Assessment of natural radioactivity levels in phosphate rocks from Wadi Qena and Abu-Tartor mine in Egypt

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Abstract

The activity concentration of ²³⁵U, ²³⁸U, ^{234m}Pa, ²²⁶Ra, ²³²Th and ⁴⁰K were determined using gamma ray spectrometry with a high-purity germanium (HPGe) detector for eighteen samples of phosphate rocks collected from Wadi Qena and Abu-Tartor mine. To obtain the exact information about the measurement of ²³⁸U by gamma-ray spectroscopy, it is essential that any one of the daughters of ²³⁸U should exist in equilibrium with ²³⁸U. This condition is fulfilled by taking the average of all present measurements of daughters of ²³⁸U, namely ^{234m}Pa, ²²⁶Ra, ²¹⁴Bi and ²¹⁴Pb. The average activity concentration for phosphate rocks of Wadi Qena are 52.31, 864.69, 54.14 and 87.39 for ²³⁵U, ²³⁸U, ²³²Th and ⁴⁰K, respectively, while that of Abu-Tartor are 24.80, 419.13, 48.21 and 115.95 for ²³⁵U, ²³⁸U, ²³²Th and ⁴⁰K, respectively. The results were compared with different locations in Egypt and the world abroad reported in the literature. The radiation hazard to the occupational workers and public, the radium equivalent activities in Bq/kg, external and internal hazards and dose rate in nGy/hr are calculated. It seems that Abu-Tartor phosphate of sedimentary origin.

Keywords: Natural Radioactivity; HPGe Gamma-Ray Spectrometry; Phosphate; Egypt.

Introduction

Phosphate rocks constitute the bulk of the raw materials for the manufacture of phosphate fertilizers and some phosphorus based chemicals. It is well known that natural phosphates contain various stable and radioactive elements that could be of environmental concern to the public. Naturally occurring radioactive materials NORM, may contain any of the primordial radio nuclides or radioactive elements as they occur in nature, such as uranium, thorium, their radioactive decay products, and potassium that are distributed in the earth's crust in available amounts(**Chang et al ., 2008**). Radiation levels presented by NORM are generally referred to as a component of natural background radiation. Phosphate ores are present normally in the form of calcium phosphates $Ca_3(Po_4)_2$ (phosphorites) which are very old marine deposits associated with fossils. This form represents 85% of the world wide production. The second type of phosphate material is apatite that is igneous origin (**Schmidt., 1993**),(**EFMA., 1997**). Sedimentary phosphate ores tend to have high concentrations of uranium, where as magmatic ores, such as apatite do not. Typical activity concentrations of 238 U are 1500 Bq/kg (**UNSCEAR., 1993**) in sedimentary phosphate deposits and 70 Bq/Kg in apatite. Uranium-238 is generally found in radioactive equilibrium with its decay products, such as 226 Ra.

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The activity concentration of ²³²Th and ⁴⁰K in sedimentary phosphate rocks are much lower than those of ²³⁸U, and comparable to those normally observed in soil. Hazards could arise from usage of the rock materials in industrial plants especially in the manufacturing of phosphoric acid and fertilizers, due to the release of dust and polluted waters into the environment. The accumulation of these effluents in vitally important media such as water, soil and food are undesirable (**Isherwood ., 1992**).

Because of the increasing use of phosphate in industry world wide, it is interesting to investigate the potential radioactivity exposure of phosphate ores. In the present work natural radioactivity of Egyptian phosphate ores in Wadi Qena and Abu-Tartor mines, have been investigated.

2.Methods and Materials

Experimental procedure

Sampling and sample preparation for gamma spectrometers

Nine samples were collected from Wadi Qena from (West of G.El Missikat-W.El Gedmi) and nine samples were collected from Abu-Tartor mine. Figures 1 and 2 show the map of Egypt and locations investigated. The samples were then placed for drying at $110 \,^{\circ}$ C for 48 h to ensure that moisture is completely removed. A total of eighteen samples were crushed, homogenized and sieved to 100 mesh by a crushing machine. The mass of the prepared samples was weighted. The samples were collected in self-locking polyethylene bottles of 250 cm³ volume. The bottles were carefully sealed for more than one month to reach secular equilibrium between ²³²Th and ²³⁸U and its short lived daughter products (Mollah et al ., 1986).

Experimental set up

Spectra for different samples were measured with a high-purity germanium (HPGe) detector of high resolution gamma-ray spectrometer. The individual samples were placed on the detector manually and each sample was analyzed for a time of about 70000 sec.





Fig. 1 Location map for Abu-Tartor mine.Fig. 2 Location map for Wadi Qena mine

The detector has a resolution of 1.85 keV for the 1332.5 keV gamma-ray transition of ⁶⁰Co with relative efficiency 30%. To reduce the gamma ray background, a cylindrical lead shield with a fixed bottom and movable cover shield the detector. The lead shield contained two inner concentric cylinders of copper and cadmium. The detection system was calibrated by using standard point sources (²⁴¹Am, ⁶⁰Co and ²²⁶Ra). The efficiency curve of the HPGe detector in the energy range from 186 to 2450 keV was obtained through two stages. In the first stage, the relative efficiency curve of the detector was performed using ²²⁶Ra point source. In the second stage, the average relative curve of the detector was normalized to an absolute efficiency curve using potassium chloride solution having the same nature of the investigated samples (El-Tahawy et al., 1992). In order to determine the background distribution in the environment around detector, an empty bottle was counted in the same manner and in the same geometry as the samples. The background spectra were used to correct the areas of gamma rays of measured isotopes.

3. Results and discussion of natural radioactivity

Results and discussion of natural radioactivity

Calculation of activity concentrations in samples under investigation

The radioactivity concentration of the different identified radio nuclides was calculated with the following relation (**Tsoulfanidies.**, **1983**).

A = Net area (Cps) / I_{γ} . ζ . M

where, **A** is the activity concentration of gamma spectral line in Bq/kg, (**Cps**) is the net detected counts per second corresponding to the energy, ζ is the detector efficiency of the specific γ -ray energy. **M** is the mass of sample in kg and I_{γ} is the absolute transition probability of the gamma decay.

The activity concentrations (in Bq/Kg) for several measured radio nuclides of ²³⁵U, ²³⁸U series represented by ^{234m}Pa, ²²⁶Ra, ²¹⁴Bi and ²¹⁴Pb, Th-series represented by ²²⁸Ac, ²⁰⁸Tl and the gamma-ray line of ⁴⁰K are determined for all collected samples. Table 1 and Table 2 illustrate the activity concentrations in Bq/kg for the measured radionuclides ²³⁵U, ²³⁸U-series and ²³²Th-series for Wadi Qena and Abu-Tartor mine respectively. We see from Table 1 that the values of ²³⁵U vary between (44.93 and 58.26) and for ²³⁸U-series, the values of ^{234m}Pa vary between (679.48 and 1137.72) Bq/kg, the values of ²²⁶Ra vary between (669.52 and 1086.09) Bq/kg, the values of ²¹⁴Bi vary between (675.46 and 908.21) Bq/kg and the values of ²¹⁴Pb vary between (673.25 and 905.73) Bq/kg. For ²³²Th-series the values of ²²⁸Ac vary between (28.26 and 69.77) Bq/kg and the values of ²⁰⁸Tl vary between (20.75 and 70.37) Bq/kg. Table 2 also shows that, the ²³⁵U values vary between (21.99 and 28.09) Bq/kg and for ²³⁸U-series, the values of ^{234m}Pa vary between (352.63 and 658.87) Bq/kg, the values of ²²⁶Ra vary between (365.58 and 534.86) Bq/kg, the values of ²¹⁴Bi vary between (332.46 and 407.99) Bq/kg, the values of ²¹⁴Pb vary between (328.27 and 406.88) Bq/kg. For ²³²Th-series the values of 228 Ac vary between (30.30 and 52.74) Bq/kg and the values of 208 Tl vary between (28.95 and 72.38) Bq/kg.

Table 1

Sample	U-235		U-238	U-238 series Th-232 series			
No.	U-235	Pa-234m	Ra-226	Bi-214	Pb-214	Ac-228	T1-208
WQ-1	44.93	693.20	885.71	725.25	730.17	42.21	62.33
WQ-2	57.53	1067.74	925.15	838.89	843.50	57.34	70.37
WQ-3	58.26	991.22	885.89	842.93	848.75	66.94	63.99
WQ-4	54.69	1137.72	1081.43	908.21	905.73	46.97	68.39
WQ-5	47.74	696.27	975.23	771.34	761.89	28.26	62.69
WQ-6	47.32	880.32	926.74	763.67	764.22	58.59	54.17
WQ-7	52.88	970.41	1086.09	884.88	895.15	69.77	54.13
WQ-8	55.95	919.76	1053.39	871.11	899.37	52.24	56.74
WQ-9	51.49	679.48	669.52	675.46	673.25	38.54	20.75

Activity concentration (in Bq/kg) of the measured natural radionuclides isotopes for ²³⁵U, ²³⁸U- and ²³²Th-series in Wadi Qena phosphate samples.

Table 2

Activity concentration (in Bq/kg) of the measured natural radionuclides isotopes for ²³⁵U, ²³⁸U- and ²³²Th-series in Abu-Tartor phosphate samples.

	U-235		U-238	T	h-232 series		
Sample							
No.	U-235	Pa-234m	Ra-226	Bi-214	Pb-214	Ac-228	T1-208
AT-1	23.07	404.96	489.25	374.43	385.82	50.46	58.16
AT-2	23.95	426.28	534.86	395.31	406.88	37.72	40.95
AT-3	21.99	473.34	443.08	357.85	362.04	38.42	37.77
AT-4	24.14	465.88	463.14	362.82	370.14	38.12	28.95
AT-5	27.37	352.63	429.59	373.44	373.64	51.19	68.27
AT-6	24.73	530.37	383.32	332.46	328.27	33.45	71.89
AT-7	24.13	658.87	365.58	381.82	377.96	52.74	72.38
AT-8	28.09	513.60	415.54	407.99	404.99	30.30	58.91
AT-9	25.77	452.71	487.36	399.81	402.66	40.40	57.66

The values of the average activity concentration for ²³⁵U, ²³⁸U, ²³²Th and ⁴⁰K for Wadi Qena and Abu-Tartor samples were illustrated in Table 3 and 4, respectively. To obtain the exact information about the measurement of ²³⁸U by gamma-ray spectroscopy, it is essential that any one of the daughters of ²³⁸U should exist in equilibrium with ²³⁸U. This condition is now fulfilled by taking the average of all present measurements of daughters of ²³⁸U, namely ^{234m}Pa, ²²⁶Ra, ²¹⁴Bi and ²¹⁴Pb. Table 3 gives the activity concentration of ²³⁵U which vary between (44.93 and 58.26) Bq/kg with average 52.31 Bq/kg, ²³²Th vary between (29.65 and 65.47) Bq/kg with average 54.14 Bq/kg and ⁴⁰K vary between (25.55 and 123.53) Bq/kg with average 87.39 Bq/kg for Wadi Qena phosphate samples. Table 4 gives the activity

concentration of ²³⁵U which vary between (21.99 and 28.09) Bq/kg with average 24.80 Bq/kg, ²³⁸U which vary between (382.33 and 446.06) Bq/kg with average 419.13 Bq/kg, ²³²Th vary between (33.54 and 62.56) with average 48.21 Bq/kg and ⁴⁰K vary between (54.66 and 189.58) Bq/kg with average 115.95 Bq/kg for Abu-Tartor samples.

Table 3

The average activity concentrations of 235 U, 238 U, 232 Th and 40 K for Wadi Qena phosphate samples.

Sample	²³⁵ U	²³⁸ U	²³² Th	⁴⁰ k
No.	(Bq/kg)	(Bq/kg)	(Bq/kg)	(Bq/kg)
WQ-1	44.93	758.58	52.27	104.10
WQ-2	57.53	918.82	63.86	123.53
WQ-3	58.26	892.19	65.47	94.98
WQ-4	54.69	1008.27	57.68	114.27
WQ-5	47.74	801.18	45.48	84.38
WQ-6	47.32	833.74	56.38	109.68
WQ-7	52.88	959.13	61.95	91.04
WQ-8	55.95	935.91	54.49	38.99
WQ-9	51.49	674.43	29.65	25.55

Table 4

The average activity concentrations of 235 U, 238 U, 232 Th and 40 K for Abu-Tartor phosphate samples.

Sample	²³⁵ U	²³⁸ U	²³² Th	⁴⁰ k
No.	(Bq/kg)	(Bq/kg)	(Bq/kg)	(Bq/kg)
AT-1	23.07	413.62	54.31	92.88
AT-2	23.95	440.83	39.34	71.69
AT-3	21.99	409.08	38.10	59.91
AT-4	24.14	415.49	33.54	54.66
AT-5	27.37	382.33	59.73	183.45
AT-6	24.73	393.61	52.67	120.19
AT-7	24.13	446.06	62.56	189.58
AT-8	28.09	435.53	44.61	141.64
AT-9	25.77	435.64	49.03	129.56

The results show that the activity concentrations of ²³⁸U in Wadi Qena phosphate deposits are higher than that Abu-Tator mine. There are rocks of granite in Wadi Qena area, they considered as a source of uranium which lead to the increase of the concentration of uranium in this region. It is obvious that, the main radioactivity content of phosphate rocks is due to ²³⁸U and its decay products because it is widely believed that the radioactivity associated with phosphate rocks of sedimentary origin is formed by the adsorption and coprecipitation of uranium with calcium. The activity concentrations of ²³²Th, ²³⁵U and ⁴⁰K occur are low, hence their contribution to natural radioactivity is relatively low. Figure 3

shows the comparison of activity concentration for 238 U, 232 Th and 40 K for Wadi Qena and Abu-Tartor mine.



Fig. 3. Comparison of activity concentration for ²³⁸U, ²³²Th and ⁴⁰K in Bq/kg for Wadi Qena and Abu-Tartor mine.

Table 5 and 6 show the activity concentrations of 238 U, 232 Th in (ppm) and 40 k in (%) for Wadi Qena and Abu-Tartor samples, respectively. The specific activity of a sample containing 1ppm by weight of 238 U is 12.35 Bq/kg, 232 Th is 4.06 Bq/kg and 1% of 40 K is 313 Bq/kg (IAEA., 1989). We see from table 5 that the concentration of 238 U, 232 Th and 40 K vary between (54.61 and 81.64) ppm, (7.30 and 16.13) ppm and (0.08 and 0.39) %, respectively in Wadi Qena phosphate samples. We see also from table 6 that the concentration of 238 U, 232 Th and 40 K vary between (30.96 and 36.12) ppm, (8.26 and 15.41) ppm and (0.17 and 0.60) %, respectively in Abu-Tartor mine. The (232 Th/ 238 U) for all samples are lower than the Clark's value (3.5) which means that the samples are U-enrichment.

Table 5

Sample	²³⁸ U	²³² Th	⁴⁰ K%	²³² Th / ²³⁸ U
No.	(PPm)	(PPm)		
WQ-1	61.42	12.87	0.33	0.21
WQ-2	74.39	15.73	0.39	0.21
WQ-3	72.24	16.13	0.30	0.22
WQ-4	81.64	14.21	0.37	0.17
WQ-5	64.87	11.20	0.27	0.17
WQ-6	67.51	13.89	0.35	0.21
WQ-7	77.66	15.26	0.29	0.19
WQ-8	75.78	13.42	0.12	0.18
WQ-9	54.61	7.30	0.08	0.13

Activity concentrations of 238 U, 232 Th (in ppm), 40 K (in %) and (232 Th/ 238 U) Clark's value in Wadi Qena phosphate samples

Table 6

Activity concentration of 238 U, 232 Th (in ppm), 40 K (in %) and (232 Th/ 238 U) Clark's value in Abu-Tartor samples.

Sample	²³⁸ U	²³² Th	⁴⁰ K%	²³² Th / ²³⁸ U
No.	(PPm)	(PPm)		
AT-1	33.49	13.38	0.29	0.39
AT-2	35.69	9.69	0.23	0.27
AT-3	33.12	9.38	0.19	0.28
AT-4	33.64	8.26	0.17	0.25
AT-5	30.96	14.71	0.59	0.48
AT-6	31.87	12.97	0.38	0.41
AT-7	36.12	15.41	0.60	0.43
AT-8	35.27	10.99	0.45	0.31
AT-9	35.27	12.08	0.41	0.34

Calculation of γ - ray radiation hazard indices Radium equivalent activity

Radium equivalent activity (**Ra**_{eq}) is the most widely used radiation hazard index the radium equivalent is a weighed sum of activities of the ²²⁶Ra, ²³²Th and ⁴⁰K based on the assumption that 370 Bq/kg of ²²⁶Ra, 259 Bq/kg of ²³²Th and 4810 Bq/kg of ⁴⁰K produce the same γ -ray dose rates . The Radium equivalent was calculated from the following equation (Beretka and Mathew ., 1985)

$Ra_{eq} = C_{Ra} + 1.43 C_{Th} + 0.077 C_{K}$

where C_{Ra} , C_{Th} , C_K are the activity concentration in (Bq/kg) for ²²⁶Ra, ²³²Th and ⁴⁰K respectively.

Radiation level index

The representative level index radiation may be defined as (NEA ., 1979):

$I_{\gamma} = C_{Ra} \, / 150 \, + C_{Th} \, / 100 \, + C_{K} \, / 1500$

It is used to estimate the level of gamma radiation hazard associated with the natural radio nuclides in the samples.

External and internal hazard index

External hazard index H_{ex} measure the radiation exposure due to the radioactivity from the samples under investigation. In order to keep the radiation hazard to be insignificant, the value of this index must be less than unity. The external hazard is given by the following equation (Tufail et al .,1992):

$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810$

The internal exposure to 222 Rn and its daughter products is controlled by an internal hazard index, H_{in} which is defined as :

$H_{in} = C_{Ra}/185 + C_{Th}/259 + C_K/4810$

The values of the radiation level index I_{γ} and the maximum value of H_{ex} , H_{in} indices must be less than unity for the radiation hazard to be negligible. The maximum value of Ra_{eq} must also be less than 370 Bq/kg (UNSCEAR., 2000).

Calculation of the absorbed dose rate

The absorbed dose rate in air express the received dose in the open air from the radiation emitted from radio nuclides concentration in environmental materials. This factor is important quantity to assess when considering radiation risk to a bio system. The absorbed dose rate D_R in air one meter above the ground level owing to the concentration of 238 U, 232 Th and 40 K is given as:

$\mathbf{D}_{\mathbf{R}} = \mathbf{D}_{\mathbf{U}}\mathbf{C}_{\mathbf{U}} + \mathbf{D}_{\mathbf{Th}}\mathbf{C}_{\mathbf{Th}} + \mathbf{D}_{\mathbf{K}}\mathbf{C}_{\mathbf{K}}$

where D is the absorbed dose rate (in nG/h) and C_{Ra} , C_{Th} and C_K are the concentration of 238 U, 232 Th and 40 K (in Bq/kg). The D_U, D_{Th} and D_K are the dose rate coefficients (conversion factors) 0.4299, 0.666 and 0.042 expressed in (nGy/hr per Bq/kg) for U, Th and K, respectively (Ajayi and Kuforiji ., 2000) (El-Arabi et al ., 2006).

Finally in order to make a rough estimate for the annual effective dose outdoors, one has to take into account the conversion coefficient from absorbed dose in air to effective dose and the outdoor occupancy factor. In the UNSCEAR [1993, 2000] recent reports, the committee used 0.7 Sv Gy⁻¹ for the conversion coefficient from the absorbed dose in air to effective dose received by the adults and 0.2 for the outdoor occupancy factor. Effective dose rate in units of μ Sv/year is calculated by the following formula (Michalis et al., 2003).

Effective dose rate $(\mu Svy^{-1}) = Dose rate (nGyh^{-1}) \times 24h \times 365.25dx 0.2$ (occupancy factor) x 0.7 Sv Gy⁻¹(conversion coefficient)x10⁻³

Table 7, 8 tabulate the radiological parameters results which illustrates the radium equivalent in (Bq/kg), representative level index, external hazard, internal hazard, absorbed dose rate (nGy/hr) and in door and out door effective dose rate (μ S/y) in Wadi Qena and Abu-Tartor, respectively. From table (7) we found that the radium equivalent activity vary between (713.89 and 1181.69) Bq/kg which is higher than the recommended value for all samples (370 Bq/kg), a modified quantity of radium equivalent activity is the external and internal hazard indices, these indices must be lower than unity in order to keep the radiation hazard insignificant, the calculated values of the external hazard index vary between (1.93 and 3.19) and the internal hazard index vary between (3.74 and 6.13), the results of external and internal hazard indices are higher than the recommended limit. The values of radiation level index vary between (4.78 and 7.92) which is higher than the recommended value which is unity. The absorbed dose rates due to ²³⁸U, ²³²Th and ⁴⁰K in phosphate rocks from Wadi Qena vary between (310.76 and 476.67) nGy/h with average (411.46) nGy/h, The workers in Wadi-Qena are being exposed to radiation via two pathways: one is due to the external γ -radiation and the other is due to the inhalation of long-lived a-emitters and Rn-daughters associated with dust particles.

Taking an outdoor occupancy factor of 0.2 and a conversion factor of 0.7 Sv Gy⁻¹ we can estimate the average value of the outdoor effective dose rate from phosphate rocks in Wadi Qena is 504.96 μ Sv/y, This value is about 50.49% of the 1.0 mSv/y recommended by the International Commission on Radiological Protections (ICRP-60., 1990) as the maximum annual dose to members of the public. Taking the indoor occupancy factor of 0.8 and a conversion factor of 0.7 Sv Gy⁻¹ (UNSCEAR., 1988). to convert the γ -ray absorbed dose to effective equivalent for workers (that is, for a working period of 1820 h in a year), the average value of the in door effective dose rate is 419.36 μ Sv/y for Wadi Qena which is far below the world allowed dose of 20 mSv/y for workers (ICRP-60., 1990) .

Table 7: The values of radium equivalent in (Bq/Kg), external hazard index and internal hazard index, radiation level index, absorbed dose rate in (nGy/h), in door effective dose rate in (μ Sv/yr) and out door effective dose rat in (μ Sv/yr) in Wadi-Qena samples.

Sample No.	Ra _{eq}	H _{ex}	\mathbf{H}_{in}	Iγ	D _R	E _{eff(in)}	E _{eff(out)}
WQ-1	968.47	2.62	5.01	6.49	365.29	372.31	448.31
WQ-2	1025.98	2.77	5.27	6.89	442.72	451.22	543.32
WQ-3	986.83	2.67	5.06	6.62	431.14	439.42	529.118
WQ-4	1172.71	3.17	6.09	7.86	476.67	485.82	584.99
WQ-5	1046.76	2.83	5.46	7.01	378.26	385.52	464.22
WQ-6	1015.81	2.75	5.25	6.82	400.58	408.27	491.61
WQ-7	1181.69	3.19	6.13	7.92	457.41	466.19	561.35
WQ-8	1134.31	3.06	5.91	7.59	440.28	448.73	540.32
WQ-9	713.89	1.93	3.74	4.78	310.76	316.724	381.37

Table 8: The values of radium equivalent in (Bq/Kg), external hazard index and internal hazard index, radiation level index, absorbed dose rate in (nGy/hr), in door effective dose rate in (μ Sv/yr) and outdoor effective dose rat in (μ Sv/yr) in Abu-Tartor mine.

Sample No.	Ra _{eq}	H _{ex}	\mathbf{H}_{in}	\mathbf{I}_{γ}	D _R	E _{eff(in)}	E _{ff(out)}
AT-1	574.07	1.55	2.87	3.87	217.89	222.07	267.39
AT-2	596.64	1.61	3.06	4.01	218.72	222.92	268.43
AT-3	502.18	1.36	2.55	3.37	203.75	207.67	250.06
AT-4	515.31	1.39	2.64	3.46	203.25	207.16	249.44
AT-5	529.13	1.43	2.59	3.58	211.85	215.92	259.99
AT-6	467.89	1.26	2.30	3.16	209.34	213.36	256.91
AT-7	469.64	1.27	2.26	3.19	241.39	246.02	296.24
AT-8	490.24	1.32	2.45	3.31	222.89	227.17	273.54
AT-9	567.45	1.53	2.85	3.83	225.38	229.70	276.59

From table (8) we found that the radium equivalent activity varies between (467.89 and 596.64) Bq/kg which is higher than the recommended value for all samples (370 Bq/kg), a modified quantity of radium equivalent activity is the external and internal hazard indices, these indices must be lower than unity in order to keep the radiation hazard insignificant, the calculated values of the external hazard vary between (1.26 and 1.61) and the internal hazard indices are higher than the recommended limit. The values of the radiation level index vary between (3.16 and 4.01) which is higher than the recommended value which is unity. Figure (4) show the comparison of H_{ex} , H_{in} and I γ for Wadi-qena and Abu-Tartor mine.



Fig. 4: Comparison of H_{ex} , H_{in} and I_{γ} for Wadi Qena and Abu-Tartor samples.

The absorbed dose rates due to ²³⁸U, ²³²Th and ⁴⁰K in phosphate rocks from Abu-Tartor mine vary between (203.25 and 241.39) nGy/h with average (217.16) nGy/h, Taking an outdoor occupancy factor of 0.2 and a conversion factor of 0.7 Sv Gy⁻¹ we can estimate the average value of out door effective dose rate from phosphate rocks in Abu-Tartor is 266.51 μ Svy⁻¹, This value is about 26.65 % of the 1.0 mSv/y recommended by the International Commission on Radiological Protection as the maximum annual dose to members of the public. Taking the indoor occupancy factor of 0.8 and a conversion factor of 0.7 Sv Gy⁻¹ to convert the γ -ray absorbed dose to effective equivalent for workers (that is, for a working period of 1820 h in a year), the average value of the indoor effective dose rate is 221.33 μ Sv/y for Abu-Tartor mine which is far below the world allowed dose of 20 mSv/y for workers. From the calculated values it can be seen that the phosphate samples have level index above the proposed acceptable level in areas under study. The total absorbed doses (nGy/hr) are very high due to the higher radium contents in phosphate fertilizers. The acceptability of risks from ionizing radiation is decreasing this tendency leads to more regulation.

For comparison purposes, the activity concentration of ²²⁶Ra, ²³⁸U, ²³²Th and ⁴⁰K in phosphate rocks from different countries are given in Table 9. The increase of the natural uranium concentration can be attributed to leaching effects. Actual radionuclide concentrations will vary in location because of varying geological characteristics of phosphate ores in different regions as well as variation in processes used for phosphate mining and production. Also, we can conclude that the radiation dose to member of the public resulting from the use of Abu-Tartor phosphate rocks and Wadi Qena were negligible compared to the average annual effective dose from natural sources (1 mSv/y) even where assuming the complete accumulation of radio nuclides in soil over many years. Ventilation of the industrial area so as to avoid radon accumulation should be carefully considered.

Table 9

Activity concentration of ²²⁶ R	1, ²³⁸ U,	²³² Th	and	40 K	and radiation	hazard	R _{eq}	in	phosphate
rocks from different countries									

Country	²²⁶ Ra	²³⁸ U	²³² Th	⁴⁰ K	R _{eq}	Reference
Wadi Qena	943.24	864.69	54.14	87.39	1027.38	This work
Abu-Tartor	445.74	419.13	48.21	115.95	523.61	This work
Abu-Zaabal plant (Egypt)	514	523	37	19	568	(Hussein ., 1994)
Morocco	1600	1700	20	10	1629	(Guimond ., 1990)
Tiba Togo (calcined)	1100	1300	30	4	1143	(Guimond ., 1990)
Bu-Croa (Western Sahara)	900	900	7	30	912	(Guimond ., 1990)
USSR (Kola)	30	40	80	40	147	(Guimond ., 1990)
Egypt (El Mahamid)	567		217	217	921	(Abbady et al., 2005)
Egypt (W.El-Mashash)	666		329	329	1182	(Abbady et al., 2005)
USA (Florida)	1600	1500	20		1629	(Guimond ., 1990)
USA (Western)	1000	1000	20		1029	(Guimond ., 1990)
Jordan	1044		2	8	1048	(Olszewska ., 1995)
Tunisia	821		29	32	865	(Olszewska ., 1995)
Algeria	619		64	22	712	(Olszewska ., 1995)
Israel	1852		11		1868	(Olszewska ., 1995)
Sudan (Uro)	2263	2598	2.5	51.7	2270	(Sam and Holm., 1995)
Sudan (Kurun)	555	684	0.83	23	558	(Sam and Holm., 1995)
Tanzania (Arusha)	5022	4641	717	286	6069	(Makweba and Holm .,
Egypt (El-Sibaiya)	538		25	N.F	574.4	(El-Taher ., 2003)
Egypt (El-Quseir)	358		38	N.F	412	(El-Taher ., 2003)
Pakistan (Hazara)	440		50	207	527	(Khan et al ., 1998)
Finland	10		10	110	36	(Khan et al ., 1998)
Brazil	256	1313	3238	1202		(Daconceicao and
Morocco	1700	1700	30	10		(Azouazi et al ., 2001)
Nigeria/Sokoto	558		16	40		(Ogunleye et al ., 2002)
Saudi Arabia		519	40	250		(Akosy et al ., 2002)
Tanzania/Arusha	5760		350	280		(Banzi et al ., 2000)
Turkye	625	557	26	256		(Akyuz et al ., 2000)
Togo/Hahatoe	968		90			(Ogunleye et al ., 2002)
South Korea	355		4	49		(Chang et al ., 2008)
Pakistan	511	550	52	206		(Sabiha., 2010)

Conclusion

The gamma spectrometric analysis indicate that phosphate samples contain²²⁶Ra, ²³⁸U exhibits highest values of activity concentration as compared with ²³²Th, ⁴⁰K. The contribution of radioactivity to agriculture lands due to the application of phosphate fertilizers is the second concern for radiation protection point of view. However this contribution is not easily quantified, since the quantity of radioactivity spread along with fertilizers in the

agriculture fields depends upon the quantity of fertilizers used, the type of crop and area of its cultivation. From the present results, it seems that Abu-Tartor phosphate rocks have lowest activity levels than Wadi Qena, where the presence of Granite rocks in Wadi Qena leads to the contribution of uranium.

Acknowledgments

The authors are grateful to the nuclear physics laboratory, Faculty of Women for Arts, Science and Education, Ain shams university for the provision of laboratory facilities. Thanks are also extending to faculty of science, physics department, Cairo university for their kind assistance in samples analysis. Thanks to Nuclear Material Authority.

References

- Abbady, A.G.E., Uosif M.A.M., El-Taher A., (2005). "Natural radioactivity and dose assessment for phosphate rocks from Wadi El-Mashash and El-Mahamid Mines" Egypt. Journal of Environmental Radioactivity, V. 84, PP. 65-78.
- Ajayi, T.R. and Kuforiji, O.O, (2000)." Natural radioactivty measurments in rock samples of Ondo and Ekiti states in Nigeria". Radiation measurmens, V.33. PP.13-16.
- Aksoy A., Ahemed M., Matter W.S.A., El-Naggar Z.R., (2002) "Gamma-ray spectroscopic and PIXE analysis of selected samples from the phosphorite deposits of Northwestern Saudi Arabia", J. Radioanal. Nucl. Chem., V. 253, PP. 517-521.
- Akyuz T., Akyuz S., Varinlioglu A., Kose A., (2000). "Radioactivity of phosphate ores from Karatas-Mazidag phosphate deposit of Turkey". J. Radioanal. Nucl. Chem., V. 243, PP. 715-718.
- Azouazi M., Ouahidi Y., Fakhi S., Andres Y., Abbe J. Ch., Benmansour M., (2001) "Natural radioactivity in phosphates, phosphogypsum and natural waters in Morocco", J. Environ. Radioact., V. 54, PP. 231-242.
- Banzi F.P., Kifanga L.D., Bundala F.M., (2000). "Natural radioactivity and radiation exposure at the Minjingu phosphate mine in Tanzania" J. Radiol. Prot., V. 20, PP. 41-51.
- Beretka J., and Mathew P.J., (1985) "Natural radioactivity of Australian building materials, industrial wastes and by products " Health Phys., V. 48, PP. 87-95
- Chang B.U., Koh S.M., Kim Y. J., Seo J. S., Yoon Y.Y., Row J.W., lee D. M., (2008)."Nation wide survey on the natural radionuclides in industrial raw minerals in south Korea". J. Environ. Radioact. V. 99, PP. 455-460.
- International Atomic Energy Agency (IAEA) (1989). Construction and use Caliration Facilities for Radiometric Field Equipment . Technical Reports Series No. 309.Vienna

- Daconceicao F.T., Bonotto D.M., (2006) "Radionuclides, heavy metals and fluorine incidence at Tapira phosphate rocks". Brazil, and their industrial (by) products, Environ. Pollut., V. 139, PP. 232-243.
- El-Arabi A.M, Abbady A.E, Hussein A.S., (2006), "Gamma-Ray mesurments of natural radioactivity in sedimentray rocks from Egypt"., Nuclear Science and Techniques, V.17(2), PP. 123-128.
- El-Tahawy M. S., Farouk M. A., Hamad F.H., Ibrahim N.M., (1992). "Natural potassium as a standard source for the absolute efficiency calibration of Ge detector". Nucl. Sci. J. V.29, PP. 361.
- El-Taher A., (2003) . "Elemental studies of environmental samples from Upper Egypt by Neutron Activation Analysis". PhD thesis, Al-Azher University, Assuit, Egypt.
- Guimond R.J., (1990). "Radium in fertilizers". Technical Report No. 310 International Atomic Energy Agency (IAEA), Environmental Behavior of Radium, PP. 113–128.
- Hussein E. M., (1994). "Radioactivity of phosphate ore, super phosphate and phosphogypsum in Abu-Zaabal phosphate plant". Egypt . Journal of Health Phys., V. 67, PP. 280–283.

International commision on radiological protection publication (ICRP-60) (1990) . Oxford: Pergamon Press.

Isherwood K. F., (1992). "phosphate industry and the environment. In : J.J. Schultz (Ed.) Phosphate fertilizer and the environment" (PP. 115-124). Muscle shoals, Al: IFDC.

- Khan K., Khan H.M., Tufail M., Khatibeh A.J.A.H., Ahmad, N., (1998). "Radiometric analysis of hazard phosphate rock and fertilizers in Pakistan". Journal of Environmental Radioactivity, V. 35 (1), PP. 7-84.
- Makweba M. M., and Holm E., (1993). "The natural radioactivity of the rock phosphates, phosphatic products and their environmental implications". The Science of the Total Environment, V. 133, PP. 99–110.

Michalis T., Haralabos T., Stelios C and George C (2003). "Gamma-ray measurements of naturally occurring radioactive samples from Cyprus characteristic geological rocks". Radiation measurements, V. 37, PP. 221-229.

Mollah A.S., Ahmed G.U., Hussain S.R., Ruhman M.M.,(1986)."the natural radioactivity of some building materials used in Bangladesh" Health Phys. V. 50, PP. 849-851.

Nuclear energy agency (NEA) (1979). "Exposure to Radiation from natural radioactivity building materials", report by NEA Group export, OECD. Paris.

- Ogunleye P.O., Mayaki M.C., Amapu I.Y., (2002) "Radioactivity and heavy metal composition of Nigerian phosphate rocks": possible environmental implications, J. Environ. Radioact., V. 62, PP. 39-48
- Olszewska-Wasiolek M., (1995). "Estimates of the occupational radiological hazard in phosphate fertilizers industry in Poland". J. Radiation Protection Dosimetry, V. 58, PP. 269–276.
- Sabiha J., Tufail M., Asghar M., (2010) "Hazard of NORM from phosphorite of Pakistan". Journal of Hazardous Materials, V. 176, PP. 426-433
- Sam A. K., Holm E., (1995). "The natural radioactivity in phosphate deposits from Sudan". The Science of the Total Environment,l. 16, PP. 173–178.
- Schmidt G., (1993)." Handling of radium and uranium contaminated waste piles and other wastes from phosphate ore processing". Contract NETNU CT32-0084.
- European fertellizer manufactures Association (EFMA) (1997)."Report on discharges of radioactive substances by non nuclear industries", SPAR COM, Oslo and Paris commissions.
- Tsoulfanidies N., (1983). "Measurements and detection of radiation". (MC Graw- Hill Book Company , chap.16.

Tufail M., Ahmed N., Mirza S.M., and Khan H.A. (1992). The Science of the Total Environment, V. 121, PP. 283 -291.

- UNSCEAR (1988), United Nations Scientific Committee on the effects of Atomic Radiation Report. "Ionizing radiation: sources and biological effects".
- UNSCEAR (1993), United nations scientific Committee on the effects of Atomic radiation Report ."Ionizing radiation: sources and Biological effects".
- UNSCEAR (2000), United nations scientific Committee on the effects of Atomic radiation Report. "Ionizing radiation: sources and Biological effects". New York, United Nations publication.

تقييم مستويات النشاط الاشعاعي الطبيعي في صخور الفوسفات من وادي قنا ومنجم ابوطرطور في مصر

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ومتوسط تركيز النشاط الاشعاعي لصخور الفوسفات في وادي قنا هي (87.39, 54.14, 864.69, 52.31) بيكريل / كجم لكلا من اليورانيوم 235 واليوانيوم 238 والثوريوم 232 والبوتاسيوم 40 علي التتابع في حين في منجم ابو طرطور هي (115.95, 48.21, 419.13, 24.80) بيكريل / كجم لكلا من اليورانيوم 235 واليورانيوم 238 والثوريوم 232 والبوتاسيوم 40 علي التتابع

تم مقارنه هذه النتائج بالمستويات العالمية للاشعاع وحساب الجرعات الاشعاعية نتيجة التعرض الخارجي لاشعة جاما وحساب نشاط الراديوم المكافي Ra_{eq} لجميع العينات وحساب معامل الاخطار الخارجي H_{ex} ومعامل الاخطار الداخلي H_{in} ووجد أن الفوسفات ذات الاصل الرسوبي من منجم ابو طرطور لديه ادني مستوي من النشاط الاشعاعي .