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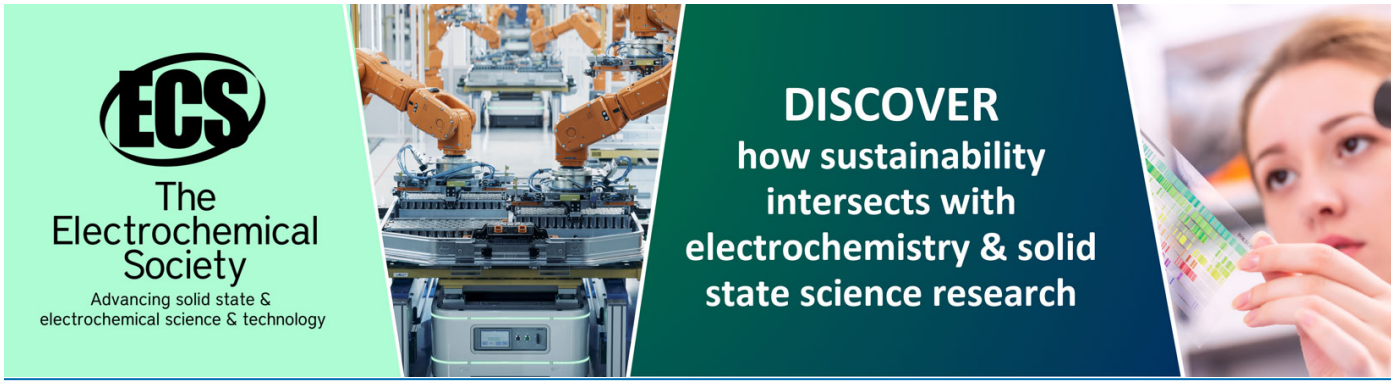

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An assessment of absorbed dose and radiation hazard index from soil around repository facility at Bukit Kledang, Perak, Malaysia

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Abstract. The process of natural decay of radionuclides that emit gamma rays can infect humans and other living things. In this study, soil samples were taken at various locations which have been identified around the Long Term Storage Facility (LTSF) in Bukit Kledang, Perak. In addition, the respective dose rates in the sampling sites were measured at 5cm and 1m above the ground using a survey meter with Geiger Muller (GM) detector. Soil samples were taken using a hand Auger and then brought back to the laboratory for sample pre-preparation process. The measuring of radioactivity concentration in soil samples were carried out using gamma spectrometer counting system equipped with HPGe detector. The obtained results show, the radioactivity concentration ranged from 11.98 – 29.93 Bq/kg for Radium-226 (^{226}Ra), 20.97 – 41.45 Bq/kg for Thorium-232 (^{232}Th) and 5.73 – 59.41 Bq/kg for Potassium-40 (^{40}K), with mean values of 20.83 ± 5.88 Bq/kg, 32.87 ± 5.88 Bq/kg and 21.50 ± 2.79 Bq/kg, respectively. To assess the radiological hazards of natural radioactivity, radium equivalent activity (R_{aeq}), the rate of absorption dose (D), the annual effective dose and external hazard index (H_{ex}) was calculated and compared to the world average values.

1. Introduction

The human environment in this world is exposed to radiation generated from various sources including cosmic rays; natural radionuclides in water, soil and plants; man-made radioactivity from nuclear tests; and applications in medicine. Gamma radiation from natural radionuclides and cosmic radiation provides external exposure while gamma radiation derived from inhalation and intake through drinks and food provides internal exposure to humans. In 1996, the IAEA estimated that 80% of the dose received was from natural radionuclides while the remaining 20% was from cosmic radiation and nuclear processing [1]. The naturally occurring radioactive element in rock, soil and water exists in different concentrations [2]. Natural radionuclide concentrations are affected by various activities such as mining; extraction and purification of uranium minerals; phosphate and phosphate fertilizer production; charcoal burning; and oil and gas production [3].

Most of the radionuclides present in the environment are primordial radionuclides, ^{238}U, ^{232}Th, ^{40}K and their decay products [4]. This natural radionuclide contributes to external exposure risk due to the release of radon (gas) radiation and its decaying product that releases alpha radiation. Natural environmental radioactivity and the associated external exposure due to gamma radiation depend

primarily on the geological and geographical conditions, and appear at different levels in the soils of each region in the world [5, 6]. Therefore the assessment of gamma radiation dose from natural sources is of particular importance as natural radiation is the largest contributor to the external dose of the world population. Knowledge of specific activity or concentration and radionuclide distribution, particularly in the soil, is an interesting thing because it provides useful information in monitoring environmental radioactivity.

The aims of this study are to determine the level of natural radionuclide concentration (^{232}Th, ^{40}K and ^{226}Ra) in the soil around the area near the repository facility and to determine the radiation hazard index in the vicinity of the repository facility by calculating radium equivalent activity (Ra_{eq}), the rate of absorption dose (D), the annual effective dose and external hazard index (H_{ex}) [7].

2. Experimental Procedure

2.1. Sampling Area

A total of 12 soil samples were collected consisting 10 samples from around the facility and 2 samples from selected residential area near the repository facility in Bukit Kledang, Perak. Originally, this facility was the Long Term Storage Facility (LTSF), which was upgraded once the decontamination and disposal process (D&D) was completed. This area is the radioactive waste storage site for the Asian Rare Earth Factory (ARE). Thorium waste and contaminated material that may contain some minor amounts of thorium hydroxide were disposed in this facility. The sampling sites are shown in figure 1 and figure 2.

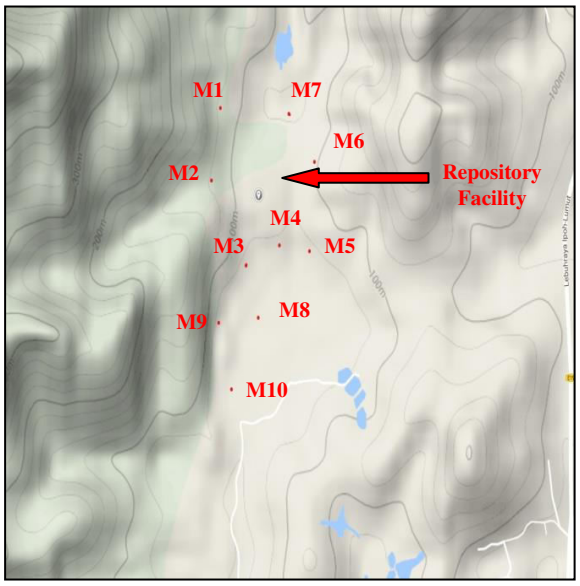

Figure 1. Location of sampling station around the repository facility.

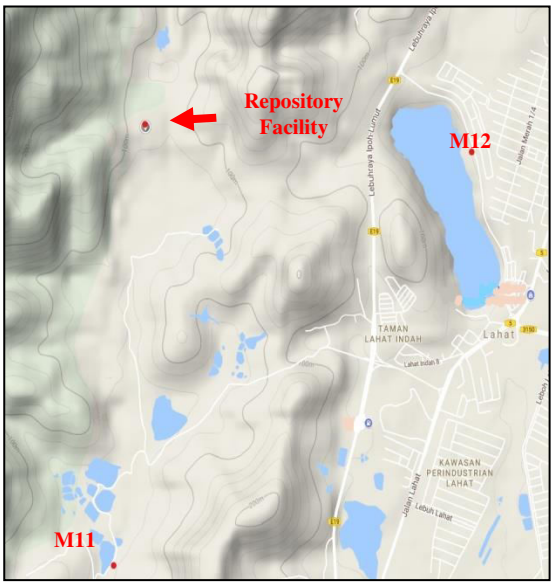

Figure 2. Location of sampling station near the residential area.

* Source: Google Map 2017.

2.2 Samples Collection and Preparation

Soil samples were collected from different locations around the repository facility. Dose monitoring was carried out using survey meter with Geiger Muller (GM) detector (µSv/h) and Sodium Iodide (NaI) detector (kcps converted to mR/h and converted to µSv/h, using conversion factor unit = 0.009335) to detect alpha, beta and gamma. In this study, the measurement had been done according to

a specified distance of 5 cm and 1 m above the ground. All the samples were collected at a depth of 0-10 cm from the soil-surface using hand Auger, then were carried in dried cleaned polyethylene bags with sample codes and transferred to the laboratory. After removing the stones and vegetation, all soil samples were dried in the oven at 85^0C, sieved (500 micron), placed in 250g marinelli container and left for four weeks to reach radioactive equilibrium.

2.3. Samples Analysis

Three Marinelli beakers were prepared for measuring the average natural radioactivity. The preparation of soil samples was made according to the procedures proposed by Johar & Embong [8]. A same amount of CRM IARMA-001 (IARMA Limited, United Kingdom) prepared in the same manner was used as standard reference material. The measurements were made using a gamma ray HPGe counting system calibrated using ^{57}Co, ^{137}Cs, ^{133}Ba, ^{85}Sr, ^{54}Mn, ^{88}Y and ^{65}Zn for two hours. The spectra were analyzed using the GENIE 2000 program. The counting time interval was 43200 seconds. The activities of ^{40}K were determined from its 1460 keV γ-line, while the specific activity of ^{226}Ra will be determined based on the average concentration of the ^{214}Pb at 352 keV and ^{214}Bi at 609 keV. As for ^{232}Th, the concentrations will be determined from the average concentrations of ^{208}Tl at 583 keV and ^{228}Ac at 911.21 keV [9]. The background spectrum was recorded immediately after or before the sample counting. The recommended values of ^{226}Ra, ^{232}Th and ^{40}K concentrations contained in the CRM IARMA-001 were 44.1, 40.0 and 440 Bq/kg, respectively, at confidence intervals of 40.7 – 47.5, 33.8 – 46.2 and 410 – 470 Bq/kg, respectively. The measurements were carried out using facilities in Malaysian Nuclear Agency, Bangi.

2.4. Activity Concentration Calculation

The activity concentration of individual nuclides in the soil samples were calculated using the following equation 1 [10]:

$$A = \frac{N_{net}}{I_\gamma \varepsilon m t} \tag{1}$$

where,

A : calculated gamma activity (Bq/kg),

N_{net} : net peak area count subtract background of the sample (counts),

ε : absolute efficiency of the detector,

I_γ : emission probability of a specific energy photo peak,

t : sample count time (sec),

m : sample weight/volume.

This calculation was done by assuming that all correction factors are equal to unity. The distribution of the activity mass concentrations due to the radionuclides ^{226}Ra, ^{232}Th and ^{40}K is not uniform throughout the soils. The incompatibility with radiation exposure has been defined in terms of radium equivalent activity (Ra_{eq}) used to represent the specific activity value of the three radionuclides by only single quantity. It is calculated using the following equation 2 [11][12]:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.07C_K \tag{2}$$

where,

Ra_{eq} : radium equivalent activity in Bq/kg,

C_{Th} : ^{232}Th activity concentration in Bq/kg,

C_{Ra} : ^{226}Ra activity concentration in Bq/kg,

C_K : ^{40}K activity concentration in Bq/kg.

While defining Ra_{eq} activity, it is assumed that 10 Bq/kg of ^{226}Ra, 7Bq/kg of ^{232}Th and 130 Bq/kg of ^{40}K produce equal gamma ray dose [13]. According to [12], the maximum value of this parameter should not exceed 370 Bq/kg in order to limit the annual exposure 1.5 mGy.

From previous studies [14,15], the external gamma absorbed dose rate (D) in the air at 1 m above ground level was calculated using the factor 0.042 nGy/h per Bq/kg for ^{40}K, 0.462 nGy/h per Bq/kg for ^{226}Ra and 0.604 nGy/h per Bq/kg for ^{232}Th. The calculations were performed according to the following equation 3 [7]:

$$D = 0.462\mathrm{C}_{\mathrm{Ra}} + 0.604\mathrm{C}_{\mathrm{Th}} + 0.042\mathrm{C}_{\mathrm{K}} \tag{3}$$

where,

D: External gamma absorbed dose rate in nGy/h,
C_{Th} : ^{232}Th activity concentration in Bq/kg,
C_{Ra} : ^{226}Ra activity concentration in Bq/kg,
C_{K} : ^{40}K activity concentration in Bq/kg.

To calculate the effective dose rate, the conversion coefficient for the dose rate absorbed to the effective dose rate and external occupancy factor should be taken into account. The absorbed dose rate found to be converted into effective dose rate using 0.7 Sv/Gy conversion factor recommended by UNSCEAR (2000) and external occupancy factor of 0.2 assuming that the man spends 20% of their time outdoors [11]. The annual effective doses are calculated using the following equation 4:

$$\text{Annual effective dose (Sv)} = D \times 8760 \times 0.7 \times 0.2 \tag{4}$$

where,

D: External gamma absorbed dose rate in nGy/h,
0.7: conversion factor in Sv/Gy,
8760: time in a year (hour),
0.2: external occupancy factor.

External hazard is an index hazard widely used in the study as it represents external exposure to humans. The value of this index must be less than unity (unity value = 1) in order to keep the radiation hazard insignificant. The maximum value of H_{ex} equal to unity corresponds to the upper limit of radium equivalent activity (370 Bq/kg) [11]. The external hazard indexs, H_{ex}, are calculated using the following equation 5 [7]:

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \tag{5}$$

where,

C_{Ra} : ^{226}Ra activity concentration in Bq/kg,
C_K : ^{40}K activity concentration in Bq/kg,
C_{Th} : ^{232}Th activity concentration in Bq/kg.

3. Results And Discussions

The results of measurement for 12 soil samples collected at different locations around the repository facility are summarised in table 1. The three most common primordial radionuclides determined in the study areas were ^{226}Ra, ^{232}Th and ^{40}K. The natural activity of ^{226}Ra and ^{232}Th of radionuclides is obtained from the most abundant photopeaks activity of uranium and thorium series decay products, while the concentration of ^{40}K natural activity is a single peak decision [13].

Table 1. Specific activities of radionuclides in soil samples at different locations around repository facility.

Samples Location	GPS Coordinates		Specific activity of radionuclides (Bq/kg)		
	Latitude (DMS)	Longitude (DMS)	^{226}Ra	^{232}Th	^{40}K
M1	N 04° 33.338'	E 101° 00.790'	17.87 ± 3.12	34.96 ± 4.66	7.59 ± 0.73
M2	N 04° 33.195'	E 101° 00.755'	16.30 ± 3.07	36.41 ± 6.54	6.51 ± 0.60
M3	N 04° 32.999'	E 101° 00.838'	20.63 ± 3.81	30.14 ± 6.52	27.95 ± 5.49
M4	N 04° 33.294'	E 101° 00.554'	22.14 ± 4.00	41.43 ± 6.80	9.30 ± 0.70
M5	N 04° 33.044'	E 101° 00.965'	25.49 ± 4.29	35.48 ± 7.25	26.96 ± 5.50
M6	N 04° 33.154'	E 101° 00.580'	**29.92 ± 5.81***	38.71 ± 8.14	**59.40 ± 1.15***
M7	N 04° 33.284'	E 101° 00.961'	24.90 ± 4.56	33.91 ± 7.00	50.84 ± 8.54
M8	N 04° 32.538'	E 101° 00.525'	17.30 ± 3.15	**41.44 ± 6.69***	10.55 ± 2.29
M9	N 04° 32.848'	E 101° 00.767'	**11.98 ± 2.20***	25.10 ± 5.52	**5.73 ± 0.50***
M10	N 04° 32.442'	E 101° 00.462'	20.80 ± 3.98	**20.97 ± 4.45***	18.33 ± 4.03
M11	N 04° 31.468'	E 101° 00.598'	21.55 ± 3.97	33.70 ± 6.92	22.31 ± 4.33
M12	N 04° 32.526'	E 101° 02.144'	12.07 ± 2.10	22.12 ± 4.18	12.47 ± 2.24
Range			11.98 – 29.92	20.97 – 41.44	5.73 – 59.407
Mean			20.83 ± 3.59	32.870 ± 5.88	21.49 ± 2.79

* The bold characters represent the minimum and maximum values.

Figure 3 shows the specific activities of ^{226}Ra was ranged from 11.98 – 29.92 Bq/kg, ^{232}Th was ranged from 20.97 – 41.44 Bq/kg and ^{40}K was ranged from 5.73 – 59.40 Bq/kg, with a mean value for ^{226}Ra, ^{232}Th and ^{40}K were 20.83 ± 3.59 Bq/kg, 32.87 ± 5.88 Bq/kg and 21.49 ± 2.79 Bq/kg, respectively. The highest ^{226}Ra and ^{40}K concentrations were recorded at M6 with a value of 29.92 ± 5.81 Bq/kg and 59.40 ± 1.15 Bq/kg, respectively, while the lowest concentrations for both radionuclides were recorded at M9 with a value of 11.98 ± 2.20 Bq/kg for ^{226} Ra and 5.73 ± 0.50 Bq/kg for ^{40}K. As for ^{232}Th, the highest and the lowest concentration were recorded at M8 and M10 with a value of 41.44 ± 6.69 Bq/kg and 20.97 ± 4.45 Bq/kg, respectively.

Hussain [16] reported that the ratio of ^{232}Th to ^{226}Ra in the earth's crust is generally greater than one. This statement supports the results of this study, where the mean activity value of ^{232}Th was higher than ^{226}Ra. The specific activity concentrations show varying values and vary according to location likely to be affected by several factors such as soil type and soil pH [13], latitude of area, soil texture, existence of underground rock and organic matter.

As explained earlier, this study focuses only on the surrounding area and near repository facility area. The results gained from this study only represent the area of the study. A comparison of a mean value of the specific activity of the radionuclides in soil samples obtained from this study with other location in Peninsular Malaysia and other countries are shown in table 2. The mean activity concentrations of ^{226}Ra and ^{232}Th were comparable with worldwide mean data [7]. However, as for ^{40}K, the value was lower (21 Bq/kg) compared to worldwide mean value (474 Bq/kg). It is also found that the average value of the three radionuclides is lower than the average value recorded by previous studies [17][18][19] for certain areas in Peninsular Malaysia.

The values obtained for radium equivalent activity, gamma dose rate, annual effective dose and external hazard index are presented in table 3.

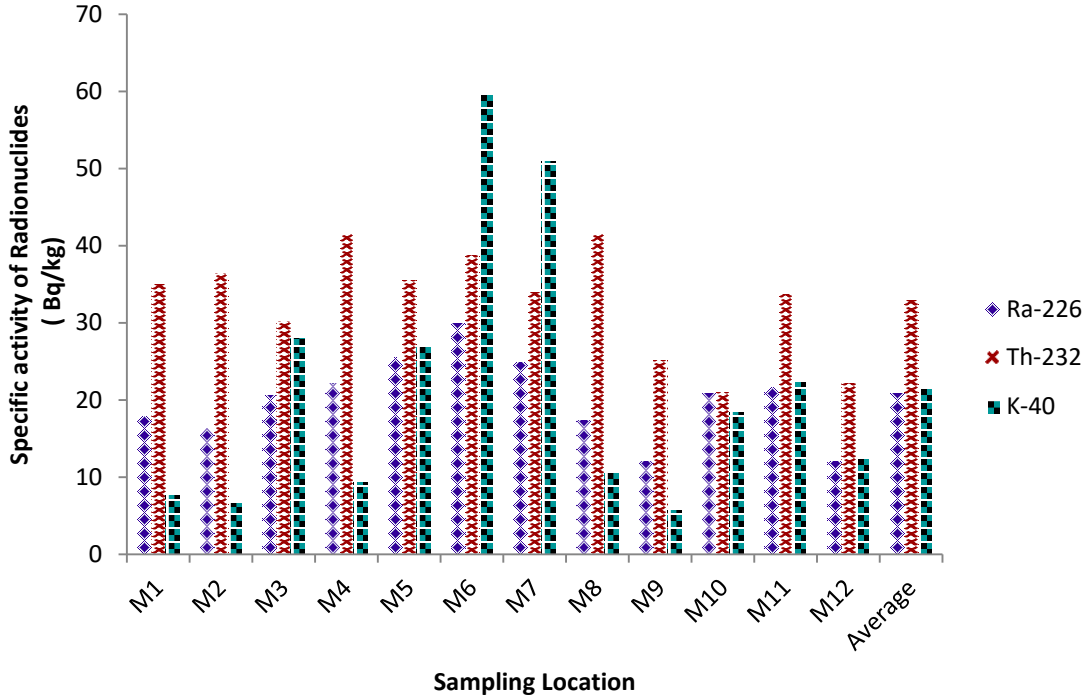

Figure 3. Specific activity concentration for ^{226}Ra, ^{232}Th and ^{40}K in soil samples

Table 2. Comparison of mean specific activities of radionuclides in soil samples at around the repository area with other location in Peninsular Malaysia and other countries [7][17][18][19].

Samples	**Specific activity of radionuclides (Bq/kg)**			**Reference**
	^{226}Ra	**^{232}Th**	**^{40}K**	
Repository Facility, Bukit Kledang, Perak.	**21**	**33**	21	**Present study**
Kinta District, Perak	**-**	246	-	Lee *et al.* [17]
Pulau Pinang	64 - 799	16 - 667	-	Almayahi *et al.* [18]
Dengkil, Selangor	-	27 - 103	-	Yasir, S. M. *et al.* [19]
Malaysia	67	82	310	UNSCEAR Report, 2000 [7]
Bangladesh	34	-	350	
Thailand	48	40	400	
Japan	33	28	310	
China	32	41	440	
India	29	64	400	
Egypt	17	18	320	
Iran	28	22	640	
Belgium	26	27	380	
Denmark	17	19	460	
Switzerland	40	25	370	
Spain	32	33	470	
Argentina	-	-	650	
United States	40	35	370	
Worldwide mean	33	36	474	

3.1. Radium Equivalent Activity (Ra_{eq})

Referring to table 3, the radium equivalent activity value for M6 (89.87 Bq/kg) is the highest among other sampling locations. Though, this value is lower than the standard set value of 370 Bq/kg. According to The Organisation of Economic Cooperation and Development (OECD) (1998) [20], the value of 370 Bq/kg is the ideal value for exposure to the public as it is equal to 1.5 mSv / year. If the value of the radium equilibrium index is high, it will cause exposure to the public to exceed the safe level of the public and may have a stochastic effect and the risk of illness that arises from radiation.

3.2. Dose Rate

The mean absorbed gamma dose in air was calculated as 30.03 nGy/h with a minimum and maximum values are 19.46 nGy/h and 39.69 nGy/h, respectively. It is found to be comparable to the world average of 57 nGy/h [7]. The main contributor to the dose rate absorbed in the air at a height of 1 m is from the ground gamma radiation generated from the naturally occurring radionuclide in the soil. Therefore, the dose rate absorbed depends on the concentrations of specific radionuclide activity. According to Al-Kaabi [21], if the concentration of radionuclide in the sample area is high, absorbed dose will also be high.

3.3. Annual Effective Dose

The calculated values of annual effective dose ranged from 0.02 to 0.05 mSv, with a mean value of 0.04 mSv. The highest value is come from M6 (48.68 μSv). Even though, the value of the area is high compared to other areas, but it is lower than the world average of 0.48 mSv [7]. This confirms that all of these research areas are at a safe level for the public.

3.4. External Hazard Index

The calculated values of external hazard index obtained in this study ranged from 0.12 to 0.24 with a mean value of 0.18. The highest value is come from M6 (0.24). The external hazard index represents external exposure to humans. Therefore, when the primordial radionuclide concentrations such as ^{226}Ra, ^{232}Th and ^{40}K are high in the area the external hazard index will also be high. However, since the values from this study are lower than unity, we can make a conclusion that the radiation hazard is low at all sampling location.

Table 3. Radium equivalent activity, dose rate, annual effective dose and external hazard index at different locations around repository facility

Samples Location	Radium equivalent activity (Bq/kg)	Dose rate (nGy/h)	Annual effective dose (10^{-6} Sv)	External hazard index
M1	68.47	29.70	36.42	0.18
M2	68.89	29.80	36.55	0.19
M3	65.89	28.91	35.45	0.18
M4	82.11	35.65	43.72	0.22
M5	78.32	34.34	42.11	0.21
M6	89.87	39.69	48.68	0.24
M7	77.31	34.11	41.83	0.21
M8	77.39	33.47	41.05	0.21
M9	48.33	20.94	26.68	0.13
M10	52.21	23.04	28.26	0.14
M11	71.46	31.24	38.32	0.19
M12	44.70	19.46	23.87	0.12
Range	44.70 – 89.87	19.46 – 39.69	23.87 – 48.68	0.12 – 0.24
Mean	68.74	30.03	36.91	0.18

4. Conclusion

Soil samples collected at different locations around the repository facility area were analysed for radioactivity due to ^{226}Ra, ^{232}Th and ^{40}K isotopes. The mean activity concentration of ^{226}Ra and ^{232}Th were comparable with worldwide mean data, however, as for ^{40}K, the value was lower (21 Bq/kg) compare to worldwide mean value (474 Bq/kg). It is also found that the average value of the three radionuclides is lower than the average value recorded by previous studies for certain areas in Peninsular Malaysia. The average value of gamma dose rate obtained in this study (30.03 nGy/h) is comparable to the world average (57 nGy/h). The calculated annual effective dose with average value 0.04 mSv is lower than the worldwide average value. All values obtained for radium equivalent activity are less than 370 Bq/kg, which are acceptable for safe use OECD 1998. The calculated values of external hazard index obtained in this study range from 0.12 to 0.24 with a mean value of 0.18 (all calculated values are lower than unity), which means that the radiation hazard is insignificant for the population living in the study area. These results can be used as an additional data to represent terrestrial radioactivity baseline data for Malaysia environment. This estimation will also serve as baseline for detection of any future related activities of contamination especially around the repository facility area.

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