

Natural Radioactivity in Some Food Crops from Bangka-Belitung Islands, Indonesia

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ABSTRACT

Natural radioactivities of food crops are the main sources of internal radiation exposure in humans. Bangka Belitung islands of Indonesia has natural background radioactivity higher than normal area because of tin mining activities. The study was carried out to evaluate the natural radioactivity concentration in some food crops grown in Bangka and Belitung Islands. Food samples collected from Bangka and Belitung Islands were analysed by means of a gamma spectroscopy for natural radionuclides ^{226}Ra , ^{232}Th and ^{40}K . The annual intake of the food was estimated on the basis of their average annual consumption. Calculations were also made to determine the effective dose to an individual consuming such diets. The intakes of these radionuclides were calculated using the concentrations in Bangka Belitung foods and annual consumption rates of these food. Annual intakes of these radionuclides were as follows: ^{226}Ra = 190.00; ^{232}Th , 633.79 and ^{40}K = 2065.10 Bq/year. The annual internal dose resulting from ingestion of radionuclides in food was 0.205 mSv/year which is very much lower than annual dose limit of 1 mSv for general public. The radionuclides with highest consumption is ^{40}K followed by ^{232}Th and ^{226}Ra .

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INTRODUCTION

Bangka-Belitung is one of Indonesia provinces, includes two large islands, Bangka and Belitung, and several smaller ones, which located at east of Sumatra, northeast of South Sumatra province. Bangka-Belitung islands province is the 31st Province in Indonesia, one of the newest provinces [1]. Bangka and Belitung Islands is known to have the geological potential of mineral resources, especially tin, with accessory minerals consisting of monazite, zircon, xenotim, ilmenite, magnetite and pyrite spread in almost all regions. Meanwhile, there are some others minerals resources like: quartz sand, building construction sand, kaolin, granite, clay and mountain stone [2].

Tin mining is the most significant activity in Bangka-Belitung islands. Indonesia is the second world's largest tin producer after China. Most of Indonesia's tin production comes from the Bangka-Belitung islands. Tin mining and processing constitute a source of pollution to the environment. Environmental damage that occurred in the province of Bangka Belitung islands are very severe. This damage does not only occur on land but

also at sea. The land in Bangka Belitung is heavily damaged, with huge holes filled with standing turbid water everywhere. Bangka Belitung coast area is suffering from abrasion due to the number of holes at the bottom of the sea set off by coastal mining. The rapidly increasing number of illegal mining, is being as a large contributor to the destruction of the Bangka Belitung environment [3]. Heavy mineral processing from the tin mining exploration on earth's core has contributed a number of natural radioactivity to the environment. The mining, milling and processing of minerals lead to enhanced radiation exposures not only to the workers but also to the inhabitants of the mining and processing sites [4-7].

The analysis of radioactive minerals in foodstuff is an important part of the environmental monitoring program. The major pathways for the transfer of radionuclides to human beings are food crops. Therefore, the natural radionuclides are the largest contributor of the radiation doses received by the population who live around the tin mining activities area. Natural radionuclides are transferred and cycled through natural processes and between the various environmental compartments by entering into ecosystems and human food chains. Studies on radiation levels and radionuclide distribution in the environment provide vital

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radiological baseline information. Such information is essential in understanding human exposure from natural and man-made sources of radiation and necessary in establishing rules and regulation relating to radiation protection.

Radiation doses obtained due to the intake of food can be calculated from the amount of radionuclide deposited on foodstuff, the activity concentration of particular radionuclide in food per unit deposition, the consumption rate of the food products and the dose per unit activity ingested [8-10]. Measurement of concentrations of naturally occurring radionuclides (^{226}Ra , ^{232}Th and ^{40}K) in the food collected from Bangka and Belitung islands enables one to assess the radiation doses received by the population. With the tin mining exploration activities, the level of radiation exposure to the environment and people live in Bangka Belitung may be increase. Generally, tin leaves some natural radionuclides which easily transfers from the mining location to its surrounding environment which goes up to the land surface or is absorbed down into the land water surface.

Since waste of tin mining has become a serious issue in Bangka Belitung, knowledge of the natural radioactivity levels in food crops due to tin mining activity is an important one in the assessment of overall human exposure to natural radiation associated with ^{226}Ra , ^{232}Th and ^{40}K . The objective of this work is to investigate the natural radioactivity levels (^{226}Ra , ^{40}K and ^{232}Th) in some selected major food crops from Bangka-Belitung islands and evaluate the implication on population radiation ingestion dose. This study provide a database of natural radionuclides activity concentrations in major food crops samples from environment of Bangka Belitung islands. This database is useful for further usage and research such as for estimation of transfer factors of certain radionuclides to food crops and risk assessment to human life. The data produced from this study not only become an asset to the Bangka-Belitung province as it will serve as a reference level against future nuclear activities in the region, but also contribute to the Indonesia Radioactivity Database. The data produced from this study will serve as reference levels and will be a useful guide against any radioactive contamination from future nuclear activities in Bangka Belitung province.

EXPERIMENTAL METHODS

Sample collection

The collected samples are some food crops grown and commonly consumed by the people of

the Bangka Belitung islands. The vegetables, fruits and tuber samples were collected from local farmers and from vegetable gardens. Those samples are grown in the farms of this Bangka Belitung islands area and also available in the local markets. Each of samples was placed in the plastic bag and labeled with a permanent marker. All the samples were packed in a rigid plastic container and then transported to the environmental laboratory of the Center for Technology of Radiation Safety and Metrology for further treatments and measurement.

The fish samples were collected from local fishermen and local fish markets. The samples were cleaned to separate sands, weeds, and gravels, washed with water. Fish samples were generally gutted by the fishermen. All fish samples were then frozen and dispatched to the above Laboratory.

Sample preparation

In the laboratory, the vegetables and fruits and tuber samples were carefully washed under running water and the in distilled water to remove all the attached sand and dust particles. Those sample were cut to pieces with a knife and were allowed to dry to a constant weight at room temperature. The samples were then weighed and ashed at 450 °C to remove organic matter [11].

Fish samples were allowed to thaw out naturally. Samples were rinsed under a cold running tap to remove fish scales and any extraneous particulate material. Flesh and muscle parts were removed from bones, mixed in the chopper and prepared as samples. They were then filleted. The fillets were again rinsed to remove extraneous material, minced, dried to constant weight. Fish samples were weighed and dried in an oven at about 100 °C and reduced to ashes in a muffle furnace at 350 – 400 °C for about 24 h [12].

Radioactivity measurements

Each of the prepared sample was transferred to airtight cylindrical plastic screw-cap vial. The vials were sealed and allowed to stand for at least 4 weeks for secular equilibrium to be established between the long-lived parent nuclides of ^{226}Ra and ^{232}Th , and their short-lived daughters. The measurement of the radionuclides in the prepared samples were carried out by using ORTEC P-type coaxial high purity Germanium (HPGe) detector with a relative efficiency of 60% and a resolution of 1.95 keV (full width at half maximum) for the peak of 1.33 keV. The detector was coupled to a multi channel computer based

analyzer. The gamma ray spectrum is recorded using a Personal Computer based 4096 channel analyzer and processed using ORTEC Vission-32 Gamma spectrum analysis computer software. The detector is located inside a cylindrical lead shield of 10.1 cm thickness with internal diameter of 28 cm and height of 40 cm. The lead shield is lined with various layers of tin and copper each of 0.5 and 1.6 mm thick respectively.

The detection efficiency calibration of the system for the determination of radionuclides in the prepared samples was carried out using certified standard source (^{152}Eu) and International Atomic Energy Agency (IAEA) reference materials prepared in geometrical shape and composition to simulate the samples' matrix. The measurements were carried out in the counting room located in the basement of laboratory building. The measurement time for samples and background was 17 hours. The background counts was used to correct the net peak area of gamma rays of measured isotopes. Quality assurance was additionally guaranteed by regular participations in national and international intercompararison exercises.

The gamma energy peaks 352 keV of ^{214}Pb and 609.31 keV of ^{214}Bi were used to determine ^{226}Ra . The gamma energy peaks of 238.6 keV from ^{212}Pb , 911.2 and 969 keV gamma energy peak from ^{228}Ac and 583 keV gamma energy peak from ^{208}Tl were used to determine the ^{232}Th , and that of ^{40}K was determined from the gamma energy peak of 1460.83 keV.

The activity concentrations (A) of ^{226}Ra , ^{232}Th and ^{40}K in Bq kg^{-1} for the samples were determined using the following expression [13,14] :

$$A = \frac{N_e}{\varepsilon_f P_\gamma t_c M} \quad (1)$$

where; N_e = net counts of a peak at energy E, ε_f = the counting efficiency of the detector system at energy E, P_γ = the gamma ray emission probability (gamma yield) at energy E, t_c = sample counting time, and M = mass of sample (kg) . If there is more than one peak in the energy analysis range for a radionuclide, then an attempt to average the peak activities is made. The results is then the weighted average radionuclide activity.

Calculation of effective dose

In order to assess the ingestion dose of radionuclides, it is necessary to determine the annual activity intake per inhabitant for each ingested

radionuclide. The intake of radionuclides by ingestion is well described by the ICRP Human Alimentary Tract Model [15,16]. To derive ingested dose, the ingested annual activity of each radionuclide is multiplied by its dose conversion coefficient [17-19]. Finally, the total ingestion dose is obtained by summing the contributions of all radionuclides.

The annual intakes (Q) for ^{226}Ra , ^{232}Th and ^{40}K from foodstuffs in the present work were determined by using the activity concentrations (A) in foods and the annual food consumption rates (F) by the Bangka - Belitung islands population based on the National Socio-Economic Survey, 2012 [20]. Estimation of the annual internal effective dose (H) was calculated by using the following equations [17-19] :

$$H = A \times F \times \text{IDCF} \quad (2)$$

where H is the annual effective dose (Sv y^{-1}) due to ingestion of radionuclides from the consumption of foodstuffs, A is the concentration of radionuclides in the ingested foodstuffs (Bq kg^{-1}), F is the annual intake of foodstuff (kg y^{-1}), and IDCF is the ingested dose conversion factor for radionuclides (Sv Bq^{-1}). IDCF values used for ^{226}Ra , ^{232}Th and ^{40}K are 0.28, 0.22 and $6.2 \times 10^{-3} \mu\text{Sv Bq}^{-1}$ respectively.

RESULTS AND DISCUSSION

Radionuclides concentrations in major foods from Bangka Belitung

The results of the activity concentration measurements in the vegetables, fruits and tuber collected from Bangka Belitung province are presented in Table 1. Among the all food crops types collected and analyzed, the activity concentration of ^{226}Ra varies between $2.69 \pm 0.51 \text{ Bq/Kg}$ (Swamp cabbage) and $12.18 \pm 2.63 \text{ Bq/Kg}$ (Cassava leaf). The ^{232}Th activity in food crops varies between $8.20 \pm 1.09 \text{ Bq/Kg}$ (Swamp cabbage) and $46.00 \pm 3.53 \text{ Bq/Kg}$ (Jackfruit). The activity concentration of ^{40}K ranged from $40.12 \pm 3.81 \text{ Bq/Kg}$ (Cucumber) to $125.23 \pm 11.80 \text{ Bq/Kg}$ (Swamp cabbage). The concentration of ^{40}K was found to be very high compared to ^{226}Ra and ^{232}Th . It is may be attributed to poor migration characteristics of radium and thorium from the soil to the vegetables in the concerned environment [10]. Potassium is a macronutrient, so the concentration may be high. It may be expected that the soil characteristics favour the mobilization of potassium and its subsequent migration into the plant.

The level of ^{40}K in Jackfruit was 65.83 ± 3.93 Bq/Kg, and 85.27 ± 8.04 Bq/Kg for Banana and for cassava 40.40 ± 5.79 Bq/Kg. The activity concentration of ^{40}K was more in vegetables and fruits when compared to tubers (cassava).

Table 1. Activity concentration of natural radionuclides in vegetables, fruits and tuber.

Type of sample	Activity concentration (Bq/kg fresh-weight)		
	^{226}Ra	^{232}Th	^{40}K
Vegetables			
Spinach	7.47 ± 0.50	19.91 ± 2.72	69.80 ± 6.59
Swamp cabbage	2.69 ± 0.51	8.20 ± 1.09	125.23 ± 11.80
Mustard greens	2.77 ± 0.65	8.82 ± 1.18	110.56 ± 10.42
String beans	11.35 ± 1.62	26.37 ± 3.96	64.31 ± 6.12
Cucumber	9.55 ± 1.20	18.95 ± 2.15	40.12 ± 3.81
Cassava leaf	12.18 ± 2.63	39.19 ± 1.31	42.81 ± 10.19
Fruits			
Jackfruit	10.85 ± 1.18	46.00 ± 3.53	65.83 ± 3.93
Banana	5.45 ± 1.48	18.60 ± 3.29	85.27 ± 8.04
Tuber			
Cassava	3.68 ± 0.80	22.10 ± 2.70	40.40 ± 5.79

Nine types of fish samples were investigated for ^{226}Ra , ^{232}Th and ^{40}K and the results are listed in Table 2. It was found that the maximum level of ^{226}Ra was 11.11 ± 1.44 Bq/kg detected in Mozambique tilapia, while the minimum level of < 0.01 was detected in Milkfish. ^{232}Th was detected in all fish samples with the maximum level of 44.62 ± 4.41 Bq/kg detected in Mozambique tilapia, while the minimum level of 0.23 ± 0.05 Bq/kg was detected in Red snapper. The levels of ^{40}K in fish samples were distributed from the lowest level to the highest level. The highest concentration was 90.84 ± 8.61 Bq/kg, recorded in Mud crab, followed by Milkfish and Shrimp with values of 64.58 ± 6.20 Bq/Kg and 57.60 ± 5.57 Bq/Kg respectively. The low concentration were 15.02 ± 1.45 Bq/Kg and 20.97 ± 2.01 Bq/Kg, detected in Cockle and Catfish respectively. Mozambique tilapia was found as dominant type of fish in absorb of ^{226}Ra , ^{232}Th and also ^{40}K . This is due to the fact that the Mozambique tilapia is type of freshwater fish collected from the river. Most tin-mining areas have rich monazite and xenotime. These two minerals have rich natural radionuclides, particularly uranium, and thorium series and potassium such as ^{226}Ra , ^{232}Th and ^{40}K . Therefore, rivers passing through these areas would carry in their load traces of these radionuclides. Then the radionuclides could enter the food chain in the aquatic system which involves the phytoplankton (algae), zooplankton (microorganism) and other aquatic life such as Mozambique tilapia [21].

Table 2. Activity concentration of natural radionuclides in fish.

Type of sample	Activity concentration (Bq/kg fresh-weight)		
	^{226}Ra	^{232}Th	^{40}K
Yellow tail	0.80 ± 0.11	0.46 ± 0.10	38.83 ± 3.89
Mackerel	0.36 ± 0.01	0.74 ± 0.14	45.24 ± 12.10
Milkfish	< 0.01	17.81 ± 4.21	64.58 ± 6.20
Mozambique tilapia	11.11 ± 1.44	44.62 ± 4.41	36.48 ± 3.51
Catfish	3.81 ± 0.71	2.81 ± 0.64	20.97 ± 2.01
Red snapper	0.12 ± 0.01	0.23 ± 0.05	47.60 ± 5.57
Shrimp	4.27 ± 1.04	18.36 ± 2.32	57.60 ± 5.57
Mud crab	0.17 ± 0.05	8.06 ± 1.54	90.84 ± 8.61
Cockle	0.10 ± 0.01	2.50 ± 0.74	15.02 ± 1.45

Annual radioisotopes intakes from Bangka and Belitung islands foods

Using the results of activities concentration of vegetables, fruits, tuber and fish presented in Tables 1 and 2 and the data on food consumption rate reported the Indonesia Agency of Statistics [20], the intake of radionuclides by the population of Bangka-Belitung province was estimated. The results are presented in Table 3 and 4. The corresponding food consumption rate used to evaluate the intake of radionuclides are also given in the tables. It can be seen from the tables that the radionuclides with the highest annual intake was ^{40}K (2065.10 Bq) followed by ^{232}Th (633.79 Bq) and ^{226}Ra (190.00 Bq). The food crops with the highest amount of intake of radionuclides is vegetable, followed by tuber, fruit and fish. The results show that vegetables predominantly absorb ^{40}K , ^{232}Th and ^{226}Ra radionuclides. This is due to the fact that vegetables have the capability to absorb elemental isotopic through root uptake and from the leaves as a result of atmospheric deposit. Among the all vegetables types analyzed, the highest amount of intake is Swamp cabbage, followed by Spinach and String beans, while the fish with the highest amount of intake is Shrimp, followed by Mozambique tilapia and Milkfish.

Table 3. The annual radionuclide intake for ^{226}Ra , ^{232}Th and ^{40}K from vegetables, fruits and tuber.

Type of food	Consumption rate ^(a) (kg y ⁻¹)	Activity concentration (Bq)		
		^{226}Ra	^{232}Th	^{40}K
Vegetables				
Spinach	3.80	28.39	75.66	265.24
Swamp cabbage	4.47	12.02	36.65	559.78
Mustard greens	1.46	4.04	12.88	161.42
String beans	2.76	31.33	72.78	177.50
Cucumber	1.56	14.90	29.56	62.59
Cassava leaf	2.65	32.28	103.85	113.45
Fruits				
Jackfruit	1.20	13.02	55.20	79.00
Banana	1.61	8.77	29.95	137.28
Tuber				
Cassava	3.48	12.81	76.91	140.59
Total intake (Bq)		157.56	493.44	1696.84

(a) according to a survey conducted by the Indonesia Agency of Statistics for Bangka-Belitung province.

Table 4. The annual radionuclide intake for ²²⁶Ra, ²³²Th and ⁴⁰K from fish.

Type of food	Consumption rate (kg y ⁻¹)	Activity concentration (Bq)		
		²²⁶ Ra	²³² Th	⁴⁰ K
Yellow tail	0.36	0.29	0.17	13.98
Mackerel	0.16	0.06	0.12	7.24
Milkfish	1.14	-	20.30	73.62
Mozambique tilapia	1.25	13.89	55.78	45.60
Catfish	1.09	4.15	3.06	22.86
Red snapper	0.21	0.03	0.05	10.00
Shrimp	3.28	14.01	60.22	188.93
Mud crab	0.05	0.01	0.40	4.54
Cockle	0.10	0.01	0.25	1.50
Total intake (Bq)		32.44	140.35	368.26

Annual internal dose

Using the activity intake results presented in Table 3 and 4, the annual effective internal dose due to the ingestion of ²²⁶Ra, ²³²Th and ⁴⁰K radionuclides are calculated. The results are given in Table 5 and 6. The recommended dose conversion coefficient for ²²⁶Ra, ²³²Th, ⁴⁰K are 0.28, 0.22 and 0.0062 μSv/Bq, respectively [17-19]. The total internal dose of ²²⁶Ra, ²³²Th, ⁴⁰K in Bangka Belitung foods were estimated to be 0.053, 0.139 and 0.013 mSv/year. The total annual internal dose from ingestion of food samples in this study was 0.205 mSv/year. It can be seen that this doses is lower than annual dose limit of 1 mSv for general public [19].

Table 5. The estimated annual internal dose for ²²⁶Ra, ²³²Th and ⁴⁰K from vegetables, fruits and tuber.

Type of food	Activity concentration (μSv)		
	²²⁶ Ra	²³² Th	⁴⁰ K
Spinach	7.948	16.645	1.644
Swamp cabbage	3.367	8.064	3.471
Mustard greens	1.132	2.833	1.001
String beans	8.771	16.012	1.100
Cucumber	4.171	6.504	0.388
Cassava leaf	9.038	22.848	0.703
Jackfruit	3.646	12.144	0.490
Banana	2.457	6.588	0.851
Cassava	3.586	16.920	0.872
Total	44.116	108.557	10.520

Table 6. The estimated annual internal dose for ²²⁶Ra, ²³²Th and ⁴⁰K from fish.

Type of food	Activity concentration (μSv)		
	²²⁶ Ra	²³² Th	⁴⁰ K
Yellow tail	0.081	0.036	0.087
Mackerel	0.016	0.026	0.045
Milkfish	-	4.467	0.456
Mozambique tilapia	3.889	12.271	0.283
Catfish	1.163	0.674	0.142
Red snapper	0.007	0.011	0.062
Shrimp	3.922	13.249	1.171
Mud crab	0.002	0.089	0.028
Cockle	0.003	0.055	0.009
Total	9.082	30.876	2.283

CONCLUSION

The study on the activity concentration of radionuclides ²²⁶Ra, ²³²Th, and ⁴⁰K in some food crops that are regularly consumed by the population of Bangka Belitung province show that the highest value of concentration was vegetables, followed by fruits and fish. Among the all vegetables samples, the highest activity concentration of radionuclides was Swamp cabbage and Mustard greens, followed by String beans, Spinach and Cassava leaf. According to this study, fruits have maximum radionuclides concentration compared to tubers (Cassava). Among the all fish samples, the highest activity concentration of radionuclides was Mozambique tilapia and Mud crab, followed by Shrimp.

Based on the activity concentration of radionuclides and the data on food consumption rate, the annual intakes of these radionuclides were estimated higher in Swamp cabbage, Spinach, String beans, Cassava leaf, Cassava, Shrimp and Mozambique tilapia. For Spinach, String beans, and Cassava leaf, the high radionuclides intake values were not only due to high radionuclides concentration but also due to the high consumption rate. Meanwhile, the estimation of annual effective internal dose received by the members of the public from consumption of these food crops, the main contributors to the intake internal dose was Cassava leaf, Spinach and String beans, followed by Shrimp and Mozambique tilapia.

The annual internal dose received by general public living in Bangka-Belitung islands area due to the consumption of the common vegetables, fruits, tuber and fish is 0.205 mSv/year. This dose is lower than annual dose limit of 1 mSv for general public.

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