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A survey of natural radioactivity level in selected rock samples from Bukit bunuh, lenggong, Malaysia

Zulhilmi, W.M[1], Iskandar, S.M[1], Azhar, A.R[1], Saidin Mokhtar[2] and Naimah, N.Y[1]

[1]School of Physics, USM, Penang, Malaysia.

[2]Centre for Global Archaeological Research, USM, Penang, Malaysia.

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ABSTRACT

Radiation that coming from the natural radionuclides such as ^{226}Ra (^{238}U), ^{228}Ra (^{232}Th) and ^{40}K can cause health risks if exposed for longer terms. The activity and radiological effect parameters of these natural radionuclides were determined in rock samples from Bukit Bunuh. The results of measurements showed that the average activity concentration of ^{226}Ra varied from 22.5 to 42.3 Bq kg^{-1}, for ^{228}Ra, it varied from 1.2 to 3.5 Bq kg^{-1} and for ^{40}K, it varied from 274.3 to 438.2 Bq kg^{-1}. Based on the available data, the radiation hazard parameter is calculated. The external gamma dose rate calculated from the concentration of the three radionuclides ranged from 29.8 to 38.7 nGy h^{-1}. Three calculated parameters from the activity concentration values, i.e. the radium equivalent activity (Ra_{eq}) range between 59.3 and 78.6 Bq kg^{-1} (mean 68.1 Bq kg^{-1}), the representative level index ($I_{\gamma r}$) range between 0.46 and 0.59 (mean 0.50) and the external hazard index (H_{ex}) range between 0.17 and 0.21 (mean 0.20). This is well below the recommended value of 370 Bq kg^{-1} (for Ra_{eq}) and unity (for H_{ex}). The annual effective dose rate of the areas was determined to be between 0.037 and 0.048 mSv y^{-1}. The results show that there are no significant health hazards to humans.

Introduction

Since the exposure of human beings to ionisation radiation from natural sources is a continuing, these studies are very important for human. It also can be a baseline for natural background. Radiation exposure received by the individuals from the primordial sources in the environment is known to constitute about 85% of natural background radiation, the remaining 15% being from cosmic rays [1]. The high activity concentrations of natural radionuclides have been observed in igneous rock and phosphate rock. Many exposures to natural radiation sources are modified by human activities. The activities such as phosphate fertiliser production and minerals mining causing enhanced natural radiation exposures. For that reason, processing some of natural resources can cause increasing of activity concentration of radionuclides in the environment. This increment is a potential risk with respect to exposure of external radiation for humans [2].

Bukit Bunuh was discovered by the USM's team of archaeologist led by Associate Professor Dr. Mokhtar Saidin during the field survey to find the archaeology site in year 2000. Bukit Bunuh is situated in Lenggong valley, north of Kota Tampan, Perak. It is located in the longitude of 10° 058.5 East and latitude 5° 4.5 North. The highest point is over 180 meters above sea level. When the site covering more than 4 km² was first surveyed in the 80s, the surface was bare but a return survey in 2001 saw thousands of rocks of many types such as suevite, quartzite, quartz, and metamorphic rocks brought to the surface by agricultural activities [3].

Naturally, most of uranium presents as U-238 and thorium presents as Th-232. Both of them are less soluble compare to their decay daughter products such as Ra-226 for U-238 and Ra-228 for Th-232. Potassium (K), uranium (U) and thorium (Th) are three most abundant, naturally occurring radioactive elements. Potassium is a major constituent of most rocks whereas uranium and thorium are present in trace amounts, either as mobile or immobile elements.

Materials and Methods

All 10 rock samples (granite) were getting from the Centre for Global Archaeological Research USM. Figure 1 shows a map of sampling points in the study area and Table 1 shows the coordinates of the sampling points obtained by using global positioning system, Garmin GPSmap 76CSx. The area was chosen because of its potential of having a uranium deposit. This area also sits in the granitic region. Granitic rock usually has some amount of uranium deposit. All rock samples were oven dried at 60°C until it reached a constant weight and eventually crushed and packed. The samples were weighed (after correction were ~100 g) and stored for at least three weeks to allow for secular equilibrium between ^{226}Ra, ^{228}Ra and its decay products before gamma spectroscopy measurements were made. The period is also long enough to ensure equilibrium ^{226}Ra and its decay product in the ^{232}Th series.

Figure 1. Maps of sampling points

Tele:
E-mail addresses: wanzul88@gmail.com

Table 1. Sampling location and coordinates of the area

Sample code	Coordinates
A	N05^0 4' 13.10" E100^0 58' 34.66"
B	N05^0 4' 25.95" E100^0 58' 38.36"
C	N05^0 4' 40.16" E100^0 58'37.49"
D	N05^0 4' 16.33" E100^0 58' 18.16"
E	N05^0 4' 51.94" E100^0 58' 17.61"
F	N05^0 5' 9.89" E100^0 58' 17.07"
G	N05^0 4' 51.92" E100^0 58' 9.24"
H	N05^0 5' 1.29" E100^0 58' 11.09"
I	N05^0 5' 5.84" E100^0 58' 11.06"
J	N05^0 5' 6.35" E100^0 57' 53.99"

Gamma spectrometry measurements of ^{226}Ra, ^{228}Ra and ^{40}K activities in rock samples were made using a portable Amptek GAMMA-RAD5 76 $\times$ 76mm, coupled to a DppMCA (Version 1.0.0.5) through a photomultiplier tube, preamplifier and amplifier. The high sensitivity NaI(Tl) detector connected to a PC-based 8k multichannel analyser and the associated software. The detector was shielded by 50 mm thick lead bricks. The detector has a resolution of 6.7% full-width at half-maximum of ^{137}Cs energy of 661.6 keV. This was good enough to distinguish the gamma ray energies of interest in the present study.

The choice of radionuclides to be detected as a reference were made based on the fact that the NaI(Tl) detector used in the study had a modest energy resolution [4]. The activity concentration of ^{214}Bi (determined from its 609.31 keV gamma ray peak) was chosen to provide an estimate of ^{226}Ra (U series) in rock samples. The radionuclide ^{228}Ac (determined from its 911.2 keV gamma ray peak) was chosen as an indicator of ^{228}Ra (Th series) because the secular equilibrium was achieved between the daughter nuclides and their parent nuclides. ^{40}K was determined by measuring the 1460.83 keV gamma rays emitted during the decay of ^{40}K. The sample was placed symmetrically on top of the detector and measured for a counting period of 6 h [5]. Measurement of each rock sample was repeated three times and the mean net area (net count rate) was determined. Calculation of the activity concentration is based on the method that had been implemented by N. Füsun Çam *et. al* [6].

The activity concentrations of ^{238}U and ^{232}Th series and ^{40}K were used to calculate the external gamma dose rate, *D* in nGy h^{-1}. The following equation was used to calculate the external gamma dose rate [7].

$$D\left(\text{nGy h}^{-1}\right) = 0.461C_{Ra} + 0.623C_{Th} + 0.0414C_K \quad ..\ (1)$$

where *D* is the absorbed dose rate, C_{Ra}, C_{Th} and C_K the activity concentrations for uranium, thorium and potassium, respectively, in Bq kg^{-1}.

Radium equivalent activity (Ra_{eq}) is a sum of activities of ^{226}Ra, ^{228}Th and ^{40}K radionuclides based on assumption that 1 Bq kg^{-1} of ^{226}Ra, 0.7 Bq kg^{-1} of ^{232}Th and 13 Bq kg^{-1} of ^{40}K produce the same gamma ray dose [8]. The equation is defined as below:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad ..\ (2)$$

where C_{Ra}, C_{Th} and C_K have the same meaning as (1)

The representative level index, $I_{\gamma r}$ is defined by the following formula [9].

$$I_{\gamma r} = \frac{C_{Ra}}{150} + \frac{C_{Th}}{100} + \frac{C_K}{1500} \quad ..\ (3)$$

External hazard index due to the emitted gamma rays of the samples and determined using the following equation.

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \le 1 \quad ..\ (4)$$

The calculated average external hazard index was found to be less unity.

Annual effective dose (mSv year^{-1}) is calculated from the following equation.

$$AED = D(\text{nGy/h}) \times 8760(\text{h/year}) \times 0.2 \times 0.7(\text{Sv/Gy}) \times 10^{-6} \quad ..\ (5)$$

Discussion

Generally, all rock samples are granite rock. The presence of natural radionuclides in rock usually because of bedrock geology. To evaluate the geological background, the activity concentrations of each rock samples are measured and tabulated in Table 2. Activity concentration of three radionuclides which are ^{226}Ra, ^{228}Ra and ^{40}K have been analysed using gamma energy 609.3, 911.07 and 1460.75 keV respectively. Activity concentration of ^{226}Ra, ^{228}Ra and ^{40}K in rock samples were calculated in Bq/kg. It is also to note that the 609.3 keV is the gamma line of ^{214}Bi and the 911.07 keV is the gamma line of ^{228}Ac. At secular equilibrium, the ^{214}Bi is equivalent to the activity concentration of ^{226}Ra and the ^{228}Ac is equivalent to ^{228}Ra.

Table 2 lists the activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K measured in rock samples. The activity concentration of ^{226}Ra is in the range of 24.1-43.2 Bq/kg. Overall, most of rock samples give a lower activity concentration of ^{226}Ra if compare with other listed regions around the world. The mean value of activity concentration of ^{226}Ra is still in the range that had been appointed by UNSCEAR. This result also shows potential uranium deposits in this area.

Similarly with ^{228}Ra, the activity concentration in this rock sample is in the range of 1.2 – 3.5 Bq/kg. The pattern of activity concentration of ^{228}Ra is quite same between rock samples. Rock sample G shows a higher activity concentration of ^{228}Ra compare to other sample rocks.

Unlike ^{226}Ra and ^{228}Ra, the activity concentration of ^{40}K shows the highest values at all sampling points. The highest value of activity concentration of ^{40}K at rock sample A, with the value of 438.2 Bq/kg. Meanwhile, the rock sample B give the lowest activity concentration for ^{40}K, with the value of 274.3 Bq/kg. Generally, the activity concentration of ^{40}K does not give effect on the concentration of uranium in rock, since ^{40}K does not belong to any uranium or thorium series. ^{40}K usually exists naturally and it can be raised by the anthropogenic activities such as agriculture. Most of Bukit Bunuh area had been planted with oil palm and rubber tree. Fertilizer application that had a high potassium content in that area maybe can cause the activity concentration of rock sample is higher.

The activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K in rock samples from studying area were compared with similar investigations in other countries. Table 3 shows a comparison of the activity concentration with other studies. Nagdya (2003) studied radioactive disequilibrium in the different rock types in Wadi Wizr, the Eastern Desert of Egypt and the concentration of ^{226}Ra, ^{228}Ra and ^{40}K in the rocks are ranged from 36 to 661 Bq/kg, from 0.9 to 13.8 Bq/kg and from 11.8 to 248 Bq/kg, respectively [10].

Table 1. Sampling location and coordinates of the area

Sample code	Coordinates
A	N05^0 4' 13.10" E100^0 58' 34.66"
B	N05^0 4' 25.95" E100^0 58' 38.36"
C	N05^0 4' 40.16" E100^0 58'37.49"
D	N05^0 4' 16.33" E100^0 58' 18.16"
E	N05^0 4' 51.94" E100^0 58' 17.61"
F	N05^0 5' 9.89" E100^0 58' 17.07"
G	N05^0 4' 51.92" E100^0 58' 9.24"
H	N05^0 5' 1.29" E100^0 58' 11.09"
I	N05^0 5' 5.84" E100^0 58' 11.06"
J	N05^0 5' 6.35" E100^0 57' 53.99"

Table 2. Activity concentration of ^{226}Ra, ^{228}Ra and ^{40}K in rock samples

Sample	Activity concentration of ^{226}Ra (Bq/kg)	Activity concentration of ^{228}Ra (Bq/kg)	Activity concentration of ^{40}K (Bq/kg)
A	41.4 ± 2.2	2.4 ± 0.3	438.2 ± 74.6
B	42.3 ± 0.9	2.2 ± 0.2	274.3 ± 28.8
C	38.5 ± 2.6	3.1 ± 0.2	335.1 ± 8.1
D	29.2 ± 4.0	2.7 ± 0.7	378. 2 ± 44.4
E	37.9 ± 3.0	1.2 ± 0.2	371.9 ± 11.4
F	40.2 ± 2.2	1.5 ± 0.2	409.5 ± 11.9
G	27.9 ± 3.1	3.5 ±0.4	418.6 ± 37.5
H	34.9 ± 2.4	2.2 ± 0.2	437.2 ± 8.7
I	22.5 ± 1.6	2.6 ± 0.1	429.5 ± 12.2
J	38.1 ± 1.1	1.8 ± 0.5	339.2 ± 0.9

Table 3. Comparison of the activity concentration of ^{226}Ra, ^{228}Ra and ^{40}K in rock samples

Country	Sample	Activity concentrations of ^{226}Ra(Bq/kg)	Activity concentrations of ^{228}Ra(Bq/kg)	Activity concentrations of ^{40}K (Bq/kg)	References
Malaysia (Bukit Bunuh)	Rock granite (quartz)	22.5 – 42.3	1.2 – 3.5	274.3 – 438.2	Present study
Egypt (Wadi Wizr)	Rock	36 – 661	0.9 – 13.8	11.8 – 248	Nagdya
Egypt (Bir El-Sid)	Rock granite	57.4	53.4	1041.4	Ahmed *et al.*
Egypt (Wadi El-Gemal)	Rock granite	39	47.9	1031	Ahmed *et al.*
Germany	Rock granite	76.1	70	1465.4	Ahmed *et al.*
Egypt (W. Allaqi)	Rock granite	9 – 111	8 – 75	119 – 790	S. A. M. Issa *et al.*
Egypt (Abu Ziran	Rock granite	19 – 34	11 – 15	216 – 274	S. A. M. Issa *et al.*
India (Kaiga)	Rock	1.2 – 14.2	0.5 – 11.5	14.8 – 866.2	Patra *et al.*
Egypt (Eastern Desert)	Rock granite	9.69 – 18.97	9.99 – 17.65	298.58 – 955.78	Shaban Harb *et al.*
Turkey (Aliağa-Foça)	Rock	12 – 96	11 – 123	264 – 1470	N. Füsun Çam *et al.*
Yemen (North Sana'a)	Rock	21.79	19.15	399.3	Shaban Harb *et al.*
World	-	16 – 116	7 – 50	100 – 700	UNSCEAR (2000)

Table 4. Sample code as well as calculated external gamma dose rate, radium equivalent activity, representative level index and external hazard index

Sample code	D (nGy/h)	Ra_{eq} (Bq/kg)	$I_{\gamma r}$	H_{ex}
A	38.7	78.6	0.59	0.21
B	32.3	66.7	0.49	0.18
C	33.5	68.7	0.51	0.19
D	30.8	62.3	0.47	0.17
E	33.7	68.3	0.51	0.18
F	36.4	73.8	0.56	0.20
G	32.4	65.2	0.50	0.18
H	35.6	71.8	0.55	0.19
I	29.8	59.3	0.46	0.16
J	32.7	66.7	0.50	0.18
Mean Value	33.6	68.1	0.50	0.20

Table 5. Annual effective dose and in-situ measurement dose (surface and 1 m above the ground dose)

Sample code	Annual effective dose (mSv/yr)	In-situ surface dose		In-situ 1 m above the ground dose	
		in μSv/h	in mSv/yr	in μSv/h	in mSv/yr
A	0.048	0.36	3.16	0.37	3.24
B	0.039	0.47	4.12	0.35	3.07
C	0.041	0.27	2.36	0.24	2.10
D	0.038	0.25	2.19	0.17	1.49
E	0.041	0.25	2.19	0.25	2.19
F	0.045	0.19	1.67	0.21	1.84
G	0.040	0.32	2.81	0.29	2.54
H	0.044	0.41	3.59	0.22	1.93
I	0.037	0.41	3.59	0.25	2.19
J	0.040	0.34	2.98	0.25	2.19
Mean value	0.041	0.33	2.87	0.26	2.28

Table 6. Comparison the external gamma dose rate, radium equivalent, representative level index, external hazard index and annual effective dose for rock samples from the present study with other studies

Country	Sample	*D* (nGy/h)	Ra_{eq} (Bq/kg)	$I_{\gamma r}$	H_{ex}	AED (mSv/year)	References
Malaysia (Bukit Bunuh)	Rock granite (quartz)	29.8 – 38.7	59.3 – 78.6	0.46 – 0.59	0.17 – 0.21	0.037 – 0.048	Present study
Egypt (Wadi Wizr)	Rock	13 – 209	-	-	-	-	Nagdia
Egypt (Bir El-Sid)	Rock granite	3.4	7.3	0.05	-	-	Ahmed *et al.*
Egypt (Wadi El-Gemal)	Rock granite	-	-	-	-	-	Ahmed *et al.*
Germany	Rock granite	-	-	-	-	-	Ahmed *et al.*
Egypt (W. Allaqi)	Rock granite	24.8 – 89.2	49 – 183.2	-	0.1 – 0.5	0.030 – 0.108	S. A. M. Issa *et al.*
Egypt (Abu Ziran	Rock granite	27 – 33.4	53.8 – 68.4	-	0.2	0.033 – 0.041	S. A. M. Issa *et al.*
India (Kaiga)	Rock	1.8 – 38.0	-	-	-	0.0073 – 0.0744	Patra *et al.*
Egypt (Eastern Desert)	Rock granite	24.24 – 58.05	49.05 – 113.39	0.53 – 1.14	0.16 – 0.35	0.12 – 0.28	Shaban Harb *et al.*
Turkey (Aliağa-Foça)	Rock	36 – 179	76 – 382	-	0.21 – 1.08	0.044 – 0.220	N. Füsun Çam *et al.*
Yemen (North Sana'a)	Rock	38.39	82.2	0.3	0.21	-	Shaban Harb *et al.*
World	-	55	370	1	1	0.3 – 0.6	UNSCEAR (2000)

Ahmad *et al*. (2006) doing a comparative study of the natural radioactivity of some selected rocks from Egypt (Bir El-Sid and Wadi El-Gemal) and Germany [11]. The activity concentration for ^{226}Ra are 57.4, 39 and 76.1 Bq/kg, respectively. For ^{228}Ra and ^{40}K, the activity concentration are 53.4, 47.9, 70 and 1041.4, 1031, 1465.4 Bq/kg, respectively. S. A. M. Issa *et al.* (2011) determined the natural radionuclide concentrations in granite rocks in W. Allaqi and Abu Ziran, Egypt [12]. The rock activity ranged from 9 to 11 and 19 to 34 Bq/kg; from 8 to 75 and 11 to 15 Bq/kg; from 119 to 790 and 216 to 274 Bq/kg for ^{226}Ra, ^{228}Ra and ^{40}K, respectively. Similar investigations in other countries are also compared with the present study such as Patra *et al.* [13], Shaban Harb *et al.* [14] and N. Füsun Çam *et al.* [6].

Table 4 shows the external gamma dose rate, radium equivalent activity, representative level index and external hazard index for Bukit Bunuh area. The mean external gamma dose rate for Bukit Bunuh area is 33.6 nGy/h. This calculated values were lower than the estimate of average global terrestrial radiation of 55 nGy/h [15]. Radium equivalent activity is used to estimate the hazard posed by the different concentrations of radionuclides in materials. The mean Raeq for Bukit Bunuh area is 68.1 Bq/kg. The mean Ra_{eq} value is lower than the internationally accepted value 370 Bq/kg [8]. The mean value of representative level index for the studied samples is 0.50. The $I_{\gamma r}$ values are below the internationally accepted value 1. The mean external hazard index for Bukit Bunuh area is 0.20 which is less than 1. The external hazard index estimated for this rock samples were less than the safety limit.

Table 5 shows the corresponding annual effective dose and in-situ measurement dose for Bukit Bunuh area. The annual effective dose is calculated based on the external gamma dose rate (absorbed dose). The mean value for the annual effective dose for Bukit Bunuh area is 0.037 mSv/year. The world average range is between 0.3 – 0.6 mSv/year and the limit is 1 mSv/year [7]. A dose rate measurements were taken 1 m from the ground and surface using a RAM DA3-2000 Survey Meter with unit μSv/h for natural background radiation. A correlation study between annual effective doses with in-situ dose rate was carried out as shown in Figure 2. From the figure above, there is no strong correlation between annual effective dose with in-situ dose rate measurement as the points are scattered around and no trends are obtained from the graph. This shows that the measured surface doses and annual effective doses are not correlated. It is also shown that not only these three radionuclides that contribute to the radiation in the area but the other radionuclides that is also exists and contribute to the surface dose rate.

Table 6 shows the comparison of external gamma dose rate, radium equivalent Ra_{eq}, representative level index $I_{\gamma r}$, external hazard index H_{ex} and annual effective dose for rocks from Bukit Bunuh with the results in other countries (Nagdia [10], Ahmed *et al.* [11], S. A. M. Issa *et al.* [12], Patra *et al.* [13], Shaban Harb *et al.* [14,16] and N. Füsun Çam *et al.* [6]). It can be seen that values of external gamma dose rate for rocks obtained does not exceed from the international recommended value except in the case of Wadi Wizr, Egypt and Aliağa-Foça, Turkey. The representative level index, radium equivalent activity, external hazard index and annual effective dose from the other studies shows the values that had been estimated not exceeded the value that had been recommended.

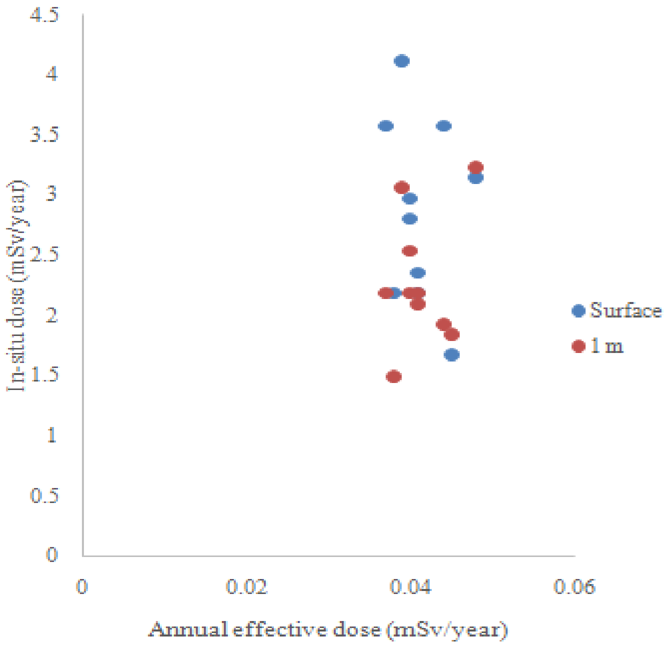

Figure 2. Correlation between annual effective dose and in-situ dose measurement

Conclusion

Selected rock samples from Bukit Bunuh were measured for their radioactivity content. The activity concentration of ^{226}Ra (uranium series), ^{228}Ra (thorium series) and ^{40}K were measured using a NaI(Tl) spectrometer. The result shows that the activity concentration of ^{226}Ra range from 22.5 – 42.3 Bq/kg for uranium series. The activity concentration of ^{228}Ra range from 1.2 – 3.5 Bq/kg for thorium series. The activity concentration for ^{40}K range from 274.3 – 438.2 Bq/kg. These results are comparable with the literature. The mean values and ranges of the external gamma dose rate (*D*), radium equivalent activity (Raeq), representative level index ($I\gamma r$) and external hazard index (Hex) for rock samples were 33.6 (29.8 – 38.7) nGy/h, 68.1 (59.3 – 78.6) Bq/kg, 0.50 (0.46 – 0.59) and 0.20 (0.17 – 0.21), respectively. The results show that the dose rate in all rock samples does not exceed from what had been recommended by UNSCEAR which is 55 nGy/h [7]. It is understood that from the USM Geophysics' group, the Bukit Bunuh area had been reliable is one of the meteorite impact areas. This study will help us to see any significant health hazard to humans in long terms especially for those that lives and work in there.

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