RADIOLOGICAL STUDIES OF NATURALLY OCCURRING RADIOACTIVE MATERIALS IN SOME MALAYSIA'S SAND USED IN BUILDING CONSTRUCTION

(Kajian Radiologi Bahan Radioaktif Tabii Beberapa Pasir Malaysia Yang Digunakan Dalam Binaan Bangunan)

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Abstract

The presence of natural radioactivity in materials used in building constructions results in external and internal radiation exposure to the dwellers. Sand is one of the main components in building construction beside cements, granites and bricks. Thus, this research has been carried out in order to investigate the levels of natural radioactivity and associated radiation hazard in some Malaysia's sand used in building constructions. Samples were obtained directly from local hardware stores. The activity concentration of ^{226}Ra (^{238}U series), ^{232}Th and ^{40}K were determined using gamma-ray spectrometry. Activity concentration of ^{226}Ra (^{238}U series), ^{232}Th and ^{40}K were found in the range of 6.45 to 107.90 Bq/kg, 7.78 to 96.67 Bq/kg and 31.05 to 1105.53 Bq/kg respectively. The mean range of representative gamma level indexes were found to be in the range of 0.189 ± 0.027 to 2.366 ± 0.057 whereas the mean range of annual equivalent doses to dwellers were in the range of 0.059 ± 0.005 mSv/year to 0.738 ± 0.018 mSv/year.

Keywords: Natural radioactivity; building construction; sand; radiation hazard

Abstrak

Kehadiran keradioaktifan tabii dalam bahan binaan bangunan mengakibatkan kepada dedahan sinaran luaran dan dalaman kepada penghuni. Pasir merupakan salah satu komponen penting dalam pembinaan bangunan selain daripada simen, batu kerikil dan batu-bata. Kajian ini dilakukan bertujuan untuk menentukan kepekatan keradioaktifan tabii serta menilai hazard sinaran dalam sampel pasir yang digunakan untuk membina bangunan di beberapa lokasi di Semenanjung Malaysia. Kepekatan aktiviti ^{226}Ra (siri ^{238}U), ^{232}Th dan ^{40}K ditentukan menggunakan spektrometri sinar gama. Hasil kajian menunjukkan julat purata kepekatan aktiviti masing-masing bagi ^{226}Ra (siri ^{238}U), ^{232}Th dan ^{40}K berada dalam julat 6.45 – 107.90 Bq/kg, 7.78 – 96.67 Bq/kg dan 31.05 – 1105.53 Bq/kg. Purata aras perwakilan sinar gama berada dalam julat 0.189 ± 0.027 hingga 2.366 ± 0.057 manakala julat purata dos setara tahunan adalah 0.059 ± 0.005 mSv/tahun hingga 0.738 ± 0.018 mSv/tahun.

Kata kunci: Keradioaktifan tabii; pembinaan bangunan; pasir; hazard sinaran

Introduction

Naturally occurring radioactive materials is a widespread substance that can be found everywhere in the environment including soil, rocks, water, air and also tissues of living things. There are no ways to avoid the presence of natural radionuclide since it presence from the formation of earth. Sand is one of the main components in building constructions are known to contain with naturally occurring radioactive materials. Sands are mineral deposits formed through weathering and erosion of either igneous or metamorphic rocks [1]. Natural radioactivity in sands contributed radiation dose to dwellers that originate from ^{238}U, ^{232}Th and their progeny and ^{40}K. For building construction purposes, sand is not just drawn out from river but it is also extracted from the abandoned mines. Yasir *et al*. [2] reported the concentration of natural radioactivity in evacuated tin mining sand located in Dengkil, Selangor, Malaysia is high compared to the world average values of natural radioactivity in soil.

The awareness of radiological impact to dwellers also increase due to the potential of the unwanted sand in tin tailings processing industry locally called 'amang' to be used in building construction. Several studied on natural radioactivity in tin tailings processing industry in Malaysia reported that the concentration was relatively high compared to the world average [3-4].

UNSCEAR [5] reported that the world average of activity concentrations for ^{226}Ra (^{238}U series), ^{232}Th and ^{40}K are 35 Bq/kg, 30 Bq/kg and 400 Bq/kg respectively. Research conducted by Omar [6] found that the activity concentration (Bq/kg) of ^{226}Ra in Malaysia's sand (abandoned mine sand and river sand) is higher than the world average as reported by UNSCEAR [5]. However, Yasir *et al*. [7] reported that the activity concentration of ^{226}Ra, ^{232}Th in abandoned mine sand are lower than the world average, except for ^{40}K which is higher than the world average value in soil. Studies on natural radioactivity concentration in sand also have been carried out by other researchers (shows the potentials of radiological hazard to dwellers although the activity concentration found were low compared to the world average [8-10]. Thus, the study of distribution of natural radionuclide in Malaysia's sand is important since the limited data have prevented the local authority board to control the building materials industry adhere to the national standards and guidelines besides to achieve 'As Low As Reasonable Achievable' (ALARA) principle. Thus, the aims of this study are to determine the activity concentrations of ^{226}Ra (^{238}U series), ^{232}Th and ^{40}K in and associated radiological hazard in Malaysia's sand used in building construction.

Method

Sampling and sample preparation

Samples were obtained from 19 different locations in four regions in Peninsular of Malaysia and were prepared in three replicated as proposed method by IAEA Technical Report 295 [11]. Samples were cleaned and grounded into powder form using a Retch PM100 planetary ball mill grinder machine. Samples then were dried in the oven for 48 hours at 105^{o}C, until a constant weight was achieved. Samples were left to cool at room temperature before being sieved with a 500μm sieve and packed into air tight acrylic counting bottle. Prior to counting, samples were kept for 30 days to attain secular equilibrium with the parents radionuclide (^{238}U and ^{232}Th) and their progeny. In secular equilibrium, both parent and progeny activity are equal, and both decay with the half-life of the parent nuclide [12].

Natural radioactivity analysis

Natural radioactivities in these samples were determined for 12 hours counting times using a gamma spectrometry system with a High-purity Germanium Detector (HPGe) coupled to a Personal Computer Analyzer (PCA) using APEX software. The HPGe detector was enclosed inside CANBERRA 747 shielding with 10 cm thickness lead coated with 1 mm and 1.6 mm of tin and copper respectively to reduce background radiation from building and cosmic rays. Energy resolution for the 1332.5 keV energy photopeak was 1.80 keV, while the relative efficiency of the detector was 30%. The gamma spectrometry system was calibrated using a mixture of ^{22}Na, ^{51}Cr, ^{57}Co, ^{60}Co, ^{85}Sr, ^{88}Y, ^{109}Cd, ^{113}Sn, ^{137}Cs, ^{123m}Te and ^{241}Am radionuclides. Activity concentration of ^{226}Ra (^{238}U series), ^{232}Th and ^{40}K were determine through the 1764.5 keV (^{214}Bi), 2614.5 keV (^{208}Tl) and 1460.3 keV energy photopeaks respectively [7,13].

Result and Discussion

Determination of radionuclide concentration

Radionuclide concentration was determined using the following equation:

$$W_s = \frac{M_{rm} \times A_s}{M_s \times A_{rm}} \times W_{rm}$$

where W_s and W_{rm} are the radionuclide concentration in the sample and reference material respectively in Bq/kg, M_s and M_{rm} are the sample and reference material mass in grams (g) while A_s and A_{rm} are the activities in cps for sample and reference material. In this study, IAEA Soil-375 was used as the reference material. The ranges of natural radioactivity concentrations in the studied samples were shown in Table 1. Activity concentration of ^{226}Ra (^{238}U series), ^{232}Th and ^{40}K were found to be in the range of 6.45 to 107.90 Bq/kg, 7.78 to 96.67 Bq/kg and 31.05 to 1105.53 Bq/kg respectively. Sand samples from Machang displayed the highest mean concentration for ^{226}Ra (^{238}U series), ^{232}Th and ^{40}K at 103.00 ± 6.05 Bq/kg, 94.56 ± 1.83 Bq/kg and 1100.54 ± 4.36 Bq/kg respectively which is much higher than the world average concentration (35 Bq/kg for ^{226}Ra, 30 Bq/kg for ^{232}Th and 400 Bq/kg for ^{40}K) reported by UNSCEAR [5]. The studied location in Machang used the fine structure of quarry dust (granite) and used as replacement for ordinary sand in building construction. Several studied on granites have proof that high concentration of natural radioactivity usually found in granite. Pavlidou *et al*. [14] reported that the natural radioactivity concentration of granite in Greece were higher than the world average reported by UNSCEAR [5] while Michalis *et al*. [15] also reported similar findings in Cyprus. Besides that, two

Table 1: Natural radioactivity concentration (Bq kg^{-1}) in studied sand samples

State	**Location**	**Sample ID**		**Radioactivity concentration (Bq kg^{-1})**		
				^{238}U	**^{232}Th**	**^{40}K**
Selangor	Banting A	1	Range	7.95 - 9.29	9.40 - 11.49	31.05 - 42.56
			Mean	9.20 ± 0.71	10.35 ± 1.13	36.89 ± 5.76
	Banting B	2	Range	8.06 - 9.67	9.53 - 9.79	310.12 - 323.40
			Mean	9.51 ± 0.87	9.63 ± 0.14	315.88 ± 6.81
	Sepang A	3	Range	9.22 - 11.96	12.46 - 13.48	372.99 - 375.42
			Mean	10.57 ± 1.56	12.48 ± 0.56	376.18 ± 1.28
	Sepang B	4	Range	10.36 - 10.34	11.29 - 12.24	329.24 - 348.88
			Mean	10.44 ± 0.39	11.31 ± 0.52	340.27 ± 9.86
	Dengkil	5	Range	9.23 - 10.54	11.41 - 12.51	338.24 - 343.53
			Mean	9.24 ± 0.75	12.47 ± 0.62	338.74 - 2.72
	Puchong	6	Range	13.44 - 16.16	17.99 - 19.13	334.11 - 350.99
			Mean	14.81 ± 1.36	18.03 ± 0.64	340.17 ± 8.50
	Serendah	7	Range	14.78 - 16.09	10.56 - 11.76	163.32 - 172.43
			Mean	14.82 ± 0.72	11.67 ± 0.66	167.50 ± 5.16
Kedah	Bukit Sinambang	8	Range	19.41 - 21.16	19.26 - 20.23	652.96 - 674.27
			Mean	19.64 ± 0.90	20.23 ± 0.51	660.52 ± 10.79
	Sungai Limau	9	Range	37.87 - 40.72	26.48 - 27.99	645.84 - 660.61
			Mean	39.67 ± 1.56	27.42 ± 0.82	653.68 ± 7.43
	Sungai Pau	10	Range	11.91 - 13.17	16.59 - 17.63	543.78 - 551.99
			Mean	13.19 ± 0.72	16.62 ± 0.58	544.82 ± 4.68
	Alor Setar	11	Range	14.41 - 15.74	13.43 - 14.51	509.62 - 538.20
			Mean	14.44 ± 0.75	14.47 ± 0.61	526.33 ± 14.34
	Padang Serai	12	Range	63.86 - 72.99	20.29 - 21.72	95.71 - 96.55
			Mean	68.77 ± 4.60	21.23 ± 0.81	96.08 ± 0.42
Perlis	Sanglang	13	Range	6.45 - 6.72	8.22 - 9.15	284.76 - 298.05
			Mean	6.56 ± 0.14	8.27 ± 0.48	294.76 ± 7.24
	Bukit Kedak	14	Range	13.03 - 14.40	13.34 - 15.43	511.95 - 518.56
			Mean	14.36 ± 0.78	14.40 ± 1.05	513.19 ± 3.35
	Beseri	15	Range	10.52 - 13.17	11.34 - 11.44	511.56 - 524.38
			Mean	11.83 ± 0.76	11.39 ± 0.05	516.72 - 6.75
Kelantan	Machang	16	Range	96.24 - 107.90	93.44 - 96.67	1097.45 - 1105.53
			Mean	103.00 ± 6.05	94.56 ± 1.83	1100.54 ± 4.36
	Pasir Putih	17	Range	20.38 - 23.31	20.25 - 23.38	667.35 - 695.15
			Mean	21.37 - 1.68	21.81 ± 1.57	683.77 ± 14.57
	Tanah Merah	18	Range	32.04 - 35.01	43.63 - 49.84	671.87 - 691.46
			Mean	33.04 ± 1.71	46.75 ± 3.10	681.61 ± 9.79
	Gua Musang	19	Range	20.41 - 23.33	7.78 - 9.35	172.19 - 175.59
			Mean	22.36 ± 1.68	8.32 ± 0.90	173.34 ± 1.95

studied samples from Sungai Limau and Padang Serai also registered higher concentration of ^{226}Ra (^{238}U series) compared to the world average while high concentration of ^{232}Th were found in Tanah Merah sands. The concentration of ^{40}K also found to be higher than the world average, which is found in Bukit Sinambang, Sungai Limau, Sungai Pau, Alor Setar, Bukit Kedak, Beseri, Pasir Putih and Tanah Merah. From these concentration values, it is possible to evaluate the representative gamma level index and the annual equivalent dose rates at 1m above the ground.

Determination of representative gamma level index ($I_{\gamma r}$)

The representative specific activities of extractive types of materials traditionally used by the building industry for ^{226}Ra (^{238}U series) and ^{232}Th is 50 Bq/kg and 500 Bq/kg for ^{40}K [16, 17]. These nuclides are of interest since they contribute to the external radiation of persons roughly according to the ratios of their specific exposure rate constant 1 : 10 : 15 [17]. The representative gamma level index ($I_{\gamma r}$) on the other hand is the quotient of the sum of three specific activities equaling to 600 Bq/kg with the denominators chosen to reflect the specific exposure rate and yields a sum equal to unity, as shown by the following equation:

$$\frac{A_{Ra}}{\left(\frac{600}{1}\right)} + \frac{A_{Th}}{\left(\frac{600}{10}\right)} + \frac{A_K}{\left(\frac{600}{15}\right)} = 1$$

where A_{Ra}, A_{Th} and A_K are refers to the concentrations of ^{226}Ra (^{238}U series), ^{232}Th and ^{40}K in Bq/kg respectively. $I_{\gamma r}$ was calculated using the following equation [17]:

$$I_{\gamma r} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} < 1$$

This equation defines the effect of radiation underestimating slightly the gamma ray exposure [17]. The $I_{\gamma r}$ was used to estimate the gamma ray hazard caused by natural radionuclide in building materials [7]. The calculated values of $I_{\gamma r}$ were shown in Table 2. The mean ranges of Iγr in the studied samples were in the range of 0.189 ± 0.027 to 2.366 ± 0.057. Although some studied samples displayed high concentration of natural radioactivity, only two samples from Machang and Tanah Merah (samples ID 16 and 18 respectively) were exceed the values of one which is the limited values permitted by NEA-OECD [17] as presented in Fig. 1.

Indoor annual equivalent dose

External exposure to gamma rays from natural radioactive elements occurs outdoors and indoors. The equivalent dose rate at 1 m above the ground (in nSv/hour) was calculated using Monte Carlo method as proposed by UNSCEAR [5]:

$$D = 0.7\left(0.462 A_{Ra} + 0.604 A_{Th} + 0.0417 A_K\right)$$

where A_{Ra}, A_{Th} and A_K are refers to the concentrations of ^{226}Ra (^{238}U series), ^{232}Th and ^{40}K in Bq/kg respectively. In the present work, the annual effective dose rates (obtained from measurements of ^{40}K, ^{232}Th and ^{226}Ra concentrations of each sand sample) were evaluated using simple calculations and the results can be used as reference values. For evaluation of the annual equivalent dose, it is important to take into account the occupancy factor, i.e. how long people spend at a particular location. UNSCEAR [18] report has proposed that the indoor occupancy factor is 0.8 on average around the world. By considering that people in Malaysia spend 80% of their times indoor within the 8760 hours of the annually accumulative time, the annual equivalent dose (mSv/year) was calculated using the following equation:

$$D_{(Indoor)} = D \times 8760 \text{ hours} \times 0.8$$

The calculated mean values of annual equivalent dose were shown in Table 2. The annual indoor equivalent dose rates in the samples were found to be in the range of 0.059 ± 0.005 mSv/year to 0.738 ± 0.018 mSv/year. The distribution of mean values of annual effective dose rates for the 19 samples analyzed in this work was presented in Fig. 2. From these results it can be observed that for some specific site (Machang), the values of the annual equivalent dose rates exceed the average worldwide exposure of 0.48 mSv year^{-1} due to natural sources [5]. However, the annual equivalent dose rates in the studied samples were still lower than the recommended limit of 1.5 mSv/year for building materials as suggested by NEA-OECD [17].

Table 2: Mean gamma level index and annual equivalent dose calculated from studied sand samples

State	**Location**	**Sample ID**	**Representative gamma level index ($I\gamma r$)**	**Annual equivalent dose (mSv year^{-1})**
Selangor	Banting A	1	0.189 ± 0.027	0.059 ± 0.005
	Banting B	2	0.370 ± 0.003	0.115 ± 0.001
	Sepang A	3	0.446 ± 0.005	0.138 ± 0.002
	Sepang B	4	0.410 ± 0.009	0.127 ± 0.003
	Dengkil	5	0.412 ± 0.011	0.127 ± 0.004
	Puchong	6	0.506 ± 0.016	0.157 ± 0.005
	Serendah	7	0.327 ± 0.001	0.102 ± 0.001
Kedah	Bukit Sinambang	8	0.774 ± 0.009	0.240 ± 0.003
	Sungai Limau	9	0.974 ± 0.014	0.305 ± 0.005
	Sungai Pau	10	0.617 ± 0.007	0.191 ± 0.002
	Alor Setar	11	0.592 ± 0.010	0.183 ± 0.003
	Padang Serai	12	0.735 ± 0.025	0.238 ± 0.009
Perlis	Sanglang	13	0.323 ± 0.002	0.100 ± 0.000
	Bukit Kedak	14	0.582 ± 0.017	0.180 ± 0.005
	Beseri	15	0.537 ± 0.009	0.166 ± 0.003
Kelantan	Machang	16	2.366 ± 0.057	0.738 ± 0.018
	Pasir Putih	17	0.816 ± 0.016	0.253 ± 0.005
	Tanah Merah	18	1.142 ± 0.035	0.353 ± 0.010
	Gua Musang	19	0.348 ± 0.003	0.111 ± 0.001

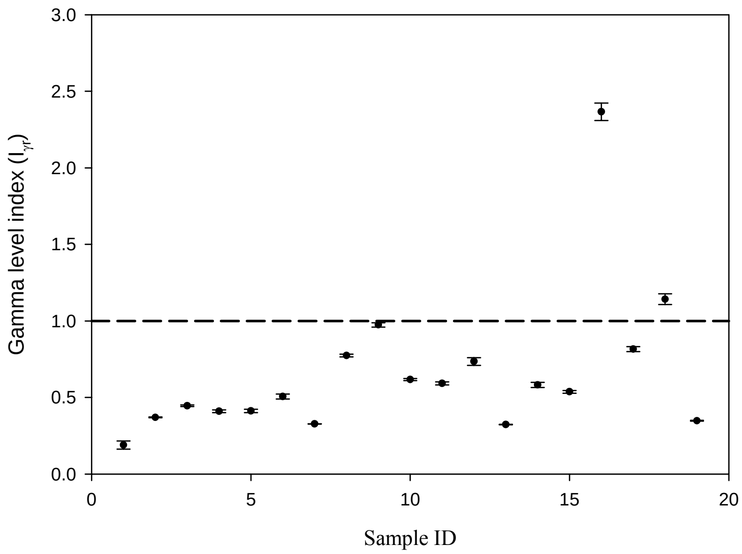

Fig. 1 The representative gamma level indexes analyzed in this work. The dashed line represents the maximum value of one as suggested by NEA-OECD (1979).

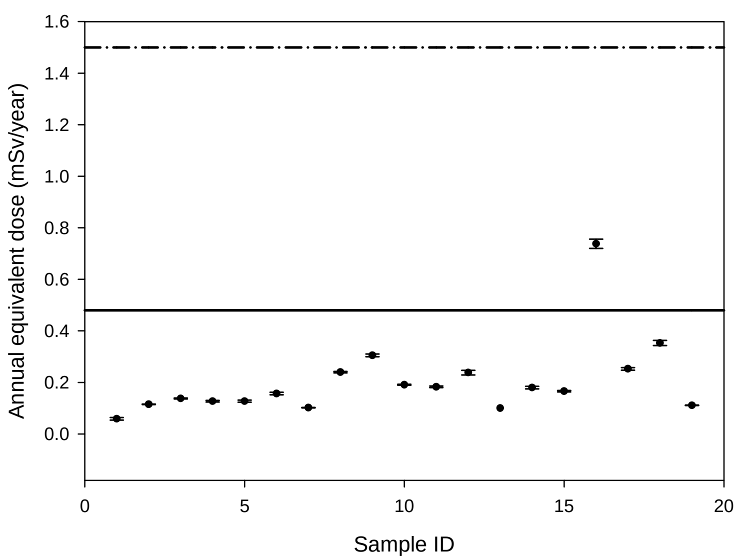

Fig. 2 The calculated annual equivalent dose in sand samples. The dotted line represents 1.5 mSv year^{-1} as limited dose to dwellers suggested by NEA-OECD (1979) while the solid line represents 0.48 mSv year^{-1} as world average of annual equivalent dose reported by UNSCEAR (2000).

Conclusion

The concentration of natural radioactivity and the associated radiation hazard levels in sand were evaluated for the 19 different locations in some state in Peninsular of Malaysia's. The mean concentration for ^{226}Ra (^{238}U series), ^{232}Th and ^{40}K were found in the range of 6.45 to 107.90 Bq/kg, 7.78 to 96.67 Bq/kg and 31.05 to 1105.53 Bq/kg respectively. The mean range of $I_{\gamma r}$ were found to be in the range of 0.189 ± 0.027 to 2.366 ± 0.057 while the annual equivalent dose were in the range of 0.059 ± 0.005 mSv/year to 0.738 ± 0.018 mSv/year. As a conclusion, the $I_{\gamma r}$ and annual equivalent dose obtained from all studied samples are lower than the recommended limit although high concentrations of natural radioactivity have been detected in some samples. Thus, this information is an important alert for local authority board to advice the local people to avoid the use of sand with high concentration of natural radioactivity in building construction.

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