1} \quad (6)$$

$$\text{AEDE}_{int}\left(\text{nSv}\cdot\text{y}^{-1}\right)=D_{int}\left(\text{nGy}\cdot\text{h}^{-1}\right)\times 8760\ \text{h}\cdot\text{y}^{-1}\times 0.8\times 0.7\ \text{Sv}\cdot\text{Gy}^{-1} \quad (7)$$

The annual effectiveness of the equivalent dose was determined using a conversion factor of 0.7 Sv·Gy^{-1}, which converts the absorbed dose in the air into an effective dose for adults. The external occupancy was 0.2 and the internal occupancy was 0.8 [2] [14].

2.3.4. Annual Gonadal Dose Equivalent

According to UNSCEAR, the organs of interest are the gonads (ovaries or testes, which generate gametes), the cells of the bone surface, and the active bone marrow [15]. The annual equivalent dose to the gonads (AGED) is estimated by using the equation provided by [16]:

$$\text{AGED}\left(\mu\text{Sv}\cdot\text{y}^{-1}\right)=3.09C_{Ra}+4.18C_{Th}+0.314C_{K} \quad (8)$$

where AGED is the Annual Equivalent Dose to the Gonads (μSv·y^{-1}), and C_{Ra}, C_{Th}, and C_K (Bq·kg^{-1}) are the radioactivity levels of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

2.3.5. Excess Lifetime Cancer Risk

The following equations have been used to calculate the lifetime excess cancer risk value (ELCR) [15]:

$$\text{ELCR}_{ext}=\text{AEDE}_{ext}\times\text{LE}\times\text{RF} \quad (9)$$

$$\text{ELCR}_{int}=\text{AEDE}_{int}\times\text{LE}\times\text{RF} \quad (10)$$

where (AEDE_{ext}) and (AEDE_{int}) are the annual effective doses, LE is the life expectancy of 68.9 years according to the report from the National Agency of Statistics and Demographics of Senegal (ANSD, 2023), and RF (Sv^{-1}) is the fatal risk factor per Sievert, which is 0.05 [15].

2.3.6. Internal Hazard Index and External Hazard Index

The internal and external hazard indices (H_{int}; H_{ext}) are calculated using the following expressions [17]:

$$H_{ext}=\frac{A_{Ra}}{370}+\frac{A_{Th}}{259}+\frac{A_{K}}{4810} \quad (11)$$

$$H_{\text{int}} = \frac{A_{\text{Ra}}}{185} + \frac{A_{\text{Th}}}{259} + \frac{A_{\text{K}}}{4810} \tag{12}$$

where A_{Ra}, A_{Th}, and A_{K} representing the specific activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq·kg^{-1}, respectively.

2.3.7. Gamma and Alpha Radioactivity Level Indices

The gamma radioactivity level index (I_γ) is used to estimate the risk rate associated with gamma radiation from natural radionuclides in the investigated samples [18]. It is defined by the following equation:

$$I_\gamma = \frac{C_{\text{Ra}}}{300} + \frac{C_{\text{Th}}}{200} + \frac{C_{\text{K}}}{3000} \tag{13}$$

where C_{Ra}, C_{Th}, and C_{K} are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

The alpha index (I_α) is used to estimate the excess alpha radiation caused by the inhalation of radon from building materials. The alpha index is determined using the following equation [17]:

$$I_\alpha = \frac{C_{\text{Ra}}}{200} \tag{14}$$

3. Results and Discussion

3.1. Activity Concentration of ^{226}Ra, ^{232}Th, and ^{40}K

The characterization of radioactive species is the first step in assessing the radiological impact associated with phosphogypsum. In order to verify if the radionuclides ^{214}Bi and ^{214}Pb are in secular equilibrium, the graphs (**Figure 2**) present the respective activity concentration ratios of the different samples (labeled EPG1, EPG2, EPG3, EPG4, EPG5, EPG6, EPG7, and EPG8). According to the results, it was found that the measured values of the descendants were in secular equilibrium with those of radium-226, as the observed discrepancy between the measurements of the descendant radionuclides is less than 2%. This slight variation confirms the secular balance, thereby validating the accuracy of the measurements taken.

Table 2 presents the activity concentrations of natural radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) measured in phosphogypsum samples using gamma spectrometry.

The results indicate a variation in activity concentrations between 408.66 ± 27.57 and 809.83 ± 54.54 Bq·kg^{-1} for ^{226}Ra, from 2.64 ± 0.61 to 6.63 ± 0.67 Bq·kg^{-1} for ^{232}Th, and from 6.55 ± 1.83 to 24.59 ± 3.11 Bq·kg^{-1} for ^{40}K. For ^{226}Ra, ^{232}Th, and ^{40}K, the average values have been determined to be 586.26 ± 129.91 Bq·kg^{-1}, 4.05 ± 1.47 Bq·kg^{-1}, and 12.22 ± 6.75 Bq·kg^{-1}, respectively. The first two samples analyzed, EPG1 and EPG2, show significantly higher radionuclide activities than those of the other samples. This disparity is reflected by the statistical standard deviation, which measures the dispersion of values around the mean, thus indicating a greater variability in these two samples compared to the others. This difference could be explained by the fact that these two samples are more recent. On

the other hand, older samples may have undergone phenomena such as leaching, leading to a reduction in their activity. This variation highlights the potential impact of time and environmental conditions on the concentrations of radionuclides in phosphogypsum.

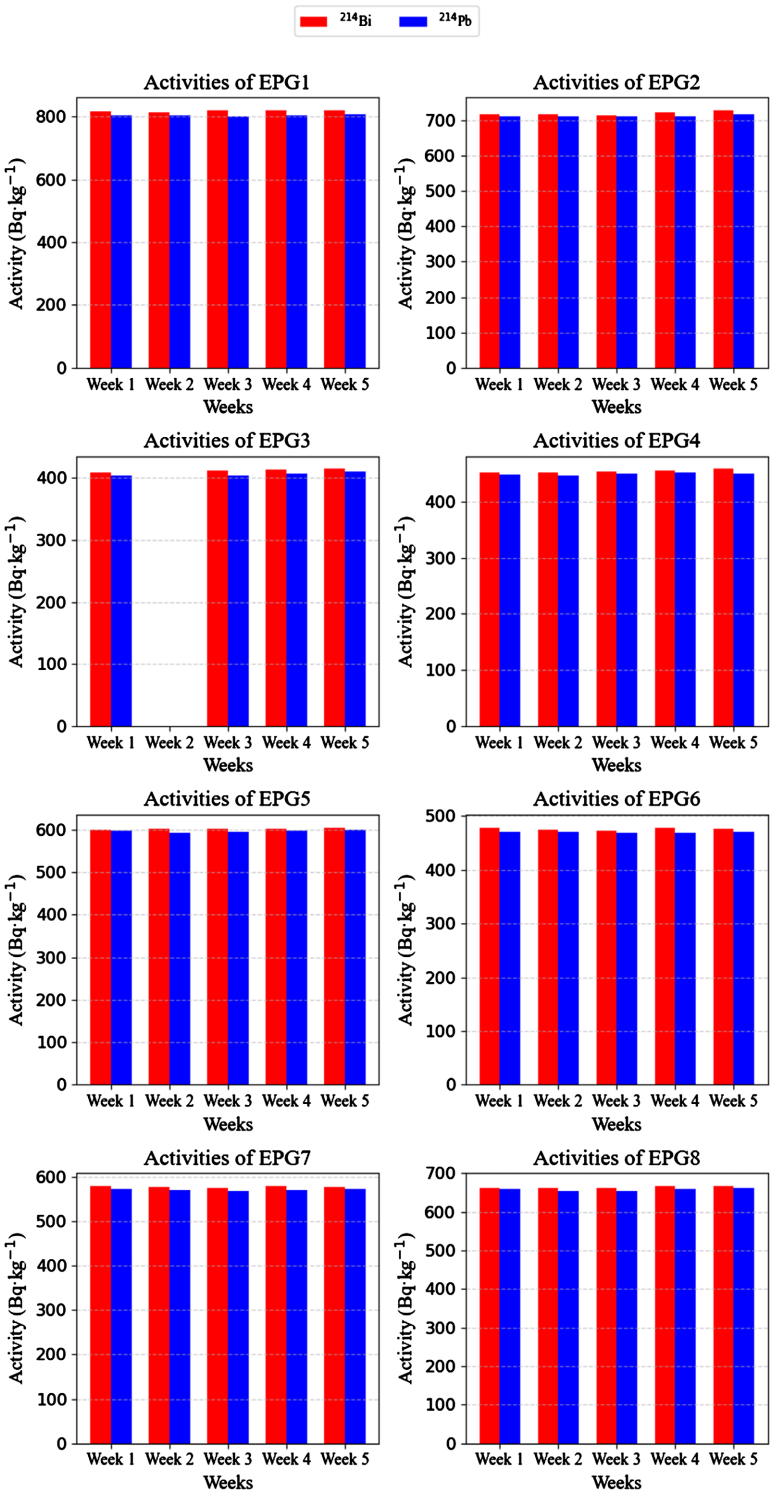

Figure 2. Secular equilibrium between ^{214}Bi and ^{214}Pb for phosphogypsum samples.

Table 2. Distribution of activity concentrations (Bq·kg^{-1}) of ^{226}Ra, ^{232}Th, ^{40}K, and radium equivalent in phosphogypsum samples.

Sample Codes	Natural Radionuclides (Bq·kg^{-1})			Radium equivalent
	^{226}Ra	^{232}Th	^{40}K	Raeq
EPG1	809.83 ± 54.54	6.63 ± 0.67	24.59 ± 3.11	821.20
EPG2	714.77 ± 48.16	6.22 ± 0.71	20.78 ± 2.71	725.26
EPG3	408.66 ± 27, 57	3.13 ± 0.46	11.02 ± 1.98	413.98
EPG4	451.34 ± 30.42	2.64 ± 0.61	10.64 ± 1.52	455.93
EPG5	598.78 ± 40.35	4.50 ± 0.57	6.66 ± 1.49	605.72
EPG6	472.37 ± 31.83	3.21 ± 0.54	6.55 ± 1.83	477.47
EPG7	573.93 ± 38.66	3.27 ± 0.45	9.67 ± 1.72	579.34
EPG8	660.42 ± 44.50	2.81 ± 0.55	7.86 ± 1.61	665.04
Min	408.66 ± 27.57	2.64 ± 0.61	6.55 ± 1.83	413.98
Max	809.83 ± 54.54	6.63 ± 0.67	24.59 ± 3.11	821.20
Mean ± SD	586.26 ± 129.91	4.05 ± 1.47	12.22 ± 6.75	592.99 ± 141.06

The decreased concentrations of radium-226 in older phosphogypsum samples compared to more recent samples could be related to the precipitation of radium-226 as radium sulfate ($RaSO_4$). Indeed, it is plausible that radium-226 gradually transforms into $RaSO_4$ over time, leading to a reduction in the levels of free radium-226 in solid samples. Several factors can explain the relatively high presence of ^{226}Ra in phosphogypsum samples compared to other radionuclides. First of all, radium has a strong chemical affinity for sulfate, which facilitates its coprecipitation in the form of radium sulfate ($RaSO_4$) during the wet process of phosphoric acid production. Consequently, ^{226}Ra tends to preferentially accumulate in the solid phase of phosphogypsum. Secondly, the geological properties of the phosphates deposits being exploited can affect the initial rate of uranium (^{238}U) and, consequently, the amount of ^{226}Ra that result from its decay. Finally, leaching process or differential movement of radionuclides during storage can also explain the greater retention of radium compared to thorium and potassium. The average activity concentrations in phosphogypsum samples compared to the global average values (50 Bq·kg^{-1} for radium-226, 50 Bq·kg^{-1} for thorium-232, and 500 Bq·kg^{-1} for potassium-40) reported by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and referenced by Ndour *et al.* (2021), have shown that the activities of thorium-232 and potassium-40 are significantly lower than the global average, while the activity of radium-226 remains significantly higher than this average.

The comparison of the average values of activity concentrations measured in this study for the analyzed samples with those reported in other countries is presented in Table 3 and Figure 3.

Table 3. Comparison of ^{226}Ra, ^{232}Th, and ^{40}K activity concentrations with those obtained in other countries.

Countries	Natural Radionuclides (Bq·kg^{-1})			References
	^{226}Ra	^{232}Th	^{40}K	
Egypt	203 ± 5.3	43.2 ± 3.5	126 ± 15	[16]
Korea	618	8. 5	24.1	[19]
Tunisia	188.0 ± 9.5	12.4 ± 1.4	<13.5	[4]
Spain	647	8	33	[20]
Jordan	376	4	40	[21]
Hungary	337.0 ± 179.3	5.4 ± 5.8	210.0 ± 122.2	[22]
Morocco	1420 ± 330	-	-	[1]
Portugal	735 ± 70	-	-	[23]
Florida	1120	3. 7	-	[24]
Senegal	586.26 ± 129.91	4.05 ± 1.47	12.22 ± 6.75	This Study

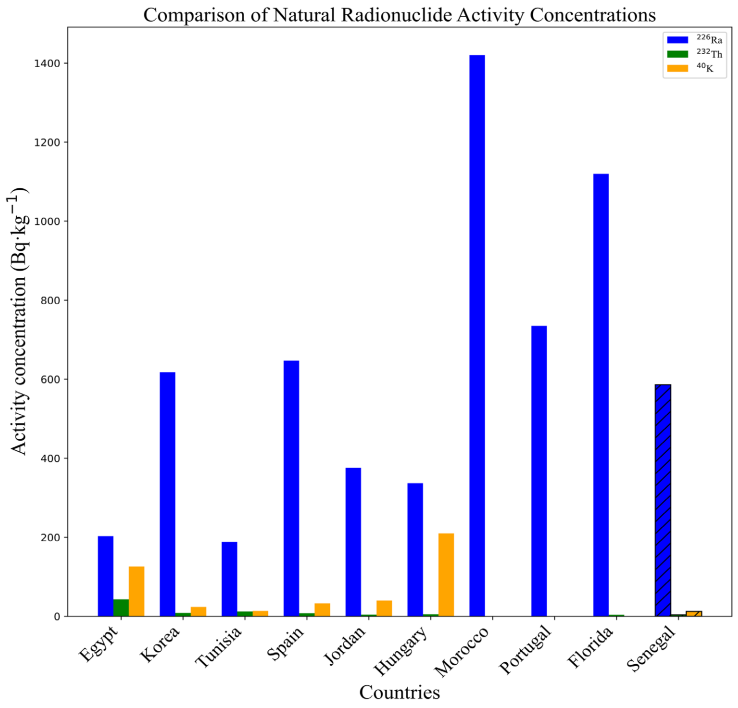

Figure 3. Comparison of activity concentrations of natural radionuclides with other countries around the world.

These results indicate that the activity concentration of ^{226}Ra in the phosphogypsum samples of this study is higher than that observed in Egypt, Tunisia, Jordan, and Hungary. However, the activity concentrations recorded in Korea, Spain, Morocco, Portugal, and Florida are higher than those observed in this study. Apart from

the concentration of ^{232}Th measured in Egypt and the concentrations of ^{40}K recorded in Egypt and Hungary, the activity concentrations of ^{232}Th and ^{40}K in this study are consistent with those observed in other countries (see **Figure 3**). This trend can be attributed to variations in industrial processes, the nature of phosphate deposits, as well as the environmental conditions specific to each region.

3.2. Dose Parameters and Radiological Risks

According to "Equations (3) to (14)", the radiological dose parameters of the phosphogypsum samples have been evaluated. Thanks to these equations, it is possible to calculate various parameters essential for assessing the radiological risks associated with these materials, such as the radiological hazard index (Raeq), the absorbed dose rate, the annual equivalent effective dose, as well as the internal and external risk indices. They also offer the possibility to measure danger levels for gamma and alpha rays. Using these equations, it was possible to measure the potential impact of phosphogypsum residues on human health and the environment. The calculations also include the assessment of lifetime cancer risk and annual equivalent gonadal doses, providing a comprehensive overview of the potential radiological hazards associated with these materials. For the various phosphogypsum samples, the Raeq values were calculated using "Equation (3)", and the results are presented in **Table 2**. The Raeq obtained for the phosphogypsum samples ranges from 413.98 to 821.20 Bq·kg^{-1}, with an average of 592.99 ± 141.06 Bq·kg^{-1}. The Raeq index exceeds the authorized limit of equivalent radium activity, which should not exceed 370 Bq·kg^{-1}, in order to ensure safe use in various civil applications [13]. **Table 4** and **Figure 4** presents the values of the absorbed dose rate outdoors, D_{ext}, which fluctuate between 191.15 and 379.17 nGy·h^{-1}, with an average of 273.81 nGy·h^{-1}.

Table 4. Radiological dose parameters, dose rate, AEDE, and AGED.

Sample Codes	Radiological Dose Parameters				
	D_{ext} (nGy·h^{-1})	D_{int} (nGy·h^{-1})	AEDE$_{ext}$ (nSv·y^{-1})	AEDE$_{int}$ (nSv·y^{-1})	AGED (μSv·y^{-1})
EPG1	379.17	754.32	465013.36	3700415.57	2537.80
EPG2	334.85	666.11	410655.21	3267677.18	2241.16
EPG3	191.15	380.30	234427.66	1865614.31	1279.30
EPG4	210.56	419.00	258226.26	2055428.75	1409.01
EPG5	279.63	556.36	342938.74	2729298.94	1871.12
EPG6	220.45	438.65	270359.28	2151826.28	1475.12
EPG7	267.53	532.39	328099.99	2611693.89	1790.13
EPG8	307.14	611.31	376674.38	2998853.80	2054.91
Mean	273.81	544.81	335799.36	2672601.09	1832.32

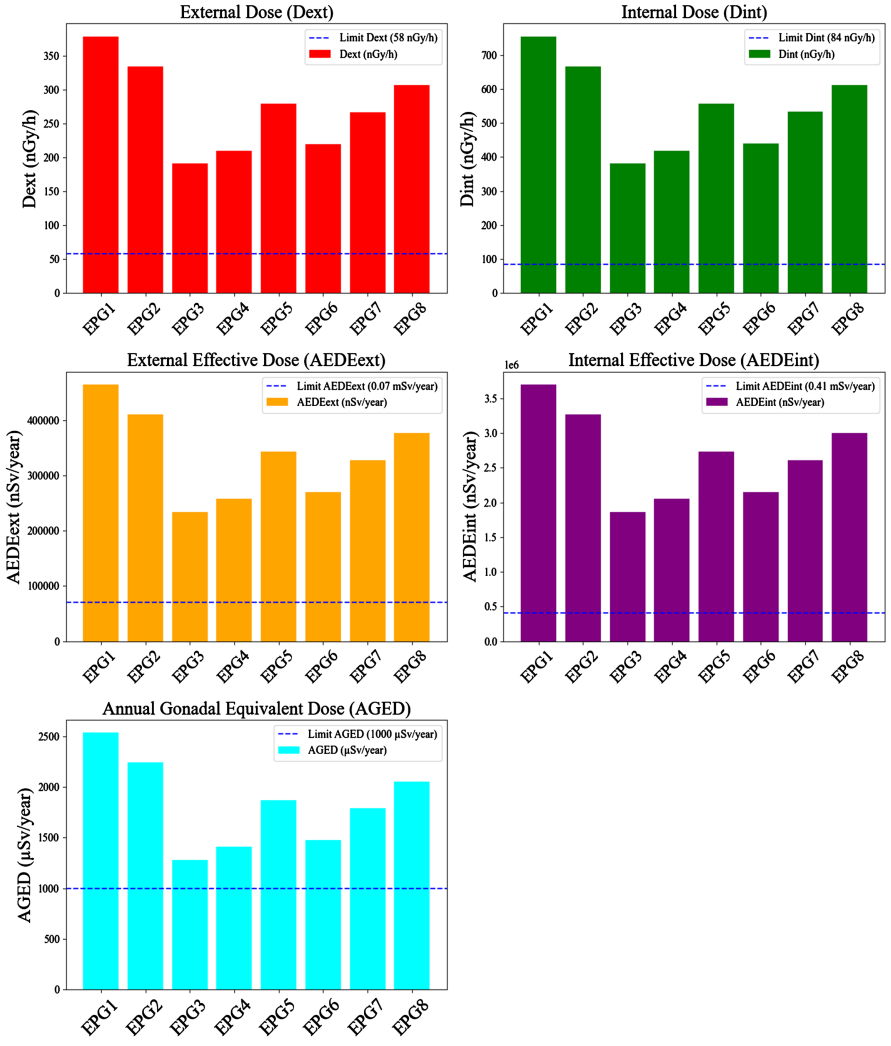

Figure 4. The different dose parameters.

Conversely, the absorbed dose rate indoors ranges from 380.30 to 754.32 nGy·h^{-1}, with an average of 544.81 nGy·h^{-1}. These values exceed the global average of 58 nGy·h^{-1} for D_{ext} and 84 nGy·h^{-1} for D_{int}, respectively [25]. The results indicate that the values of the external effective dose ($AEDE_{ext}$) for the various samples range from 0.23 to 0.46 mSv·y^{-1}, with an average of 0.33 mSv·y^{-1}, which exceeds the permitted limit of 0.07 mSv·y^{-1} (UNSCEAR, 2008). In the same way, the values of the effective internal dose ($AEDE_{int}$) for the samples range from 1.86 to 3.70 mSv·y^{-1}, with an average of 2.67 mSv·y^{-1}, thus exceeding the recommended value of 0.41 mSv·y^{-1} (UNSCEAR, 2008), as shown in **Table 4**. The values obtained for the Annual Equivalent Dose to the Gonads (AGED), which fluctuate between 1279.30 and 2537.80 µSv·y^{-1}, with an average of 1832.32 µSv·y^{-1}. This average value is higher than the globally recommended limit, which is 1000 µSv·y^{-1} for the general public [16]. These values indicate that gonad exposure levels may have significant radiological effects on the bone marrow and superficial bone cells of miners, workers and inhabitants of the area studied. In the long term, the combination of

doses at this level of exposure could endanger the reproductive organs (ovaries and testicles) of individuals living near these mining sites. "Equations (9) and (10)" are used to calculate the values of the lifetime cancer risk excess (ELCR) for outdoor and indoor exposures, as presented in Table 5 and Figure 5.

Table 5. Radiological risk indices from different phosphogypsum samples.

Sample Codes	Radiological Risk Indices						
	H_{int}	H_{ext}	I_γ	I_α	$ELCR_{int}$	$ECLR_{ext}$	ELCR total (10^{-3})
EPG1	4.41	2.22	2.74	4.05	12.75	1.6	14.35
EPG2	3.89	1.96	2.42	3.57	11.26	1.41	12.67
EPG3	2.22	1.12	1.38	2.04	6.43	0.80	7.23
EPG4	2.45	1.23	1.52	2.26	7.08	0.89	7.97
EPG5	3.26	1.64	2.02	2.99	9.40	1.18	10.57
EPG6	2.57	1.29	1.59	2.36	7.41	0.93	8.43
EPG7	3.12	1.57	1.93	2.87	8.98	1.13	10.11
EPG8	3.58	1.81	2.22	3.30	10.33	1.30	11.46
Mean	3.19	1.60	1.98	2.93	9.21	1.16	10.22

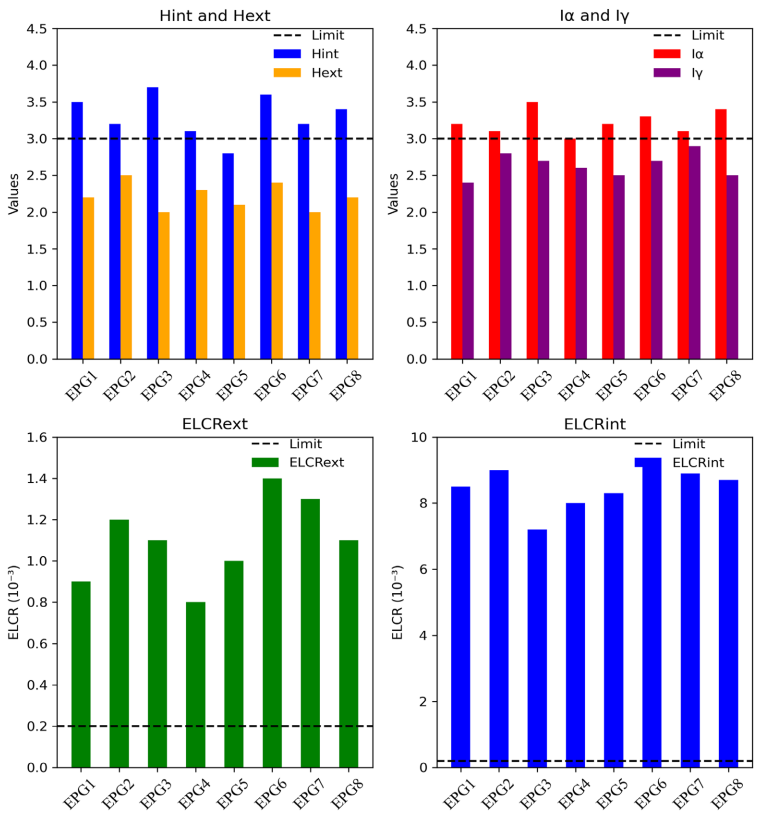

Figure 5. Risk indices compared to their limit values.

The external and internal ELCRs have been estimated at 1.16×10^{-3} and 9.21×10^{-3}, respectively, while the total average value of internal and external exposure is estimated at 10.22×10^{-3}. These are values exceeding the recommended limits, which are 0.3×10^{-3} for external ELCR and 1.2×10^{-3} for internal ELCR. Furthermore, the global ELCR for all samples exceeds the world average of 1.45×10^{-3} [3]. It was found that all the samples showed an ELCR higher than the established standards. Mohammed and Ahmed [26] indicated that ELCR values of 1, 10, 100, and 1000 $mSv{\cdot}y^{-1}$ would increase the risk of developing fatal cancer by 0.004%, 0.04%, 0.4%, and 4% respectively. Although the lifetime excess lifetime cancer risk (ELCR) observed in this study is above the global average, the increased risk of cancer over a lifetime remains negligible, even for indoor ELCR. The analysis reveals that the annual effective doses (AEDE) and excess lifetime cancer risks (ELCR) in certain cases surpass the limits recommended by the International Commission on Radiological Protection (ICRP), which stipulates a maximum public exposure of 1 $mSv{\cdot}y^{-1}$. Sustained exposure to elevated levels of radium-226 may substantially heighten the incidence of bone cancer and leukemia within the exposed populations. Although all individuals are at risks, children represent a particularly sensitive subgroup due to higher cellular proliferation rates and longer latency periods for radiation-induced pathologies. These results highlight the necessity for rigorous radiological surveillance and the implementation of protective strategies aimed at minimizing exposure for the entire population residing near phosphogypsum storage areas. The external risk index (H_{ext}), the internal risk index, the representative gamma index, and the representative alpha index, calculated respectively from "Equations (11) to (14)", are also shown in **Table 5**. It is essential that the results obtained for the H_{ext} and the H_{int} are both below 1 in order to keep the radiological risk (annual effective dose) within safe limits [27]. The averages of H_{ext}, H_{int}, I_γ, and I_α obtained are respectively 1.60; 3.19; 1.98; and 2.93. All these average values of radiological risk, namely H_{ext}, H_{int}, I_γ, and I_α, exceed the recommended limit of 1.0 for the public [28]. The internal risk index is three times higher than the allowed limit, while the gamma and alpha radiation level indices are nearly twice as high. According to the results, it has been demonstrated that phosphogypsum waste poses a radiological threat to the population, which represents a danger to human health and the environment. However, it is necessary to acknowledge certain limitation of this research, especially the limited sample, which may not represent all the existing phosphogypsum storage sites, as well as the lack of study on the migration of radionuclides in the surrounding soils and groundwater. It would be necessary to conduct additional research, with a broader spatial and temporal coverage, to validate these conclusions and more precisely assess the long-term risks.

4. Conclusions

Gamma spectroscopy was used to analyze the concentrations of natural radionuclides in eight samples of phosphogypsum. The measurements taken on ^{226}Ra,

^{232}Th, and ^{40}K have allowed for the evaluation of various radiological risk factors, such as the Absorbed Dose Rate (D), the Annual Effective Dose Equivalent (AEDE), the Radium Equivalent (R_{aeq}), the Annual Equivalent Dose to the Gonads (AGED), the Excess Lifetime Cancer Risk (ELCR), the External Risk Index (H_{ext}), the Internal Risk Index (H_{int}), the Representative Gamma Index (I_γ), and the Representative Alpha Index. Unlike the global average values for thorium-232 and potassium-40 activity (50 Bq·kg^{-1} for thorium-232 and 500 Bq·kg^{-1} for potassium), the activity of radium exceeds the recommended average value of 50 Bq·kg^{-1}. The average radium equivalent obtained in this study exceeds the recommended limit of 370 Bq·kg^{-1}. Furthermore, the other radiological risk parameters for all the samples studied are above the globally recommended thresholds.

These results highlight that the materials studied pose a significant radiological risk to the population, which requires appropriate monitoring and management to ensure safety. This assessment provides a comprehensive overview of the radiological hazards associated with the examined phosphogypsum samples, allowing for a better understanding of the potential consequences on health and the environment. It is important to monitor and manage the levels of radioactivity in construction materials to ensure public safety, as evidenced by the results obtained. It light of the high levels of radioactivity detected, the establishment of a regular monitoring program for phosphogypsum storage sites is recommended, accompanied by periodic examinations of soils, groundwater, and surrounding crops. Moreover, the development of strategies aimed at confining or controlled reuse of phosphogypsum could help minimize population exposure. Future studies should focus on analyzing the migration of radionuclides in the environment and assessing the transfer of pollutants in the food chain.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

References

[1] Azouazi, M., Ouahidi, Y., Fakhi, S., Andres, Y., Abbe, J.C. and Benmansour, M. (2001) Natural Radioactivity in Phosphates, Phosphogypsum and Natural Waters in Morocco. *Journal of Environmental Radioactivity*, **54**, 231-242. https://doi.org/10.1016/s0265-931x(00)00153-3

[2] Fávaro, D.I.T. (2005) Natural Radioactivity in Phosphate Rock, Phosphogypsum and Phosphate Fertilizers in Brazil. *Journal of Radioanalytical and Nuclear Chemistry*, **264**, 445-448. https://doi.org/10.1007/s10967-005-0735-4

[3] El-Bahi, S.M., Sroor, A., Mohamed, G.Y. and El-Gendy, N.S. (2017) Radiological Impact of Natural Radioactivity in Egyptian Phosphate Rocks, Phosphogypsum and Phosphate Fertilizers. *Applied Radiation and Isotopes*, **123**, 121-127. https://doi.org/10.1016/j.apradiso.2017.02.031

[4] Tayibi, H., Gascó, C., Navarro, N., López-Delgado, A., Choura, M., Alguacil, F.J., *et al*. (2011) Radiochemical Characterization of Phosphogypsum for Engineering Use. *Journal of Environmental Protection*, **2**, 168-174. https://doi.org/10.4236/jep.2011.22019

[5] Ndour, O., Thiandoume, C., Traore, A., Cagnat, X., Diouf, P.M., Ndeye, M., *et al*. (2021) Determination of Natural Radionuclides in Phosphogypsum Samples from Phosphoric Acid Production Industry in Senegal. *Environmental Forensics*, **24**, 197-204. https://doi.org/10.1080/15275922.2021.2006362

[6] Moutaouakil, A., Pineau, J.L. and Lahlou, K. (2003) La recherche d'un procédé viable de valorisation d'un phosphogypse provenant de l'industrie phosphatière marocaine. *Environnement, Ingénierie & Développement*, **29**, 31-35. https://doi.org/10.4267/dechets-sciences-techniques.2231

[7] Alcordo, I.S. and Rechcigl, J.E. (1993) Phosphogypsum in Agriculture: A Review. In: *Advances in Agronomy*, Elsevier, 55-118. https://doi.org/10.1016/s0065-2113(08)60793-2

[8] Saadaoui, E., Ghazel, N., Ben Romdhane, C. and Massoudi, N. (2017) Phosphogypsum: Potential Uses and Problems—A Review. *International Journal of Environmental Studies*, **74**, 558-567. https://doi.org/10.1080/00207233.2017.1330582

[9] Gezer, F., Turhan, Ş., Uğur, F.A., Gören, E., Kurt, M.Z. and Ufuktepe, Y. (2012) Natural Radionuclide Content of Disposed Phosphogypsum as TENORM Produced from Phosphorus Fertilizer Industry in Turkey. *Annals of Nuclear Energy*, **50**, 33-37. https://doi.org/10.1016/j.anucene.2012.07.018

[10] Mazzilli, B., Palmiro, V., Saueia, C. and Nisti, M.B. (2000) Radiochemical Characterization of Brazilian Phosphogypsum. *Journal of Environmental Radioactivity*, **49**, 113-122. https://doi.org/10.1016/s0265-931x(99)00097-1

[11] Reguigui, R., Sfar Felfoul, H., Ben Ouezdou, M. and Clastres, P. (2005) Radionuclide Levels and Temporal Variation in Phosphogypsum. *Journal of Radioanalytical and Nuclear Chemistry*, **264**, 719-722. https://doi.org/10.1007/s10967-005-0778-6

[12] Khan, H.M., Chaudhry, Z.S., Ismail, M. and Khan, K. (2010) Assessment of Radionuclides, Trace Metals and Radionuclide Transfer from Soil to Food of Jhangar Valley (Pakistan) Using Gamma-Ray Spectrometry. *Water, Air, & Soil Pollution*, **213**, 353-362. https://doi.org/10.1007/s11270-010-0390-4

[13] Beretka, J. and Mathew, P.J. (1985) Natural Radioactivity of Australian Building Materials, Industrial Wastes and By-Products. *Health Physics*, **48**, 87-95. https://doi.org/10.1097/00004032-198501000-00007

[14] Rahman, S.U., Matiullah, Malik, F., Rafique, M., Anwar, J., Ziafat, M., *et al*. (2010) Measurement of Naturally Occurring/Fallout Radioactive Elements and Assessment of Annual Effective Dose in Soil Samples Collected from Four Districts of the Punjab Province, Pakistan. *Journal of Radioanalytical and Nuclear Chemistry*, **287**, 647-655. https://doi.org/10.1007/s10967-010-0819-7

[15] Qureshi, A.A., Tariq, S., Din, K.U., Manzoor, S., Calligaris, C. and Waheed, A. (2014) Evaluation of Excessive Lifetime Cancer Risk Due to Natural Radioactivity in the Rivers Sediments of Northern Pakistan. *Journal of Radiation Research and Applied Sciences*, **7**, 438-447. https://doi.org/10.1016/j.jrras.2014.07.008

[16] Attallah, M.F., Metwally, S.S., Moussa, S.I. and Soliman, M.A. (2019) Environmental Impact Assessment of Phosphate Fertilizers and Phosphogypsum Waste: Elemental and Radiological Effects. *Microchemical Journal*, **146**, 789-797. https://doi.org/10.1016/j.microc.2019.02.001

[17] Joel, E.S., Maxwell, O., Adewoyin, O.O., Ehi-Eromosele, C.O., Embong, Z. and Saeed, M.A. (2018) Assessment of Natural Radionuclides and Its Radiological Hazards from Tiles Made in Nigeria. *Radiation Physics and Chemistry*, **144**, 43-47. https://doi.org/10.1016/j.radphyschem.2017.11.003

[18] Alhous, S.F., Kadhim, S.A., Alkufi, A.A. and Kadhim, B.A. (2020) Measuring the

Level of Radioactive Contamination of Selected Samples of Sugar and Salt Available in the Local Markets in Najaf Governorate/Iraq. *IOP Conference Series*: *Materials Science and Engineering*, **928**, Article ID: 072097. https://doi.org/10.1088/1757-899x/928/7/072097

[19] Song, M.H., Chang, B.U., Koh, S.M., Kim, Y.J., Kim, D.J. and Kim, G.H. (2011) Overall Natural Radioactivity of a Phosphate Fertilizer Industry in Korea. *Radioprotection*, **46**, S113-S118. https://doi.org/10.1051/radiopro/20116835s

[20] Dueñas, C., Fernández, M.C., Cañete, S. and Pérez, M. (2010) Radiological Impacts of Natural Radioactivity from Phosphogypsum Piles in Huelva (Spain). *Radiation Measurements*, **45**, 242-246. https://doi.org/10.1016/j.radmeas.2010.01.007

[21] Al-Jundi, J., Al-Ahmad, N., Shehadeh, H., Afaneh, F., Maghrabi, M., Gerstmann, U., *et al*. (2008) Investigations on the Activity Concentrations of ^{238}U, ^{226}RA, ^{228}RA, ^{210}PB and ^{40}K in Jordan Phosphogypsum and Fertilizers. *Radiation Protection Dosimetry*, **131**, 449-454. https://doi.org/10.1093/rpd/ncn214

[22] Korany, K.A., Masoud, A.M., Rushdy, O.E., Alrowaili, Z.A., Hassanein, F.H. and Taha, M.H. (2021) Phosphate, Phosphoric Acid and Phosphogypsum Natural Radioactivity and Radiological Hazards Parameters. *Journal of Radioanalytical and Nuclear Chemistry*, **329**, 391-399. https://doi.org/10.1007/s10967-021-07796-8

[23] Jose Madruga, M., Prudencio, M.I., Gil Corisco, J.A., Mihalik, J., Marques, R., Santos, M., *et al*. (2019) Distribution of Natural Radionuclides, Rare Earth Elements, Metals and Metalloids in a Phosphogypsum Stockpile. *International Journal of Waste Resources*, **9**, Article No. 363. https://doi.org/10.35248/2252-5211.19.9.363

[24] Ajam, L., Ben Ouezdou, M., Felfoul, H.S. and Mensi, R.E. (2009) Characterization of the Tunisian Phosphogypsum and Its Valorization in Clay Bricks. *Construction and Building Materials*, **23**, 3240-3247. https://doi.org/10.1016/j.conbuildmat.2009.05.009

[25] United Nations (2008) UNSCEAR 2008 Report on Sources and Effects of Ionizing Radiation. Vol. 1. https://doi.org/10.18356/a02938bf-en

[26] Abdullahi, S., Ismail, A.F. and Samat, S. (2019) Determination of Indoor Doses and Excess Lifetime Cancer Risks Caused by Building Materials Containing Natural Radionuclides in Malaysia. *Nuclear Engineering and Technology*, **51**, 325-336. https://doi.org/10.1016/j.net.2018.09.017

[27] Kolo, M.T., Aziz, S.A.B.A., Khandaker, M.U., Asaduzzaman, K. and Amin, Y.M. (2015) Evaluation of Radiological Risks Due to Natural Radioactivity around Lynas Advanced Material Plant Environment, Kuantan, Pahang, Malaysia. *Environmental Science and Pollution Research*, **22**, 13127-13136. https://doi.org/10.1007/s11356-015-4577-5

[28] Imani, M., Adelikhah, M., Shahrokhi, A., Azimpour, G., Yadollahi, A., Kocsis, E., *et al*. (2021) Natural Radioactivity and Radiological Risks of Common Building Materials Used in Semnan Province Dwellings, Iran. *Environmental Science and Pollution Research*, **28**, 41492-41503. https://doi.org/10.1007/s11356-021-13469-6