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RADIOLOGICAL RISK ASSESSMENT FROM THE INTAKE OF VEGETABLES AND FRUITS IN MALAYSIA

(Penilaian Risiko Radiologi dari Pengambilan Sayur-Sayuran dan Buah-Buahan di Malaysia)

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 $\frac{\textbf{Abstract}}{\textbf{The activity concentration of natural radionuclides}} \ ^{226}\textbf{Ra,} \ ^{232}\textbf{Th and} \ ^{40}\textbf{K} \ \text{in vegetables and fruits in central zone of Malaysia were}$ measured using gamma spectrometry. This is for the purpose of assessing the radiological risk in term of ingestion dose and cancer risk. The total ingestion dose from the consumption of vegetables were found to be 0.469 ± 0.003 , 0.326 ± 0.005 and 0.232 ± 0.003 mSv/y for infants, children and adults respectively. Whereas the total ingestion dose from the consumption of fruits for infants, children and adults were 0.349 ± 0.005 , 0.283 ± 0.007 and 0.195 ± 0.004 mSv/y respectively. The estimated cancer risk from the consumption of vegetables and fruits were 8.13×10^{-4} and 6.83×10^{-4} respectively for adults. The results obtained for activity concentration, dose and risk were found to be below the internationally recommended level. This indicates that the vegetables and fruits consumed in Malaysia would not pose any significant radiological impact to the population.

Keywords: activity concentration, gamma spectrometry, ingestion dose, natural radionuclides, risk assessment

Abstrak

Kepekatan aktiviti radionuklid semulajadi ²²⁶Ra, ²³²Th and ⁴⁰K dalam sayur-sayuran dan buah-buahan di Malaysia bahagian tengah telah diukur menggunakan spektrometer gama. Ini bertujuan untuk menilai risiko radiologi dalam istilah dos dimakan dan risiko kanser. Didapati jumlah dos dimakan dari pengambilan sayur-sayuran adalah 0.469 ± 0.003, 0.326 ± 0.005 and 0.232 ± 0.003 mSv/tahun masing-masing bagi bayi, kanak-kanak dan dewasa. Manakala jumlah dos dimakan dari pengambilan buahbuahan bagi bayi, kanak-kanak dan dewasa adalah masing-masing 0.349 ± 0.005 , 0.283 ± 0.007 and 0.195 ± 0.004 mSv/tahun. Risiko kanser dari pengambilan sayur-sayuran dan buah-buahan bagi orang dewasa masing-masing dijangkakan sebanyak 8.13 × 10⁻⁴ dan 6.83 × 10⁻⁴. Kesemua hasil yang diperolehi dalam kajian ini bagi kepekatan aktiviti, dos dan risiko didapati berada di bawah aras yang dibenarkan oleh badan antarabangsa. Ini menunjukkan bahawa sayur-sayuran dan buah-buahan yang diambil di Malaysia tidak menimbulkan bahaya radiologi yang ketara terhadap manusia.

Kata kunci: kepekatan aktiviti, spektrometer gama, dos dimakan, radionuklid semula jadi, penilaian risiko

Introduction

The total annual radiation dose received by a member of the population each year is 3.01 mSv. About 79.73% of this number is contributed by natural radiation sources (2.4 mSv/y) and the rest 20.27% by man-made radiation sources (0.61 mSv/y) [1]. It is clear that the natural radiation sources is the main contributor to the total annual radiation dose. One component of this natural radiation sources is the ingestion dose (0.29 mSv/y) that are mainly contributed by 238 U and 232 Th series (0.12 mSv/y) and 40 K (0.17 mSv/y) [2].

Ingestion dose was received by human beings from the consumption of food contaminated with different radionuclides [3]. One of important food that can contributes to better health of human is vegetables and fruits [4]. According to Ministry of Health Malaysia, vegetables and fruits are categorized into the most second important food after rice and grains. Therefore, it is recommended to take these kind of food for at least 5 servings a day (approximately 400 g/d) [5].

Since the amount of recommended intake is quite big, it is important to monitor the radiological effect from the consumption of vegetables and fruits. This matter has become a great concern as has been reported by many researchers in different part of the world. From eleven different countries, 58 types of vegetables have been studied with seven common types namely spinach, carrot, cucumber, tomato, pumpkin, green bean and potato [6-16]. However in Malaysia, only 16 types of vegetables have been studied with spinach and cucumber as the common type [17, 18]. Studies conducted in ten different countries reported that 26 types of fruits have been studied with eight common types namely apple, pear, orange, mango, banana, papaya, grapes and watermelon [6-16]. Whereas in Malaysia, only nine types of fruits have been studied i.e. apple, orange, mango, banana, papaya, star fruit, pineaple, rambutan and durian [17].

The present work is intended to (a) to determine the activities of ²²⁶Ra, ²³²Th and ⁴⁰K; (b) to assess the ingestion doses (due to these three radionuclides) incurred to the infants (1–2 years), children (7–12 years) and adults 17 years), (c) to compare these activities and doses with the previous local and international results, and (estimate the consequent health risk due to the presence of those radionuclides in the vegetables and fruits. This work is also conducted to complete the precedent researches carried out by the authors on some food products such as milk and water [19, 20].

Materials and Methods

Sample collection

Twelve different samples of commonly eaten vegetables and fruits were collected. All the samples were purchased from the local market in central zone of Malaysia. All vegetables and three fruits sample (banana, papaya and mango) were locally produced in Malaysia whereas the remaining three fruits sample i.e. apple, orange and pear were imported from China, Korea and Australia respectively.

Sample preparation

All vegetables and fruits samples were prepared following the IAEA procedure [21]. They were made into powder to follow the geometry of standard silica powder (4 cm height in 10 cm polyethilene beaker). First, they were washed and minced to get a small size. The unused part such as skin and seeds were discarded. Samples were then oven dried at 80 °C for 48 hours or until they reached constant mass. The dried sample were pulverized and sieved until they become 500 µm homogen powder. The powdered samples were then kept in a sealed tight polyethilene beaker for 30 days to achieve secular equilibrium. For every types of samples, three samples were prepared to obtained the average activity concentration. This means, 36 samples (= 12×3 samples) has been prepared for these two categories of samples.

 226 Ra, 232 Th and 40 K measurement The radionuclides 226 Ra, 232 Th and 40 K count, N in the vegetables and fruits sample were measured using gamma ray spectrometry system with high purity germanium detector (HPGe). The detector was connected to MCA card (Accuspec B) and Genie-2000 analysis software. The detector energy resolution was 1.8 keV at 1.33 keV of ⁶⁰Co gamma ray peak and the relative efficiency was 30% at the same energy peak [22]. Each measurement was carried out for 43,200 second (12 hours). The count of each radionuclides were resulted from the line energy peaks emited directly by the corresponding radionuclide (1460 keV for ⁴⁰K) or by the radionuclides daughter (352 and 609 keV for ²²⁶Ra and 239, 583 and 911 keV for ²³²) [17]. The equation 1 used to calculate the activity concentration, A and its uncertainty was expressed below [21, 23]:

$$A = \frac{N}{\varepsilon \times t \times P_{\nu} \times M_{S}} \tag{1}$$

where ε is the detector efficiency, t is measurement time, P_{γ} is the γ -ray emission probability (%) and $M_{\rm S}$ is the mass of the sample (kg). To conceal the minor photopeaks for each sample, the minimum detectable activity (MDA) was calculated using the formula originally defined by Currie [24].

$$MDA = \frac{2.71 + 4.66 \sigma}{t \times \varepsilon \times P_{\gamma}}$$
 (2)

where σ is the uncertainty of background count. The present measuring system recorded the MDA of 0.30, 0.34 and 2.88 Bq/kg for ²²⁶Ra, ²³²Th and ⁴⁰K respectively.

Dose and risk calculation

From the ²²⁶Ra, ²³²Th and ⁴⁰K activities, the ingestion dose D (mSv/y) can be calculated using the following relationship (equation 3) [25]:

$$D = A \times I \times E \tag{3}$$

where I(kg/y) is the annual intake of the vegetables and fruits [26] and E(Sv/Bq) is the ingestion dose conversion factor of the radionuclide [27]. The values of I and E for the three age categories are given in Table 1. The dose contributed from three radionuclides 226 Ra, 232 Th and 40 K were summed to obtain the total dose, D_T . If it is assumed that the life-span of an average individual, L is 70 years [28, 29] and the the cancer risk factor for low doses, RF is 5 \times 10⁻⁵ (1/mSv) [1], the cancer risk R can be estimated from:

$$R = D_T \times L \times RF \tag{4}$$

Table 1. The annual intake, I and the ingestion dose conversion factor, E for infants, children and adults

A so Cotosom	I (kg/	$E \times 10^{-8} \text{ Sv/Bq}$			
Age Category	Vegetables	Fruits	²²⁶ Ra	²³² Th	40 K
Infants (1–3 years)	12.96	23.01	96	45	4.20
Children (7–12 years)	25.92	46.02	80	29	1.30
Adults (> 17 years)	38.88	69.03	28	23	0.62

Results and Discussion
The measured activity concentration, A of 226 Ra, 232 Th and 40 K in six samples of vegetables and six samples of fruits, their ranges and their average values were shown in Table 2. It was shown that the activity concentration in some samples of vegetables and fruits were not detectable. For vegetables, the detected activity concentration ranged from 0.60 ± 0.57 to 5.64 ± 0.65 Bq/kg for 226 Ra, 0.41 ± 0.32 to 4.40 ± 0.49 Bq/kg for 232 Th and 398.85 ± 6.89 to 1072.59 ± 10.25 Bq/kg for 40 K. The average activity concentration of 226 Ra, 232 Th and 40 K were found to be 1.38 ± 0.21 , 1.06 ± 0.12 and 791.51 ± 3.66 Bq/kg respectively. For fruits, the detected ²²⁶Ra activity concentration ranged from 0.90 ± 0.64 to 3.83 ± 0.56 with an average value of 1.22 ± 0.17 , ²³²Th activity concentration ranged from 0.56 ± 0.48 to 2.22 ± 0.38 with an average value of 0.77 ± 0.12 and the ⁴⁰K activity concentration ranged from 75.39 ± 4.78 to 651.64 ± 9.75 with an average value of 288.63 ± 2.83 , all in Bq/kg.

For both types of sample, the highest activity concentration were found to be ⁴⁰K followed by ²²⁶Ra and ²³²Th. This same pattern has also been reported by many previous work as shown in Table 3. The activity concentration of previous results for vegetables samples ranged from 0.049 to 48.80, 0.001 to 25.36 and 0.79 to 1190.45 Bq/kg for ²²⁶Ra, ²³²Th and ⁴⁰K respectively. For fruits samples, they ranged from 0.009 to 8.15, 0.001 to 32.30 and 20.94 to 439.53 for ²²⁶Ra, ²³²Th and ⁴⁰K respectively. In comparison with this range, the present results are in agreement with the previous results. This is because all the averaged value of the present results lie clearly within the range of the previous results.

Table 2.	226 Ra.	²³² Th and ⁴⁰ K	measured activ	rity concentration.	A from	vegetables and fruits
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C	Name	Activity concentration, $A \pm \Delta A$ (Bq/kg)					
Sample	Name -	²²⁶ Ra	²³² Th	⁴⁰ K			
Vegetables	Spinach	5.64 ± 0.65	4.40 ± 0.49	998.04 ± 9.13			
	Carrot	ND^{a}	ND	398.85 ± 6.89			
	Broccoli	1.29 ± 0.62	ND	664.54 ± 8.75			
	Pumpkin	ND	0.41 ± 0.32	682.90 ± 6.92			
	Tomato	0.76 ± 0.63	ND	932.11 ± 11.02			
	Cucumber	0.60 ± 0.57	1.55 ± 0.44	1072.59 ± 10.25			
	Range	0.60 - 5.64	0.41 - 4.40	398.85 - 1072.59			
	Average	2.07 ± 0.31^{b}	2.12 ± 0.24^{b}	791.51 ± 3.66			
Fruits	Apple	2.58 ± 0.58	1.84 ± 0.42	151.49 ± 5.69			
	Banana	ND	ND	401.59 ± 5.85			
	Orange	ND	ND	279.74 ± 7.70			
	Pear	3.83 ± 0.56	2.22 ± 0.38	75.39 ± 4.78			
	Papaya	0.90 ± 0.64	0.56 ± 0.48	651.64 ± 9.75			
	Mango	ND	ND	171.95 ± 6.61			
	Range	0.90 - 3.83	0.56 - 2.22	75.39 - 651.64			
	Average	2.44 ± 0.34^{b}	1.54 ± 0.25^{b}	288.63 ± 2.83			

^{a)}ND = Not detectable (MDA for ²²⁶Ra, ²³²Th and ⁴⁰K were 0.30, 0.34 and 2.88 Bq/kg respectively). ^{b)}Due to the ND, respectively two and three out of six ²²⁶Ra and ²³²Th activities of vegetables and respectively three and three out of six ²²⁶Ra and ²³²Th activities of fruits were undetected. Therefore, this average was calculated from detected values only.

Table 3. The measured activity concentration, *A* in vegetables and fruits in comparison with the previous reported values

	Activity concentration (Bq/kg)					
	Vegetables Fruits					
²²⁶ Ra	²³² Th	⁴⁰ K	²²⁶ Ra	²³² Th	⁴⁰ K	[References]
_	_	93.29	-	_	86.93	Spain [6]
_	_	0.79	_	_	34.91	Syria [7]
0.18	0.20	48.15	_	_	_	Iran [8]
1	_	116.04	0.74	-	42.00	Egypt [9]
0.075	0.012	-	0.027	0.001	_	China [10]
_	_	74.30	_	_	52.90	Lebanon [11]
0.049	0.001	68.39	0.009	0.001	20.94	Korea [12]
0.51	1.33	57.64	0.46	1.62	69.88	India [13]
1.90	2.40	80.30	_	_	_	India [14]
2.60	2.92	163.40	1.30	1.56	111.00	Italy [15]
7.67	20.24	75.47	8.15	32.30	75.55	Indonesia [16]
1.22	1.95	1190.45	1.61	1.22	439.53	Malaysia [17]
48.80	25.36	1017.94	_	_	_	Malaysia [18]
0.049-	0.001-	0.79–	0.009-	0.001-	20.94–	Range of previous study
8.80	25.36	1190.45	8.15	32.30	439.53	[6-18]
2.07	2.12	791.51	2.44	1.54	288.63	Present study

	_	Ingestion Dose (mSv/y)						
Type	Age Category		$D \pm \Delta D$					
		²²⁶ Ra	²³² Th	⁴⁰ K	$D_T \pm \Delta D_T^{(a)}$			
Vegetables	Infants	0.026 ± 0.004	0.012 ± 0.001	0.431 ± 0.002	0.469 ± 0.003			
	Children	0.043 ± 0.006	0.016 ± 0.002	0.267 ± 0.001	0.326 ± 0.005			
	Adults	0.023 ± 0.003	0.019 ± 0.002	0.191 ± 0.001	0.232 ± 0.003			
Fruits	Infants	0.054 ± 0.008	0.016 ± 0.003	0.279 ± 0.003	0.349 ± 0.005			
	Children	0.090 ± 0.013	0.021 ± 0.003	0.173 ± 0.002	0.283 ± 0.007			
	Adults	0.047 ± 0.007	0.024 ± 0.004	0.124 ± 0.001	0.195 ± 0.004			

Table 4. Calculated ingestion dose, D from the intake of 226 Ra, 232 Th and 40 K in vegetables and fruits for three age categories

Upon comparing these two values, it was found that for vegetables: (a), the D due to intake of 226 Ra and 232 Th for three age categories were below the UNSCEAR annual ingestion dose. Hence, it was automatically below the annual radiation dose from natural sources, (b) only the D due to intake of 40 K for children and adults were below the UNSCEAR annual ingestion dose, whereas for infants, the D value were higher. However, it is still below the value of annual radiation dose from natural sources, (c) the D due to total intake of three radionuclides for infants and children are higher than the UNSCEAR annual ingestion dose by a factor of 1.57 and 1.04 respectively, however they are still below the value of annual radiation dose from natural sources.

The comparison results for fruits: (a) the D due to intake of 226 Ra, 232 Th and 40 K for three age categories were below the UNSCEAR annual ingestion dose. Hence, it was automatically below the annual radiation dose from natural sources, (b) the D due to total intake of three radionuclides were also below the UNSCEAR annual ingestion dose except for infants (higher by a factor of 1.08). However, it is still below the value of annual radiation dose from natural sources. To provide a better understanding of these results, the dose comparison were shown in Figure 1.

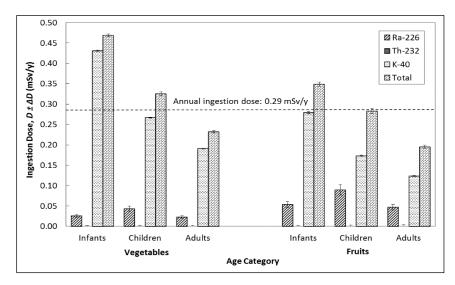


Figure 1. Ingestion dose of infants, children and adults from the intake of radionuclides 226 Ra, 232 Th and 40 K in vegetables and fruits. The dose contributed by 232 Th is in the range of 0.012 - 0.019 mSv/y for vegetables and 0.016 - 0.024 mSv/y for fruits, therefore they hardly can be seen.

 $^{^{(}a)}D_T = D_{\text{Ra-226}} + D_{\text{Th-232}} + D_{\text{K-40}}$

The D for infants and children for both vegetables and fruits were found to be higher than adults despite the lower value of intake I (Table 1). This is due to the higher value of E (ingestion dose conversion factor) for each radionuclide. The E value for infants were found to be higher by adults in the range of 1.96 - 6.77, whereas for children in the range of 1.26 - 2.86. Following the formula of $D = A \times I \times E$, for the same A, it makes sense that the D is higher when the E is higher.

The cancer risk R of 8.13×10^{-4} and 6.83×10^{-4} were estimated corresponding to the adult total ingestion dose of 0.232 and 0.195 mSv/y for vegetables and fruits respectively. These risk are significantly lower than the risk estimated by UNSCEAR from total natural radiation dose of 2.4 mSv/y i.e. 8.4×10^{-3} . The reduction by a factor of 10.34 and 12.31 respectively for vegetables and fruits deduces that the vegetables and fruits in Malaysia are safe for human consumption.

Another approach from Cohen and Lee [32] were also used to estimate the health risk posed from the intake of vegetables and fruits. The avoid confusion, these authors estimated the health risk in terms of day loss. It is estimated that an average loss of 9.9 days from continuos exposure of 1 mSv/y throughout life. Based on the total dose of 0.215 and 0.159 mSv/v, the estimated life expectancy for adults is shorten by only 2.13 and 1.57 days due to the consumption of vegetables and fruits respectively. It is therefore concluded that the vegetables and fruits consumed by Malaysian population would not pose any significant radiological impact on health and cancer risk to the population.

were found to be below natural radiation exposure reported by UNSCEAR. It was also comparable to published value reported by other researchers. The estimated risk has no significant health hazard as they were also below the UNSCEAR cancer risk factor. Therefore, it is concluded that the vegetables and fruits available in Malaysia are radiologically safe for human consumption. The data reported here will provide a baseline radiometric values for natural radionuclides in vegetables and fruits and also the radiological risk indices associating with them.

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