Variations in radioactivity of phosphate rocks from different sites in Central Eastern Desert of Upper Egypt

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Abstract Natural radionuclides in the phosphate rock samples collected from Wadi Batur, Wadi Hegaza and Gabal Abu Had at Eastern Desert were determined using a high resolution γ-ray spectrometer. The experimental results concerning ^{226}Ra, ^{232}Th and their daughters are presented. Analyses of the measured spectra show that ^{226}Ra is distinguished with remarkable activities with average values ranging from 22.4 to 558 Bq/kg, while ^{232}Th activity concentration is in the range of 9.7 to 92.8 Bq/kg. The results of the analyses were found to be in a good agreement with the data obtained by others.

Keywords Phosphate rock, ^{226}Ra, ^{232}Th

CLC numbers P597^{+}.1, P598 A

1 INTRODUCTION

The Egyptian phosphates are widely distributed between latitudes 23° 10 and 29° 30 N. They are exposed along the escarpment Abu Tartur plateau, along the Nile Valley (Idfu and El Sibaiya) and the western coast of the Red Sea between Safaga and Quseir. The rugged relief of the Red Sea region is one of its characteristic features and is mainly attributed to faulting. The phosphate deposits are widely distributed and extend as a belt along the western coast of the Red Sea between Safaga and Quseir. These deposits were the subject of several authors.[1-3] They dealt with the topography, structure, stratigraphy and petrology of the phosphate deposits. These deposits are poor in the phosphate content as compared with those of the Western Desert. The latter have P_2O_5 contents ranging from 19.5% to 30.5% with an average of 28%, while the phosphate deposits of the Red Sea have P_2O_5 contents ranging from 20% to 22%.[2]

Phosphate ores contain an important amount of natural radioactive elements, specially ^{238}U and descendants, which depend on its geographical and geological origin. There are two major types of phosphate rocks: (1) sedimentary phosphate, which represent about 85% of the phosphate rock, were formed mainly from organic residues; they contain 100–200 μg/g uranium and 2–20 μg/g thorium,[1] and (2) igneous phosphates,

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which supply the remaining part of the phosphate rock, are of volcanic origin; they contain less than 10 ppm uranium but contain an appreciable amount of thorium and rare earth elements.[1]

Sedimentary and igneous phosphate ores are used as raw material for the production of phosphoric acid, and, consequently, fertilizers for agricultural purposes. Over 70% of the phosphate rocks are used in making phosphoric acid by a wet process. The results of this process are phosphoric acid and gypsum residue. One part of the uranium originally present in the phosphate rock is transferred to the phosphoric acid. Moreover, during the calcinations step an important part of the uranium (about 30%) becomes insoluble in sulfuric acid and will be removed with gypsum residue.[1] Large-scale production of phosphoric acid results in the redistribution of huge amounts of natural radioactivity. Depending on the technology used to produce fertilizer or phosphoric acid, the distribution of radioactive elements among the various products will be different.[4] Sometimes fertilizers and a common by-product, phosphogypsum, contain such high activities of natural radioactive elements that the resulted radiological impact should be considered carefully.[5,6] Generally there is a positive correlation between uranium and P_2O_5 content in the sedimentary phosphate.[1,7] The area under investigation, which is located to the east of Qena and Idfu cities in the western Central Eastern Desert of Egypt and covers more than 8000 km^2, lies principally between latitudes 25° 10 N–26° 30 N and between longitudes 32° 50 E–33° 30 E as shown in Fig.1.

The purpose of this study is to investigate the field relationship between Egyptian phosphate deposits and radioactive minerals and to compare the mode of occurrence of uranium mineralization in the Egyptian phosphate with that found in the other countries. This study would also report the types of radioactive minerals found in the Egyptian phosphate from central Eastern Desert area at Wadi Batur, Wadi Hegaza and at Gabal Abu Had.

2 EXPERIMENT PROCEDURE

Environmental radiation measurements are usually made to identify the radionuclide sources in the environment, quantify their concentrations, and estimate their dose contributions. This is most readily done by spectrometric technique, but simpler measurements can often yield relevant information. In each case, an essential part of the system is methodology associated with detector calibration, measurement procedure, and data analysis and interpretation.

^{226}Ra, ^{232}Th and ^{40}K activity concentration (Bq/kg) in the phosphate rock samples were measured using an HPGe γ spectrometer. Uranium is calculated from the counts received by the γ-ray detector in the energy window corresponding to ^{214}Bi. This

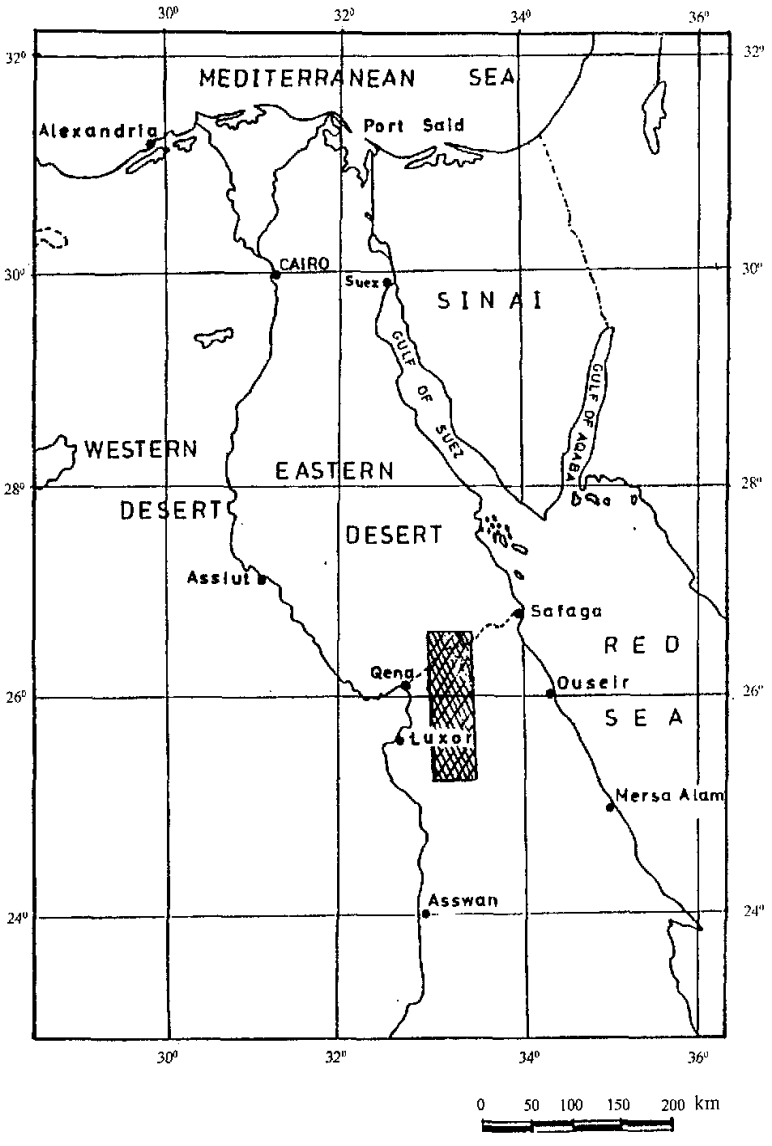

Fig.1 Location map of the studied area

technique assumes that uranium and its decay products are in secular equilibrium. The same technique is used to estimate ^{40}K and equivalent ^{232}Th. The applied low level γ-ray spectrometer consists basically of a high purity germanium detector and its electronic circuits. The detector is coaxial closed end, closed facing window geometry with a vertical dipstick (500–800 μm). The detector is located in a shielded chamber of four layers

starting with Plexiglas's (10 mm thick), copper (30 mm thick), lead (100 mm thick) and finally cadmium (3 mm thick) from outside to inside. In the present work, the spectra of standard sources with known γ-ray emission rates, usually with few γ lines, were measured as a function of two parameters, efficiency-energy dependence and efficiency-sample volume dependence.

Sixty samples of phosphorite and phosphatic rocks were collected from Duwi Formation (Maestrichtion age)[3] at Central Eastern Desert area in Wadi Batur, Wadi Hegaza and at Gabal Abu Had (see Fig.1). The rock samples were crushed to small pieces and dried at 105°C. The dried samples were ground to a grain-size of about 100 mesh. Every powdered sample was then mixed many times using an electric shaker to be sure that the sample is homogeneous. After preparing the phosphate samples, each sample was put in a container of 40 mL volume. The samples were sealed and the measurements were made one month later to assure secular equilibrium between the ^{226}Ra and its daughters. Then the sample was put face to face over the detector for at least 10 h. The spectra were either evaluated with the computer software programme Maestro (EG&G ORTIC), or manually by using a spread sheet (Microsoft Excel) to calculate the natural radioactivity. In the ^{238}U chain, the ^{226}Ra activity of the samples was determined through the intensity of the 351.9 keV and 609.3 keV γ line. Thorium activity was monitored through the ^{208}Tl, and ^{228}Ac emissions at 583.1 keV and 911.1 keV, respectively. Samples were chosen representing the high radioactivity rocks. These samples were analysed chemically to find the relation between the elements contained in phosphate rocks. Several reviews have been published covering both the applications and the constructions of semiconductor detectors, the necessary analytical accuracy is obtained through a special experimental technique-methodical scheme.[8–10]

3 RESULTS AND DISCUSSIONS

Although the radium content is quite small, it still represents a pollution hazard because gypsum is in the form of a fine powder and may result in dust blowing when stockpiled in the exterior. Radium is the most dangerous decay product of uranium because it disintegrates into the radioactive gas radon, whose half-life is 3.8 d. The results obtained from the γ spectrometric analysis indicate that the main radioisotopes in the rock phosphate and phosphate concentrate (ground rock phosphate) samples were ^{226}Ra and a small amount of ^{232}Th in some samples. ^{137}Cs could not be detected. The distribution of the activity concentrations in the different layers fluctuates between maximum and minimum values. In Wadi Batur for ^{226}Ra the maximum value is 538±27 Bq/kg and the minimum value is 60.7±13 Bq/kg with an average value of 278±94 Bq/kg, while the corresponding values of the ^{232}Th were found to be 9.7±6 Bq/kg

to 92.8±12 Bq/kg with an average value of 39.9±23 Bq/kg. In Wadi Hegaza the maximum value of ^{226}Ra is 558±29 Bq/kg and the minimum value is 22.4±9 Bq/kg with an average value 257.5±88 Bq/kg, while the corresponding values of ^{232}Th were found in the range of 9.7±6 Bq/kg to 74±14 Bq/kg with an average value of 44.4±28 Bq/kg. On the other hand, the activity concentrations in Gabal Abu Had ranged from a maximum value of 485±25 Bq/kg to a minimum value of 61.3±14 Bq/kg with an average value of 200.9±84 Bq/kg for ^{226}Ra, while the corresponding values of ^{232}Th were found in the range of 16.8±9 Bq/kg to 78.4±16 Bq/kg with an average value of 44.6±27 Bq/kg. Table 1 summarizes the calculated values of the activity concentration (Bq/kg) of ^{226}Ra and ^{232}Th for all measured phosphate rock samples in the area under investigations.

The variable values in the results are related to the different components and regions of these samples. Some samples contain high concentration of uranium, which is due to the higher phosphate oxide, and calcite content, but phosphatic rock samples (limestone, sandstone) have a small amount of uranium. An extensive use of the ground rock phosphate and phosphate fertilizers was assessed in relation to their radiological implications to the farmers.

Figs.2 and 3 show the relation between the ^{226}Ra, ^{232}Th activity concentrations

Table 1 Natural radioactivity in phosphate rock samples from the studied area (Bq/kg)

Sample	Wadi Batur		Wadi Hegaza		Gabal Abu-Had	
	^{232}Th	^{226}Ra	^{232}Th	^{226}Ra	^{232}Th	^{226}Ra
s1	90.0±11	504±17	57.3±12	318±22	16.8±9	61.3±14
s2	14.9±8	365±22	20.1±9	122±19	24.0±11	68.2±16
s3	92.8±12	538±27	9.7±6	22.4±9	52.3±13	254±18
s4	62.5±11	343±23	67.8±11	354±13	61.8±17	384±21
s5	73.4±13	509±21	73.7±9	418±18	38.2±12	179±19
s6	48.0±9	295±19	48.2±11	223±17	41.7±13	199±16
s7	37.3±8	361±17	26.7±13	82.3±14	52.6±11	146±17
s8	10.2±6	70.2±21	23.9±10	93.1±16	34.8±16	138±18
s9	70.6±11	382±19	20.8±9	105±13	53.4±12	237±23
s10	9.7±6	61.4±13	56.3±13	233±21	38.1±14	172±19
s11	22.9±11	456±21	70.7±12	415±18	30.3±13	124±11
s12	37.0±9	280±25	64.2±9	384±19	78.4±16	485±25
s13	31.9±11	63.5±17	31.7±11	454±23	62.8±13	383±23
s14	18.1±7	61.4±19	24.2±9	432±27	38.9±16	179±21
s15	24.3±9	67.1±21	23.8±11	46.0±18	62.1±14	197±19
s16	32.2±12	254±23	62.7±9	342±23	34.3±13	145±21
s17	62.4±11	282±19	40.2±12	205±28	34.5±14	138±19
s18	38.0±9	178±21	74.0±14	558±29	52.4±11	237±16
s19	27.2±11	219±18	56.7±12	281±17	37.8±16	172±21
s20	32.4±9	166±21	32.1±9	63.0±15	48.3±12	124±13
Mean	39.9±23	277.7±91	44.4±28	257.5±88	44.6±27	200.9±84

(Bq/kg) and the layers in the area under investigation, and Fig.4 illustrates the proportional relation between ^{226}Ra and ^{232}Th, the correlation factor was found 0.714, 0.871 and 0.735, respectively. Since the distribution of the isotopes in the different regions and layers is exempted from regulatory control, the values of their activity concentration spread in a wide range for all regions under investigation. The recorded high activity concentrations are considered to be indications of the existence of phosphate-bearing sediments of increased uranium content. It deserves to be mentioned that the phosphate outcrops striking NW-SE representing the hard and resistant varieties, possess a higher radioactivity level of uranium content, whereas the NE-SW striking exposures are a relatively lower radioactivity level, which represent the normal type of phosphate-bearing sediments. The lower activity level connected with the lower phosphate-bearing member could be easily related to the alternation of gray quartz sandstone with the poor normal phosphate and fragments of silicified wood. Meanwhile, the higher activity level related to the middle/upper phosphate-bearing member could be attributed to the coarse-grained, hard and resistant phosphate beds and in some places to intercalations by clay beds.[11]

The natural radioactivity in phosphate rocks depends on its origin and its geological properties. In sedimentary rocks it is much higher than in volcanic rocks.[12] Some typical concentration values are given in Table 2 besides the results of the present work, indicating the presence of a fair agreement between them.

An increase in the quantities of ^{226}Ra can be attributed to the increase in the concentration of natural uranium in samples under test. The increase of the natural uranium concentration may be due to the leaching effect.

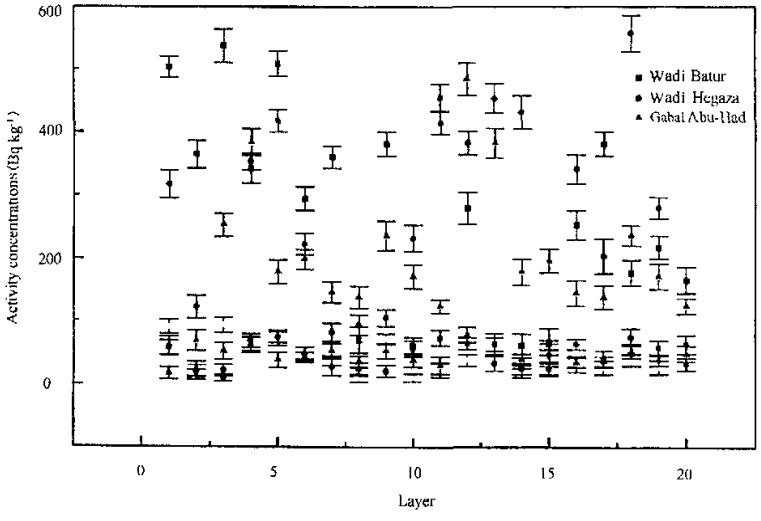

Fig.2 ^{226}Ra activity concentration in the areas under studies

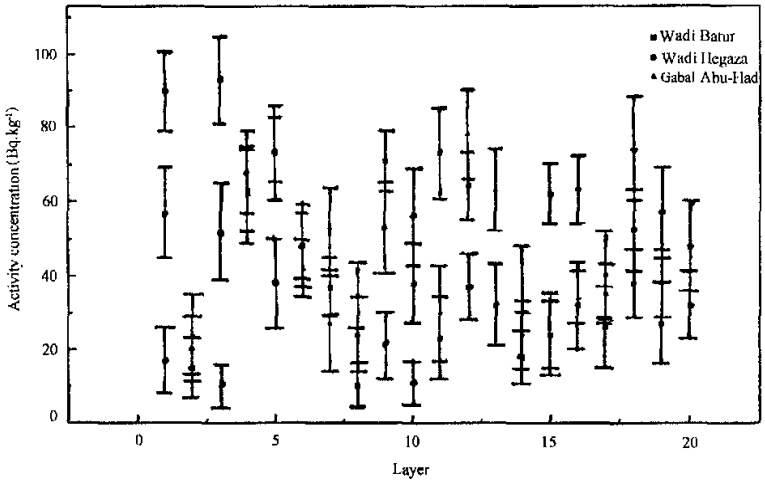

Fig.3 ^{232}Th activity concentration in the areas under studies

Table 2 Some typical values of activity concentrations in phosphate rock (Bq/kg)

Origin	Average concentration			Reference
	^{226}Ra	^{232}Th	^{40}K	
Eastern Desert	272	43	···	Present work
Gabal Owina (Nile Valley)	300	15	5	[13]
Gabal Abu-Tartur (Western Desert)	424	14	47	[13]
UK	1529	13.2	19	[14]
Florida	1600	16	···	[15]
Morocco	1700	30	···	[15]
Kola	40	90	···	[15]
China	150	25	···	[15]

Thorium is also usually more abundant in the suspended load than in the bottom sediments. This is interesting because the chemical behavior of thorium in natural waters is quite different from that of uranium. Thorium is essentially insoluble in normal surface waters, and so thorium transport is within particulate matter rather than in solutions.[16] Unlike uranium and thorium, potassium is a major element in rock forming minerals occurring mainly in alumino silicates such as feldspars and micas. Uranium does not constitute rock forming minerals but it occurs as a trace element in accessory minerals, as UO_2 precipitates, and as a chemical absorption element. Uranium forms chemical ion complex bonding with some oxides e.g. Fe_2O_3, Al_2O_3, K_2O and SiO_2 and a positive correlation with them is often found. Absolute variations in the amount of U therefore reflect the rock type. Like uranium, thorium is a common trace element in most geological

environment. Heavy accessory minerals are generally rich in thorium.[17] The increase of uranium with Fe_2O_3, Al_2O_3, K_2O and SiO_2 may be due to the fact that these constituents are acting as effective adsorbing agents for uranium.

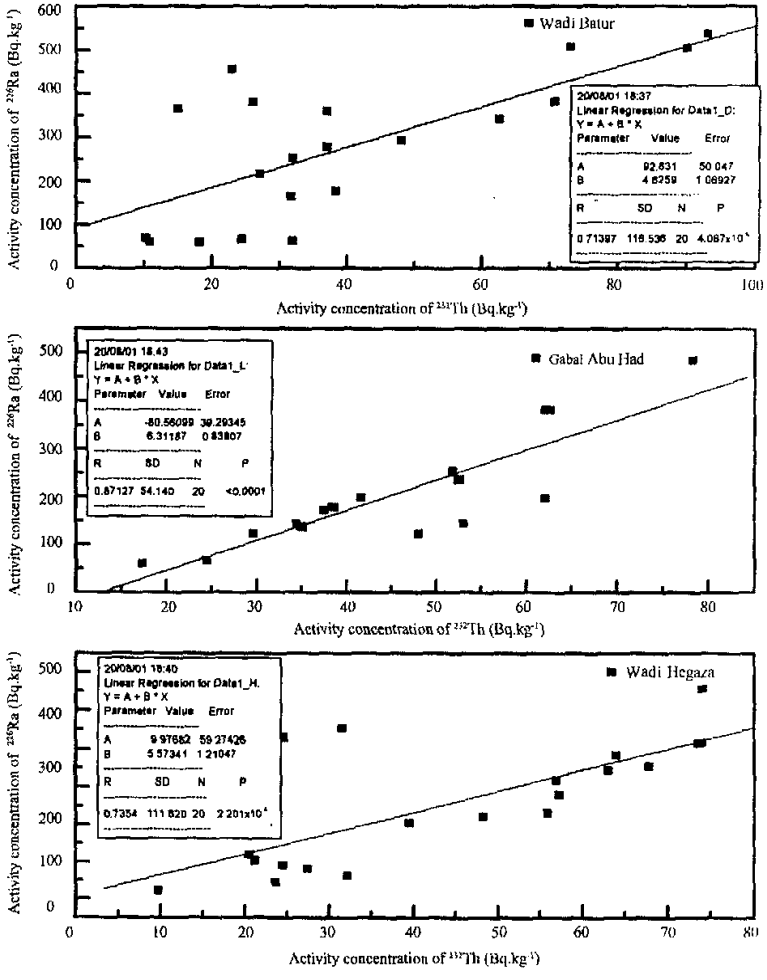

Fig.4 The relation between the activity concentrations (Bq/kg) of ^{226}Ra and ^{232}Th in the area under investigation

4 CONCLUSION

The radiochemical measurement of the uranium content of the phosphate rock is very important with respect to the protection of environment, since the phosphate rock

is a potential source of pollution. This study of the phosphorite deposit at Wadi Btur, Wadi Hegaza and Gabal Abu-Had in Upper Egypt reveals the following results. (1) All publications about uraniferous phosphorite deposits indicate that U ions are deeply involved in the chemistry of the phosphate pellets alone and are the result of their replacement of Ca ions. (2) The dissemination of U in phosphate pellets and carbonate fragments shows that there is no preferential affinity between the phosphate and U in the Upper Egypt phosphorite deposit. (3) The present result for the U-bearing phosphorite in Upper Egypt are in a fair agreement with the other investigations.

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