MEASUREMENT OF ^{226}Ra, ^{228}Ra AND ^{40}K IN SOIL IN DISTRICT OF KUALA KRAI USING GAMMA SPECTROMETRY

(Pengukuran ^{226}Ra, ^{228}Ra dan ^{40}K Di Dalam Taneh Daerah Kuala Krai Menggunakan Spektrometri Gama)

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Abstract

The granitic region is known to have high natural radionuclides content. The natural background of the area will be elevated and the exposure rate also will be higher as compared to other region. The present study is focusing on the presence of natural uranium isotopes using its progenies in soils belong to the river basin of the granitic region of Kuala Krai district, Malaysia. Granitic characteristics of the region were believed to produce significant concentrations of natural radionuclide such as uranium and thorium. This paper presents the results of measurement activity concentration ^{226}Ra, ^{228}Ra and ^{40}K in soil using Gamma Spectrometry to estimate activity concentration of radionuclides in fourteen soil samples collected from this study area. The range of activity concentration of ^{226}Ra, ^{228}Ra and ^{40}K is 40.2-264.0, 49.2-312.9 and 491.1-1184.2 Bq/kg respectively. These results were used to estimate the hazards index and annual exposure rate to the member of public.

Keywords: ^{238}U activity concentration, granitic region, gamma spectrometer

Abstrak

Kawasan granatik telah diketahui mempunyai kandungan radionuklida tabii yang tinggi. Kadar sinaran latar kawasan tersebut juga akan tinggi dan kadar pendedahan terhadap manusia juga akan lebih tinggi berbanding dengan kawasan lain. Kajian ini memberikan fokus kepada pengesanan isotop uranium tabii menggunakan progeninya di dalam taneh yang di ambil dari kawasan granit di Daerah Kuala Krai, Malaysia. Sifat-sifat granitik kawasan ini dipercayai mempunyai kepekatan radionuklida tabii seperti uranium dan torium yang signifikan. Kertas kerja ini membentangkan hasil kajian pengukuran ^{226}Ra, ^{228}Ra dan ^{40}K di dalam taneh menggunakan spektrometri gama bagi menentukan kepekatan aktiviti radionuklida di dalam empat belas sampel taneh dari kawasan kajian. Julat pengukuran bagi ^{226}Ra, ^{228}Ra dan ^{40}K masing-masing 40.2-264.0, 49.2-312.9 dan 491.1-1184.2 Bq/kg. Keputusan ini telah digunakan untuk mengira indeks berbahaya dan kadar pendedahan tahunan terhadap penduduk setempat.

Kata kunci: Kepekatan aktiviti ^{238}U, kawasan granit, spektrometer gama

Introduction

Kuala Krai is one of the districts in Kelantan and the second largest after Gua Musang District. Kuala Krai has an area of 2.329 km^2, bordering with Machang in the North and Gua Musang in the South, while the East is adjacent to the Terengganu and Jeli in the West. It is located on granites set which is an undifferentiated intrusive rock. It can be categorized into two sets which are coarse grain and fine grain. Coarse grain consists of pink granite, adamellite, diorite and granodiorite. Fine grain consists of rhyolite and dacite. Besides, there are lots of non-metallic mineral (clay, bell clay, kaolin, silica, barytes, serpentine, limestone, and dolomite), light metals in heavy minerals (e.g. ilmenite rutile) and other metals such as iron, manganese, chromium, copper, zinc, lead, gold and even uranium [1].

Information of distribution pattern of radionuclides in the soils is absolutely necessary in controlling radiation exposure level and can give some details or changes in the background on natural sources of radionuclides [2,3,4]. The natural radioactivity of soil and sediment depends on the soil and sediment formation and transport process that involving chemical and biochemical interactions that influence the distribution patterns of uranium, thorium and their decay products [5]. Although widely distributed, their concentration in the environment is believed to depend on the local geological conditions [6].

There were several studies on the measurement of radionuclides in Malaysia such as a few studies done by ESCAN (Environmental Studies using Conventional and Nuclear Techniques) group from UiTM. They have done studies on low level radionuclides in palm oil area in Jengka, Pahang, former tin mining area in Kg. Gajah, Perak and tourism beach areas on the peninsular side in Malaysia [7,8,9]. Those studies can be used as a guide as well as risk assessment to evaluate the possible exposure level to the natural radiation. However, until now, no similar study has been done in Kelantan, thus, this paper is aimed to set a baseline data for any possible environmental conditions which can be used for a future reference for any anthropogenic activities.

Measurement of ^{226}Ra, ^{228}Ra dan ^{40}K in soil using Gamma Spectrometer is reliable and preferably because it is an established method. Gamma Spectrometry is a convenience method with simple preparation procedure and simultaneously determined many radionuclides in a sample [10,11].

The objectives of the present study, are to measure the activity concentration of ^{226}Ra, ^{228}Ra dan ^{40}K in soil samples collected from district of Kuala Krai in Kelantan, to estimate the radium equivalent activity (Ra_{eq}), total absorbed dose rate (D), external hazard index (H_{ex}) and annual effective dose. The finding will be used as a baseline data in estimating the exposure to the background radiation for the Kelantan's population.

Method

Sampling site

The top soil samples were collected along Sungai Kelantan and its tributaries in the district of Kuala Krai. Figure 1 shows a map of sampling points in the study area, while Table 1 shows the coordinates of the sampling point obtained by using GPS.

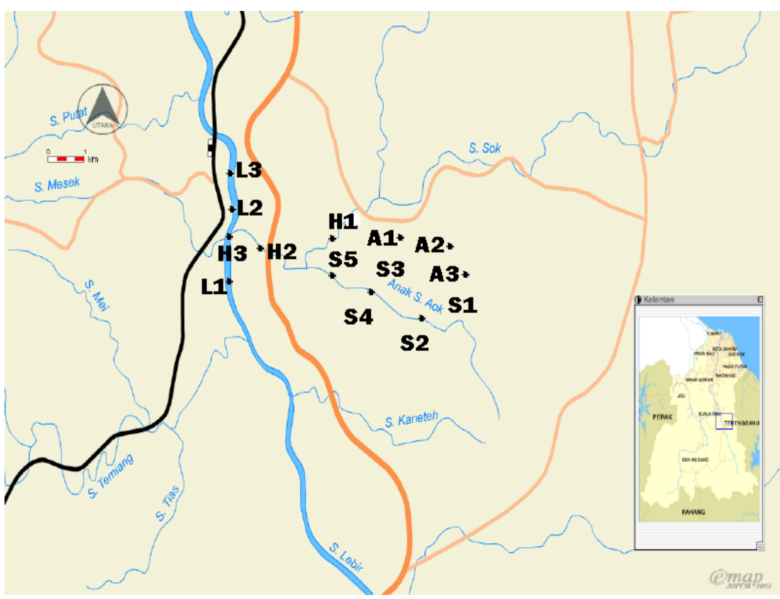

Figure 1: Maps of Sampling Points

Samples were collected at two types of locations, the one at hill and the other is along river. The area was chosen because of its potential of having uranium deposit and as well rich with other minerals. This area also is sitting on the granitic, giving another reason of choosing this area, because granitic rock usually has some amount of uranium deposit. The coordinates of the sampling points are given in the Table 1.

Table 1: Sampling location, condition and the coordinates of the area.

Sample Code	Location	Coordinates
A1	Hilly area	-
A2		N05°21.372’ , E102°17.554’
A3		N05°21.379’ , E102°17.339’
S1	Small river	N05°22.231’ , E102°15.042’
S2		
S3		N05°22.262’ , E102°14.984’
S4		
S5		N05°22.311’ , E102°15.184’
H1	Small river	N05°22.514’ , E102°16.605’
H2		N05°22.789’ , E102°16.186’
H3		N05°22.486’ , E102°14.141’
L1	Main river	N05°22.133’ , E102°14.233’
L2		N05°22.680’ , E102°14.332’
L3		N05°23.253’ , E102°14.195’

Sampling and samples preparation

Soil samples were taken at 15 cm depth using hand auger at five different holes at each sampling point as to represent the location. All the stones, grass, root and the non-soil were taken out before transferred it into the plastic bags. The samples were then dried at 60°C until the constant mass. The dried samples were then ground using Agate bowl mill and sieved through 250 µm aperture mesh to homogenize them. After that, those samples were transferred into 500 ml plastic container and sealed for at least three weeks to reach the secular between radium and the corresponding daughters.

Instrumentation

All measurements was performed with gamma spectrometer on an ORTEC coaxial HPGe detector having 1.85 keV energy resolution with a relative photo peak efficiency of 25%, at 1332 keV ^{60}Co gamma ray. The associated electronics consisted of a multi channel analyzer (MCA) allowing the determination of the uranium and thorium series radionuclide. For γ-analysis, the samples were placed directly over a coaxial HPGe detector. The sample counting time was 21600 seconds. The integrated counts for energy peaks of ^{226}Ra, ^{228}Ra and ^{40}K were analysed. Spectra analysis was done using Gamma Vision software. The efficiency calibration of the spectrometer was obtained using analytical grade UO_3 ore in KCl matrix prepared in UiTM laboratory [12].

Results and Discussion

Activity concentration of three radionuclides which are ^{226}Ra, ^{228}Ra and ^{40}K have been analyzed using gamma energy of 609, 911.2 and 1460 keV respectively. These energy peaks were chosen because of its high intensity which is 0.461, 0.29 and 0.107 respectively. Activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K in samples were calculated in Bq/kg. For this measurement, the Minimum Detectable Activity (MDA) was estimated to be about 1.5 Bq/kg [7].

Table 2 lists the activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K measured in 14 soil samples. The result shows a pattern whereby the hilly areas have the activity concentration of about two times higher than along the rivers. The activity concentration of ^{226}Ra (uranium series) is in the range of 143.4-264.0 Bq/kg at the hilly areas while 40.2-

182.1 Bq/kg along the river. It shows the hilly area has more uranium content in the soil, we need to do a thorough study in this area in order confirm that it has a potential uranium deposits.

Similarly with thorium series, the activity concentration of ^{228}Ra in the samples from the hilly area especially A1 and A3 also give higher value than others, i.e. 312.9 and 273.6 Bq/kg respectively. Generally, Th always presents with U in soil and give normal ratio about 1-3. Another sampling point that is H2 gives higher value of 284.4 Bq/kg. The ratio of ^{228}Ra/^{226}Ra shows that from this study area is in the range of 0.6-1.6.

Unlike ^{226}Ra and ^{228}Ra, the activity concentrations of ^{40}K show the highest value at H1 sampling point, with the value of 1184.2 Bq/kg. Activity concentration of ^{40}K does not give any effect on the concentration on uranium in soil, since ^{40}K does not belong to any uranium or thorium series. ^{40}K exists naturally and it can be elevated by the anthropogenic activities, such as fertilizer application in the agricultural area. Generally ^{40}K activity concentrations is higher in the samples collected along the river than the hilly area. This could be due to river system where it was flooded during the monsoon season and will deposits it potassium content along the river bank.

Table 2**:** Activity concentration of ^{226}Ra, ^{228}Ra and ^{40}K in soil samples

Sample Code	Activity Concentration ^{226}Ra (Bq/kg)	Activity Concentration ^{228}Ra (Bq/kg)	Activity Concentration ^{40}K (Bq/kg)	^{228}Ra/^{226}Ra ratio
A1	264.0±16.2	312.9±17.7	649.5±25.5	1.2
A2	143.4±12.0	166.8±12.9	559.9±23.7	1.2
A3	251.1±15.8	273.6±16.5	491.1±22.2	1.2
S1	124.7±11.2	168.6±13.0	1005.4±31.7	1.1
S2	159.6±12.6	192.6±13.9	932.8±30.5	1.4
S3	120.6±11.0	160.0±12.6	988.6±31.4	1.3
S4	162.9±12.8	213.2±14.6	820.7±28.6	1.3
S5	130.5±11.4	199.1±14.1	646.1±25.4	1.5
H1	122.7±11.1	148.5±12.2	1184.2±34.4	1.2
H2	182.1±13.5	284.4±16.9	671.6±25.9	1.6
H3	47.1±6.9	54.2±7.4	498.4±22.3	1.2
L1	122.7±11.1	71.7±8.5	671.0±25.9	0.6
L2	75.0±8.7	100.4±10.0	599.9±24.5	1.3
L3	40.2±6.3	49.2±7.0	650.4±25.5	1.2
*Malaysia average	67(38-94)	82(63-110)	310(170-430)	1.3
* Recommended Value	35	30	400	-

*UNSCEAR (2008), in bracket is the range value

Table 3 shows the corresponding total absorbed dose rate (D), radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}) and annual effective dose for each samples. The calculation was done using the reviewed equations below. The absorbed dose rate of gamma-rays is the estimate of amount of radiation receive or deposited onto human, therefore there is a need to calculate this value since there are villages along the rivers where the population are exposed to the gamma radiation. The radium equivalent activity and external hazard index are calculated to assess the radiological hazard index of soils. The annual effective dose is also higher than other places, thus it may affect the health of the human.

The contribution of natural radionuclides in absorbed dose rate is depending on the activity concentration of ^{226}Ra (uranium series), ^{228}Ra (thorium series) and ^{40}K. It is a total ionizing dose and is divided by the time it takes to deliver the dose. The high dose rate usually can cause more harm and damage to the human body than low dose rates. The absorbed dose rate for area under investigation is tabulated in Table 3. The highest value of absorbed dose rate is from A1 and A3 which are 343.5 and 306.5 nGy/hr respectively. The value from this study is much higher than value found by Kurnaz [13], where the absorbed dose rate in air at Firtina Valley is ranging from 19.1 to 149.6 nGy/hr. It is also beyond the international recommended value which is 55 nGy/hr [14].

$$\text{Absorbed Dose Rate} = 0.461C_{Ra} + 0.623C_{Th} + 0.0414C_K \quad (1)$$

Radium equivalent activity (Ra_{eq}) is a term of radiation hazard that is widely used to set a limit or regulatory regarding total activity of radionuclides. It is assumed that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K produce the same gamma-ray dose [15,16,17]. S3 shows the biggest value which is 761.4 Bq/kg suggested that total activity of radionuclides from this place are the highest among others. S5 and H2 also give slightly higher than the other places with 680.2 and 640.5 Bq/kg respectively. Thus, the exposure of radiation to human that lives around the area should be monitored as it can affect the health of people.

$$\text{Radium equivalent} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (2)$$

External hazard index (H_{ex}) is another one of index that has been widely used to set a limit of exposure to human. The external hazard index is ranging from 0.43 to 2.02 with the highest value at points A1 and S1. It can be considered as a high value because the limit of external hazard index is 1.00, thus will affect the health of public or villagers around this area.

$$\text{External Hazard Index} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \quad (3)$$

Annual effective dose is an estimate of the stochastic effect of radiation on human. It takes into account that people spend the times outdoor only 20% of the day, the dose received from natural radioactivity range from 0.09-0.42 mSv/yr.

$$\text{Annual Effective Dose} = D\ (\text{nGy/h}) \times 8760\ (\text{h/year}) \times 0.2 \times 0.7\ (\text{Sv/Gy}) \times 10^{-6} \quad (4)$$

Table 3: Sample codes as well as calculated absorbed dose rates, radium equivalent, external hazard index and Annual Effective Dose and In-situ surface dose.

Sample Code	D (nGy/hr)	Ra_{eq} (Bq/kg)	H_{ex}	Annual Effective Dose (mSv/yr)	In-situ surface dose (µSv/hr)	In-situ 1m above the ground dose (µSv/hr)
A1	343.5	443.2	2.06	0.25	0.58	0.57
A2	193.2	506.8	1.15	0.28	0.50	0.54
A3	306.5	425.5	1.84	0.24	0.55	0.54
S1	204.1	530.9	2.20	0.30	0.53	0.55
S2	232.2	465.0	1.37	0.26	0.49	0.46
S3	196.2	761.4	1.15	0.42	0.41	0.50
S4	241.9	425.1	1.43	0.24	0.57	0.56
S5	211.0	680.2	1.26	0.38	0.72	0.68
H1	198.1	426.3	1.15	0.24	0.46	0.60
H2	288.9	640.5	1.73	0.35	0.18	0.21
H3	76.10	163.0	0.44	0.09	0.17	0.22
L1	129.0	276.9	0.75	0.16	0.39	0.34
L2	121.9	264.7	0.71	0.15	0.32	0.31
L3	76.10	160.7	0.43	0.09	0.38	0.39

A correlation study between annual effective doses with in-situ dose rate was carried out, as shown in **Figure 2**. There is no strong correlation between them as the points are scattered around and no trends are obtained. This indicates that the measured surface doses and annual effective doses are not correlated. It is also indicates that not only these three radionuclides that contribute to the radiation in the area but other radionuclides also exist and contribute to the surface dose rate.

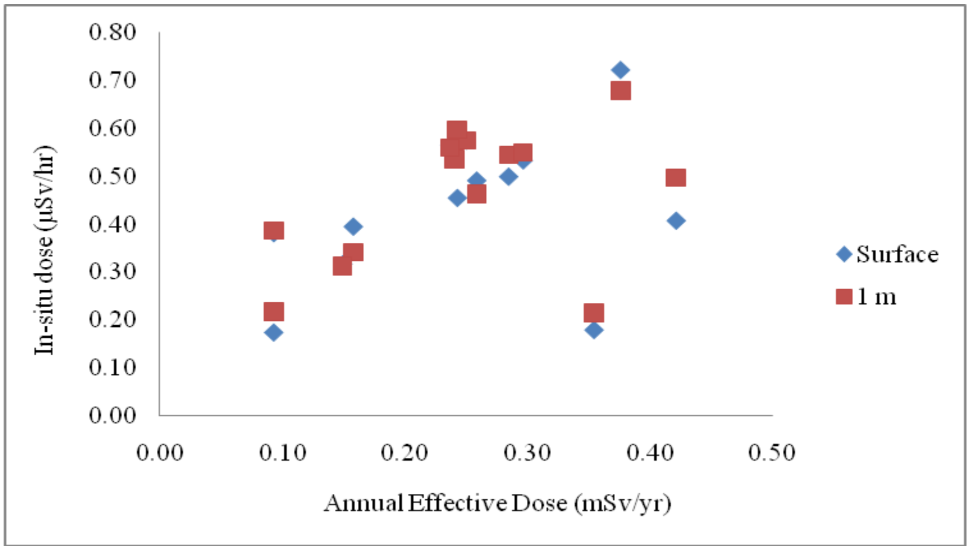

Figure 2: Correlation between Annual Effective Dose and in-situ dose measurements

Table 4 shows the comparison of the present study with the studies in Malaysia as well as in the whole world. Generally, most of the samples from the present study give values exceeded the international recommended value. It is slightly higher than the value obtained in Jengka, Pahang and beach areas. The value from Kg. Gajah, Perak, is higher than this study due to the former tin mining area with historical ore processing and smelting activities. Comparing to the world average, Kelantan area still on higher average level in terms of radiation level except for the annual effective dose.

Table 4: Comparison of the activity concentration of ^{226}Ra, ^{228}Ra and ^{40}K in soil samples as well as calculated absorbed dose rates, radium equivalent, external hazard index and absorbed dose rates with the literature.

Region	Activity Concentration (Bq/kg)			D (nGy/hr)	Ra_{eq} (Bq/kg)	H_{ex}	AED (mSv/yr)	References
	^{226}Ra	^{228}Ra	^{40}K					
Kuala Krai, Kelantan, Malaysia	40.2-264.0	49.2-312.9	491.1-1184.2	76.1-343.5	160.7-761.4	0.43-2.02	0.09-0.42	Present study
Tourism Beach Areas, Malaysia	7.3-51.0	5.9-58.4	32-1293	9.05-90.09	18.3-189.3	0.05-0.51	0.05-0.55	Ahmad *et al.*, (2011)
Jengka 15, Pahang, Malaysia	16.6-22.1	22.4-33.8	55.5-243	25.76-42.41	53.5-91.4	0.16-0.25	0.78×10^{-4}-1.42×10^{-4}	Masitah Alias *et. al.*, (2008)
Kg. Gajah, Perak, Malaysia	199-12932	1215-32330	21-9304	558.7-35951.2	-	3.43-220.36	-	Zaini Hamzah *et. al.*, (2008)

Turkey (Firtina Valley)	15.29-188.26	-	105.35-1234.65	19.12-149.58	44.92-385.85	0.12-1.04	0.0234-0.1834	Kurnaz *et al.* (2007)
Kinta District, Perak, Malaysia	12-426	19-1377	19-2204	39-1039	52-2227	-	-	Lee *et al.* (2009)
Burullus Lake, Eqypt	6.1-40	4.3-44.3	224-341	14.6-51.6	30.1-112.7	-	-	El-reefy *et al.* (2006)
Dhaka City, Bangladesh	21-43	34-81	402-750	-	-	-	-	Miah *et. al.*, (1998)
Aldama, Chihuahua, Mexico	44.6-460.5	41.9-77.0	807.5-1766.2	220.4-83.8 (nSv/hr)	-	-	0.44-0.73	Sujo *et. al.*, (2004)
Inshaa, Cairo	5.3-7.7	10.7-17.0	152-202	-	-	-	-	Higgy & Pimpl, (1998)
*World Average Range	16-116	7-50	100-700	44	370	-	0.3-0.6	UNSCEAR (2000).

*UNSCEAR (2000)

Conclusion

The environmental radioactivity monitoring of 14 soil samples from the region of having the potential uranium deposits showed high activity concentration of ^{226}Ra range from 40.2-264.0 Bq/kg. The activity concentration of ^{228}Ra ranges from 49.2-312.9 Bq/kg and activity concentration of ^{40}K 491.1-1184.2 Bq/kg, which are comparable with literature. This maybe attributed from the granitic region of that area that been believed to have a potential uranium deposit. From the activity concentration of these radionuclides, external hazard index (H_{ex}) of the area range is 0.43-2.02. This study can be used as the baseline data and can be used as reference data for monitoring any pollution for future study in that area or neighboring place.

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