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RADIOLOGICAL RISK ASSESSMENT OF PHOSPHATE MINING IN EL-SEBAIYA LOCALITY, ASWAN ZONE, EGYPT

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Abstract. *In the present work, the specific activity concentrations of natural radionuclides of ^{238}U and ^{232}Th chain members, as well as ^{40}K were measured in phosphate samples using a gamma-ray spectrometric technique based on high-resolution hyper-pure germanium detectors (HPGe). Samples were collected from the El-Sebaiya area at the Aswan zone, Egypt. The external hazard index(Hex), the external absorbed dose rates(D), the annual effective doses (E) and the excess lifetime cancer risk (ELCR) due to gamma radiation from these samples have been calculated and compared with the corresponding average worldwide values. The evaluations of the associated radiological hazards from these materials on the workers during mining processes in the El-Sebaiya area were carried out.*

Keywords: *Annual effective dose E, gamma dose rate D, Excess Lifetime Cancer Risk ELCR, radiological hazards*

1. INTRODUCTION

The natural terrestrial gamma radiation dose rate is an essential contribution to the average dose rate received by the world's population. Estimation of the radiation dose distribution is important in assessing the health risk to the population and serves as a reference in documenting changes to environmental radioactivity due to anthropogenic activities [1]. Radionuclides with half-lives comparable with the age of the earth or their corresponding decay products existing in terrestrial material such as ^{238}U and ^{232}Th chains members, as well as ^{40}K are of great interest. Since these radionuclides are not uniformly distributed, the knowledge of their distribution in soil and rocks plays an important role in radiation protection and measurement [2]. Gamma radiation from these radionuclides represents the main external source of radiation exposure to the human body. The concentrations of these radionuclides in soil and rock are determined by their radioactivity that depends on the nature of the formation of these materials [3]. Therefore, these radionuclides generate a significant component of the background radiation exposure to the population.

Distribution of these radionuclides depends on geological and geographical conditions, but they can be redistributed as a result of human activities such as drilling for oil and gas, extracting uranium and phosphate and mining operations. So the exposure to natural sources is enhanced by technological activities. Generally, some of the non-nuclear industrial processes considerably contribute to the radiological pollution of the environment [4, 5].

The common example of these processes is extracting the phosphate ore. The phosphate ore is a complex material which contains calcium, phosphate, fluoride, carbonate and other elements or groups bound together in a crystal lattice. It is important for the natural sources of fertilizers, and also widely used in chemical industries. Phosphate rocks are used extensively, mainly as a source of phosphorus for phosphoric acid and other special chemicals. In addition, it usually contains relatively high concentrations of useful elements such as uranium, fluorine, potassium, and vanadium. Phosphate ores are present normally in the form of calcium phosphates Ca3 (PO4)2 (phosphorites), which are very old marine deposits associated with fossils. The second type of phosphate material is apatite Ca5 [(PO4)3(F)], which is of igneous origin. Also, phosphates are rich, typically, in uranium, and represent one of the sources of technologically enhanced natural radiation, which might increase exposure of people to natural radionuclides [6].

The major form of the phosphate ore is the sedimentary phosphate which represents 85% of worldwide production. It tends to have high concentrations of uranium isotopes (^{238}U) ranging from 50 to 200 mg/kg [7]. In sedimentary rocks, ^{238}U is generally found in radioactive equilibrium with its decay products, such as ^{226}Ra. While the activity concentrations of ^{232}Th and 40K are much lower than those of ^{238}U, they are comparable to those normally observed in soil. Phosphate deposits in Egypt occur

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mainly in the Duwi Formation, which is a phosphate-bearing unit that occupies a stratigraphic position at the top of the Quseir shale and underlies the Dakhla shale [8].

The main task of this study includes a measurement of the natural radioactivity level due to the decay of ^{238}U and ^{232}Th chains members, as well as ^{40}K using a Hyper-Pure Germanium detector (HPGe) on the phosphate samples collected from the El-Sebaiya locality, Aswan governorate, Egypt. From the radioactivity measurements of samples the external hazard index(Hex), the external absorbed dose rates (D), the annual effective doses (E) and the excess lifetime cancer risk (ELCR) have been calculated and compared with the average worldwide values to evaluate the associated radiological hazards from these materials on the workers during the mining processes in the El-Sebaiya area.

2. Materials and methods

2.1. Sampling (Collection and Samples Preparation)

Samples were collected from the phosphate rocks in the west of Nile River representing (the Nile valley) the El-Sebaiya areas, Idfo, Aswan government. The areas of study are located between 25.195 N longitude and 32.78 latitude. A total of nineteen represented samples of phosphate were collected from the El-Sebaiya area, Aswan governorate, Egypt. The samples were thoroughly crushed and pulverized to powders. The powders were sieved through a 200 μm mesh, which is the optimum size for substances rich in heavy minerals. The samples have been put in containers of cylindrical shapes. Containers were made of polypropylene with a density 0.946 g/cm^3. A container that had an inner diameter of about 6.5 cm with a wall thickness of 1.0 cm and a height of 6.0 cm and a tare weight of about 22.0 grams. The containers were tightly sealed for 4 weeks to avoid the escape of ^{222}Rn gas in order to ensure the secular equilibrium between ^{226}Ra and their respective progenies [9].

2.2. Experimental setup for γ-Ray Detection

High efficiency γ-spectrometer includes a hyper-Pure Germanium (HpGe) detector for 43, 200 S. The HPGe detector (EG& G Ortec Model GEM100P4) with a 100 % relative efficiency and a 2.1 keV full width at half-maximum (FWHM) at the 1.33 MeV gamma transition of 60Co was used. The samples gamma lines were analyzed for their emitters based on the HPGe γ- spectrometer using the Gamma vision Ortec software (Model A66-B32, version 6.00) for data analysis.

The average specific activity concentrations of ^{238}U chain members were calculated based on the energy transitions, which are direct γ-ray emitters, 186.21 keV (3.59%) for 226Ra, for 214Pb 295.1 keV (19.30 %) and 351.99 keV (37.60 %), and 214Bi activity determined from the 609.31 keV (46.10 %), 1120.28 (15.12%) and 1764.49 keV (15.40 %) emission gamma-lines. The gamma-line 186.21 keV was separated from the 185.72 keV (57. 20%) peak of ^{235}U using a method that was discussed in earlier works [10, 11].

The average specific activity concentrations of ^{232}Th chain members were calculated based on the energy transitions of 338.32 keV(11.27%), 911.21 keV(25.84%) and 968.97 keV(15.83%) for^{228}Ac, 238.63 keV(43.30%) for ^{212}Pb , 727.33 keV(6.58%) for ^{212}Bi and 2614.53 keV (13.10%) for ^{208}Tl. The activity of ^{40}K was determined from the 1460.7 keV(10.7%) emission gamma-line.

3. Results and discussions

3.1. Specific radioactivity

The results for the specific activity concentrations of the natural radionuclides ^{238}U and ^{232}Th chains members, as well as ^{40}K in the phosphate samples are reported in tables (1, 2).

Table 1. Specific activity concentrations of the natural radionuclides ^{238}U chain members in the collected samples.

Sample Number	Ra-226 Bq/Kg	Bi-214 Bq/Kg	Pb-214 Bq/Kg
1	749.11 ± 41.99	523.69 ± 4.91	594.00 ± 5.41
2	948.29 ±35.28	654.92 ± 5.57	758.15 ± 6.1
3	762.16 ±40.47	516.49 ± 4.76	580.26 ± 5.39
4	795.12 ± 27.99	569.61 ± 5.34	643.86 ± 5.66
5	939.17 ±38.22	627.04 ± 5.94	710.28 ± 5.87
6	802.27 ± 37.39	569.62 ± 5.15	654.3 ± 5.75
7	789.28 ± 35.99	549.19 ± 5.01	628.87 ± 5.01
8	810.26 ± 32.73	503.43 ± 4.89	574.84 ±5.51
9	791.85 ± 36.5	543.68 ± 4.96	616.56 ± 5.41
10	746.76 ± 34.13	543.95 ± 4.98	613.14 ± 5.5
11	791.85 ± 23.76	555.11± 5.14	630.96 ± 5.7
12	792.26 ± 27.49	561.64 ± 5.05	636.93 ± 5.49
13	986.41 ±66.58	687.11 ±10.10	780.73 ±10.32
14	812.84 ± 30.16	552.38 ± 4.93	623.36 ± 5.64
15	826.00 ±28.25	594.90 ± 5.15	698.75 ± 5.92
16	805.04 ± 27.13	585.75 ± 5.10	663.29 ± 5.71
17	799.87 ± 31.51	538.07 ± 5.00	615.93 ± 5.58
18	791.85 ±30.09	558.30 ±5.05	641.11 ± 5.55
19	766.95 ±33.98	544.04 ± 4.92	616.11 ± 5.35
Min.	746.76 ± 34.13	503.43 ± 4.89	574.84 ± 5.51
Max.	986.41 ±66.58	687.11 ± 10.10	780.73 ±10.32
Mean	816.17 ± 34.72	567.31 ±5.37	646.40 ± 5.83

The world average radioactivity concentrations and their ranges for ^{226}Ra, ^{232}Th (^{228}Ac) and ^{40}K in soils and rocks are 32 (17-60) Bq/kg, 45(11-64) Bq/kg and 420 (140-850) Bq/kg, respectively, (UNSCEAR 2000, 2008) [4, 12].

The average specific activity concentrations of ^{238}U chain members ^{226}Ra, ^{214}Pb and ^{214}Bi are 816.17 ± 34.72, 646.40 ± 5.83 and 567.31± 5.37 Bq/kg (dry weight), respectively.

The average specific activity concentrations of ^{232}Th chain members ^{228}Ac, ^{212}Pb, ^{212}Bi and ^{208}Tl are 19.51 ± 2.98, 20.98 ± 1.84, 23.26 ± 10.30 and 6.57 ± 0.74 Bq/kg (dry weight), respectively. The average specific activity concentration of ^{40}K is 79.89 ± 11.64 Bq/kg (dry weight).

It is clear that the average radioactivity concentrations of ^{232}Th (^{228}Ac) and ^{40}K in the samples are 2.31 and 5.26 times lower than the world average and the average radioactivity concentration of ^{226}Ra is 25.51 times higher than the world average as shown in Figure 1. These results can be interpreted as resulting from the geochemical behavior of the radionuclides during the development of the phosphate samples in the El-Sebaiya area, as was investigated in earlier work [10].

Table 2. Summary of the specific activity concentrations of the natural radionuclide ^{232}Th chain members, as well as ^{40}K in the collected phosphate samples

Sample Number	Ac-228 Bq/Kg	Pb212 Bq/Kg	Bi-212 Bq/Kg	Tl-208 Bq/Kg	K-40 Bq/Kg
1	21.84 ± 2.87	20.85 ± 1.69	25.46 ±12.19	6.65 ± 0.69	78.95 ± 7.81
2	17.33 ± 3.78	17.23 ± 1.82	15.18 ±11.57	5.39 ± 0.68	67.43 ± 11.09
3	18.16 ± 2.32	20.2 ± 1.70	24.88 ±11.12	5.99 ± 0.66	80.63 ± 10.56
4	18.12 ± 2.21	20.28 ± 1.74	28.4 ±10.99	6.87 ± 0.75	84.53 ± 9.34
5	13.59 ± 2.75	17.84 ± 1.80	21.37 ±11.93	5.38 ± 0.65	53.99 ± 9.43
6	19.85 ± 2.41	21.14 ± 1.83	31.01 ±12.45	7.05 ± 0.76	82.17 ± 7.13
7	21.12 ± 2.81	22.95 ± 1.79	28.09 ± 11.01	7.13 ± 0.75	77.45 ± 9.14
8	21.27 ± 2.67	21.58 ± 1.76	29.78 ± 9.51	6.62 ± 0.73	77.22 ± 10.53
9	19.58 ± 2.45	21.55 ± 1.77	20.24 ± 9.4	7.20 ± 0.79	84.72 ± 9.79
10	25.97 ± 3.52	22.12 ± 1.73	22.66 ± 6.85	7.08 ± 0.70	79.79 ± 9.60
11	18.93 ± 2.53	22.40 ± 1.78	11.4 ± 5.20	6.40 ± 0.76	97.29 ± 10.80
12	19.12 ± 2.75	22.33 ± 1.76	22.53 ± 9.36	6.71 ± 0.72	89.3 ± 12.72
13	20.13 ± 4.51	17.53 ± 3.20	24.35 ±16.50	5.75 ± 1.18	74.8 ± 21.18
14	19.45 ± 2.68	22.05 ± 1.74	22.83 ±10.92	6.53 ± 0.66	84.91 ± 12.46
15	20.52 ± 3.67	22.55 ± 1.79	21.25 ± 7.86	7.01 ± 0.70	82.06 ± 8.82
16	16.94 ± 2.70	20.66 ± 1.79	28.96 ±11.62	7.15 ± 0.72	77.21 ± 13.66
17	20.75 ± 3.69	21.84 ± 1.76	17.05 ± 9.42	6.72 ± 0.77	68.98 ± 13.91
18	19.71 ± 2.81	22.42 ± 1.86	24.19 ± 8.69	7.13 ± 0.70	86.62 ± 22.01
19	18.34 ± 3.57	21.11 ± 1.72	22.41 ± 9.09	6.16 ± 0.70	89.83 ± 11.13
Min.	13.59 ± 2.75	17.23 ± 1.82	11.4 ± 5.20	5.38 ± 0.65	53.99 ± 9.43
Max.	25.97 ± 3.52	22.95 ± 1.79	31.01 ±12.45	7.20 ± 0.79	97.29 ± 10.80
Mean	19.51 ± 2.98	20.98 ± 1.84	23.26 ±10.30	6.57 ± 0.74	79.89 ± 11.64

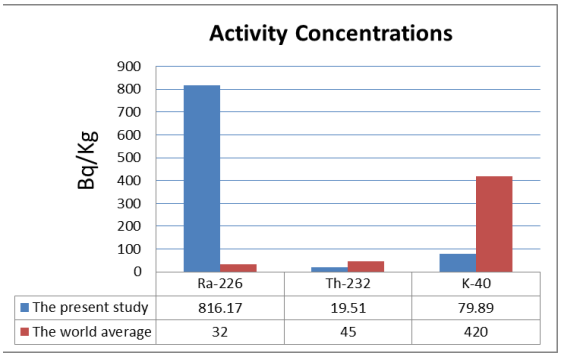

Figure 1. The Average activity concentrations in the collected phosphate samples

3.2. Radiological hazard parameters

3.2.1 External Hazard Index (H_{ex})

In order to assess the external radiological hazards, we use the index (H_{ex}). H_{ex} considers only the exposure risks due to gamma-ray and is defined as [13]

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (1)$$

where A_{Ra}, A_{Th}, and A_K are the specific activities of ^{226}Ra, ^{232}Th (^{228}Ac) and ^{40}K, respectively.

The external hazard index for the samples ranges from 2.13 to 2.76, with an average value of 2.30. The external hazard index for the studied samples as represented in Table 3 is higher than unity.

Table 3. Outdoor and indoor external absorbed dose rates (D), the annual outdoor (surface) and indoor (pits) effective doses (E) and the total excess lifetime cancer risk (ELCR) for the workers in the studied phosphate samples.

Sample No.	H_{ex}	$D_{outdoor}$ nGy/h	D_{indoor} nGy/h	$E_{surface}$ mSv/y	E_{pit} mSv/y	E_{total} mSv/y	ELCR x10^{-3}
1	2.13	224.35	314.10	0.21	0.18	0.39	0.64
2	2.64	276.46	387.05	0.26	0.22	0.48	0.78
3	2.15	220.24	308.33	0.21	0.17	0.38	0.62
4	2.24	242.46	339.45	0.23	0.19	0.42	0.69
5	2.60	263.14	368.40	0.25	0.21	0.45	0.75
6	2.26	243.41	340.77	0.23	0.19	0.42	0.69
7	2.23	235.09	329.13	0.22	0.19	0.41	0.67
8	2.29	216.36	302.90	0.20	0.17	0.37	0.61
9	2.23	232.35	325.30	0.22	0.18	0.40	0.66
10	2.14	233.39	326.74	0.22	0.18	0.40	0.66
11	2.23	237.03	331.84	0.22	0.19	0.41	0.67
12	2.23	239.63	335.49	0.23	0.19	0.41	0.68
13	2.76	290.07	406.10	0.27	0.23	0.50	0.82
14	2.29	235.60	329.84	0.22	0.19	0.41	0.67
15	2.33	254.44	356.21	0.24	0.20	0.44	0.72
16	2.26	248.63	348.08	0.23	0.20	0.43	0.70
17	2.26	229.78	321.70	0.22	0.18	0.40	0.65
18	2.23	238.86	334.41	0.22	0.19	0.41	0.68
19	2.16	232.07	324.90	0.22	0.18	0.40	0.66
Min.	2.13	216.36	302.90	0.20	0.17	0.37	0.61
Max.	2.76	290.07	406.10	0.27	0.23	0.50	0.82
Mean	2.30	241.76	338.46	0.23	0.19	0.42	0.69

3.2. Dosimetric estimation

3.2.1. Terrestrial γ-Dose Rates

The external absorbed dose rates, D (nGy/h), in the outdoor air at 1 m above the ground surface can be calculated from the activities of terrestrial radionuclides according to the following formula [4]

$$D_{outdoor} = \sum A_E * Cf \quad (2)$$

where A_E is the specific activity concentrations in Bq/Kg and C_f is the concentration to dose conversion factor in units of nGy/h per Bq/Kg. The conversion factors for ^{238}U chain members ^{226}Ra, ^{214}Pb and ^{214}Bi are C_{Ra-226} = 0.00175, C_{Pb-214} =0.0501 and C_{Bi-214} = 0.348 nGy/h per Bq/Kg, for ^{232}Th chain members ^{228}Ac, ^{212}Pb, ^{212}Bi and ^{208}Tl are C_{Ac-228}=0.213, C_{Pb-212} =0.0286, C_{Bi-212} = 0.0205 and C_{Tl-208} =0.306 nGy/h per Bq/Kg and for ^{40}K is C_{K-40} = 0.041 nGy/h per Bq/Kg [14].

So, the above Equation 2 can be written as

$$D_{outdoor(nGy/h)} = \sum C_U A_U + \sum C_{Th} A_{Th} + C_{K-40} \cdot A_K \quad (3)$$

According to the worldwide values [4, 12], the indoor contribution is 1.4 times higher than the outdoor dose, so the indoor dose rate was calculated from the following formula:

$$D_{indoor}(nGy/h)=1.4*D_{outdoor} \quad (4)$$

As presented in Table 3, by using Equations 3 and 4, $D_{outdoor}$ and D_{indoor} values for the samples range from 216.36 to 290.07 nGy/h, and from 302.90 to 406.10 nGy/h, with the average values of 241.76 and 338.46 nGy/h, respectively. The average worldwide values for the outdoor and indoor external absorbed dose rates are 58 (50-59) nGy/h and 84 (20-200) nGy/h (UNSCEAR 2000, 2008) [4, 12], respectively, so the average and range values of the outdoor and indoor external absorbed dose rates in the samples are 4.17 (3.73-5.0) and 4.0 (3.6-4.8) times higher than the world average, respectively, as shown in Figure 2.

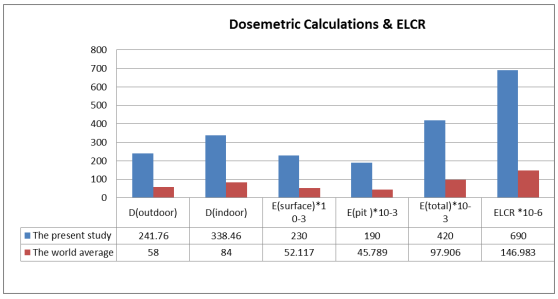

Figure 2. The Average parameters of dosimetric calculations & ELCR in the collected phosphate samples.

3.2.2. Terrestrial Annual γ- Effective Dose Rate

The annual effective dose rate, E (mSv/ y), to personnel from external exposure was calculated using the following equation:

$$E_i = D_i * OF * C_F \quad (5)$$

Where Ei is the annual effective dose due to the radionuclide, CF is the conversion factor for the absorbed dose in the air to the external effective dose in adults and is given as 0.7 Sv/Gy (UNSCEAR 2008) [12]; Di is the absorbed dose rate at 1m above the ground due to the radionuclide, and OF is the occupancy factor.

When workers are assigned to work in the pits (indoor), they usually spend about 3 h in the pit and 5 h on the surface (outdoor). Hence, their effective dose E can be calculated as follows:

$$E_{surface} = [D_{surface} * OF_{surface} * CF] * 10^{-6} \quad (6)$$

$$E_{pit} = [D_{pit} * OF_{pit} * CF] * 10^{-6} \quad (7)$$

$$E_{total} = E_{surface} + E_{pit} \quad (8)$$

where, $OF_{surface}$ = 5hr/day *5 day/week * 52.177 week/yr and for the indoor is OF_{pit} = 3hr/day *5 day/week * 52.177 week/yr.

By using equations 6, 7 and 8, the values of the annual surface and pit effective dose of the studied samples for the worker range from 0.20 to 0.27 mSv/y and from 0.17 to 0.23 mSv/y, with the average values of 0.23 and 0.19 mSv/y, respectively, and values of the total annual effective dose range from 0.37 to 0.50 mSv/y with an average value of 0.42 mSv/y, as represented in Table 3, (see Table 3).

The average worldwide values of the terrestrial gamma annual outdoor and indoor effective doses and the average total annual effective dose for adults are 0.07 mSv, 0.41 mSv, and 0.48 mSv, respectively, as reported in UNSCEAR 2000 and 2008 [4, 12]. By investigating these values it was found that they were calculated based on the occupancy factor of (8760 h ×0.2) and (8760 h ×0.8) for the annual outdoor and indoor effective doses, respectively. So, in comparing the present values with the average worldwide values we should take into account the present occupancy factor. That means the used average worldwide values of the annual outdoor (surface), indoor (pits) effective doses and the average total annual effective dose for workers are 0.0521 mSv, 0.0458 mSv, and 0.0979 mSv, respectively. So, the average and range values of the annual outdoor (surface), indoor (pits) effective doses and the average total annual effective dose in the samples are 4.36 (3.90 to 5.23), 4.17 (3.73 to 5.00) and 4.27(3.82 to 5.12) times higher than the world average values, respectively, as shown in Figure 2, (see Fig. 2).

3.2.3. Excess Lifetime Cancer Risk (ELCR)

Excess lifetime cancer risk (ELCR) was calculated, based upon the calculated values of the total annual effective dose (AED), using the following equation [15]:

$$ELCR = AED * DW * RF \quad (9)$$

where DW is the duration of work (40 years) and RF (Sv-1) is the risk factor, fatal and non-fatal cancer risks per Sievert. For stochastic effects after exposure to radiation at a low dose rate, ICRP 103 uses values of 0.041 for the adult workers [16].

The calculated range of ELCR is from $0.61x10^{-3}$ to $0.82x10^{-3}$, with an average of $0.69x10^{-3}$, as given in Table 3. The average worldwide value of the total (ELCR) is $1.45x10^{-3}$ [15]. By considering other corresponding values given in references [15,17and 18], it was found that the calculated value is based on the occupancy factor of 8760 h ×0.2 and 8760 h ×0.8 for the annual outdoor and indoor effective doses, respectively, with the duration of life (~66 years) and a risk factor, fatal cancer risk 0.05 Sv-1 based on the ICRP 60 values [19]. So, in comparing the present values with the average worldwide values, we should take into account the present occupancy factor, risk factor and the duration of work, not life. This means that the used average worldwide value of the total (ELCR) for workers is 0.14698x10-3, the average values of the total (ELCR) for workers in the samples are 4.66, ranging also from 4.17 to 5.59 times higher than the world average, respectively, as shown in Figure 2.

4. Conclusion

The results of the present work are summarized as:

1- The average radioactivity concentrations of ^{232}Th (^{228}Ac) and ^{40}K in the samples are lower than the world average, and the average radioactivity concentration of ^{226}Ra is higher than the world average. So most of the external absorbed dose results from the decay of the

natural radionuclides ^{238}U chain members and this is expected based on the formation of the sedimentary phosphate.

2-The external hazard index for the studied samples is more than twice the recommended value (unity).

3-The average and range values of the outdoor and indoor external absorbed dose rates (D), the annual outdoor (surface) and indoor (pits) effective doses (E) and the total excess lifetime cancer risk (ELCR) for the workers in the studied samples are several times higher than the world average.

Accordingly, from the results of this study, one can recommend:

(1) Doing a wide study to determine radiological hazards due to the inhalation and ingestion of the phosphate dust and the inhalation of ^{222}Rn.

(2) Taking the radiological protection measures by reducing the exposure time and wearing the appropriate clothes, shoes, and gags.

(3) Making regular, periodic cancer records and medical examinations for the safety of workers.

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