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PRIMORDIAL RADIONUCLIDES CONTENT AND RADIATION RISK ASSESSMENT OF SURFACE SOILS SURROUNDING TEMENGGOR RESERVED FOREST, GERIK, PERAK

(Kandungan Radionuklid Primordial dan Penilaian Risiko Radiasi Tanah Permukaan di Sekitar Hutan Rizab Temenggor, Gerik, Perak)

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Abstract

Natural radioisotopes' occurrence on the earth's surface and the crust might raise concerns for the environment in terms of their radiation risk. Their existence in the environment can be measured through the evaluation of soil from the targeted areas. This study aimed to assess the radionuclides content including thorium-232, uranium-238, and potassium-40 in surface soil samples from Temenggor Reserved Forest areas. The radiation assessment of the environment was done by calculating the Radium Equivalent (Ra_{eq}), Absorbed Dose Rate (AD), Outdoor Annual Effective Dose (AED), and External Hazard Index (H_{ex}). The surface soil samples were collected from twelve locations surrounding Temenggor Reserved Forest using a hand auger. The radionuclides content was measured using the Energy Dispersive X-Ray Fluorescence (EDXRF) Spectrometer and was converted to activity concentration for the risk assessment calculation. The activity concentration of ^{232}Th, ^{238}U and ^{40}K was found to be in the range of 28.92-179.92, 32.56-110.10 and 214.45-1083.38 Bq/kg, respectively. The values of Ra_{eq}, AD, AED and H_{ex} that were obtained were in the range of 103.90-312.17 Bq/kg, 49.99-149.65 (nGy/h), 0.06-0.18 mSv/y and 0.29-0.86, respectively. The results showed that the activity concentration and radiation risk were higher than the limit set by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR 2000) at certain study locations. However, the AED and H_{ex} were very low compared to the UNSCEAR 2000 limit. The high content of radionuclides might be caused by the types of soils and rocks as well as the anthropogenic sources from past activity in the targeted area.

Keywords: energy dispersive X-ray fluorescence, Perak, radionuclides, risk assessment, surface soil

Abstrak

Kewujudan radionuklid semulajadi di permukaan bumi dan di dalam kerak bumi mungkin menimbulkan kebimbangan kepada alam sekitar dari segi risiko radiasi mereka. Kewujudan mereka di dalam alam sekitar boleh diukur melalui penilaian tanah dari kawasan yang di kenalpasti. Kajian ini menyasarkan untuk menilai kandungan radionuklid termasuk torium-232, uranium-238 dan potasium-40 didalam sampel tanah permukaan di kawasan Hutan Rizab Temenggor. Penilaian risiko radiasi di kawasan sekitar telah dijalankan menggunakan penggiraan *Radium Equivalent (Ra_{eq}), Absorbed Dose Rate (AD), Outdoor Annual Effective Dose (AED) and External Hazard Index (H_{ex})*. Sampel-sampel tanah permukaan telah dikumpulkan dari dua belas (12) lokasi disekitar kawasan Hutan Rizab Temenggor dengan menggunakan gerimit tangan. Kandungan radinuklid telah diukur dengan menggunakan Energy Dispersive X-Ray Fluorescence (EDXRF) Spektrometer dan ditukar kepada aktiviti kepekatan untuk penggiraan risiko radiasi mereka. Aktiviti kepekatan tiga radionuklid ^{232}Th, ^{238}U dan ^{40}K telah direkodkan masing-masing dalam julat 28.92-179.92, 32.56-110.10 dan 214.45-1083.38 Bq/kg. Keputusan aktiviti kepekatan dan risiko radiasi yang direkodkan adalah melebihi had yang telah ditetapkan oleh United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR 2000) di beberapa lokasi kajian. Walaubagaimanapun, keputusan AED dan H_{ex} adalah sangat rendah berbanding had yang ditetapkan oleh UNSCEAR 2000. Kandungan radionuklid yang tinggi mungkin disebabkan oleh jenis tanah dan batu serta sumber antropogenik daripada aktiviti terdahulu di kawasan kajian.

Kata Kunci: energy dispersive x-ray fluorescence; perak; radionuklid, penilaian risiko, tanah permukaan

Introduction

Primordial radionuclides are long-lived radionuclides that are present on earth since their formation [1,2]. These radionuclides exist in various geological formations in the environment including the earth's crust, rocks, soils, plants, water and air. Radionuclides in the ^{238}U and ^{232}Th decay series and non-series of ^{40}K are called primordial radionuclides. Every radionuclide emits radiation at a specific rate, which is calculated as its half-life. Half-life is the time for the half-radionuclides that are present to decay, thus forming another type of radioisotope. Therefore, the radionuclides emit some radiation during this process. Exposure to a high level of radiation can cause acute health effects such as skin burns and acute radiation syndrome as well as long-term effects such as cancer and cardiovascular disease. However, exposure to low-level radiation does not cause immediate health effects but can be a minor contributor to an overall cancer risk [3]. Thus, the activity concentrations of these radionuclides play an important role in environmental public health.

Soils play a vital role in transporting radionuclides into the environment, and act not only as the source but also as a medium to transport radionuclides into other environmental media such as air and water. Thus, from the perspective of radiation protection, the assessment of the level of radionuclides in the environment and the consequent radiation exposure to humans is mostly carried out in soil [1]. However, the distribution of these radionuclides in the soil is not uniform; it varies according to the soil type, mineral content, geological features and geological conditions [4-8]. The types of rock also elevate the concentrations of the radionuclides in the soil. Higher concentrations of radionuclide are mostly found in igneous rocks such as granite whereas the lower concentration is discovered in sedimentary rocks [1, 2]. Moreover, anthropogenic activities in an area can alter the distribution of radionuclides in such an environment.

Many countries around the world have begun to achieve the Sustainability Development Goals (SDGs) that has been launched by the United Nation (UN), whose agenda aims to protect the environment for future generations. Correspondingly, Malaysia is no exception in jointly making the agenda a success. To ensure the sustainability of the environment, early data should be documented to allow other research or appropriate steps to be taken to achieve SDGs. To date, the radionuclides data of Temenggor areas are still not comprehensively available. Thus, this study aims to evaluate the activity concentration of ^{238}U, ^{232}Th and ^{40}K in the surface soil of the surrounding Temenggor Reserved Forest area. Furthermore, Temenggor Reserved Forest is part of Belum-Temenggor Forest Complex (BTFC) [9] which is well known for its nature attractions. During the peak

season, BTFC will be busy with tourists for activities such as bird watching, water activities, fishing and others. Moreover, the Temenggor area has lithosolos and shallow red-yellow podzolic solids with s-types granitoid soil [10, 11]; granite, phosphate and igneous rocks contain a higher concentration of uranium and thorium. Thus, another objective of this study is to estimate their possible radiation risk to the environment in terms of Radium Equivalent (Ra_{eq}), Absorbed Dose Rate (AD), Outdoor Annual Effective Dose (AED), and External Hazard Index (H_{ex}). These predictions allowed the researches to observe and categorised the sampling locations according to their risk exposure to humans. The calculated risk was compared to the limit set by UNSCEAR 2000 as well as other research studies.

Materials and Methods

Study area

Temenggor Reserved Forest is situated in the North part of Peninsular Malaysia. Located in Gerik, Perak, Temenggor Reserved Forest is a part of the bigger Belum-Temenggor Forest Complex (BTFC) and is one of Malaysia's biggest premier ecotourism spots. The main attractions of this ecotourism area include bird watching, Rafflesia tracking and water activities. This reserved forest consists of several small islands such as Pulau Tali Kail, Pulau TNB, and Pulau MARA. Several Orang Asli localities are also located in the surrounding the Temenggor Reserved Forest.

Sampling

The surface soil samples were collected from twelve (12) points surrounding the Temenggor Reserved Forest. The locations were determined based on their accessibility and random judgmental method. The exact sampling sites were marked using the Global Positioning System (GPS) as tabulated in Table 1, and illustrated in a map as shown in Figure 1. About 1 kg of each sample was collected using a hand auger from a 15 cm depth, at the marked sampling site. The samples were collected from a 1 m^2 area of five (5) different holes and mixed in a labelled plastic bag to represent one (1) sample.

Table 1. GPS of 12 sampling sites for soil samples

Location	GPS	
	N	E
S1	05º37'02.7	101º17'44.8
S2	05º35'47.5	101º18'53.6
S3	05º32'58.4	101º21'16.0
S4	05º27'47.1	101º17'34.3
S5	05º29'40.3	101º18'51.1
S6	05º30'37.2	101º21'24.4
S7	05º28'30.0	101º20'51.5
S8	05º28'01.8	101º21'26.4
S9	05º29'46.9	101º23'09.0
S10	05º30'11.4	101º23'17.2
S11	05º31'28.2	101º23'23.5
S12	05º32'55.9	101º22'08.0

Sample preparation and analysis of radionuclides

The wet soil samples were dried in an oven at 60-70°C for about 24-72 hours until they were completely dried. Before drying, unnecessary materials were removed from the soil to avoid contamination. The dried soil was grounded into a powder form using an agate type planetary ball mill and sieved through 250 µm sieves. About 2 g of sample was then placed in a clean pellet mold using a Fluxana Vaneox Technology Pressing machine and pressed with 15 tons pressure. The pellet form samples were stored in a desiccator prior to analysis. The concentration of uranium (U), thorium (Th) and potassium (K) were measured using an Energy Dispersive X-Ray Fluorescence (EDXRF) Spectrometer (Model of EPSILON 3XL by PANalytical), and reported in the unit of mg/kg. The measured concentration was then converted into activity concentration with the unit of Bq/kg using the conversion factors in equations 1-3 [12, 13].

$$^{23}\ \mathrm{Th}\left(\frac{\mathrm{Bq}}{\mathrm{kg}}\right) = 0.246\ \mathrm{ppm\ Th} \qquad \text{(Eq. 1)}$$

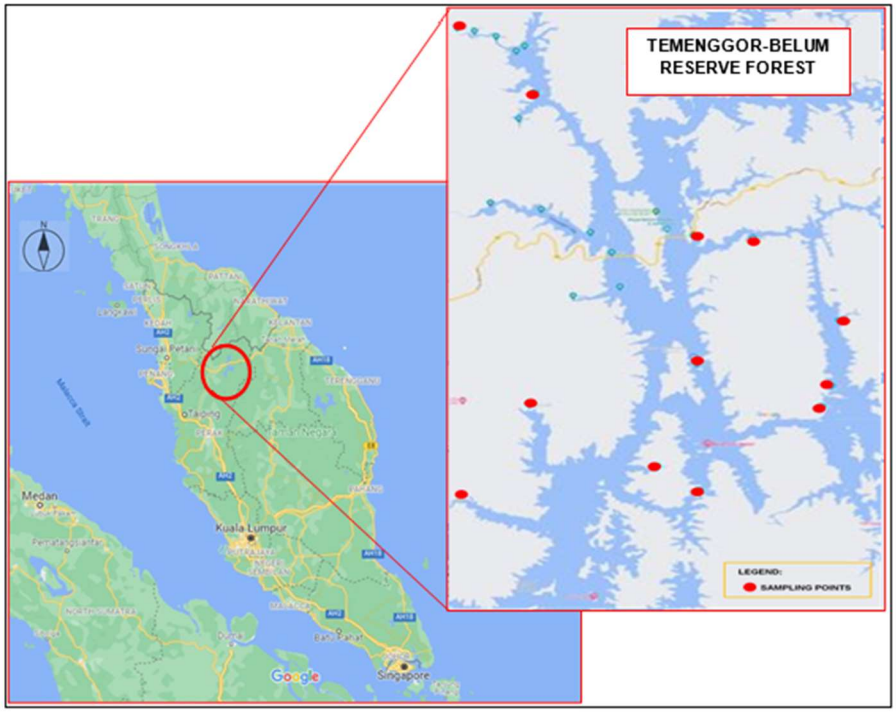

Figure 1. Maps showing the surface soil sampling sites

$$^{238}\mathrm{U}\left(\frac{\mathrm{Bq}}{\mathrm{kg}}\right) = 0.081 \text{ ppm U} \quad \text{(Eq. 2)}$$

$$^{40}\mathrm{K}\left(\frac{\mathrm{Bq}}{\mathrm{kg}}\right) = 32.2 \text{ ppm K} \quad \text{(Eq. 3)}$$

Method validation

The accuracy of the EDXRF was performed by analyzing the standard reference material (JLK-1, SRM 2709a and SO-7) for soil. The results obtained were compared with the established method of ICPMS. For this purpose, the samples were analyzed in the Agensi Nuklear Malaysia laboratory.

Radiology assessment

The content of ^{238}U, ^{232}Th and ^{40}K in soil are not uniformly distributed. The uniformity concerning radiation exposure can be expressed as Radium Equivalent Dose Rate (Ra_{eq}) measured in Bq/kg [14]. This calculation is for the comparison of the specific activity concentration level of constituents including radium, thorium and potassium as a constituent in varying amounts [10]. Before that, the activity concentration of ^{238}U was converted into ^{226}Ra activity concentration using Equation 4. Then, the Ra_{eq} was calculated using Equation 5. In addition, the Annual Effective Dose Rate (AED) is estimated to access the health effects of the absorbed dose. The AED used a conversion coefficient (0.7Sv/Gy) to transform the absorbed dose in the air into the absorbed dose received by humans with an outdoor occupancy factor (0.2) for adults (refer to Equation 7) [14, 15], whereby the absorbed dose rate (D) was calculated using Equation 6. On the other hand, multiple radionuclides contribute to the γ-dose, therefore the radiological hazards are presented as a single quantity that is known as the hazard index [10]. The hazard index is divided into two-internal and external hazard index. This study has only calculated the external hazard index to observe the radiological health risks of this locality since the

exposure to radiation from the soil is externally measured. The external hazard index was calculated using Equation 8 [14, 16, 17];

$$^{226}\mathrm{Ra}\left(\frac{\mathrm{Bq}}{\mathrm{kg}}\right) = 0.76 + 0.84\ (\mathrm{A}_{^{238}\mathrm{U}}) \quad \text{(Eq. 4)}$$

$$\mathrm{Ra}_{\mathrm{eq}}\left(\frac{\mathrm{Bq}}{\mathrm{kg}}\right) = \mathrm{A}_{^{226}\mathrm{Ra}} + 1.43\ \mathrm{A}_{^{232}\mathrm{Th}} + 0.077\ \mathrm{A}_{^{40}\mathrm{K}} \quad \text{(Eq. 5)}$$

$$\mathrm{D\ (nGy/h)} = 0.427\ \mathrm{A}_{^{238}\mathrm{U}} + 0.662\ \mathrm{A}_{^{232}\mathrm{Th}} + 0.0432\ \mathrm{A}_{^{40}\mathrm{K}} \quad \text{(Eq. 6)}$$

$$\mathrm{AED}\left(\frac{\mathrm{mSv}}{\mathrm{yr}}\right) = \mathrm{D}\left(\frac{\mathrm{nGy}}{\mathrm{h}}\right) \mathrm{x}\ 8760\ (\mathrm{h})\ \mathrm{x}\ 0.2\ \mathrm{x}\ 0.7\left(\frac{\mathrm{Sv}}{\mathrm{Gy}}\right) \mathrm{x}\ 10^{-6} \quad \text{(Eq 7)}$$

$$\mathrm{H_{ex}} = \frac{\mathrm{A}_{^{226}\mathrm{Ra}}}{370} + \frac{\mathrm{A}_{^{232}\mathrm{Th}}}{259} + \frac{\mathrm{A}_{^{40}\mathrm{K}}}{4810} \quad \text{(Eq. 8)}$$

where $\mathrm{A}_{^{226}\mathrm{Ra}}$ = Activity concentrations of ^{226}Ra, $\mathrm{A}_{^{238}\mathrm{U}}$ = Activity concentration of ^{238}U, $\mathrm{A}_{^{232}\mathrm{Th}}$ = Activity concentration of ^{232}Th, $\mathrm{A}_{^{40}\mathrm{K}}$ = Activity concentrations of ^{40}K, D (nGy/h) is the total air absorbed dose rate in the outdoors, 8760h is the number of hours in one year; 0.2 is the outdoor occupancy factor (AELB standard); 0.7 Sv/Gy is the conversion coefficient from absorbed dose in the air to the effective dose that is received by adults (AELB standard); 10^{-6} is the conversion factor between nano- and milli measurements.

Results and Discussion

In this study, the EXDRF method was validated using a variety of standard reference materials (SRM) including JLK-1, SRM 2709a and SO-7. Good recoveries were obtained in the range of 87-115%. The accuracy of the EDXRF method was also compared with the inductively coupled plasma-mass spectrometer (ICPMS) in the Agensi Nuklear Malaysia laboratory using similar samples. The results obtained were relatively comparable with the recoveries in the range of 70-136 %, showing the reliability of the EXDRF method that was used for the analysis of elements in the soil samples.

The activity concentrations of three (3) radionuclides of ^{238}U, ^{232}Th and ^{40}K from twelve (12) locations surrounding the Temenggor area are presented in Table 2. The measured concentration from the EDXRF in the unit of mg/kg was converted into activity concentration with the unit of Bq/kg. As tabulated in the table, the activity concentrations ^{238}U, ^{232}Th and ^{40}K were in the range of 32.56-110.10, 28.92-179.92 and 214-1083 Bq/kg with the mean of 52.55, 79.49, and 476 Bq/kg, respectively. The lowest level of ^{238}U was observed at site S05 with a value of 32.56 Bq/kg and the highest was at site S03 (110.10 Bq/kg). Meanwhile, for ^{232}Th the lowest was at site S01 (28.92 Bq/kg) and the highest was at site S11 (179.92 Bq/kg). Site S11 gave the lowest concentration with a value of 214 Bq/kg, while S03 contained the highest activity concentration of ^{40}K (1083 Bq/kg). The difference in concentrations was most probably caused by the type of soil and the surrounding environment of the sampling site. According to Sanusi et al., granite soil has higher radionuclides content compared to other types of soil [11]. Temenggor areas are made up of lithosolos and shallow red-yellow podzolic solids with s-types granitoid soil [10]. Factually, this can explain the high concentrations of radionuclides in this area. Moreover, the additional anthropogenic sources might elevate the natural content of radionuclides. It was found that site S03 was located near the small jetty for a private boat, a site that has become a pickup and drop-off point for goods and other amenities. This site is also a place for small boats to refuel their engine oil. Furthermore, vehicles can also access to this location from the mainland. Thus, radionuclides can be released into the environment

through transportation as radionuclides are one of the burning fuel products [18, 19]. On the other hand, site S11 is located in a place named Tanah Banun (according to local people). This location has an open space which is a habitat for most of the wild animals to roam as well as rest. The transfer of radionuclides from their origin to the environment could be through the food chain. Plants absorb nutrients and other contaminants from the soil and are eaten by animals. Additionally, soil can also be ingested by animals through the licking or preening of their fur, feathers, or offspring [20]. Then, the absorbed radionuclides can be excreted in urine or endogenously excreted in the faeces and absorbed back into the soil of the areas [21]. This might explain the elevated concentrations of radionuclides in location S11. Furthermore, Site S11 is also near one of the Orang Asli localities in the Temenggor area. While samples from other locations have mostly been collected from the small island that has not been occupied anymore.

The standard permissible limit for radionuclides in soil that is reported by the UNSCEAR is 35, 30, and 400 Bq/kg for ^{238}U, ^{232}Th, and ^{40}K, respectively as shown in Table 2. The majority of the sampling sites have higher radionuclides content if compared to the world limit [3]. Additionally, the activity concentrations of the three radionuclides in this study were compared to other reported studies of soil around Malaysia. Mohammad Syazwan et al. [22] studied the influence of geological and soil formation on natural gamma (γ) radiation exposure. The study focused to obtain the baseline data of γ radiation exposure and radioactivity that were associated with soil in the granitic territory in the Western Region of Peninsular Malaysia. The study stated ^{238}U and ^{232}Th activity concentrations to be in the range of 7-554 and 23-1809 Bq/kg, respectively. Temenggor area has lower activity concentrations when compared to the Western Region of Peninsular Malaysia despite having the same soil types [22]. Another reported study of radionuclides from the soil samples that were collected in Bangi by Mei-Wo and Ali showed a different pattern [23]. The activity concentrations of ^{32}Th and ^{40}K are relatively in the range of the Temenggor area but there is a slightly higher range for ^{238}U. The author has stated that the noticeable difference may be attributed to the geochemical composition and origins of the soil types in the particular study areas. Moreover, the higher values of the activity concentrations belong to soil samples that may be attributed to soil types which were probably radioactive-rich granite, phosphate, sandstone, and quartzite [23]. Abdul Aziz and Khoo studied the assessed absorbed dose and radiation hazard index from the soil in Bukit Kledang, Perak [24]. The obtained results were in the lower range than this study for the three radionuclides, as shown in Table 2. The authors stated that the varied values that were obtained varied according to the location. It is most likely to be affected by several factors such as soil type, soil pH, the latitude of area, soil texture, the existence of underground rock, and organic matter [24].

The findings of activity concentrations surround Temenggor area was also compared to some studies from oversea. The vertical distributions of forty-four soil samples from Qatar were studied by Ahmad et al. [7]. The mean concentrations of this study are presented in Table 2 and have lower concentrations compared to this study. The authors agreed that the different levels of radionuclides in the soil depend primarily on the concentration of radionuclides in bedrocks from which the soil originates. Besides, the author said that the natural radionuclides levels (^{226}Ra, ^{232}Th, and ^{40}K) in the soil might be affected by the physiochemical characteristic of the soil [7]. Another reported study of activity concentrations for the three natural radionuclides (^{226}Ra(^{238}U), ^{232}Th, and ^{40}K) using soil samples was carried out in Korea [8]. The in-situ and laboratory γ-ray spectroscopy techniques were compared to evaluate the activity concentration of natural radionuclides in soil. Table 2 shows the results of the in-situ measurement using gamma spectroscopy techniques. The authors reported that all-natural radionuclide concentrations were lower on Jeju Island due to its geological origin of quarternary basalt/trachyte, while they were higher in Chuncheon due to its geological origin of Jurassic granite. Moreover, a strong correlation was observed between the techniques with linear regression lines of 99% for ^{226}Ra and ^{232}Th, whereas for ^{40}K the correlation was 97.5% [8]. All of the reported studies have agreed that the difference and high concentrations of radionuclides

are most likely caused by the original, geochemical, and physiochemical types of the soil itself.

Table 2. The activity concentrations of ^{238}U, ^{232}Th and ^{40}K

Location	Activity Concentrations (Bq/kg)		
	^{238}U	^{232}Th	^{40}K
S01	33.31	28.92	418
S02	70.97	72.59	752
S03	110.10	86.75	1083
S04	46.86	28.95	290
S05	32.56	38.69	263
S06	44.73	102.42	339
S07	51.90	44.43	657
S08	53.86	160.51	411
S09	41.49	41.79	311
S10	51.21	71.67	405
S11	34.50	179.92	214
S12	59.17	97.28	562
Mean	**52.55**	**79.49**	**476**
UNSCEAR 2000 [14]	35	30	400
Western region Peninsula Malaysia [22]	7-554	23-1806	-
Bukit Kledang, Perak [24]	11.98 – 29.93	20.97 – 41.45	5.73 – 59.41
Bangi [23]	17.2-85.4	23.9-89.9	98.4-499.4
Qatar [7]	17	10	201
Korea [8]	13-188	14.6-230	241-1328

Temenggor area has Ra_{eq} values between 103.90 - 312.17 Bq/kg. The Ra_{eq} of the study area is presented in Figure 2. It can be observed that this study has a lower concentration than the limit that is set by UNSCEAR 2000 of 370 Bq/kg. Meanwhile, it is relatively comparable with the reported values of other studies [1,14,22,25,26,27].

The absorbed dose date of radionuclides was calculated in the unit of nGy/h. This is the measurement of presented radionuclides in the air exposed to humans. The absorbed dose rate at sampling sites varies from 49.99 to 149.65 nGy/h (Figure 3). The highest value in the Temenggor area that has been observed at site S03 is about three times higher in value than the world limit (55 nGy/h). However, Temenggor areas have absorbed dose rates in a range comparable with other reported studies [1, 14, 25, 27].

The estimated outdoor absorption of radiation was calculated using the annual effective dose (AED) in the unit of mSv/y. The calculated AED for the Temenggor area was about four times lower than the reported world limit by UNSCEAR 2000 as illustrated in Figure 4. The value was between 0.0621 to 0.1835 mSv/y. The reported value from Nigeria in 2019 [26] has a higher AED than the Temenggor area, while the values in other reported studies were comparable with the study areas [1, 14, 25, 26, 27].

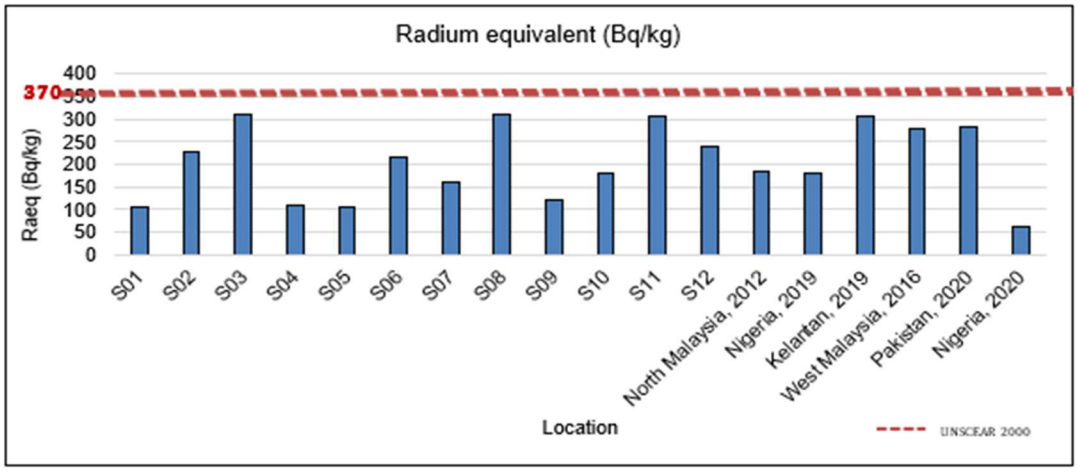

Figure 2. The radium equivalent (Ra_{eq})

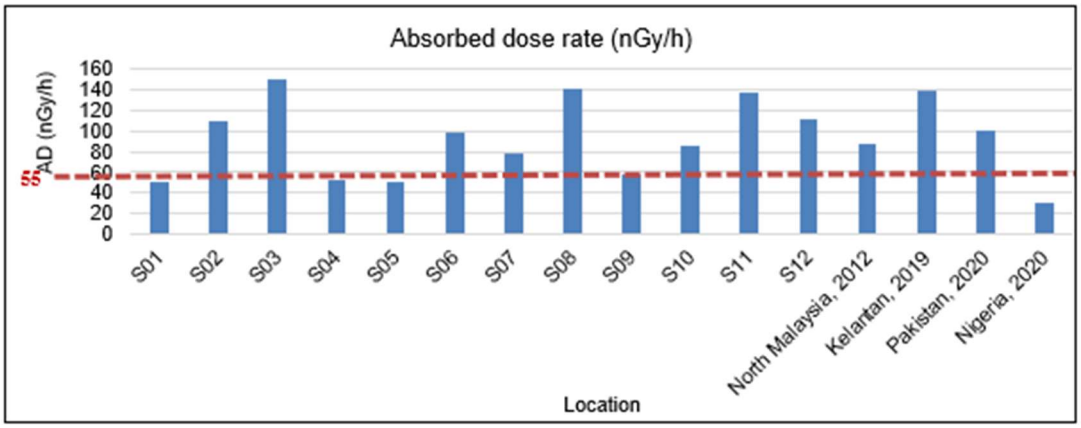

Figure 3. The absorbed dose rate (AD)

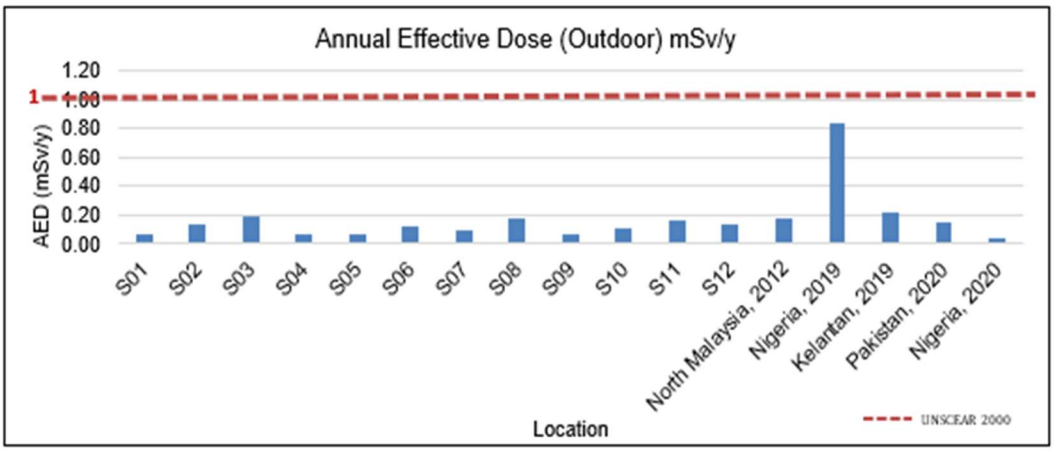

Figure 4. The annual effective dose (AED)

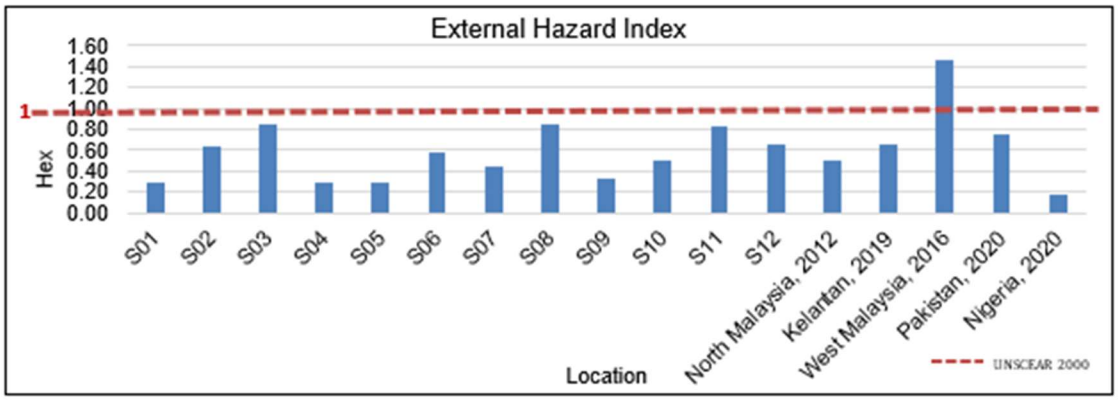

Figure 5. The external hazard index (H_{ex})

The total estimation of γ -dose radiation was calculated using the hazard index. In this study, researchers used the external hazard index since the estimation was based on the radiation in the soil. Figure 5 shows the distribution of the external hazard index for the Temenggor area and a comparison between the world limit and other reported studies. Temenggor area has a hazard index in the range of 0.2885-0.8577. All locations have a lower value than the world limit that is set by UNSCEAR 2000 with the value of 1. This implies that exposure to the gamma radiation from the soil has no significant consequence to health. The other reported values [1, 14, 25, 27] were also in-between the Temenggor area; except for West Malaysia [22] where it has a higher value.

Conclusion

A study to observe the distribution of three radionuclides- including ^{238}U, ^{232}Th and ^{40}K, has been conducted in the Temenggor Reserved Forest area. The mean activity concentrations of ^{238}U, ^{232}Th, and ^{40}K are 52.55, 79.49, and 476 Bq/kg, respectively. Some of the locations have higher activity concentrations than the world limit that is released by UNSCEAR 2000. Comparing the activity concentrations with the reported studies around Malaysia, the Temenggor area has a lower activity concentration. The type of soil in the study area contributes to the high activity concentrations of radionuclides. On the other hand, the radiation risk assessment has shown that the study areas do not pose a significant risk to humans. This is based on the calculation of Radium Equivalent (Ra_{eq}), Absorbed Dose Rate (AD), Outdoor Annual Effective Dose (AED), and External Hazard Index (H_{ex}).

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