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Natural radioactivity in virgin and agricultural soil and its environmental implications in Sungai Petani, Kedah, Malaysia

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ABSTRACT: A study on natural radioactivity in virgin and agricultural soil samples collected from Sungai Petani was conducted using high-purity germanium. The mean activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in virgin soils were 51.06±5.83, 78.44±6.42, and 125.66±7.26 Bq kg^{-1}, respectively, while those in agricultural soils were 80.63±5.78, 116.87±7.87, and 200.66±18.24 Bq kg^{-1}, respectively. The corresponding activity concentrations in agricultural soils were higher than those in virgin soils and those reported for other countries of the world. The average values of radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}), outdoor annual effective doses (E_{out}), and indoor annual effective doses (E_{in}) in agricultural soils were 258.38 Ba kg^{-1}, 0.708, 0.925, 0.162 mSv y^{-1}, and 0.669 mSv y^{-1}, respectively. The average values of outdoor external dose (D_{out}) and indoor absorbed dose (D_{in}) rate in agricultural soils were 116.04 and 218.46 nGy h^{-1}, respectively, which were higher than the permissible limit. Soil with H_{ex} and H_{in} less than unity are suitable for use as building materials and in agriculture.

Keywords: HPGe, natural radioactivity, radiological hazard, soil.

INTRODUCTION

Terrestrial gamma radionuclide is a significant part of the total dose in the form of natural sources. Only nuclides with half-lives comparable with the age of the earth and present in terrestrial materials such as ^{232}Th, ^{238}U, and ^{40}K are of great interest as they cause external and internal hazards because of gamma ray emissions(Ahmad et al., 2014). We currently live in a world of radionuclides, where we inhale and ingest these radioactive substances daily in the form of food, water, and air as no place on the earth is free of radioactivity (Abbady, 2005). Soil is one of the most important sources of radioactivity. It is used as a raw material in the construction of roads, buildings, landfills, and playgrounds. Numerous studies conducted worldwide have shown that ^{238}U, including its decay products in soils and rocks, and ^{232}Th in monazite sands are the main sources of high natural background area (Al-Jundi et al., 2003).

The main sources of phosphorus for fertilizers and the primary material for the production of phosphate products are phosphate rocks. Fertilizers are generally used in improving the properties of crops and in reclaiming land. Phosphate fertilizers contain elevated natural uranium and its decay products (Bolca et al., 2007). Radionuclides in phosphate rocks can enter the environment through different ways, such as usage of phosphogypsum in building materials and agriculture or fertilization of agricultural lands (Hussein,

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1994). A previous study reported that the concentration levels of natural radioactivity increase with the use of phosphate rocks, thereby enhancing radiation dose and causing diseases to the surrounding population (Righi et al., 2005). Therefore, knowledge on radionuclide distribution in rocks and soils is very essential in controlling health risks to populations. The present study was conducted to measure the concentration of natural radioactivity and assess the radiation dose from virgin and agricultural soils collected from Sungai Petani, Kedah, Malaysia. Agricultural soils were collected from palm oil, banana, and chili farms. This study aimed to compare the radioactivity levels between virgin and agricultural soil samples collected from Sungai Petani.

Sungai Petani is the capital of Kala Muda district in the state of Kedah situated in the northern part of Peninsular Malaysia. It covers an area of 925 km^2(Noresah and Ruslan, 2009) and is located at 5º 38´ 49´´ N and 100º 29´ 15´´ E. Sungai Petani is the largest town in the Kedah with a population of 443,458 in 2010. Sungai Petani was selected as the study area because of its agricultural activities and geological setting (from Silurian to Devonian) as its soil mainly consists of schist, hyllite, slate and limestone and minor intercalations of sandstone. Different types of fertilizers are used in reclaiming land and improving crop growth in the study area. Figure 1 shows the location of study area.

MATERIALS AND METHODS

Virgin and agricultural soil samples were collected from different locations of Sungai Petani, Kedah, Malaysia. Soil samples were pulverized and sifted through a 0.249mm sieve after drying in an oven for 24 h at 110°C to remove the moisture content. 1 kg of the dried sample of soil was packed in a Marinelli beaker and then stored for one month before gamma spectrometric analysis to achieve secular equilibrium between ^{226}Ra and ^{232}Th and their respective progenies (Shanbhag et al., 2005; Ramola et al., 2008). The radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in soil samples were determined using high-purity germanium spectroscopy shielded with 10 cm-thick Pb, with inner lining of Al, Cu, and Perspex. The system was connected to apre-amplifier MCA card with a built-in spectroscopy amplifier and ADC (7070) installed in a Windows-operated PC with GAMMA-W software. The background spectrum with a counting time of 36,000 s was tripped from the soil spectra. The reference material (Soil-375) obtained from the IAEA was used for efficiency calibration of the detector. The efficiency calibration curve is shown in Figure 2. The ^{226}Ra concentration was determined through the photo peaks of its daughters ^{214}Pb (351.9 keV) and ^{214}Bi (609.3, 1120.2, and 1764.5 keV) while the activity concentration of ^{232}Th was assessed through ^{212}Pb (338.0, 911.1, and 968.9 keV) and ^{208}Tl (583.1 keV). The activity concentration of ^{40}K was assessed directly from its 1460.8 KeV gamma ray peak. The efficiencies of the spectrum peaks of interest were determined from the fit peak equation obtained from the curve shown in Figure 2.

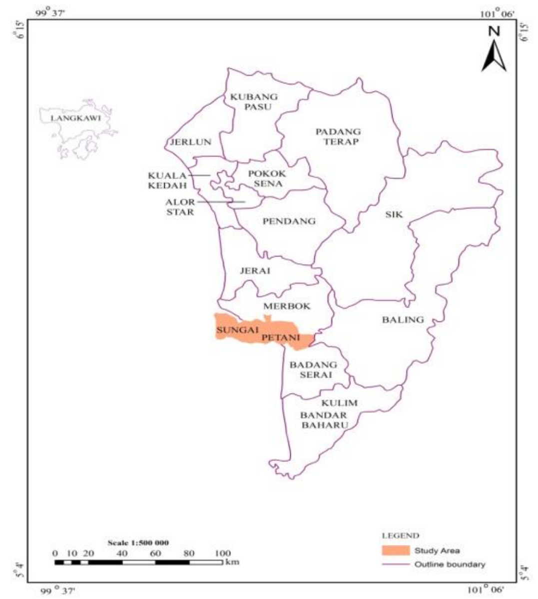

Fig. 1. Location map showing study area, Sungai Petani, Kedah, Malaysia

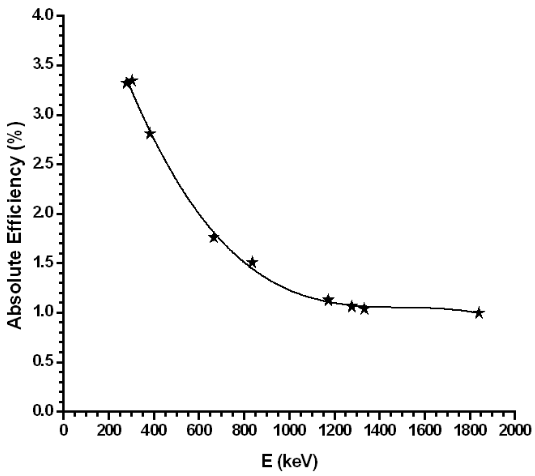

Fig. 2. Efficiency calibration curve for the HPGe detector

RESULTS AND DISCUSSION

The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K measured in virgin and agricultural soil samples from the study areas are summarized in Table 1. The activity concentration of ^{226}Ra in virgin soils varied from 27.07±12.30 Bq kg^{-1} to 83.32±12.20 Bq kg^{-1} (mean: 51.06±5.83 Bq kg^{-1}) while that in agricultural soils ranged from 37.75±2.49 to 193.23±16.56 Bq kg^{-1} (mean: 80.63±5.78 Bq kg^{-1}). The activity concentration of ^{232}Th in virgin soils ranged from 55.91±8.56 to 102.04±5.10 Bq kg^{-1} (mean: 78.44±6.42 Bq kg^{-1}) while that in agricultural soils ranged from 56.69±4.30 Bq kg^{-1} to 182.82±6.68 Bq kg^{-1} (mean: 116.87±7.87 Bq kg^{-1}). The concentration obtained for ^{40}K in virgin soils ranged from 83.66±15.87 Bq kg^{-1} to 165.50±15.05 Bq kg^{-1} (mean: 125.66±7.26 Bq kg^{-1}), while that in agricultural soils ranged from 90.10±30.66 Bq kg^{-1} to 318.68±32.73 Bq kg^{-1} (mean: 200.66±18.24 Bq kg^{-1}). The average activity concentrations of ^{226}Ra and ^{232}Th in virgin and agricultural soils measured in the present study were higher than the world average concentrations of 35 and 45 Bq kg^{-1}, respectively, except for ^{40}K, which exhibited a lower activity concentration than the world average value of 420 Bq kg^{-1}. The typical ranges for ^{226}Ra, ^{232}Th, and ^{40}K are 16–116 , 7–50 and 100–700 Bq kg^{-1}, respectively (UNSCEAR, 2000; Almayahi et al., 2012a; b). In the comparison of virgin and agriculture soils, a significant increasing trend was observed in the measured activity concentrations of agricultural soils. This variation reflected the fertilizers used for reclaiming land and improving the properties of crops in the agricultural area understudy. Phosphate rocks contain substantial amounts of ^{238}U, ^{226}Ra, ^{232}Th, and ^{226}Ra decay products (Skorovarov et al., 1996). The order of distribution of activity concentrations was ^{226}Ra< ^{232}Th< ^{40}K (Table 1). The ^{232}Th/ ^{226}Ra ratios in virgin soil ranged from 0.89 to 2.43 (mean: 1.66), whereas those in agricultural soils ranged from 0.84 to 2.91 (mean: 1.63).The ^{40}K/ ^{226}Ra ratios in virgin soils ranged from 1.33 to 5.14 (mean: 2.76), whereas those in agricultural soils ranged from 1.64 to 6.84 (mean: 2.86). The ^{40}K/^{232}Th ratios in virgin soil ranged from 1.06 to 3.39 (mean: 1.66) while those in agricultural soils ranged from 0.92 to 4.54 (mean: 1.84). The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the earth's crust vary with their half-lives (that is, the greater the half-life, the higher the activity concentration of the radionuclide). The ratios summarized in Table 1 may be used as an indicator of the relative abundance of radionuclides. Figure 3 shows the relative contribution of activities caused by radium, thorium, and potassium. ^{40}K usually has high concentrations in soil and rock samples.

The activity concentrations measured in this study were compared with those from other countries (Table 2).The ^{226}Ra activity concentration for virgin soils in our study area was higher than those in Yemen, Nigeria, China, Botswana, and Malaysia (Potain) but not those in Bangladesh, Turkey, and Malaysia (Penang), where its value in agricultural soils was higher than all countries mentioned in Table 2. The ^{232}Th activity concentration for virgin soil in the study area was higher than those in all countries, except Turkey and Malaysia (Penang) while its value in agricultural soils was higher than those in all countries. The ^{40}K activity concentration for virgin soil in our study area was lower than those in all countries while its value in agricultural soils was lower than those in all countries, except Egypt.

Table 1. Specific activity of ^{226}Ra, ^{232}Th and ^{40}K, along with ratios in soil samples collected from Sungai Petani

Sample locations	Specific activity (Bq kg^{-1})			Ratio		
	^{226}Ra	^{232}Th	^{40}K	^{232}Th/^{226}Ra	^{40}K/^{226}Ra	^{40}K/^{232}Th
Virgin soil						
Industrial Area	41.53±8.09	98.92±6.94	165.5±15.05	2.37	3.98	1.67
Kampung Kilang Makau	45.99±2.51	73.47±9.51	113.73±4.50	1.59	2.47	1.54
Kampung Kubang Sapi	62.69±7.67	55.91±8.56	83.66±15.87	0.89	1.33	1.49
Kampung Bakar Kapor	48.21±4.80	60.32±3.93	129.31±1.37	1.25	2.68	2.14
Kampung Pantai Cicak	59.77±5.07	81.28±8.33	145.55±7.51	1.35	2.43	1.79
Taman Seri Baiduri	83.32±12.20	102.04±5.10	124.47±0.44	1.22	1.49	1.21
Taman Sinar Permata	39.99±4.02	97.56±5.14	103.90±8.22	2.43	2.59	1.06
Kumpung Tanah Lincin	27.07±12.30	58.25±3.89	139.23±5.12	2.15	5.14	3.39
Average	51.06±5.83	78.44±6.42	125.66±7.26	1.66	2.76	1.66
Agricultural soil						
Kampong Pulau Tigai Layar (Palm Oil)	53.43±4.95	116.99±8.71	117.40±23.09	2.18	2.19	1.00
Kampong Bukit Lembu Sungai Lalang (Palm Oil)	37.75±2.49	56.69±4.30	152.84±13.26	1.50	4.04	2.69
Taman Seri Baya (Palm Oil)	44.07±4.37	66.46±5.96	301.89±28.07	1.50	6.84	4.54
Kampong Kelang Ketapan (Palm Oil)	40.36±2.74	65.84±4.36	90.10±30.66	1.63	2.23	1.36
Kampong Guar Stati (Palm Oil)on	50.93±3.65	73.22±5.51	98.76±28.26	1.43	1.93	1.34
Kampong Padang Setol (Palm Oil)	110.6±9.45	99.31±8.89	196.92±1.03	0.89	1.78	1.98
Taman cempaka (Palm Oil)	67.05±4.07	127.56±8.35	257.64±15.03	1.90	3.84	2.01
Kampong Ban Sungai Badak (Palm Oil)	63.70±4.10	110.66±5.40	208.73±2.23	1.73	3.27	1.88
Sungai Layar Hujang (Banana)	170.83±2.42	143.87±7.01	298.91±27.24	0.84	1.74	2.07
Kampong Pulau Tiga Sungai Layar (Banana)	59.71±3.80	173.82±12.87	226.32±7.11	2.91	3.79	1.30
Kampong Bukit Lembu Sungai Lalang (Banana)	63.34±4.07	122.32±14.83	113.68±24.12	1.93	1.80	0.92
Kampong Guar Station (Banana)	193.23±16.56	169.46±10.95	318.68±32.73	0.87	1.64	1.88
Kampong Jelatang (Banana)	72.74±5.13	127.35±6.03	172.85±7.71	1.75	2.37	1.35
Kampong Kapala Bukit (Banana)	101.56±13.16	182.40±6.68	254.54±14.94	1.80	2.50	1.39
Average	80.63±5.78	116.87±7.84	200.66±18.24	1.63	2.86	1.84

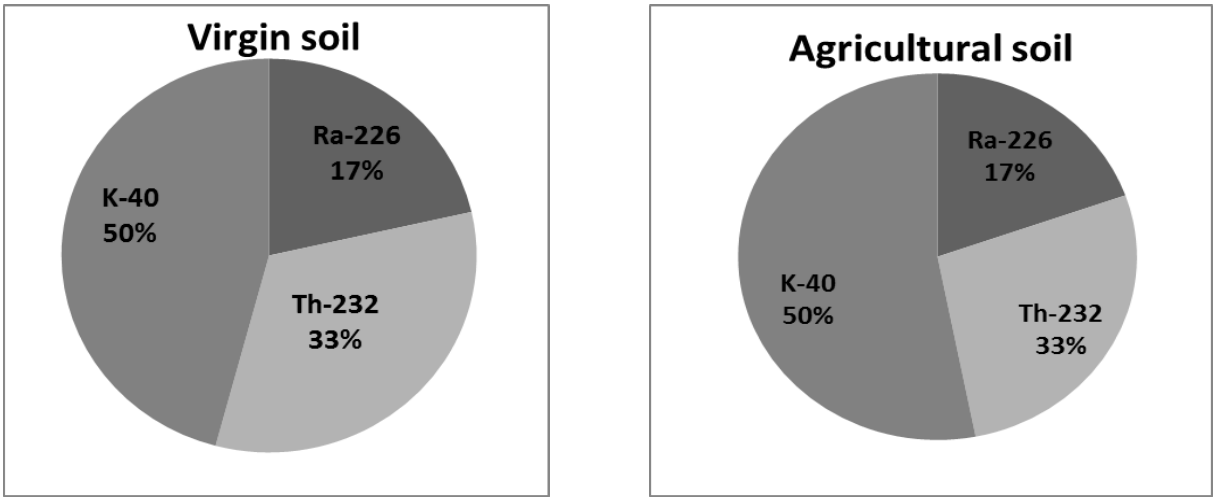

Fig. 3. Relative contribution to total activity due to ^{226}Ra, ^{232}Th and ^{40}K in virgin and agricultural soil understudy

Table 2. Comparison of radioactivity levels in soil samples collected with other countries of the world

Country	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	Reference
		Virgin soil		
Yemen	44	58	822	(Abd El-mageed et al., 2011)
Bangladesh	60.20	60.80	928.00	(Alam et al., 1999)
Nigeria	18	22	210	(Agbalagba and Onoja, 2011)
China	38	57.6	838	(Ziqiang et al., 1988)
Turkey	115	192	1207	(Merdanoğlu and Altınsoy, 2006)
Botswana	34.8	41.8	432.7	(Murty and Karunakara, 2008)
Malaysia (Penang)	396	165	835	(Almayahi et al., 2012a; Ahmad et al., 2014)
Malaysia (Pontian)	37	53	293	(Saleh et al., 2013)
Malaysia	51.06	78.44	125.66	Present study
		Agriculture soil		
Yugoslavia	39.3	53	454	(Bikit et al., 2004)
Pakistan	-	50.6-64	560.2-635.6	(Akhtar et al., 2005)
Pakistan	30	56	602	(Tufail et al., 2006)
Egypt	43	54	183	(Issa, 2013)
Algeria	53.2	50.03	311	(Boukhenfouf and Boucenna, 2011)
Malaysia	80.63	116.87	200.66	Present study

In soil and rocks, the distribution of ^{226}Ra, ^{232}Th, and ^{40}K is not uniform; therefore, a common index called radium equivalent activity (Ra_{eq}) was used to compare their combined radiological effects. Its value must be less than 370 Bq kg^{-1} for safe use, as suggested by the Organization of Economic Cooperation and Development (OECD, 1979; Almayahi et al., 2012a).

The radium equivalent activity (Ra_{eq}) was calculated using Equation (1) according to Beretka and Mathew (1985) and results are shown in Table 3.

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (1)$$

As shown in Table 3, the calculated values of Ra_{eq} for virgin soils ranged from 120.9 to 238.6 Bq kg^{-1} (mean: 172.7 Bq kg^{-1}), whereas these values for agricultural soils ranged from 130.49 to 459.50 Bq kg^{-1} (mean: 258.36 Bq kg^{-1}). The average values of Ra_{eq} for both soil samples were lower than the recommended maximum admissible limit.

Table 3. Results of hazards indices for soil of Sungai Petani

Sample locations	Ra_{eq} Bq kg^{-1}	H_{ex}	D_{out} (nGy h^{-1})	H_{in}	D_{in} (nGy h^{-1})	E_{out} mSv y^{-1}	E_{in} mSv y^{-1}
	Virgin soil						
Industrial Area	195.2	0.52	85.70	0.640	160.02	0.120	0.491
Kampung Kilang Makau	159.6	0.431	70.36	0.555	132.22	0.098	0.405
Kampung Kubang Sapi	148.9	0.402	66.22	0.572	125.86	0.092	0.386
Kampung Bakar Kapor	144.3	0.390	64.09	0.520	121.05	0.089	0.371
Kampung Pantai Cicak	187.0	0.505	82.77	0.667	156.04	0.115	0.478
Taman Seri Baiduri	238.6	0.64	105.31	0.870	198.85	0.147	0.610
Taman Sinar Permata	187.3	0.50	81.73	0.614	152.41	0.114	0.468
Kumpung Tanah Lincin	120.9	0.327	53.49	0.400	100.11	0.074	0.307
Average	172.7	0.464	76.20	0.604	143.32	0.106	0.439
`	**Agricultural soil**						
Kampong Pulau Tigai Layar (Palm Oil)	229.58	0.620	100.24	0.764	187.23	0.140	0.574
Kampong Bukit Lembu Sungai Lalang (Palm Oil)	130.49	0.352	58.05	0.454	109.31	0.081	0.335
Taman Seri Baya (Palm Oil)	162.23	0.438	73.09	0.557	137.80	0.102	0.422
Kampong Kelang Ketapan (Palm Oil)	141.34	0.382	62.17	0.491	116.76	0.087	0.358
Kampong Guar Stati (Palm Oil)on	163.12	0.440	71.87	0.578	135.29	0.100	0.415
Kampong Padang Setol (Palm Oil)	267.61	0.723	119.30	1.02	226.74	0.167	0.695
Taman cempaka (Palm Oil)	269.09	0.727	118.76	0.908	222.61	0.166	0.682
Kampong Ban Sungai Badak (Palm Oil)	237.84	0.642	104.97	0.814	197.02	0.146	0.604
Sungai Layar Hujang (Banana)	339.35	1.07	178.28	1.54	339.33	0.249	1.04
Kampong Pulau Tiga Sungai Layar (Banana)	320.43	0.86	139.70	1.01	259.64	0.195	0.796
Kampong Bukit Lembu Sungai Lalang (Banana)	246.82	0.667	107.88	0.838	201.91	0.151	0.619
Kampong Guar Station (Banana)	459.50	1.24	204.91	1.76	389.67	0.286	1.19
Kampong Jelatang (Banana)	267.96	0.724	117.73	0.920	220.83	0.164	0.677
Kampong Kapala Bukit (Banana)	381.71	1.03	167.70	1.30	314.43	0.234	0.964
Average	258.36	0.708	116.04	0.925	218.46	0.162	0.669

The external hazard index (H_{ex}) was calculated using Equation (2) (Beretka and Mathew, 1985).

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4,810} \qquad (2)$$

The calculated values of H_{ex} (Table 3) for virgin soils were between 0.327 and 0.64 (mean: 0.464), while those for agricultural soils were between 0.352 to 1.24 (mean: 0.708). Radioactivity may cause harm to the population if the calculated value of H_{ex} is higher than unity.

The outdoor dose (D_{out}) was calculated using Equation (3), and results are shown in Table 3 (UNSCEAR, 2000).

$$D_{out} = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K \qquad (3)$$

The average values for virgin and agricultural soils were 76.20 (range: 53.49–105.31 nGy h^{-1}) and 116.04 nGy h^{-1} (range: 58.05–204.91 nGy h^{-1}), respectively. These values were higher than the mean value of 51 nGy h^{-1}recommended in the UNSCEAR (2000) report.

The results obtained for the internal hazard index (H_{in}) in virgin and agricultural soil are summarized in Table 3.

Equation (4) was used to calculate the values of H_{in} (Beretka and Mathew, 1985).

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4{,}810} \quad (4)$$

The average values of H_{in} for virgin and agricultural soil were 0.604 (range: 0.400–0.870) and 0.925 (range: 0.454–1.76), respectively. The average values of H_{in} in both virgin and agricultural soil were less than unity.

The indoor absorbed dose rate (D_{in}) for virgin and agricultural soils were calculated using Equation (5) (Beretka and Mathew, 1985) and the results are summarized in Table 3.

$$D_{in} = 0.92C_{Ra} + 1.1C_{Th} + 0.08C_K \quad (5)$$

The average values of D_{in} for virgin and agricultural soils were 143.32 (range: 100.11–198.85 nGy h^{-1}) and 218.46 nGy h^{-1} (range: 109.31–389.67 nGy h^{-1}), respectively. These values were higher than the recommended value of 70 nGy h^{-1} (UNSCEAR, 1988).

For the general public who work outside and inside houses, the annual effective dose was calculated in terms of outdoor (E_{out}) and indoor (E_{in}) annual effective doses using Equations (6) and (7), respectively. The conversion factor (0.7 Sv Gy^{-1}) and outdoor (0.2) and indoor occupancy factors (0.8) were used to estimate E_{out} and E_{in} (UNSCEAR, 2000).

$$E_{outdoor} = absorbed\ dose\ (Gy / h) \times 8766\ h / y \times 0.7\ (Sv / Gy) \times 0.2 \times 10^{-6} \quad (6)$$

$$E_{indoor} = absorbed\ dose\ (Gy / h) \times 8766\ h / y \times 0.7\ (Sv / Gy) \times 0.8 \times 10^{-6} \quad (7)$$

where 8766 h y^{-1} is number of hours in one year (leap year was taken in account), and 10^{-6} is the conversion factor between nano and milli.

The results obtained for E_{out} and E_{in} are shown (Table 3).The average values of E_{out} for virgin and agricultural soils were 0.106 (range: 0.074–0.147 $mSvy^{-1}$) and 0.162 mSv y^{-1} (range: 0.081–0.286 mSv y^{-1}), respectively. These values were less than the lower limit of 20 mSv y^{-1} for radiation workers and even lower than the recommended level of 1 mSv y^{-1} for the general population (ICRP, 1991). The average values of E_{in} for virgin and agricultural soils were 0.439 (0.307–0.610 mSv y^{-1}) and 0.669 mSv y^{-1} (0.335–1.19 mSv y^{-1}), respectively. The average values of E_{in} were lower than the recommended level of 1 mSv y^{-1} for the general population.

CONCLUSIONS

Virgin and agricultural soil samples from Sungai Petani, Kedah, Malaysia, were investigated for their radioactivity content. The results revealed that the mean activity concentration values of ^{226}Ra, ^{232}Th and, ^{40}K in agricultural soils were 80.63±5.78, 116.87±7.84, and 200.66±18.24 Bq kg^{-1}, respectively and higher than those in virgin soils (51.06±5.83, 78.44±6.42, and 125.66±7.26 Bq kg^{-1}, respectively). The average activity concentrations of the measured radionuclides in both virgin and agricultural soils were higher than the world recommended values, except for ^{40}K.

The Ra_{eq} activity concentrations for all samples were less than the recommended level of 370 Bq kg^{-1}, except for soil collected from Kampong Gure Station and Kampong Kapala Bukit. The measured values of outdoor and indoor external hazard indices for virgin and agricultural soils were lower than the recommended values, except for soil collected from Kampong Padang Setol (H_{in}>1), Sungai Layar Hujang and Kampong Guar Station (H_{ex}, H_{in}, E_{in}>1), and Kampong Kapala Bukit (H_{ex}, H_{in}>1). Values obtained for D_{out} and D_{in} were higher than the permissible limits at 50 and 70nGy h^{-1}, respectively.

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