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Determination of Natural Radioactivity and Radiological Hazards of ^{226}Ra, ^{232}Th, and ^{40}K in the Grains Available at Penang Markets, Malaysia, Using High-purity Germanium Detector

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Abstract– **In the present study, the concentrations of ^{226}Ra, ^{232}Th, and ^{40}K and their radiological hazards in 18 types of grain samples, collected from local markets in Penang, Malaysia, are investigated using high-purity germanium detector (HPGe). The results indicated that the concentration of ^{226}Ra, ^{232}Th, and ^{40}K in grain samples was ranged from 56.97 to 86.13 Bq.kg^{-1}, from 34.71 to 52.14 Bq.kg^{-1}, and from 517.05 to 997.59 Bq.kg^{-1}, respectively. The results of the average annual ingestion dose of natural radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K were found to be 66.555, 35.199, and 15.328 µSv y^{-1}, respectively. This results are below the standard worldwide value (290 µSv y^{-1}) that was reported by UNSCEAR. Therefore, the studied samples are considered safe in terms of the radiological health hazards, and there is no health hazard from the grain in this region.**

Index Terms— **^{226}Ra, ^{40}K, Grain gamma rays, Natural Radioactivity, radiological hazards.**

I. Introduction

Natural radionuclides are present in every human environment; earth material, water, air, foods, and even our own body contain naturally occurring radioactive. The main natural radioactive sources of ionizing radiation are the long-lived ^{238}U, ^{232}Th, and their decay series and the ^{40}K (Tawalbeh et al., 2011). Radioactive elements such as uranium and thorium are also present in the atmosphere of cement plants (Adil et al., 2018). Analysis of these radionuclides in foodstuff is an important part of the environmental monitoring program. These natural radioactive sources are the largest contributor of the radiation doses received by humanity (Abdulaziz et al., 2013). Naturally occurring potassium ^{40}K is present virtually in all foodstuff as primary constituent of cellular material (Abdulaziz et al., 2013; Rohit et al., 2014; Awudu et al., 2012; Cumhur and Mahmut, 2013). Radionuclides can enter the human body through inhalation and ingestion. The ingested radionuclides could be concentrated in certain parts of the body (Tawalbeh et al., 2011); therefore, ingestion of radionuclides through food intake may account for a substantial fraction of the average radiation doses to various organs of the body, and this may also represent one of the important pathways for long-term health considerations (Jibiri et al., 2007; Al-Masri et al., 2004). For example, it has been estimated that at least one-eighth of the mean annual effective dose due to natural sources can be attributed to the intake of food (Jibiri et al., 2007; Gabdo et al., 2015).

Food is known to contain natural and artificial radionuclides that, after ingestion, contribute to an effective internal dose. It has been estimated that a large portion, at least one-eighth, of the mean annual dose due to natural sources is caused by the intake of food. Average radiation doses to various organs of the body also represent an important pathway for long-term health considerations. ^{232}Th, ^{238}U (^{226}Ra), and ^{40}K are three long-lived naturally occurring radionuclides present in the earth crust. They generally enter the human body through the food chain (Rafat and Fawzia, 2013). Measurements of natural radioactivity in environmental elements have been carried out in different countries to establish baseline data from the natural radiation levels (Ahmad et al., 2015). The data on the radioactivity of radium, thorium, and potassium in food are directly related with the safety of population; therefore, this study aimed to provide the basic radiometric data of radioactive in the grain food. The primary purpose of this study is to determine the activity concentration levels of ^{226}Ra, ^{232}Th, and ^{40}K in the different types of grain that are available in Penang markets, Malaysia, to ensure that food safety is not compromised and the effective doses due to ingestions are within the specified safety limits. Several studies have been performed in different countries to determine the radionuclide concentration in different food samples and dose assessment

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from consumption of that foodstuff by the population (Awudu et al., 2012). As grain and its products are the main component of daily serving such as breads, rice, and pasta, it is considered as a staple food. The levels of radioactive materials in some grains consumed by population need to be carefully measured so as to forecast any possible associated radiological risk.

II. Methodology

A. Sample Collection and Preparation

In the present study, to determine grain radionuclides' concentration, an experiment was carry out on 18 types of grain samples collected from local markets of Penang city, Malaysia. Afterward, the samples were immediately brought to laboratory within 1 day to prepare the grain samples and to keep them accordingly. Each sample was crashed into fine powder form by blender and passed through sieve with mesh to produce particle sizes of <0.249 mm and hence to obtain uniform sample powder (homogenous), which is in line with the study of Shafaei et al., 2011. 300 g of each grain sample was weighted using electrical balance; then, the samples put in small plastic tube and sealed to prevent the leakage of radon gas and then stored separately for 1 month to allow radioactive equilibrium stage between ^{226}Ra and ^{232}Th and its short-lived decay products before performing radioactivity measurements (Murat et al., 2010; Usikalu et al., 2014). Then, the radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K in grain samples were measured using high-purity germanium spectroscopy (HPGe detector) (Bashir et al., 2012).

B. Statistical Analysis

Statistical descriptions were performed using SPSS (Statistical Package for the Social Sciences) for Windows, standard version 22.0. Analysis of the data was carried out by frequency distributions (Pearson correlation) to assess the statistical significance in the three radionuclides measured in the grain samples.

C. Gamma Spectrometry Analysis

In the present work, the measurements of natural radioactivity levels were performed by gamma-ray spectrometry, using a HPGe detector connected to a multichannel analyzer. A high power supply that generates a high voltage (0–1500 V) to the detector through an amplifier at 1332 keV of ^{60}Co source, having ability to differentiate the gamma-ray energies, was utilized which is in agreement with the study of Gordana et al., 2015. The gamma spectrometry was shielding by a thick shield (5 cm) of lead encasing the HPGe detector (the inner diameter is 10 cm, and height is 50 cm). The background radiation was determined using an empty container with dimensions similar to that of the samples. The analysis was fixed at the duration of 86,400 s to produce a gamma spectrum that is agree with previous studies (Mohammed et al., 2015; Augustine et al., 2015). The samples were then placed on the top of the detector and were counted for 86,400 s in an attempt to attain minimum counting error in accordance with the study of Matthew et al., 2015.

D. Efficiency and Energy Calibrations

In this study, before the analysis of the samples, the calibrations of gamma energy and efficiency calibration for the system were performed using standard sources from the International Energy Agency IAEA, such as ^{60}Co, ^{137}Cs, ^{22}Na, ^{241}Am, and ^{226}Ra, that is in agreement with the study of Hossain et al., 2012. Certified standards of known activities were used to derive the calibration curve for energy and the efficiency of the HPGe detector since the efficiency is an important parameter of HPGe detector (Khandaker, 2011). The efficiency calibration curve of HPGe detector is shown in Fig. 1. Furthermore, the absolute efficiency of the HPGe detector for gamma-ray energies was calculated using the following equation (Njinga et al., 2015):

$$\varepsilon = \frac{CPS}{A_t \times I_\gamma} \times 100\% \quad (1)$$

Where CPS is counts per second, A_t presents activity of the source, and Iγ is gamma-ray intensity per decay.

The analyses of radionuclides of the grain samples were carried out based on the energy peaks of the progenies. The concentrations of the decay products of ^{214}Pb (295.224 keV, 18.7% and 351.932 keV, 35.8%) and ^{214}Bi (609.312 keV, 45%; 1120.287 keV, 14.8%; and 1764.494 keV, 15.65%) were taken to indicate^{226}Ra, whereas the specific activity of ^{232}Th has been calculated based on the energy peaks of ^{212}Pb (238.632 keV, 47.3%), ^{228}Ac (911.204, 29% and 968.971 keV, 17.5%), and ^{208}Tl (583.191, 84.5%), but the activity concentration of ^{40}K was assessed directly from its gamma-ray peak of 1460.83 KeV, 10.67% (IAEA, 1989) which is in compliance with studies made by Darwish et al., 2015; Raymond et al., 2016; and Mohammad et al., 2015. After correcting the background, the concentrations of the radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K of the grain samples were calculated by subtracting the area of prominent gamma-ray energy from the background radiation using the following equation (Nisar, 2015; Banzi et al., 2017):

$$\text{Concentration (Bq.kg}^{-1}) = (C-C_{background})/tp_\gamma \varepsilon w \quad (2)$$

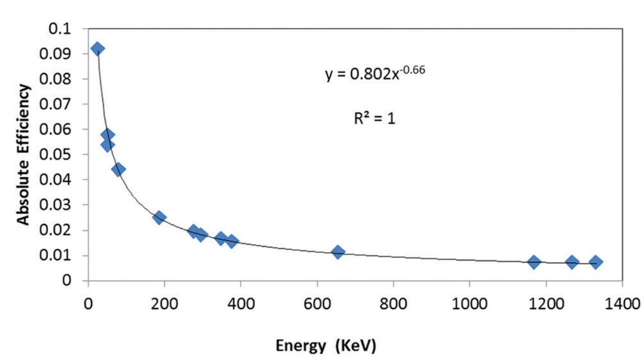

Fig. 1. Efficiency calibration curve for the high-purity germanium detector.

Where C is net area under peak, $C_{background}$ is net area of background radiation, t is time of counting (sec), P_{γ} is the absolute transition probability, ε is detector efficiency for the corresponding peak, and w is weight of the grain sample in kg (Njinga et al., 2015).

III. Calculation Of Concentration of Radionuclide and Hazard Indices

A. Concentration of Radionuclides

The concentration of radionuclides of ^{226}Ra, ^{232}Th and ^{40}K in a unit of $Bq.kg^{-1}$ has been calculated using the relation in the study of Murtadha et al., 2017, and Nisar, 2015.

B. Hazard Indices

Assessment of radiological hazard

The relationship between radiation risk and natural radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K can be determined by different radiation hazard indices. In the presented study, three hazard indices were considered, which are as follows (Okeme et al., 2017):

Radium equivalent activity: The radium equivalent activity (Raeq), which is a single index, used to describe the gamma output from different mixtures of radium, thorium, and potassium in the material. It was calculated from the following equation (Nisar, 2015; Al-Hamed et al., 2017):

$$Raeq = C_{Ra}+1.43C_{Th}+0.077C_{K} \quad (3)$$

Where C_{Ra}, C_{Th}, and C_{K} are activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

Alpha index: The excess alpha radiation due to the radon inhalation originating from the grain samples is assessed through alpha index, and it was <1. Alpha index ($I\alpha$) was calculated according to the following equation (Gordana et al., 2015).

$$I\alpha = C_{Ra}/200 \quad (4)$$

Annual ingestion dose: The annual ingestion dose (E_{ING}) for human was coming from consumption of grain, owing to the ingestion of radionuclides.The average consumption of grain product is (3.3) kg y^{-1} that was reported by the United Nations Scientific Committee on Effects Atomic Radiations in 2000 (UNSCEAR, 2000). Therefore, 3.3 kg per year has been considered for the estimation of radiation dose to the adult population in Penang (Kritsananuwat et al., 2014). The E_{ING} was calculated using the following equation (Murtadha et al., 2017; Rafat and Fawzia, 2013; Adjirackor et al., 2014):

$$E_{ING} = A_{I}\times C\times FDC_{ING} \quad (5)$$

Where E_{ING} is the annual ingestion dose (μSv y^{-1}), A_{I} is the activity concentration ($Bq.kg^{-1}$) of the investigated radionuclides in the vegetables, C is the consumption rate (3.3) kg y^{-1} (Kritsananuwat et al., 2014; Ononugbo et al., 2017) depending on the type of samples, and FDC_{ING} is the ingestion dose coefficient of the ^{226}Ra, ^{232}Th, and ^{40}K which was 0.2, 0.23 (μSv Bq^{-1}), and 6.2 (nSv Bq^{-1}), respectively (Murtadha et al., 2017).

IV. Results and Discussion

The concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were successfully measured through gamma-ray spectrometry in different types of grain. Eighteen samples of grain (different types) were analyzed using high resolution gamma-ray spectrometry with high pure germanium detector. The concentration distribution of ^{226}Ra, ^{232}Th, and ^{40}K in the samples found in $Bq.kg^{-1}$ was divided among various sources which include cereals: Wheat, Oats, rice, maize, kamut, buckwheat, barley, rye, and millet and legumes: Clover, alfalfa, beans, mesquite, lentils, peas, soybeans, lupins, and carob, as given in Table I. The radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K in grains are not uniformly distributed, and hence, the radionuclide concentration (Raeq) in $Bq.kg^{-1}$ is used to compare the specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th, and ^{40}K. It was calculated using the formula given by Equation (2). The concentration of ^{226}Ra, ^{232}Th, and ^{40}K in grain samples ranged between minimum and maximum values as follows: 56.97 $Bq.kg^{-1}$ and 86.13 $Bq.kg^{-1}$, 34.71 $Bq.kg^{-1}$ and 52.14 $Bq.kg^{-1}$, and 517.05 $Bq.kg^{-1}$ and 997.59 $Bq.kg^{-1}$, respectively. Table I also shows that the maximum values of concentration were found in^{40}K among the three natural radionuclides studied in the grain samples. The mean concentration was observed to be highest for ^{40}K, followed by ^{232}Th, and the lowest mean concentration was for ^{226}Ra (766.44>72.03>46.38) because ^{40}K is an essential element for living organisms; therefore, the ^{40}K radioactivity cannot be avoided.

The concentration for natural radionuclides indicates that concentrations for all radionuclides are higher than

TABLE I
The Concentration of ^{226}Ra, ^{232}Th, and ^{40}K in Grains, Cereals, and Legumes Samples

Code of samples	Types of grain samples	Specific activity of ^{226}Ra, ^{232}Th, and ^{40}K (Bq.kg−1)		
		^{226}Ra	^{232}Th	^{40}K
	Cereals			
GS01	Wheat	64.87	49.57	517.05
GS02	Oats	70.52	48.44	995.90
GS03	Rice	86.13	51.82	618.13
GS04	Maize	67.12	47.61	992.81
GS05	Kamut	66.87	52.14	809.32
GS06	Buckwheat	71.21	50.34	814.59
GS07	Barley	56.97	34.71	986.02
GS08	Rye	66.81	48.25	643.52
GS09	Millet	73.69	41.81	602.06
	Legumes			
GS10	Alfalfa	77.09	43.51	855.70
GS11	Clover	68.41	44.21	611.36
GS12	Beans	72.72	43.85	954.44
GS13	Peas	78.82	47.13	620.18
GS14	Lentils	69.32	42.47	572.36
GS15	Mesquite	74.22	45.35	997.59
GS16	Carob	81.06	50.09	636.79
GS17	Lupins Carob	78.58	49.97	963.16
GS18	Soybeans	72.12	43.51	605.02
Ave		72.03	46.38	766.44
Mix		86.13	52.14	997.59
Min		56.97	34.71	517.05

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the world median values except for ^{226}Ra which is agreed with the previous study reported by Matthew et al., 2015. Lowest concentration of ^{226}Ra (56.97 Bq.kg^{-1}) and ^{232}Th (34.71 Bq.kg^{-1}) was found in Barley samples, but Barley sample has highest radionuclides of ^{40}K (986.02 Bq.kg^{-1}). Therefore, the lowest concentration of ^{226}Ra was found in Barley (56.97 Bq.kg^{-1}) and highest concentration of ^{226}Ra was found in rice (86.13 Bq.kg^{-1}) with the average concentration of ^{226}Ra (72.03 Bq.kg^{-1}), and the lowest concentration of ^{232}Th was found in Barley (34.71 Bq.kg^{-1}) and highest concentration of ^{232}Th was found in kamut (52.14 Bq.kg^{-1}) with the average concentration of ^{232}Th (46.38 Bq.kg^{-1}); furthermore, the lowest concentration of ^{40}K was found in wheat (517.05 Bq.kg^{-1}) and highest concentration of ^{40}K was found in mesquite (997.59 Bq.kg^{-1}), with the average of concentration for ^{40}K (766.44 Bq.kg^{-1}). The mean concentration of ^{232}Th (46.38 Bq.kg^{-1}) was slightly higher than the World's average (30 Bq.kg^{-1}) (UNSCEAR, 2000). The highest activity concentration of ^{232}Th was found in kamut (52.14 Bq.kg^{-1}). The highest activity concentration of ^{226}Ra was found in rice (86.13 Bq.kg^{-1}) and the highest activity concentration of ^{40}K was found in the both mesquite (997.59 Bq.kg^{-1}) and oats (995.90 Bq.kg^{-1}), whereas lowest concentration of ^{232}Th was found in barley (34.71 Bq.kg^{-1}), lowest concentration of ^{226}Ra was found in barley (56.97 Bq.kg^{-1}), and lowest concentration of ^{40}K was found in wheat (517.05 Bq.kg^{-1}) among the samples when compared to other samples in this study, as shown in Table I and Fig. 2.

In Table II, the activity ratio of ^{232}Th- ^{226}Ra was ranged between 0.564 in alfalfa sample and 0.779 in kamut sample with an average value of 0.646, the activity ratio of ^{40}K- ^{226}Ra was ranged between 7.17 in rice sample and 17.31 in barley sample with an average value of 10.77, and the activity ratio of ^{40}K- ^{232}Th was ranged between 10.43 in wheat sample and 28.41 in barley sample with an average value of 16.74**.**

In addition, the external irradiation radon and its short-lived products are also hazardous to the respiratory organs. The alpha radiation producing from the grain is estimated through alpha index should be less than 1 and is calculated according to the equation (Awudu et al., 2012) and average value of Iα is 0.360 Bq.kg^{-1} that is <1, as shown in Table III, which is in agreement with the study of Gordana et al., 2015, because the recommended limit concentration of ^{226}Ra is 200 Bq.kg^{-1} for which I_a=1 (Gordana et al., 2015).

It is clear from the results of Table IV that the average annual ingestion dose of natural radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K was 66.555, 35.199, and 15.328 µSv y^{-1}, respectively. These obtained values are significantly below the total worldwide annual effective ingestion dose of ^{226}Ra, ^{232}Th, and ^{40}K that was 290 µSv y^{-1}, reported by UNSCEAR, 2000, and Kritsananuwat et al., 2014; Asaduzzaman et al., 2014). Furthermore, the average annual ingestion dose of grain was found to be below the values recommended, 250–400 µSv y^{-1}, as reported by the WHO, 2011. The differences could be due to the variation in the consumption of the grain and the natural environment in these countries. However, the average total annual ingestion dose in the grain samples is low and, therefore, is not harmful to human health.

TABLE II
The Ratio of ^{232}Th, ^{226}Ra, and ^{40}K in the Grain Samples Under Study

Code of samples	Types of grain samples	The ratio of specific activity of ^{226}Ra, ^{232}Th, and ^{40}K		
		^{232}Th-^{226}Ra	^{40}K-^{226}Ra	^{40}K-^{232}Th
	Cereals			
GS01	Wheat	0.764	7.97	10.43
GS02	Oats	0.687	14.12	20.56
GS03	Rice	0.601	7.17	11.92
GS04	Maize	0.709	14.79	20.85
GS05	Kamut	0.779	12.11	15.52
GS06	Buckwheat	0.707	11.44	16.18
GS07	Barley	0.609	17.30	28.41
GS08	Rye	0.722	9.63	13.33
GS09	Millet	0.567	8.17	14.41
	Legumes			
GS10	Alfalfa	0.564	11.09	19.66
GS11	Clover	0.646	8.93	13.83
GS12	Beans	0.603	13.12	21.76
GS13	Peas	0.597	7.86	13.16
GS14	Lentils	0.613	8.25	13.47
GS15	Mesquite	0.611	13.44	21.99
GS16	Carob	0.618	7.85	12.71
GS17	Lupins	0.636	12.25	19.27
GS18	Soybeans	0.603	8.38	13.91
Ave		0.646	10.77	16.74
Mix		0.779	17.31	28.41
Min		0.564	7.17	10.43

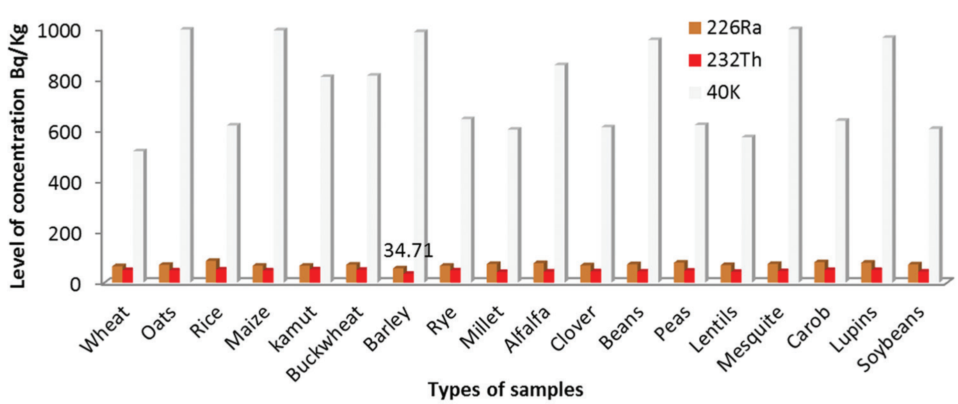

Fig. 2. The concentration of radionuclides in the samples as the function of the types of samples.

TABLE III
Radiation Hazard Indices Of Gamma Ray in the **Grain** Samples

Code of samples	Raeq (Bq.kg^{-1})	Iα (Bq.kg^{-1})
GS01	175.568	0.324
GS02	216.485	0.352
GS03	207.840	0.431
GS04	211.646	0.335
GS05	203.752	0.334
GS06	205.929	0.356
GS07	182.525	0.285
GS08	185.367	0.334
GS09	179.832	0.368
GS10	205.211	0.385
GS11	178.697	0.342
GS12	208.928	0.363
GS13	193.970	0.394
GS14	174.132	0.346
GS15	215.893	0.371
GS16	201.738	0.405
GS17	224.209	0.393
GS18	180.939	0.360
Ave	197.370	0.360
Mix	224.209	0.430
Min	174.131	0.285

TABLE IV
The Annual Ingestion Dose Estimate in the Grain Samples

Code of samples	Types of grain samples	Annual ingestion dose of ^{226}Ra, ^{232}Th, and ^{40}K (μSv y^{-1})		
		^{226}Ra	^{232}Th	^{40}K
	Cereals			
GS01	Wheat	59.939	37.623	10.341
GS02	Oats	65.160	36.765	19.918
GS03	Rice	79.584	39.331	12.362
GS04	Maize	62.018	36.136	19.856
GS05	Kamut	61.787	39.574	16.186
GS06	Buckwheat	65.798	38.208	16.299
GS07	Barley	52.640	26.345	19.720
GS08	Rye	61.732	36.621	12.870
GS09	Millet	68.089	31.734	12.041
	Legumes			
GS10	Alfalfa	71.231	33.024	17.114
GS11	Clover	63.210	33.555	12.227
GS12	Beans	67.193	33.282	19.088
GS13	Peas	72.829	35.771	12.403
GS14	Lentils	64.051	32.234	11.447
GS15	Mesquite	68.579	34.420	19.959
GS16	Carob	74.899	38.018	12.736
GS17	Lupins Carob	72.608	37.927	19.263
GS18	Soybeans	66.638	33.024	12.100
Ave		66.555	35.199	15.3289
Mix		79.584	39.574	19.959
Min		52.640	26.345	10.341

A. Correlation between Laboratory Data Analysis and the Radionuclides (^{226}Ra, ^{232}Th, and ^{40}K)

From Table V, it can be observed that positive correlation exists among the three radionuclides and annual ingestion dose. Pearson correlation showed significant strong positive correlations (1.000**, *p* value <0.001) for each annual ingestion dose of ^{226}Ra with the concentration ^{226}Ra, annual ingestion dose of ^{232}Th with the concentration ^{232}Th, and annual ingestion dose of ^{40}K with the concentration ^{40}K. This correlation among variables indicates similar source and behavior in the environment, but not significant correlations (*P* = 0.479) were found between ^{226}Ra and ^{40}K and no significant correlations (*P* = 0.603) were found between ^{232}Th and ^{40}K. Therefore, the correlation of^{226}Ra and^{232}Th was more stronger than the correlation of^{40}K, as shown in Table V.

TABLE V
Pearson Correlation Among Radionuclides Parameters and Laboratory Data

Laboratory data				
Variables	Correlations	^{226}Ra	^{232}Th	^{40}K
^{222}Ra concentration	Pearson correlation	1.000	0.459	−0.173
	P value	<0.001	0.055	0.479
^{232}Th concentration	Pearson correlation	0.459	1.000	−0.131
	P value	0.055	<0.001	0.603
^{40}K concentration	Pearson correlation	−0.173	−0.131	1.000
	P value	0.479	0.603	<0.001
Annual ingestion dose of ^{226}Ra	Pearson correlation	1.000**	0.459	−0.173
	P value	<0.001	<0.055	0.479
Annual ingestion dose of ^{232}Th	Pearson correlation	0.459	1.000**	−0.131
	P value	0.055	<0.001	0.603
Annual ingestion dose of ^{40}K	Pearson correlation	−0.173	−0.131	1.000**
	P value	0.479	0.603	<0.001

**Correlation is high significant at the 0.01 level (two-tailed), correlation is significant at the 0.05 level (two-tailed)

V. Conclusions

A study of natural radioactivity in the grain samples is usually done to gain information about the levels of harmful of radioactivity in environment and to understand the behavior of natural radionuclides. Therefore, the grain samples for eating are considered to be safe for inhabitants. It is suggested that the values reported in the current study can be considered as within the "normal level" of radiation and is below the worldwide averaged value. The baseline data of this type will almost certainly be of importance in making estimations of population exposure. The purpose of this study is to analyze the level of radioactivity of ^{40}K, ^{226}Ra, and ^{232}Th in the different types of grain that is available in Penang markets, Malaysia to determine the effective doses of ^{40}K, ^{226}Ra, and ^{232}Th in the samples due to ingestions which are within the specified safety limits. Hence, there is no radiological risk in the grain samples in the Penang market, and the results of this study indicated that radionuclide intake due to grain consumption has no consequence on public health. Hence, results concluded that all the three radionuclides contribute significantly to gamma-ray emission at the sampling points.

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