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Radioactivity levels of ^{238}U and ^{232}Th, the α and β activities and associated dose rates from surface soil in Ulu Tiram, Malaysia

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A survey was carried out to determine terrestrial gamma radiation dose rates, the concentration level of ^{238}U and ^{232}Th and α and β activities for the surface soil in Ulu Tiram, Malaysia. A 125 measurements were performed using a NaI(Tl) gamma-ray detector with crystal size of 1"×1" on 15 soil samples collected from the site area about 102 km^2. ^{238}U and ^{232}Th concentrations were determined in soils by using hyper pure germanium (HPGe) gamma-ray spectrometry. The activity of α and β from the surface soil was counted by using alpha beta counting system. The average value of ^{238}U and ^{232}Th concentrations in soil samples collected are 3.63±0.39 ppm within the range of 1.74±0.20 to 4.58±0.48 and 43.00±2.31 ppm within the range of 10.68±0.76 to 82.10±4.01 ppm, respectively. The average estimate of α and β activity in soil samples collected are 0.65±0.09 Bq·g^{-1} and 0.68±0.08 Bq·g^{-1}, respectively. The average of terrestrial gamma-radiation dose rates measured in Ulu Tiram was found to be 200 nGy·h^{-1}, within the range of 96 to 409 nGy·h^{-1}. The population weighted outdoor annual effective dose was 1.2 mSv.

Introduction

The concern about absorbed dose rates at environmental level is rising due to its possible contribution to radiation stochastic effects. Naturally occurring radionuclides of terrestrial origin are present in various degrees in all media in the environment, including in the human body itself. The terrestrial gamma-radiation dose rates exposures vary widely depending on locations. The concentrations of ^{238}U and ^{232}Th may be elevated in localized areas; the exposure can also vary as result of human activities and practices.[1] The UNSCEAR report[1] concludes that more data on exposures from natural occurring radionuclides are still not properly quantified.

A survey was carried out to determine the terrestrial gamma-radiation dose rates, concentration levels of ^{238}U and ^{232}Th and its α and β activities from the surface soil of Ulu Tiram, Malaysia. This study also will investigate the influence of geological structure and soil types on the concentrations of ^{238}U and ^{232}Th in the soil and its effect to the terrestrial gamma-radiation dose rates in the environment.[2] Survey of terrestrial gamma-radiation dose rates also is of interest to the explorer of mineralization especially ^{238}U.

Ulu Tiram is located at the south of Peninsular Malaysia between latitudes 1° 35'and 1° 40' North and longitudes 103° 44' and 103° 51' East. It has an area of approximately 102 km^2. Seventy percent of the area is covered by forest and the main use of the land currently is for agriculture. Ulu Tiram is a rural area of Johor Bahru city which will be developed as an industrial centre and housing estate. Therefore, survey of the terrestrial gamma-radiation dose rates in Ulu Tiram is important to provide a data base to determine an environmental radiological safety status.[2] Ulu Tiram can be divided into two major geological areas of different geological age.[3] The geological formations overlaid are igneous rock or granite structured consists of acid with undifferentiated granite rock and the intermediate including syenite, tonalite and diorite. The other one is Quaternary consists of mainly recent alluvium. Igneous rock or granite is more abundant in Ulu Tiram. These two geological structures are different as one acidic and the other is basic. This is an important factor for mineralization concentration especially for ^{238}U and ^{232}Th.[4]

Ulu Tiram is overlaid by three groups of soil types as classified by FAO/UNESCO.[5] The soil types are:

(a) *Dystric Fluvisols*, this group consists of flood plains and alluvial soil and the local name is Rusila. Most of these groups are found on the coastal plain, mostly in tidal swamps covered by mangrove.

(b) *Dystric Nitosols* is a type of soil of shiny pad surfaces and the local names is Renggam and it is the most abundant soil type in Ulu Tiram.

(c) *Ferric Acrisols* is an acidic soil of low base saturation and the local names is Harimau Tampoi or Durian.

Experimental

Ulu Tiram is divided into 125 stations by aligning the grid along the latitudinal and longitudinal lines (1'×1'). Each station measures around 0.9 km×0.9 km. Soil types with underlying geological formations for each station were determined.[6] Measurements of the terrestrial gamma-radiation dose rate are conducted with

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gamma-ray detectors manufactured by Ludlum (USA) Model 19, Micro R Meter. The equipment uses a 2.54×2.54 cm^2 NaI(Tl) crystal. The smallest scale division for the instrument is 1 $\mu R \cdot h^{-1}$ (~9 $nGy \cdot h^{-1}$). The instrument had almost a flat energy response to gamma-radiations between 40 keV to 1.2 MeV. The low response of the instrument to high energy gamma-radiation implies that a contribution from cosmic sources is not considered. It is suitable for environmental gamma-radiation measurements.[7] It covers a majority of significant gamma-radiations emitted from terrestrial sources. The uncertainty of reading observed on the maximum scale of the instrument is of the order of 10%. The instrument was calibrated by the Malaysian Institute of Nuclear Technology Research (MINT). It is a certified institution of instrument calibration laboratory.

The terrestrial gamma-radiation dose rate measurements were conducted at Ulu Tiram, away from sites of developments such as road, building and foundation soils. The locations for each sampling point were established by global positioning system (GPS).[8] A total of 125 measurements were conducted.

Fifteen soil samples were collected at a depth of 10 cm from surface soils.[6] They were dried in an oven at 100–110 °C until a constant dry weight was obtained. The samples were then crushed into fine powder until all samples passed through 150 μm sieves to be homogenized and to remove stones, pebbles and other macros impurity. The weight of each soil sample was approximately 500 g and was carefully sealed in airtight PVC containers for 30 days to ensure equilibrium between ^{226}Ra and its daughters; and ^{228}Ra and its daughters before gamma-radiation spectrometric analysis.[9] Soil sample containers were then placed into a shielded high-purity germanium (HPGe) detector and measured for 6 hours.

The naturally occurring radionuclides considered in the present analysis are: ^{212}Pb (with a main γ-radiation energy at 239 keV and a γ-radiation yield of 43.1%), ^{214}Pb (with the main γ-radiation energy at 352 keV and a γ-radiation yield of 37.1%), ^{214}Bi (with the main γ-radiation energy at 609 keV and a γ-radiation yield of 46.1%) and ^{228}Ac (with the main γ-radiation energy at 911 keV and a γ-radiation yield of 29%).

Since secular equilibrium was reached between ^{232}Th and ^{238}U and their decay products, the concentration of ^{232}Th was determined from the average concentration of ^{212}Pb and ^{228}Ac in the samples, and that of ^{238}U was determined from the average concentration of ^{214}Pb and ^{214}Bi decay products.[9] Thus an acceptable radionuclides concentration of ^{232}Th and ^{238}U were obtained. The environmental gamma-ray background at the laboratory site has been determined by using empty Marinelli beaker under identical measurement condition.

The energy resolution (FWHM) achieved in the calibration measurement was 1.8 keV at the 1.33 MeV reference transition of ^{60}Co. Depending on the background peak the minimum detectable activity (MDA) for ^{238}U and ^{232}Th were calculated to be 0.03 ppm and 0.30 ppm, respectively, for the 6-hour counting.

Results and discussion

Standard samples were measured for the purpose of quality assurance. Soil material IAEA-326 was used as a standard reference material. Table 1 shows the result and the certified values. This result shows good performance of the measurement and analysis technique utilized. Table 2 shows the distribution of gamma-radiation dose rates for each soil sample collected at the area with underlying soil type and geological formation. Data on concentration of ^{238}U and ^{232}Th and α and β activities in soil samples collected from each chosen station are given in Table 3. Table 2 clearly shows that soil types and geological formations influence the measured gamma-radiation dose rates. This result is similar to other studies carried out in many countries in the world such as Brazil,[10] Egypt,[11] Italy,[12] Spain,[13] Kuwait,[14] India,[15] Pontian, Malaysia[16] and Kota Tinggi, Malaysia.[17] As such, it might be possible to predict the terrestrial gamma-radiation dose rate for other areas based upon the information on the geology and soil types.

The highest terrestrial gamma-radiation dose-rate measured that is 409 $nGy \cdot h^{-1}$ was in area covered by *Dystric Nitosols* soil types. This soil type is underlain by acid with undifferentiated granite rock and the intermediate including syenite, tonalite and diorite formations. Both of these geological formations are abundant in granites and extensively intruded by schist, shale, quartzite and siltstone. Such rocks contain high concentrations of naturally occurring radionuclides such as ^{238}U, ^{232}Th and ^{40}K.[18] Most of radioactive occurrences in the base rocks of Ulu Tiram are in the granites. The high level of radioactivity of this rock is attributed to the presence of accessory minerals like zircon, monazite, thorite, uranothorite and allanite.[1]

The lowest terrestrial gamma-radiation dose rates were measured in an area underlain by geological formation of alluvium and covered by soil type *Ferric Acrisols*; it is 95.7 $nGy \cdot h^{-1}$. The terrestrial gamma-radiation dose rates are lowest in Quaternary areas formed from peat, humic clay and silt. Low dose rates were registered on alluvial sands in certain locations along the coast and the river.[17] Natural terrestrial gamma-radiation dose rate in *Dystric Histosols* (Peat areas) is very low. The dose rates here depend on the thickness and purity of the peat.[19]

Dystric Nitosols soil types with underlying geological formation of acid intrusive rock are the most abundant in Ulu Tiram. These geological formation and soil types contain highest concentration of primordial radionuclides especially ^{238}U and ^{232}Th.[20] This result is similar to those from other studies nearby the districts such as Kota Tinggi,[17] Pontian[16] and some other places in Johor state, Malaysia. ^{238}U and ^{232}Th are generally enriched in the youngest, most felsic and most potassic members of comagmatic suites of igneous rocks.[21] This caused the higher of gamma-radiation dose rates measurements obtained in Ulu Tiram. The mean of the terrestrial gamma-radiation dose rates obtained from the area covered by *Dystric Nitosol* with underlying of acid intrusive rock is 211±11 nGy·h^{-1}.

Table 3 shows the concentration of ^{238}U and ^{232}Th in surface soil and α and β activities in soil samples. From Tables 2 and 3, it could be seen that the terrestrial gamma-radiation dose rates is depending on the concentration of ^{238}U and ^{232}Th in surface soil. The concentration of ^{238}U and ^{232}Th are influenced by the underlying of geological formation and soil type at the area.

The highest concentrations of ^{238}U and ^{232}Th were obtained from sample S5, which is from soil type *Dystric Nitosols* with underlying acid intrusive rock it was 4.64±0.50 ppm and 81.81±3.90 ppm, respectively. The sample with the highest concentration of ^{238}U and ^{232}Th also give the highest α and β activities. Sample S2 contain the lowest concentration of ^{238}U and ^{232}Th that is 1.74±0.20 ppm and 10.68±0.76 ppm, respectively. Soil sample S2 also has the lowest α and β activities that is 0.20 Bq·g^{-1} and 0.24 Bq·g^{-1}, respectively.

Table 1. Quality control measurement using the reference Soil Material IAEA-326

Radioelement	Mean value	95% confidence level	Certified values
^{232}Th, ppm	9.71	9.26–10.15	10.32 ± 0.53
^{238}U, ppm	2,38	2.28–2.49	2.53 ± 0.15

Table 2. Information on soil samples

Sample	Geological structure	Soil types	Terrestrial gamma-radiation dose rate, nGy·h^{-1}
S1	Undifferentiated granite	Dystric Nitosols	139
S2	Undifferentiated granite	Dystric Nitosols	113
S3	Undifferentiated granite	Dystric Nitosols	348
S4	Undifferentiated granite	Dystric Nitosols	313
S5	Undifferentiated granite	Dystric Fluvisols	409
S6	Undifferentiated granite	Dystric Fluvisols	226
S7	Quaternary	Ferric Acrisols	131
S8	Quaternary	Dystric Fluvisols	148
S9	Quaternary	Dystric Fluvisols	244
S10	Quaternary	Ferric Acrisols	261
S11	Undifferentiated granite	Ferric Acrisols	174
S12	Undifferentiated granite	Dystric Nitosols	322
S13	Undifferentiated granite	Dystric Nitosols	287
S14	Undifferentiated granite	Dystric Nitosols	244
S15	Undifferentiated granite	Dystric Nitosols	261

Table 3. Data on ^{238}U and ^{232}Th concentration and the α and β activities in soil samples collected at Ulu Tiram, Malaysia

Sample	U, ppm	Th, ppm	Ratio Th/U	Alpha activity, Bq/g	Beta activity, Bq/g
S1	3.50 ± 0.24	21.20 ± 1.31	6.05	0.34	0.52
S2	1.74 ± 0.20	10.68 ± 0.76	6.12	0.22	0.24
S3	3.96 ± 0.43	69.16 ± 3.38	17.47	1.13	1.01
S4	4.47 ± 0.50	77.87 ± 3.74	17.43	1.08	1.08
S5	4.64 ± 0.50	81.81 ± 3.90	18.84	1.02	1.17
S6	3.84 ± 0.42	61.19 ± 3.46	15.95	0.52	0.54
S7	3.43 ± 0.36	13.70 ± 0.96	3.99	0.29	0.58
S8	4.58 ± 0.48	22.97 ± 1.43	5.02	0.43	0.54
S9	3.00 ± 0.32	16.45 ± 1.04	5.48	0.36	0.34
S10	3.83 ± 0.42	37.92 ± 2.09	9.90	0.67	0.58
S11	2.46 ± 0.29	19.82 ± 1.43	8.05	0.41	0.50
S12	3.27 ± 0.38	32.83 ± 1.88	10.06	0.54	0.55
S13	4.57 ± 0.52	81.12 ± 4.01	17.95	1.18	1.15
S14	3.40 ± 0.39	41.96 ± 2.33	12.36	0.73	0.67
S15	4.10 ± 0.47	55.37 ± 2.90	13.51	0.84	0.73

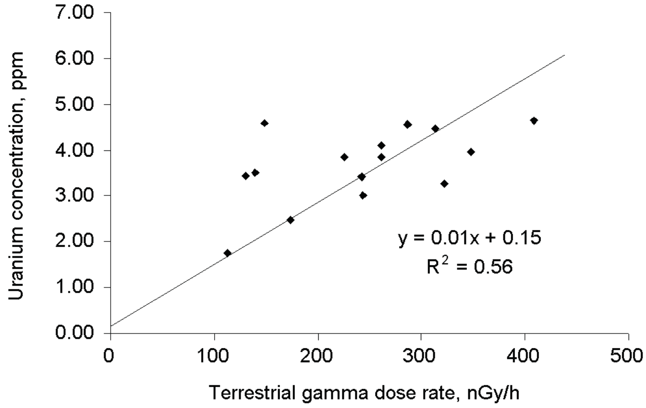

Fig. 1. Correlation between ^{238}U concentration and terrestrial gamma-radiation dose rates

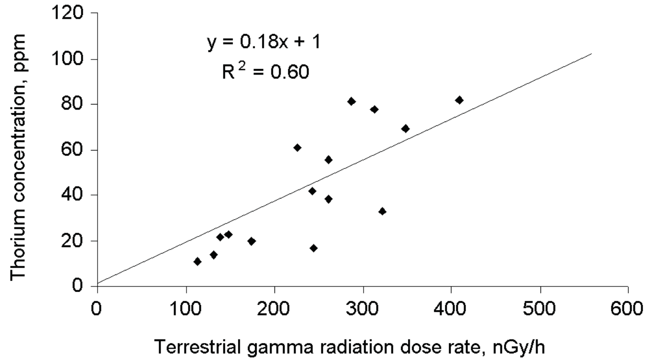

Fig. 2. Correlation between ^{232}Th concentration and terrestrial gamma-radiation dose rates

Figures 1 and 2 show that there is a linear relationship between radionuclide concentrations and the terrestrial gamma-radiation dose rates. The correlations between concentrations of ^{238}U and ^{232}Th with terrestrial gamma-radiation dose rates were computed from the results given in Tables 2 and 3. The correlation coefficient between ^{238}U and ^{232}Th terrestrial gamma-radiation dose rates was found to be 0.56 and 0.60, respectively.

Conclusions

Gamma-radiation dose rates measured due to terrestrial sources were greatest in areas covered by soil types which were formed from granite formation. The maximum value is 409 nGy·h^{-1}. The lowest gamma-radiation dose rate is 98 nGy·h^{-1}. Close relationship between terrestrial gamma-radiation dose rate and underlying soil types was established. It might be possible to statistically predict the terrestrial gamma-radiation dose rate from geological and soil type information.

The gamma-radiation spectrometric analysis revealed that high radioactivity in soil studied was due to ^{238}U and ^{232}Th. Higher concentrations of ^{238}U and ^{232}Th in surface soil lead to higher α and β activities. Further study on ingestion and radionuclides migration is needed to assess health implication to individuals. The solubility of ^{238}U in soil also has direct implications to the body, specifically, as the solubility of ingested ^{238}U increases; more ^{238}U will be absorbed into the blood stream and distributed to body organs especially the kidneys.[23]

The average terrestrial gamma-radiation dose rate in Ulu Tiram is 200 nGy·h^{-1}. The Malaysian average[1] is 92 nGy·h^{-1} and world average is 59 nGy·h^{-1}. Using the conversion factor[1] of 0.7 Sv·Gy^{-1} the average dose from such terrestrial gamma-radiation dose rate to an individual assuming a tropical rural setting is estimated to be 1.23 mSv per year, which is considered to be within the normal range for doses from natural sources. It is not expected to cause any statistically significant radiological health impact.

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