

Natural Radioactivity Measurement in Sedimentary Rock Samples Collected From the Bahariya Oasis, Western Desert, Egypt

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Abstract. The specific activities of natural radionuclides in ten sedimentary rocks samples collected from the Bahariya oasis area of Western Desert district, Egypt, have been evaluated. Experimental results were obtained by using a high-purity germanium (HPGe) detector and gamma-ray spectrometry analysis system. It was found that, the sedimentary rocks specific activity ranges from 6.9 to 43.5 Bq kg⁻¹ for ²³⁸U, between 0.5 and 33.9 Bq kg⁻¹ for ²³²Th and between 6.5 and 1007.7 Bq kg⁻¹ for ⁴⁰K. The activity of the Bahariya Formation rises from base to top and reaches 1007.7 Bq kg⁻¹ for ⁴⁰K which records the highest in the whole area at all in its upper most glauconitic siltstone, mudstone layer. In addition, it was found that the activity increases with the occurrence of Fe-oxide minerals as in the ferruginous sandstones, glauconite in siltstone and mudstone layers in the Bahariya and Naqb formations. The activity of the carbonate rocks shows the highest in the Naqb Formation (pink limestone), less in the Qazzun Formation and the lowest in the Khoman Chalk, particularly for Th and K activity. The average value of gamma dose rate (30.04 nGy/h) and the average annual effective dose (0.184 mSv y⁻¹) obtained in this study are lower than the worldwide dose rate average value (55 nGy/h) and the maximum annual dose to members of the public (1.0 mSv y⁻¹), respectively.

Keywords: Radioactivity; Sedimentary Rocks; Gamma Spectrometry; Bahariya Oasis.

Introduction

The Bahariya oasis is one of the five oases located nowadays in the Egyptian Western Desert (together with the Kharga, Dakhla, Farafra, and Siwa oases). It is set in a depression covering over 2000 Km² representing the lowest point in Egypt. It is located in the heart of the Western Desert of Egypt between latitudes 27° 48' and 28° 30' N and longitudes 28° 32' and 29° 10' E, about 370 km southwest of Cairo and 190 km west of Samalut in the Nile Valley (Fig. 1). Due to many populated villages and cultivated lands all over the Bahariya Oasis, the authors aimed to study the natural radioactivity of the area under consideration. This is considered as a step to establish a database reference of radioactivity background levels in the Western Desert region of Egypt.

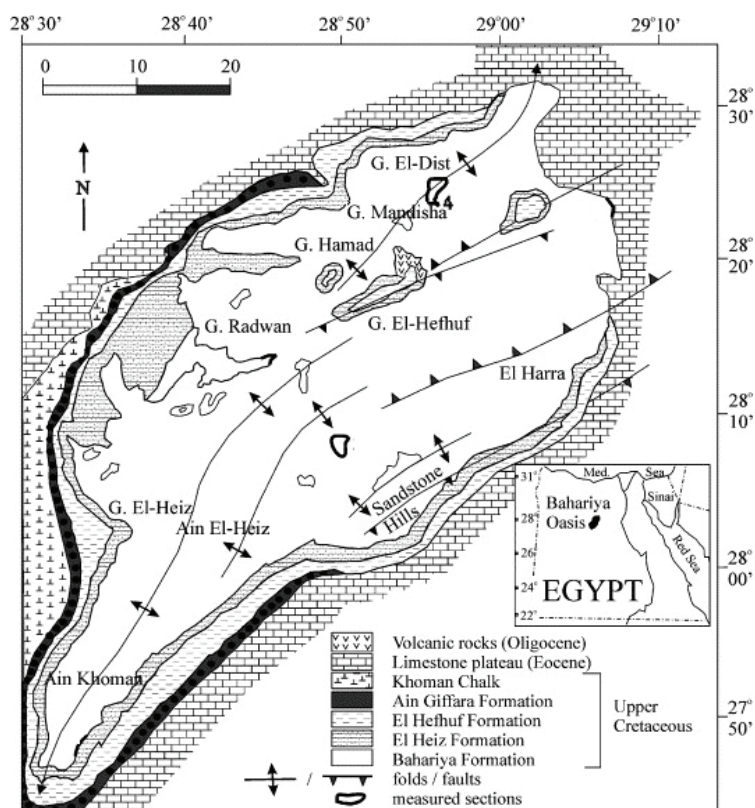


Fig. 1. Location map showing the general geology of the study area (after Catuneanu *et al.*, 2006).

Geological Setting

A brief description to the lithology of the Bahariya Formation, the Naqb Formation, the Qazzun Formation, the Khoman Chalk and the playa deposits is discussed in the following sections. Comprehensive discussions of the geology and structural setting are given elsewhere by (Ball and Beadnell 1903; El-Akkad and Issawi 1963; Hilmy *et al.*, 1984; Morsy, 1987; Said 1990; Zaghoul *et al.*, 1993; Samir 1994; El Bassyouny, 1970, 2004; Catuneanu *et al.* 2006 and online web page <http://www.nilevalleytravel.com-Geology of Egypt>).

Bahariya Formation

The Cenomanian Bahariya Formation is the oldest exposed unit in the Bahariya Depression and forms most of the cliffs and slopes surrounding the depression. Gabal El Dist, lies at lat. 28° 25' N and long. 28° 55' E, is considered as the type locality of the Bahariya Formation (Ball and Beadnell, 1903; Morsy, 1987; Said, 1990; and Catuneanu *et al.*, 2006). It is represented by dominantly a clastic succession, with lithologies ranging from light colored fine- to very fine-grained sandstones with horizontal lamination (with thicknesses of about 15m) capped by dark gray mudstones and yellow to creamy colored claystones at the base grading upwards into fossiliferous glauconitic siltstones and sandstones (Morsy, 1987; and Catuneanu *et al.*, 2006). Numerous iron-rich paleosol horizons, referred to as ferricretes by Catuneanu *et al.* (2006), are present almost throughout the Bahariya Formation, Fig. (3b). Columnar Oligocene flood basalt sheets cover the Bahariya Formation at Gebel Mandisha area which is located with the position of 28° 54' E and 28°22' N in the Bahariya oasis depression, (Fig.1 & 2).



Fig. 2. Photograph for Gabal Mandisha area, Bahariya Oasis.

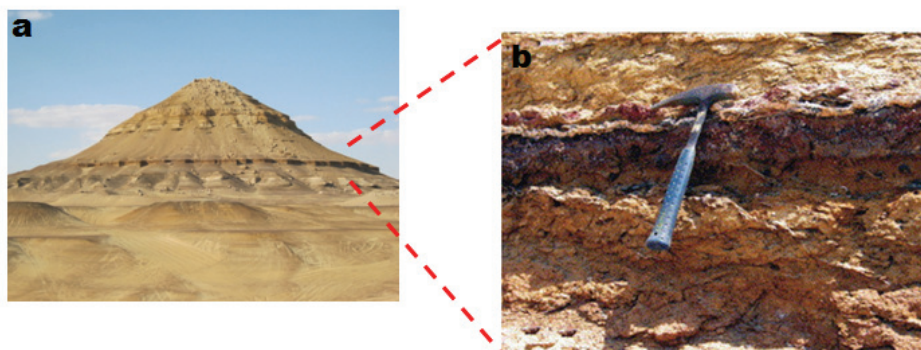


Fig. 3. Photograph for Gabal El-Dist (a), base of Bahariya Fm section shows yellow claystone capped by red iron-rich layer (b).

Khoman Formation

The Khoman Formation is made up of massive snow-white chalk and chalky limestone deposited during the Late Cretaceous (Maastrichtian), about 70 million years ago, when the sea covered the vast area of Western Desert (Ball and Beadnell, 1903; El-Akkad and Issawi, 1963; Said 1990; Zaghloul *et al.*, 1993; and Samir, 1994). The chalk, which reflects deep marine conditions, is characterized by a great number of joints filled with calcite veins, small iron-rich pipes and pyrite concretions. The floor of white desert is partly covered by small fragments of pseudomorphs of iron oxides after pyrite (<http://www.nilevalleytravel.com-Geology of Egypt>). The type section (more than 50 m (164 ft) thickness) is represented in the scarp west of Ain Khoman to the southwest of the Bahariya Oasis (Fig.1 & 4).

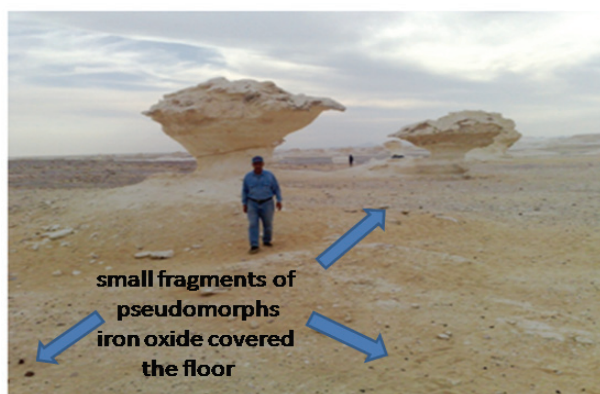


Fig. 4. Photograph for Khoman Chalk in White Desert, west of Ain Khoman, south of the Bahariya Oasis.

Naqb Formation

The Naqb Formation belongs to the Middle Eocene and is made up of a succession of pure limestone beds intercalated at the middle part of the formation with dolostones. The limestones are micritic, sparitic and intraclastic at the base, ferruginous in parts, with chalcedony replacing fossil shells and forming geodes (Hilmy *et al.*, 1984; Said, 1990; and El Bassyouny, 1970, 2004). It differentiates to two members: The lower one is mainly dark gray to pink, non-fossiliferous dolomitic and siliceous limestone. While the upper member is fossiliferous limestone beds with minor clay and conglomeratic intercalations.

Qazzun Formation

The Qazzun Formation covers a large tract of the plateau surface north and northwest of the Bahariya Oasis and conformably overlies the Naqb Fm (Hilmy *et al.*, 1984; and Said, 1990). Lagoonal limestone: white to gray to yellowish, thinly-bedded, partly crystalline, partly chalky, occasionally siliceous and/or dolomitic, common calcite pockets. The limestone includes characteristic melon-shaped concretions of siliceous limestone.

Playa Deposits

Remanent of Holocene pluvial lakes have been known in the south Western Desert of Egypt as playa deposits (Said, 1990). They lie mostly at the footslopes of the limestone plateau. Such deposits represent the lowest points of the various enclosed Bahariya drainages and are restricted to small areas on the road near Ain El Heiz village. They are clay flat surface type deposited under fluvial environment.

Materials and Methods

Samples Collection

Ten rock samples were collected from the Bahariya Oasis particularly from G. El Dist, G. Mandisha, White Desert (west Ain Khoman south the Bahariya Oasis) and surface playa deposits on the road near Ain El Heiz village (Fig. 1). Geologically, such rock samples represent the Bahariya Formation, the Naqb Formation, the Qazzun Formation, the Khoman Chalk and the playa deposits (Fig. 1- 4).

Samples Preparation

The collected samples were crushed, sieved by a 0.8 mesh, homogenized, mixed and weighed. The samples were stored in Marinelli polyethylene containers of 100 cc. Each sample was carefully sealed for 4 weeks to reach secular equilibrium between ^{220}Rn and ^{232}Th and their respective progeny (Mollah *et al.*, 1987).

Samples Measurements

For gamma radiation measurements, a coaxial HPGe detector (ORTEC 572A) of sensitive volume of 76.11 cm^3 is used. The energy resolution of HPGe detector is 1.9 keV at 1332 γ - ^{60}Co transition. The quantitative and qualitative analysis is achieved by using a Maestro H-EG&G card interfaced with an IBM PC-compatible to work as a Multi-Channel Analyzer (MCA). All gamma measurements are taken after energy calibrating the MCA with ^{241}Am , ^{60}Co , ^{133}Ba and ^{226}Ra standard sources.

The absolute efficiency calibration of the used detector is determined using potassium chloride solutions to get efficiency value for the energy values of 1460 keV emitted by ^{40}K . In addition, ^{60}Co , ^{133}Ba and ^{226}Ra standard sources having the same shape and size were used to complete the efficiencies over a wide energy range. The detector is surrounded by a lead cylindrical shield to eliminate the contribution of naturally occurring background radionuclides in the environment. However, the natural background gamma spectrum is obtained and subtracted from each sample spectrum to get more accurate results. All gamma spectra are accumulated for 24 hours and are analyzed after background subtraction.

Results and Discussion

Activity of Natural Radionuclides Estimation

The results of measurements for ten sedimentary rock samples collected at different locations in the Bahariya oasis area are presented in Table 1. The specific activity of the radionuclides ranged between 6.9 to 43.5 Bq kg^{-1} for ^{238}U , between 0.5 and 33.9 Bq kg^{-1} for ^{232}Th , and between 6.5 and 1007.7 Bq kg^{-1} for ^{40}K . These activities of the radionuclides are represented to two discriminated clastic sequence (Samples 1-6 & 10) and carbonate sequence (Samples 7-9).

With respect to the clastics of the Bahariya Formation, generally the activity rises from its base to its top (as shown from samples S1 to S6 in Table 1). Although samples S1 and S2 are white creamy clays, sample S1, which was collected from Gabal Mandisha location (Fig. 1, 2), has activity of 22.3, 28.1 and 365.1 Bq kg⁻¹ for ²³⁸U, ²³²Th and ⁴⁰K, respectively (Table 1). Whereas, sample S2, which was collected from Gabal El Dist location (Fig. 1, 3), has activity of 11.0, 16.4 and 184.9 Bq kg⁻¹ for ²³⁸U, ²³²Th and ⁴⁰K, respectively (Table 1). In other words, the activity of natural radionuclides of S1 is twice that of S2. This is may be due to that the Gabal Mandisha sample was subjected to hydrothermal alteration that was associated with the basaltic sheet intrusion. The latter covers the Bahariya Fm at Gebel Mandisha area as mentioned in the geological setting section (Fig. 1& 2).

Table 1. Specific activities of radionuclides in sedimentary rock samples at different locations in the Bahariya Oasis area.

Sample	Age	Location	Formation name and lithology	Specific activity of radionuclides (Bq/kg)								
				²³⁸ U		²³² Th		⁴⁰ K				
				Mean	Dev.	St. Dev.	Mean	Dev.	St. Dev.	Mean	Dev.	St. Dev.
S10	Holocene	near Ain El Heiz village	Palya deposits	37.9	3.14	33.9	7.52	248.9	28.37			
S9	Middle Eocene	melon-shaped field	Qazzun Fm. (Limestone)	16.6	0.99	0.50	0.12	15.6	1.77			
S8		G. El-Dist	Naqb Fm. (Limestone)	43.5	2.71	3.90	0.34	51.4	5.85			
S7	Maestrichtian	White desert S. Bahariya Oasis	Khoman Chalk	21.3	2.10	0.80	0.86	6.50	0.74			
S6			Galuconite	6.90	1.30	6.70	1.08	1007.7	114.9			
S5			Dark gray mudstone	25.6	2.58	33.9	1.73	241.1	27.49			
S4	Early Cenomanian	G. El-Dist	Bahariya Fm	Deep brown colored ferruginous S.S.	22.2	3.01	21.2	0.49	183.5	20.92		
S3				Light brown colored ferruginous S.S.	9.60	1.67	11.5	0.27	138.6	15.80		
S2				White creamy Clays	11.0	1.94	16.4	1.63	184.9	21.08		
S1				G. Mandisha	White creamy Clays	22.3	1.33	28.1	0.48	365.1	41.62	

On the other hand, the results of the average activity of ^{238}U , ^{232}Th and ^{40}K for the ferruginous sandstone samples displayed interesting results (Table 1; Fig. 5). The activities of the three radionuclides increased from the light brown colored ferruginous sandstone (S3) to the deep brown colored ferruginous sandstone (S4) (Table 1; Fig. 5). Since the Fe-Oxides content increases gradually from (S3) to (S4), the ^{238}U activity increases from 9.6 ± 1.67 to 22.2 ± 3.01 Bq kg $^{-1}$, the ^{232}Th activity increases from 11.5 ± 0.27 to 21.2 ± 1.73 Bq kg $^{-1}$ and the ^{40}K activity increases from 138.6 ± 15.8 to 183.5 ± 20.92 Bq kg $^{-1}$ (Table 1). These results indicate that there is a direct correlation between the increasing of the samples radioactivity and their own iron-oxide content. Consequently, the radioelements and the iron minerals are deposited under the same physicochemical-depositional environmental process. Then, the average activity of the dark gray mudstone (S5) is still in continuous increasing for the three radioelements ^{238}U , ^{232}Th and ^{40}K as 25.6, 33.9 and 241.1 Bq kg $^{-1}$ respectively (Table 1; Fig. 5), supporting the increasing activity line from Bahariya Fm base (S2) to top (S5).

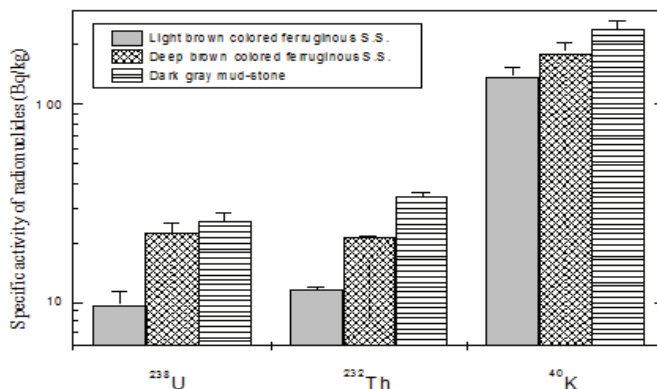


Fig. 5. Ranges of ^{238}U , ^{232}Th and ^{40}K specific activity concentration for ferruginous sandstones (light and deep brown color) and dark gray mud-stone samples.

The highest K-activity, over the whole area, was recorded for the upper most layer of the Bahariya Formation (S6); it was 1007.7 Bq kg $^{-1}$ (Table 1). Such layer is composed of glauconitic siltstone and mudstone. It is worth to mention, that glauconite, which has chemical formula $(\text{K},\text{Na}) (\text{Fe}^{3+},\text{Al},\text{Mg})_2(\text{Si},\text{Al})_4\text{O}_{10}(\text{OH})_2$, is considered a diagnostic mineral that develops as a consequence of diagenetic alteration of sedimentary deposits. The upnormality of potassium content could be

attributed to the presence of potassium and iron constituents and also to diagenetic alterations.

The activity among the carbonate rocks, which are represented by three formations; Khoman Chalk (S7), Naqb Fm (S8) and Qazzun Fm (S9), Naqb Fm (S8) recorded the highest one (Fig. 6). As shown from (Table 1; Fig. 6) Naqb Fm sample (S8) exhibited an activity of 43.5, 3.9 and 51.4 Bq kg⁻¹ for ²³⁸U, ²³²Th and ⁴⁰K, respectively. At the same time, Qazzun Fm sample (S9) exhibited an activity of 16.6, 0.5, and 15.6 Bq kg⁻¹ for ²³⁸U, ²³²Th and ⁴⁰K, respectively. Although both Naqb Fm and Qazzun Fm samples are carbonate rocks and both belong to the same age (middle Eocene), the radioactivity levels of the Naqb Fm are higher about three times than the Qazzun Fm samples for ²³⁸U and ⁴⁰K and about eight times for ²³²Th (Table 1& Fig. 6). These differences can be attributed to their content of accessory mineral constitutes. As regards to the mineralogical content, limestone of the Naqb Fm sample (S8) is partly siliceous and occasionally dolomitic. It is partly marly with 3.5 m clay bed near the base of the section. In addition, it acquires its pink color due to the including ferruginous material with disseminated glauconitic grains. Whereas, the Qazzun Fm sample (S9) is composed of nummulitic limestone beds, that are occasionally siliceous and/or dolomitic particularly south of the Bahariya Oasis, but loss of Fe-oxide minerals as in the case of the Naqb Fm.

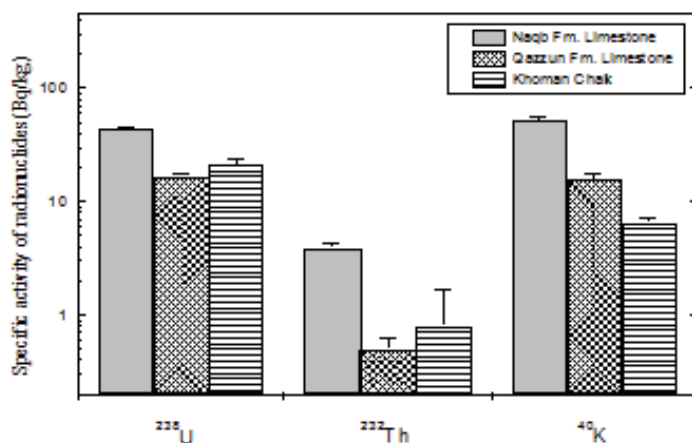


Fig. 6. Ranges of ²³⁸U, ²³²Th and ⁴⁰K specific activity concentration for carbonate rocks; Khoman Chalk, Naqb Fm, and Qazzun Fm.

On the other hand, Khoman Chalk sample (S7) displayed average activities of ^{238}U , ^{232}Th and ^{40}K as 21.3, 0.8 and 6.5 Bq kg⁻¹, respectively. These activity values are lower than those of both the Naqb Fm (S8) and the Qazzun Fm (S9) particularly the thorium and potassium contents (Table 1; Fig. 6). Although the three formations; Naqb, Qazzun and Khoman Chalks belong to carbonate rocks, these lower activity values can be attributed to its deep marine facies nature since it has Maestrichtian age. In addition, although the uranium activity of the Khoman Chalk (S7) is still lower than the Naqb Fm (S8), it is higher than the Qazzun Fm (S9) (Table 1; Fig. 6). The increasing of the uranium content in the Khoman Chalk, relative to the Qazzun Fm, may be due to the same physicochemical-depositional process associated with the included small iron-rich pipes, pyrite concretions and the small fragments of pseudomorphs of iron oxides.

Finally, the fluvial environmental mudstone playa deposits sample (S10) revealed considerable high activities of the three radioelements ^{238}U , ^{232}Th and ^{40}K ; 37.9, 33.9 and 248.9 Bq kg⁻¹ in sequence. These higher activities could be owing to its depositional nature which yields radioelements leaching in the lowest points of the various enclosed Bahariya drainages at the footslopes of the limestone plateau neighboring rocks.

Gamma Dose rate (D_r) and Annual Effective Dose (E_{eff}) Estimation

Approximately 87% of the radiation dose received by mankind is due to natural radiation sources and the rest is due to anthropogenic radioactivity (Kannan *et al.*, 2002). Natural radioactivity is associated mainly to primordial radionuclides, including the elements belonging to the ^{238}U , ^{232}Th and ^{40}K series. People of the Bahariya Oasis are frequently using natural stones in a wide range for construction purposes without knowing how much level of radioactivity is present in these stones. Therefore, the estimation of the risks to the human health caused by the radioactivity of Bahariya Oasis rocks should be carried out. The values of the external gamma absorbed dose rate in the air and annual effective dose rate can be calculated in terms of the activities of ^{238}U ,

^{232}Th and ^{40}K in collected samples. The calculations were performed according to the following equation UNSCEAR (2000):

$$D_r = 0.4299 A_U + 0.666 A_{Th} + 0.042 A_K \dots\dots\dots(1)$$

Where, D_r is the dose rate in nGy h^{-1} and A_U , A_{Th} & A_K are the specific activities (Bq kg^{-1}) of ^{238}U , ^{232}Th and ^{40}K , respectively.

The calculated D_r can be used to assess the annual effective dose rate equivalent (E_{eff}) by the following relation:

$$E_{eff} = D_r (\text{nGy h}^{-1}) \times Q \times T \times \frac{10^3 (\text{mSv})}{10^9 (\text{nGy})} \dots\dots\dots(2)$$

where D_r is the absorbed dose rate (in nGy/h), Q is 0.7 ($\text{Sv Gy}^{-1} \text{y}^{-1}$) for environmental exposure to gamma rays of moderate energy and T is the time in hours in one year, *i.e.*, 8760 h. To deduce the indoor occupancy (*i.e.* human effective dose) and outdoor exposure to gamma rays, the E_{eff} results should be multiplied by factors of 0.8 and 0.2 adopted by the UNSCEAR (2000).

It is noticeable from Table (2) that the highest absorbed dose values are recorded for the collected samples of glauconite (S6), playa (S10), dark gray mud-stone (S5) and white creamy clays of G. Mandisha (S1). The lowest values are recorded for the carbonate rocks; Qazzun Fm (S9) and Khoman Chalk (S7), as well as the light brown colored ferruginous sandstone (S3). In addition, Table (2) shows that, the absorbed dose rate values of the Bahariya area vary from 8.1 to 49.8 nGy h^{-1} , with a mean value of 30.04 nGy h^{-1} . These mean values are lower than the estimate of average global terrestrial radiation of 55 nGy h^{-1} (UNSCEAR, 1988) and are lower than the mean values of sedimentary rocks from Upper Egypt (Abbadly, 2004). The calculated annual effective dose with average value (0.184 mSv y^{-1}) is lower than the worldwide recommended value to members of the public (1.0 mSv y^{-1} is the maximum permissible annual effective dose limit according to ICRP, 1990).

Table 2. The absorbed dose rate, annual effective dose, indoor and outdoor absorbed dose.

Sample	Absorbed Dose in Air (D_r) (nGy/h)		Annual Effective Dose Rate (E_{eff}) (mSv/y)		Absorbed dose indoor (mSv/y)		Absorbed dose outdoor (mSv/y)	
	mean	st. dev.	mean	st. dev.	mean	st. dev.	mean	st. dev.
S10	49.3	115.2	0.30	0.71	0.242	0.565	0.060	0.141
S9	8.1	2.0	0.05	0.01	0.040	0.010	0.010	0.002
S8	23.5	6.5	0.14	0.04	0.115	0.032	0.029	0.008
S7	10.0	2.4	0.06	0.01	0.049	0.012	0.012	0.003
S6	49.8	114.9	0.31	0.70	0.244	0.564	0.061	0.141
S5	43.7	27.7	0.27	0.17	0.214	0.136	0.054	0.034
S4	31.4	21.1	0.19	0.13	0.154	0.104	0.038	0.026
S3	17.6	15.9	0.11	0.10	0.086	0.078	0.022	0.019
S2	23.4	21.2	0.14	0.13	0.115	0.104	0.029	0.026
S1	43.6	41.6	0.27	0.26	0.214	0.204	0.054	0.051
Average	30.04		0.184					

Conclusion

Gamma-ray spectrometry has been utilized to determine the ^{238}U , ^{232}Th and ^{40}K specific activity concentration in ten samples of different types of sedimentary rocks collected from Bahariya Oasis. The specific activity of the radionuclides ranged between 6.9 to 43.5 Bq kg⁻¹ for ^{238}U , between 0.5 and 33.9 Bq kg⁻¹ for ^{232}Th and between 6.5 and 1007.7 Bq kg⁻¹ for ^{40}K . It is noteworthy that, the activity increases with the occurrence of Fe-oxide minerals as in the ferruginous sandstones, glauconite in siltstone and mudstone layers in the Bahariya Formation and the pinky limestone Naqb Formation. Estimation of the external gamma absorbed dose rate in the air at 1 meter above ground level and annual effective dose rate are carried out. The average value of gamma dose rate (30.04 nGy/h) and the average annual effective dose (0.184 mSv y⁻¹) obtained in this study are lower than the worldwide dose rate

average value (55 nGy/h) and the maximum annual dose to members of the public (1.0 mSv y^{-1}), respectively.

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قياس النشاط الإشعاعي الطبيعي لعينات من الصخور الرسوبية تم جمعها من الواحات البحرية، الصحراء الغربية، مصر

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^١قسم الجيوفيزياء، ^٢قسم الفيزياء، كلية العلوم، جامعة عين شمس، القاهرة
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المستخلص. تناول البحث تقييم أنشطة محددة من العناصر المشعة الطبيعية في عشر عينات من الصخور الرسوبية التي تم جمعها من منطقة الواحات البحرية بالصحراء الغربية، مصر. تم الحصول على النتائج التجريبية باستخدام مجس الجرمانيوم عالي النقاء (HPGe) ونظام التحليل الطيفي لأشعة جاما. فقد وجد أن النشاط المحدد يتراوح ما بين ٦,٩ إلى ٤٣,٥ بيكريل كجم-١ لليورانيوم^{٢٣٨}، وما بين ٠,٥ إلى ٣٣,٩ بيكريل كجم-١ للثوريوم^{٢٣٢}، وبين ٦,٥ إلى ١٠٠٧,٧ بيكريل كجم-١ للبتواسيوم^{٤٠}. ويرتفع نشاط متكون البحرية من القاعدة إلى القمة ويصل إلى ١٠٠٧,٧ بيكريل كجم-١ للبتواسيوم^{٤٠} والتي تسجل أعلى المعدلات في المنطقة كلها على الإطلاق في طبقة الغرين والطين العليا الغنية بالجلوكونيت. بالإضافة إلى ذلك، فقد وجد أن النشاط يزداد بوجود معادن أكاسيد الحديد كما في الحجر الرملي الحديدي، وطبقات الغرين والطين الغنية بالجلوكونيت في مكونات البحرية والنقب. ويبين نشاط صخور الكربونات أن أعلى المعدلات متمثلة في تكوين النقب (الحجر الجيري ذو اللون الوردى)، وأقل في تكوين الكازون، وأدناها في تكوين الخومن الطباشيري، ولا سيما نشاط الثوريوم والبتواسيوم. ووجد أن متوسط قيمة معدل جرعة جاما (30.04 nGy/h) ومتوسط الجرعة الفعالة السنوية

(0.184 mSv y⁻¹) اللتين تم الحصول عليهما في هذه الدراسة هي أقل من قيمة الجرعة المتوسطة للمعدل العالمي (55 nGy/h) والجرعة السنوية القصوى لأفراد الجمهور (1.0 mSv y⁻¹) على التوالي.