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Radiological and environmental studies on the metamorphosed sandstones at Wadi Sikait area, southeastern desert, Egypt

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Abstract. Wadi Sikait area lies in the southern part of the Eastern Desert of Egypt along the upper stream of Wadi.Sikait. The exposed rock units in Wadi Sikait area are ophiolitic mélangé rocks, metamorphosed sandstones, and porphyritic granites, lamprophyre dykes, in addition to fluorite and quartz veins. The activity concentrations of various radionuclides are measured by NaI-Tl activated detector and the radioactive mineralization were determined by Alpha track technique using CR-39 films. Radiometric investigations indicate that ^{238}U activity concentrations range between 37.2 and 520.4 with average 148.8 BqKg^{-1} , ^{232}Th activity concentrations vary between 8.08 and 366.99 BqKg^{-1} with 76.75 BqKg^{-1} as an average. The studied metamorphosed sandstones have higher values relative the world concentration levels. High uranium content of the metamorphosed sandstone is attributed to presence radioactive minerals like uranophane and autonite, in addition to accessory minerals like allanite, zircon and monazite. Other U-bearing minerals are also recorded as biotite, muscovite, iron oxides and clays. Absorbed Dose Rate (D), annual effective dose equivalent (AEDE), radium equivalent activity (Raeq), external (H_{ex}) and internal (H_{in}) hazard index, in addition to activity gamma index (I_{γ}) caused by gamma emitting natural radionuclide are determined from the obtained values of ^{226}Ra , ^{232}Th and ^{40}K . Fairly, many of the studied metamorphosed sandstone do not satisfy the universal standards.

Keywords. Radiological, Environment, Uranium, Thorium, Egypt

1. Introduction

The natural environmental radiation depends mainly on geological and geographical conditions (Florou and Kritidis, 1992). Higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks. There are exceptions, however, as some shales and phosphate rocks have relatively high content of radionuclides (UNSCEAR, 1993). The study area lies in the southern part of the Eastern Desert of Egypt along the upper stream of Wadi.Sikait area is covered by moderate to high mountains with rugged topography. The high peak is Gabal Sikait (769 m a.s.l), it is formed of ultramafic rocks, which are often associated with beryl mineralization. The Sikait area was geologically investigated by Hassan and Hashad (1990); Hashed and El Redy (1979); Hegazy (1984); Mohamed and Hassanen (1997); Saleh (1998); Ibrahim et al., (1999) and Omar (1999) among others. Assaf et al. (2000) studied the polyphase folding in Nugrus-Sikait area and reported that the lithological constitution of the area comprise a sequence of dismembered ophiolites, ophiolitic mélangé association and arc assemblage. The older rocks are intensively deformed and intruded by intracratonic association, which, within the mapped area, is ophiolitic mélangé, metamorphosed sandstones with porphyritic granites. Detailed spectrometric and radiometric studies (Ibrahim et al. 2007) for metamorphosed sandstones site at Wadi.Sikait indicate that eU range from 15 to 100 ppm, but chemically range from (60 – 480 ppm); whereas eTh up to 85 ppm. Also they reported that the emplacement of both lamprophyre dykes and porphyritic granites may



play an important role as a heat source, which lead to U- mobilization from hot granitic magma, transported (along deep fault and banding) and redeposit in metamorphosed sandstones under suitable conditions.

The metamorphosed sandstones occur in two locations in Wadi. Sikait. The first location (Sikait-1) lies at the upper stream of Wadi. Sikait and the second location (Sikait-2) occur west the bend of Wadi. Sikait figure 1 , figure 2a and 2b. The metamorphosed sandstone rocks are fine- to medium- grained, white color, highly sheared, sometimes bedded, cross- cut by lamprophyre dykes (NW-SE and NNE-SSW), quartz veins and left strike slip faults (NW-SE , NNE- SSW& N-S) figure 2 a. Sikait-1 is the largest outcrop of metamorphosed sandstones at Wadi. Sikait with low to moderate peaks, elongated in NW-SE direction (1.4 km in length and range in width from 120 m to 300 m). These rocks are varying in color from pale white to milky white figure and show relics of primary bedding; banding and obvious foliations in NW-SE with angle of dip 35° /SW. Metamorphosed sandstones in this location are dissected by three types of left strike slip faults, N-S and NNE-SSW and NW-SE figure, so they are highly tectonized. The NW-SE is the largest and oldest one. These faults especially NNE- SSW characterized by mylonitization and many types of alterations as silicification and Fe-Mn oxy-hydroxides figure. Mineralization occurs along zones of these faults visible by naked eyes.

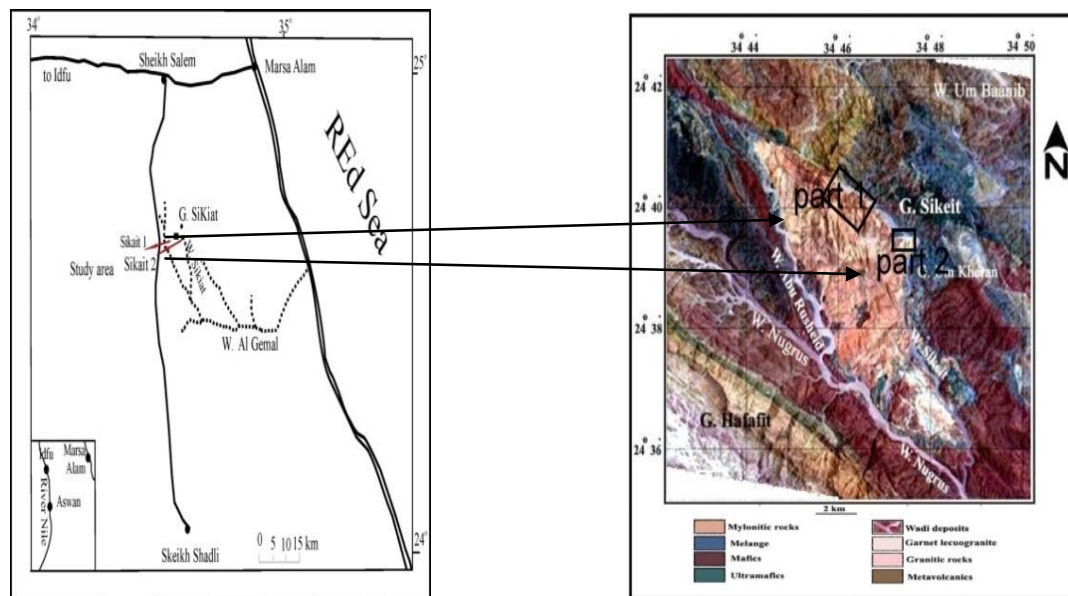
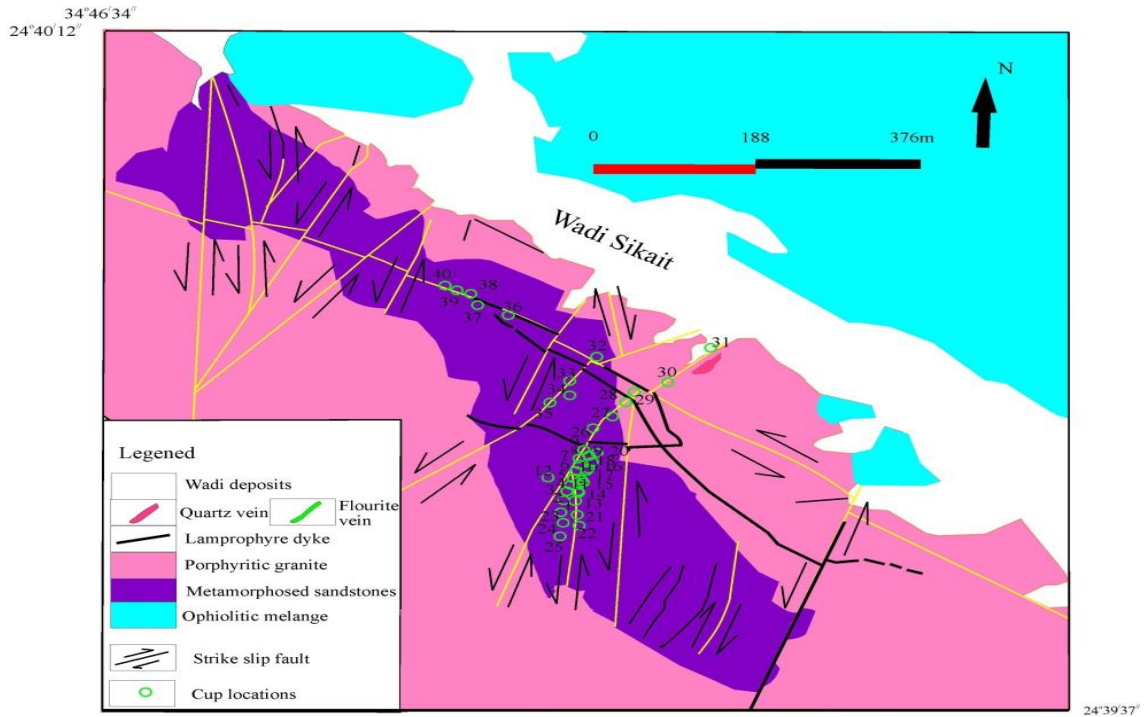


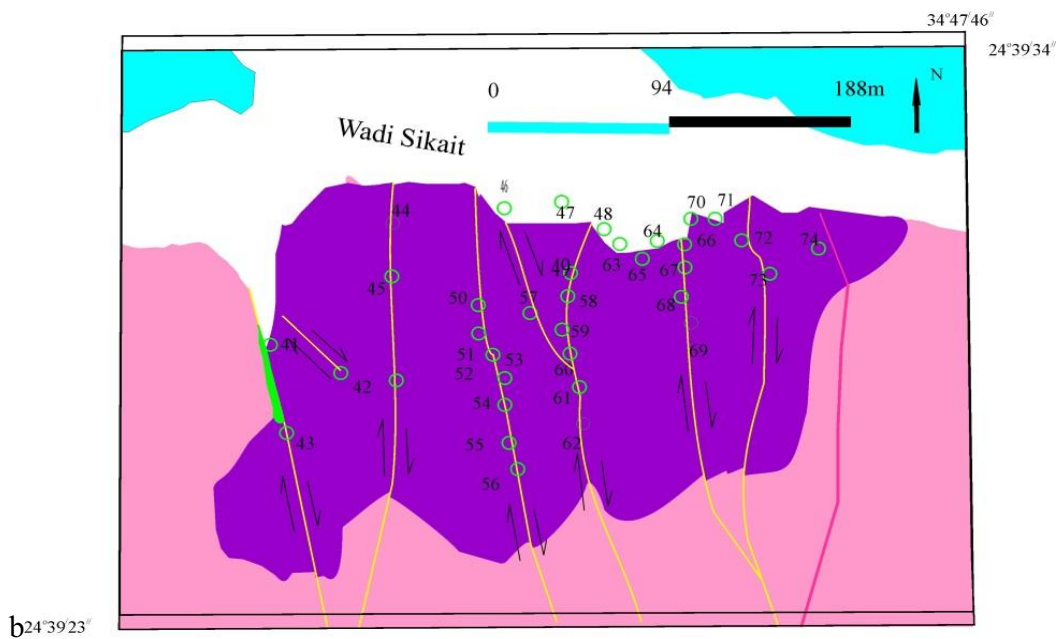
Figure 1. Location map of the study area and false color composite map TM-image that showing the location of metamorphosed sandstone.

Sikait-2 covers a small area where its length is about 450 m and maximum width about 230 m, forming low terrain, highly sheared, dissected by branches of strike slip fault running in N-S with left movements and frequently curved to N direction. Branch zones of the strike slip fault characterized silicification alteration. Many mineralizations are associated with quartz found along zones of fault branches as fluorite at west this location and wolframite and cassiterite which visible by naked eyes. Metamorphosed sandstones are generally uniform in texture and composed from fused quartz grains. Semi-angular and elongated rock fragments of older rocks are enclosed in the metamorphosed sandstones. The present work aims to study the radioactivity and environmental impacts of Wadi. Sikait metamorphosed sandstone that used as building material or as ornamental stones.



a

Figure 2 a. Detailed geologic map of Sikait-1 modified after Ibrahim et al. (2010).



b

Figure 2 b. Detailed geologic map of Sikait-2 modified after after Ibrahim et al. (2010).

2. Sampling and analytical techniques

Representative grab samples were collected from the two-mica granite (seven samples) and from the metamorphosed sandstone to represent the highest values of anomalous field radioactivity. These samples were prepared for gamma-ray spectrometric analysis in order to determine their uranium, thorium, radium and potassium contents by using a multichannel analyzer equipped with a γ -ray detector (Gamma-Spectrometer technique). The instrument used in determination of the four radioactive elements consists of a bicron scintillation detector NaI (Tl) 76×76 mm, hermetically sealed with the photomultiplier tube in aluminum housing. The tube is protected by a copper cylinder measured at 0.6 cm in thickness against induced X-ray and a chamber of lead bricks against environmental radiation. Uranium, thorium, radium and potassium are measured by using four energy regions representing Th-234, Pb-212, Pb-214 and K-40 at 93, 239, 352 and 1460 keV for uranium, thorium, radium and potassium, respectively. The measurements were carried out in plastic sample containers, cylindrical in shape, 212.6 cm³ in volume, 9.5 cm in average diameter and 3 cm in height. The rock sample was crushed as fine as about 1 mm in grain size, and then the container was filled with about 300–400 gm of the crushed sample, sealed well and left for at least 21 days to accumulate free radon to attain radioactive equilibrium. The relation between the percentage of Rn-222 accumulation and time increased till reaching the steady state in about 38 days (Matolin, 1991).

3. Results and discussion

Firstly, the metamorphosed sandstone were investigated by spectrometric method and then were confirmed by α -track as a suitable technique for tracing radioactive and radioelement-bearing minerals.

3.1 Particle track analyses

Some samples from the different mineralized parts of metamorphosed sedimentary rocks were submitted to α -track analyses as a mean of micromapping the radioelement distribution. It is denoted that mineralization is mainly located along grain boundaries, in addition to being contained in crystal lattice of allanite, zircon and monazite figure 3a and 3b. It is clear that uranium mineralization increase in the highly fractured portions of the rock with remarkable epidotization and sericitization of plagioclase. The biotite and muscovite grains are enriched in uranium mineralization along their outer boundaries and their cleavage planes figure 3a, 3b, 3c and 3d). It appears that biotite acts as reductant precipitating the uranium. Chlorites, sericitized plagioclase and highly fractured quartz grains also contain uranium mineralization figure 3e and 3f. Most of the rock forming minerals is stained with iron oxides which occasionally adsorb uranium causing intense uranium mineralization figure 3g and 3h.

The highly uranium content of the metamorphosed sandstone is attributed to the presence of radioactive minerals like uranophane and autonite figure 3a and 3b) In addition accessory minerals like allanite, zircon and monazite figure 3c, 3d, 3e and 3f. Other U-bearing minerals are also recorded as biotite, muscovite, iron oxides and clays figure 3i and 3j. In the figure 3a and 3b, radioactive- mineralization along grain boundaries and in crystal lattice of allanite, zircon and monazite and its corresponding dense alpha track image showing radioactive minerals. Figure 3c, 3d, 3e and 3f) are biotite and muscovite grains which enriched in uranium mineralization along their outer boundaries and their cleavage planes and its corresponding dense alpha track image showing radioactive minerals. Figure 3g and 3h are Chlorites, sericitized plagioclase and highly fractured quartz grains also contain uranium mineralization. Figure 3i and 3j) are Iron oxides stained with uranium causing intense uranium mineralization and its corresponding dense alpha track image showing dense and disseminated radioactive minerals.

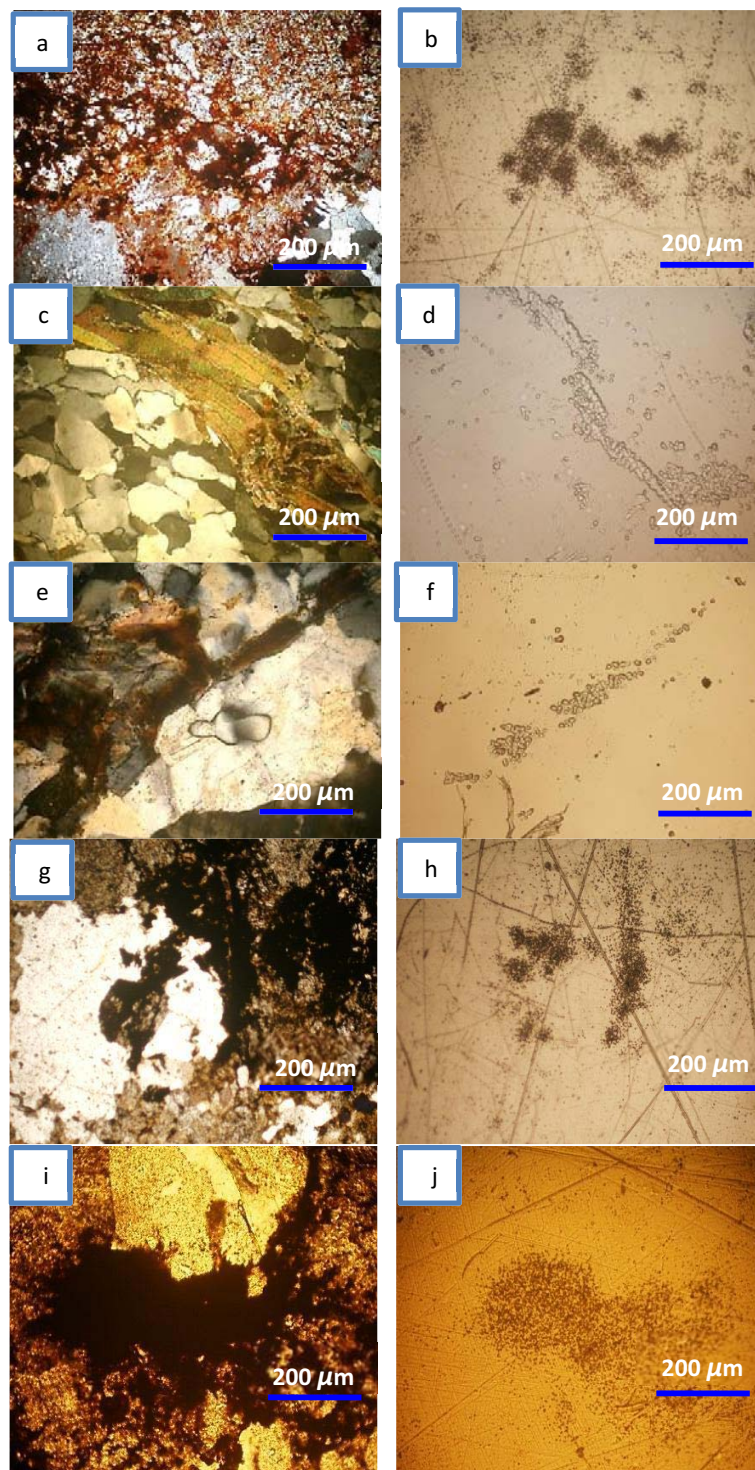


Figure 3. Radioactive minerals and their mirror image by alpha tracks.

3.2. Activity concentrations the metamorphosed sandstone

Activity concentrations of ^{238}U , ^{232}Th , ^{226}Ra , ^{40}K were measured in the metamorphosed sandstone samples as in table (). The ^{238}U activity concentrations range between 37.2 and 520.4 with average 148.8 BqKg^{-1} , ^{232}Th activity concentrations vary between 8.08 and 366.99 BqKg^{-1} with 76.75 BqKg^{-1} as an average. ^{226}Ra ranges between 11.1 and 624.04 BqKg^{-1} with average 118.05 BqKg^{-1} . ^{40}K ranges between 11.1 and 1774.71 BqKg^{-1} with average 995.42 BqKg^{-1} . The world concentration limits of ^{238}U , ^{232}Th and ^{40}K are equal to 35, 30 and 400 BqKg^{-1} , respectively ⁹⁴. The studied metamorphosed sandstones have higher values relative that recorded in the UNSCEAR (2000).

3.3. Assessment of Hazard Indices

3.3.1 Absorbed Dose Rate in Air (D)

The absorbed gamma dose rates in air at 1m above the ground surface for the uniform distribution of radionuclides (^{226}Ra , ^{232}Th and ^{40}K) were calculated by using equation 1 on the basis of guide lines provided by UNSCEAR (2000) and Örgün et al. (2007).

$$D (\text{nGy h}^{-1}) = 0.462A_{\text{Ra}} + 0.604A_{\text{Th}} + 0.0417A_{\text{K}} \quad (1)$$

Where: A_{Ra} , A_{Th} , and A_{K} are the average specific activities of ^{226}Ra , ^{232}Th and ^{40}K in Bq/kg , respectively. The estimated indoor gamma dose rate values for metamorphosed sandstones samples are shown in table 1. The DR values for metamorphosed sandstones samples range from 22.07 to 463.98 nGy h^{-1} with a mean of 156.60 nGy h^{-1} . The mean DR values for all metamorphosed sandstone samples exceed the worldwide average value of soils (55 nGy h^{-1}), (Yang et al. 2005).

3.3.2 Annual Effective Dose Equivalent (AEDE)

The annual effective dose equivalent (AEDE) was calculated from the absorbed dose by applying the dose conversion factor of 0.7 Sv/Gy and the outdoor occupancy factor of 0.2, (UNSCEAR, 2000; Örgün et al. 2007).

Furthermore, the average values of annual effective dose for all metamorphosed sandstone samples were also listed. The values obtained varied between 0.03 and 0.57 mSvy^{-1} . The mean value 0.19 found to be less than 0.48 mSvy^{-1} recommended by UNSCEAR (2000) as the worldwide average of the annual effective dose.

3.3.3 Radium Equivalent Activity (Ra_{eq}).

The radium equivalent activity for the samples was calculated. The exposure to radiation (Tufail et al. 1992) can be defined in terms of the radium equivalent activity (Ra_{eq}), which can be expressed by the following equation:

$$Ra_{\text{eq}} = A_{\text{Ra}} + 10/7A_{\text{Th}} + 10/130A_{\text{K}} \quad (2)$$

Where: A_{Ra} , A_{Th} and A_{K} are the specific activities of Ra, Th and K, respectively, in Bq/kg . The maximum value of Ra_{eq} in metamorphosed sandstone samples must be less than 370 Bqkg^{-1} for safe use, i.e., to keep the external dose below 1.5 mSv y^{-1} (UNSCEAR, 1993; UNSCEAR, 2000). The range and the mean value of Ra_{eq} of metamorphosed sandstone samples are presented in table 1. The mean Ra_{eq} values for metamorphosed sandstone samples (304.26 Bqkg^{-1}) are below the criterion limit of 370 Bqkg^{-1} except for some samples in the N-S shear zone figure 3. From the results it is evident that there are considerable variations in the Ra_{eq} of the same rock type originating from different shear zones. This fact is important from the point of view of selecting suitable materials for use in building and construction especially concerning those which have large variations in their activities. Large variation in radium equivalent activities may suggest that it is advisable to monitor the radioactivity levels of materials from a

new source before adopting it for use as a building material (Kumar et al. 1999; Ngachin et al. 2007). The recommended maximum level of Radium equivalents for building materials used for homes is 370 Bqkg^{-1} and for industries is $370\text{-}740 \text{ Bqkg}^{-1}$, (Oresegun and Babalola, 1998).

3.3.4. External and Internal Hazard Index (Hex and Hin).

To limit the annual external gamma-ray dose (Saito, and Jacob, 1995; Saito et al. 1998; UNSCEAR, 2000) to 1.5 Gy for the samples under investigation, the external hazard index (Hex) is given by the following equation:

$$H_{\text{ex}} = A_{\text{Ra}}/370 + A_{\text{Th}}/259 + A_{\text{K}}/4810 \quad (3)$$

The internal exposure to ^{222}Rn and its radioactive progeny is controlled by the internal hazard index (H_{in}), which is given by

$$H_{\text{in}} = A_{\text{Ra}}/185 + A_{\text{Th}}/259 + A_{\text{K}}/4810 \quad (4)$$

These indices must be less than unity in order to keep the radiation hazard insignificant. Values of eU, eTh and Ra(eU) in ppm as well as K in %, were converted to activity concentration, Bq kg^{-1} , using the conversion factors given by Polish Central Laboratory for Radiological Protection (Malczewski et al. 2004).

The value of this index must be less than unity for the radiation hazard to be negligible, i.e. the radiation exposure due to radioactivity in construction materials must be limited to 1.5 mSv y^{-1} . The values of external and internal hazard indices H_{ex} and H_{in} for the studied metamorphosed sandstone samples range between 0.13 and 2.77 and 0.23 to 3.76 respectively. External and internal hazard indices are higher than unity for some of the studied samples indicating that many of these samples cannot be used as building or decorative material of dwelling.

3.3.5. Activity Concentration Index (I_γ).

Another radiation hazard index called the representative level index I_{γ} is defined as follows (NEA-OECD and Nuclear Energy Agency, 1979).

$$I_{\gamma} = A_{\text{Ra}}/150 + A_{\text{Th}}/100 + A_{\text{K}}/1500 \quad (5)$$

Where: A_{Ra} , A_{Th} and A_{K} are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K , respectively in Bq/kg (Abbady et al. 2005). The safety value for this index is ≤ 1 (El-Galy et al. 2008; El-Aassy et al. 2012).

The values of the range and mean gamma-index (I_{γ}) are presented in table 1, for raw materials. It is observed that the mean I_{γ} values (2.42) of the metamorphosed sandstone samples which are higher dose criterion (0.3 mSv/y) and correspond to an activity concentration index of $2 \leq I_{\gamma} \leq 6$ proposed by EC (1999), for materials used in bulk construction.

Contribution of ^{238}U , ^{232}Th and ^{40}K radionuclides for the absorbed dose rate within the studied rocks are plotted in figure 4. It is clear that ^{40}K plays the main and most important role in dose rate contribution.

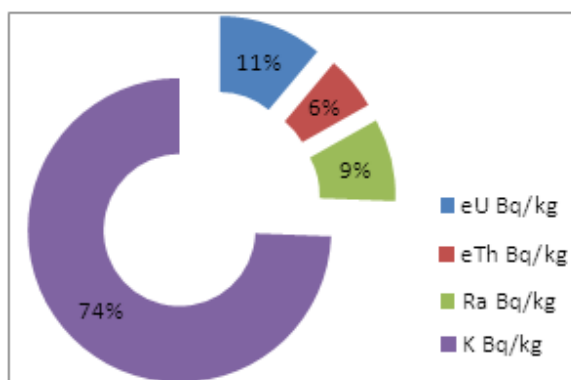


Figure 4. Contribution of ^{238}U , ^{232}Th and ^{40}K radionuclides for the absorbed dose rate within the studied rock samples.

Table 1. Results of radionuclide concentrations, the dose rate (D), the annual effective dose equivalent (AEDE), radium equivalent activity (Ra_{eq}), external (H_{ex}), internal (H_{in}) hazard indices and gamma index (I_γ) for metamorphosed sandstone samples.

S. No.	eU Bq/kg	eTh Bq/kg	Ra Bq/kg	K Bq/kg	Abs. Dose nGy/h	Eff. Dose mSv	Ra_{eq}	H_{ex}	H_{in}	I_γ
1	86.80	44.44	44.40	1208.18	117.32	0.14	200.82	0.66	0.89	1.83
2	49.60	36.36	55.50	1048.55	88.60	0.11	188.10	0.49	0.63	1.39
3	62.00	28.28	44.40	873.27	82.14	0.10	151.97	0.46	0.63	1.28
4	106.39	67.43	88.91	845.10	125.12	0.15	250.24	0.72	1.01	1.95
5	62.00	8.08	55.50	478.89	53.49	0.07	103.88	0.30	0.47	0.81
6	99.20	40.40	44.40	1079.85	115.26	0.14	185.18	0.65	0.92	1.79
7	37.20	52.52	55.50	1452.32	109.47	0.13	242.25	0.61	0.71	1.74
8	364.31	366.99	624.04	1774.71	463.98	0.57	1284.83	2.77	3.76	7.28
9	86.80	20.20	44.40	1026.64	95.11	0.12	152.23	0.53	0.76	1.47
10	134.54	80.96	94.13	1136.19	158.44	0.19	297.19	0.91	1.28	2.46
11	103.17	40.32	108.56	738.68	102.82	0.13	222.98	0.59	0.87	1.58
12	130.32	81.33	110.67	1161.23	157.75	0.19	316.17	0.91	1.26	2.46
13	243.41	157.12	191.70	1264.52	260.08	0.32	513.42	1.53	2.19	4.04
14	37.20	8.08	11.1	11.1	22.07	0.03	11.54	0.13	0.23	0.33
15	89.40	68.11	79.48	920.22	120.82	0.15	247.57	0.70	0.94	1.89
16	248.00	181.80	133.20	112.68	229.08	0.28	401.58	1.40	2.07	3.55
17	114.08	87.26	97.13	754.33	136.87	0.17	279.81	0.80	1.11	2.14
18	234.36	208.22	130.43	237.88	243.96	0.30	446.18	1.49	2.12	3.80
19	248.00	163.82	158.18	410.03	230.62	0.28	423.75	1.39	2.06	3.56
20	74.40	60.60	77.70	870.14	107.26	0.13	231.21	0.62	0.82	1.68
21	520.43	21.98	536.13	694.86	282.69	0.35	620.98	1.64	3.04	4.15
22	198.40	68.68	99.90	973.43	173.74	0.21	272.89	1.00	1.54	2.66
23	124.00	60.60	111.00	867.01	130.04	0.16	264.26	0.75	1.08	2.01
24	212.54	120.84	169.05	1690.20	241.66	0.30	471.69	1.39	1.97	3.75
25	308.14	11.47	179.93	729.29	179.70	0.22	252.42	1.03	1.86	2.66
26	62.00	84.84	88.80	1302.08	134.18	0.16	310.16	0.77	0.93	2.13
27	86.80	8.08	55.50	1599.43	111.68	0.14	190.08	0.60	0.83	1.73
28	86.80	8.08	55.50	1414.76	103.98	0.13	175.87	0.56	0.79	1.60
29	210.80	64.64	177.60	109.55	141.00	0.17	278.37	0.84	1.41	2.12
30	104.28	50.90	100.12	970.30	119.39	0.15	247.48	0.68	0.96	1.85
31	186.00	88.88	77.70	635.39	166.11	0.20	253.55	0.98	1.48	2.55
32	86.80	52.52	66.60	1230.09	123.12	0.15	236.25	0.69	0.93	1.92
33	186.00	76.76	88.80	1333.38	187.90	0.23	301.02	1.08	1.58	2.90
34	111.60	76.76	99.90	1395.98	156.13	0.19	316.94	0.89	1.19	2.44

Table 1. Continuous.

S. No.	eU Bq/kg	eTh Bq/kg	Ra Bq/kg	K Bq/kg	Abs. Dose nGy/h	Eff. Dose mSv	Ra _{eq}	H _{ex}	H _{in}	I _γ
35	148.80	84.84	88.80	1295.82	174.02	0.21	309.68	1.00	1.40	2.70
36	124.00	72.72	99.90	1295.82	155.25	0.19	303.46	0.89	1.22	2.42
37	124.00	68.68	77.70	1280.17	152.15	0.19	274.29	0.87	1.20	2.37
38	148.80	64.64	77.70	1543.09	172.14	0.21	288.74	0.97	1.37	2.67
39	111.60	88.88	122.10	1001.60	147.01	0.18	326.12	0.85	1.15	2.30
40	198.40	92.92	111.00	1061.07	192.03	0.24	325.36	1.12	1.65	2.96
av	148.8	76.75	118.05	995.42	156.60	0.19	304.26	0.91	1.31	2.42
Min	37.2	8.08	0	0	22.067	0.03	11.543	0.13	0.23	0.33
Max	520.4	366.99	624.04	1774.71	463.98	0.57	1284.83	2.77	3.76	7.28

4. Conclusion

The field mapping indicates that metamorphosed sandstones occur in two sites at Wadi. Sikait (Sikait-1 and Sikait-2) and the exposed rocks at this area can be arranged based on field observations and structural relations from older to younger as follows:

1. Veins (fluorite and quartz) youngest
2. Lamprophyre dykes
3. Porphyritic granites
4. Metamorphosed sandstones
5. Ophiolitic mélangé oldest

High uranium content of the metamorphosed sandstone is attributed to presence radioactive minerals like uranophane and autonite, in addition to accessory minerals like allanite, zircon and monazite. Other U-bearing minerals are also recorded as biotite, muscovite, iron oxides and clays. Absorbed Dose Rate (D), annual effective dose equivalent (AEDE), radium equivalent activity (Ra_{eq}), external (H_{ex}) and internal (H_{in}) hazard index, in addition to activity gamma index (I_γ) caused by gamma emitting natural radionuclide are determined from the obtained values of ²²⁶Ra, ²³²Th and ⁴⁰K. Fairly, many of the studied metamorphosed sandstone samples are higher than the international limits.

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