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Radionuclides Level and its Radiation Hazard Index in Some Drinks Consumed in the Central Zone of Malaysia

(Aras Radionuklid dan Indeks Bahaya Sinaran dalam Beberapa Minuman yang Terdapat di Zon Tengah Malaysia)

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ABSTRACT

Fifty three samples of different types of imported and locally produced drinks consumed in the central zone of Malaysia were analyzed using gamma-ray spectrometry system equipped with a high purity germanium (HPGe) detector. The activity concentrations of the natural radionuclides ^{238}U, ^{232}Th and ^{40}K present in the studied samples were measured and the radium equivalent activities Ra_{eq} were calculated. In addition, the radiation hazard index (HI) was calculated. The average concentrations of ^{238}U, ^{232}Th and ^{40}K were $0.69 \pm 7\times10^{-4}$, $1.3 \pm 8\times10^{-4}$ and $20.52 \pm 6\times10^{-4}$ Bq/Kg, respectively. The values of Ra_{eq} were between 0.002 and 10.0 Bq/kg. The HI were between 0.002 and 0.03, which is below one (the higher limit of HI). The results obtained were compared to the standard accepted international values and found to be within the acceptable limits.

Keywords: Activity concentration; NORM; radiation hazard index; radium equivalent; ^{238}U, ^{232}Th and ^{40}K

ABSTRAK

Lima puluh tiga sampel daripada pelbagai jenis minuman tempatan dan import yang terdapat di zon tengah Malaysia dianalisis menggunakan sistem spektroskopi sinar dengan pengesan germanium berketulenan tinggi (HPGe). Kepekatan radionuklid, tabii ^{238}U, ^{232}Th dan ^{40}K dalam sampel diukur dan setara radium Ra_{eq} dikira. Selain itu, indeks bahaya sinaran (HI) juga dikira. Kepekatan aktiviti ^{238}U, ^{232}Th dan ^{40}K dalam sampel berada dalam julat $0.69 \pm 7\times10^{-4}$, $1.3 \pm 8\times10^{-4}$ dan $20.52 \pm 6\times10^{-4}$ Bq/Kg. Nilai Ra_{eq} berada dalam julat antara 0.002 dan 10.0 Bq/kg. Indeks bahaya sinaran (HI) berada dalam julat antara 0.002 dan 0.03, kurang daripada satu (had atas HI). Keputusan yang diperoleh dibandingkan dengan piawai di peringkat antarabangsa dan didapati berada dalam julat yang boleh diterima.

Kata kunci: Indeks bahaya sinaran; kepekatan aktiviti; kesetaraan radium; NORM; ^{238}U, ^{232}Th dan ^{40}K

INTRODUCTION

^{238}U, ^{232}Th and ^{40}K are three long-lived naturally occurring radionuclides present in the earth crust. Generally, there are two sources of environmental radionuclide, natural (mainly from the ^{238}U, ^{232}Th series) and artificial (^{137}Cs) sources. These radionuclides can be released into the environment as a result of human activities including energy production and military operations such as nuclear weapons testing or caused by nuclear accidents (Chernobyl Disaster in 1986 and Fukushima Earthquake in 2011). Ionizing radiation is dangerous to the health, especially the charged particles and the high energy photon (Turner 1995). The main natural radioactive sources of ionizing radiation are the long lived ^{238}U, ^{232}Th and their decay series ^{40}K. The radiological hazards can be the consequences of external or internal exposure. Radionuclides can enter human body through inhalation and ingestion. The ingested radionuclides could be concentrated in certain parts of the body. For examples, ^{238}U accumulated in human lungs and kidney, ^{232}Th in lungs, liver and skeleton tissues and ^{40}K in muscles (Samat & Evans 2011). Depositions of large quantities of these radionuclides in particular organs will affect the health condition of the human such as weakening the immune system, induce various types of diseases and finally the increase in mortality rate.

The environmental radionuclides present the most risk to human health, so it is important to understand the transport, fate and effects of radionuclides moving through our drinks and foods. There are many types of drinks in Malaysia and the main components of daily serving are water, tea, coffee and chocolate drink. Generally, the central zone of Malaysia consumes a large amount of different type of drinks as shown in Table 1 (MHO 2006). Since naturally occurring radioactive materials (NORM) are present in all type of drinks commodities, the levels in some drinks consumed in central zone of Malaysia need to be established in order to forecast any possible radiological risk associated with the consumption of the drinks.

The main objectives of this research were to quantify the presence of natural radionuclides in some important

TABLE 1. Drinks consumption statistics for central zone of Malaysia

Type of drinks	EMI[1] ± SE[2] (g/day)
Plain water	1,565.07 ± 19.27
Tea	278.65 ± 7.96
Coffee	172.91 ± 6.58
Chocolate drinks	129.98 ± 5.17
Herbs drinks	90.53 ± 5.05
Carbonated drinks	64.96 ± 3.64
Fruit juices	57.95 ± 2.60
Soybean drinks	54.85 ± 2.12
Energy drinks [3]	44.14 ± 2.23
Fresh milk	37.94 ± 4.10
Sweetened condensed milk	34.11 ± 1.10
Yoghurt	16.86 ± 1.86
Wine [3]	8.62 ± 1.56
Water melon	8.3 ± 0.39

[1] Estimated mean intake
[2] Standard error of mean
Ministry of Health Malaysia (MHO) and Food Safety Quality. 2006
[3] These are imported sample

drinks consumed in the central zone of Malaysia and to estimate the radium equivalent index (Ra_{eq}) and hazard index (HI). These data can also be used as baseline on drinks radioactivity in Malaysia for future reference.

EXPERIMENTAL METHODS

MATERIALS

Fifty three types of drinks were taken as samples. All samples used were both local and imported drinks and were obtained from the local markets. The name of the samples and their number are as follows: drinking water (3), mineral water (3), isotonic drinks (4), tea (6), coffee (3), chocolate drinks (3), herbs drinks (3), carbonated drinks (3), fruit juices (4), soybean drinks (3), energy drinks (3), fresh milk (3), sweetened condensed milk (3) yoghurt (3), wine (3) and water melon (3). This makes the total number of samples collected as fifty three.

SAMPLE PREPARATION

The samples were prepared for the natural radioactivity measurement. Each liquid sample was weighted and sealed in 500 mL marinelli bottles and kept at room temperature (25°C) for at least 30 days before counting to allow reaching the secular equilibrium of ^{232}Th and ^{238}U with their respective decay products, in which the activities of all radionuclide within each series are nearly equal. The amount of samples counted is calculated by weighing the bottle without and with samples.

NATURAL RADIOACTIVITY MEASUREMENT

The measurement was conducted for 12 h using a Canberra p-type high purity germanium (HPGe) gamma spectrometer with 30% relative efficiency resolution of 1.8 keV at 1.33 MeV. The detector was connected to a computer with MCA card (Accuspec B) and Genie-2000 Analysis software of Canberra Industries, USA. A10-cm thick lead bricks shielded the detector from the background radiation from the radionuclides in the environment and cosmic rays. The system was calibrated using ^{241}Am, ^{109}Cd, ^{57}Co, ^{123}Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y,^{60}Co for their known energy (which covers the energy range from 60 keV to 1333 keV) and peak width of gamma-ray emission. The counting efficiency was determined previously for all of its counting geometry. Figure 1 shows the efficiency graph for standard water. The radionuclides were identified according to their individual photopeak, which were 609 keV (^{214}Bi) and 351.9 keV(^{214}Pb) for ^{238}U , 238 keV (^{212}Pb), 583.191, 510.80 keV (^{208}Tl) and 911keV(^{228}Ac) for ^{232}Th. 1460 keV for ^{40}K. The activity of ^{226}Ra during the equilibrium was assumed to be the same as its parent, ^{238}U. The specific activity for each radionuclide was calculated using equation proposed by Jabbar et al. (2009):

$$A_S = \frac{(C_s - C_b)}{t\, E_Y\, P_Y\, M_S}, \tag{1}$$

where A_S is the specific activity of each radionuclide in Bq/kg, C_s the count rate in cps for sample, C_b the count rate in cps for background, E_Y and P_Y are detection efficiency and emission probability of γ-ray, t is the counting time and M_S is the mass of the sample in kg.

RADIUM EQUIVALENT INDEX (Ra_{eq})

The radium equivalent activity (Ra_{eq}) concept allows a single index or number to describe the gamma output from different mixtures of ^{238}U (i.e. ^{226}Ra) ^{232}Th and ^{40}K in a material (Frame 2006). Ra_{eq} for each sample in Bq/kg, is calculated using the following formula proposed by UNSCEAR in Bq/kg (Yasir et al. 2007):

$$Ra_{eq} = A_{Ra} + 1.43\, A_{Th} + 0.077\, A_K, \tag{2}$$

where A_{Ra}, A_{Th} and A_K in Bq/kg are the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

HAZARD INDEX

This index is used to estimate the level of *Y*- radiation hazard associated with the natural radionuclides in grains samples. The hazard index (HI) is calculated from the equation (Tufail et al. 2000):

$$\mathrm{HI} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \leq 1. \tag{3}$$

RESULTS AND DISCUSSION

The activity concentration of ^{238}U (^{226}Ra) ^{232}Th and ^{40}K in Bq/kg present in the samples are given in Table 2. The average concentrations of ^{238}U, ^{232}Th and ^{40}K were 0.69 ± 7×10^{-4}, 1.3 ± 8×10^{-4} and 20.52 ± 6×10^{-4} Bq/kg, respectively.

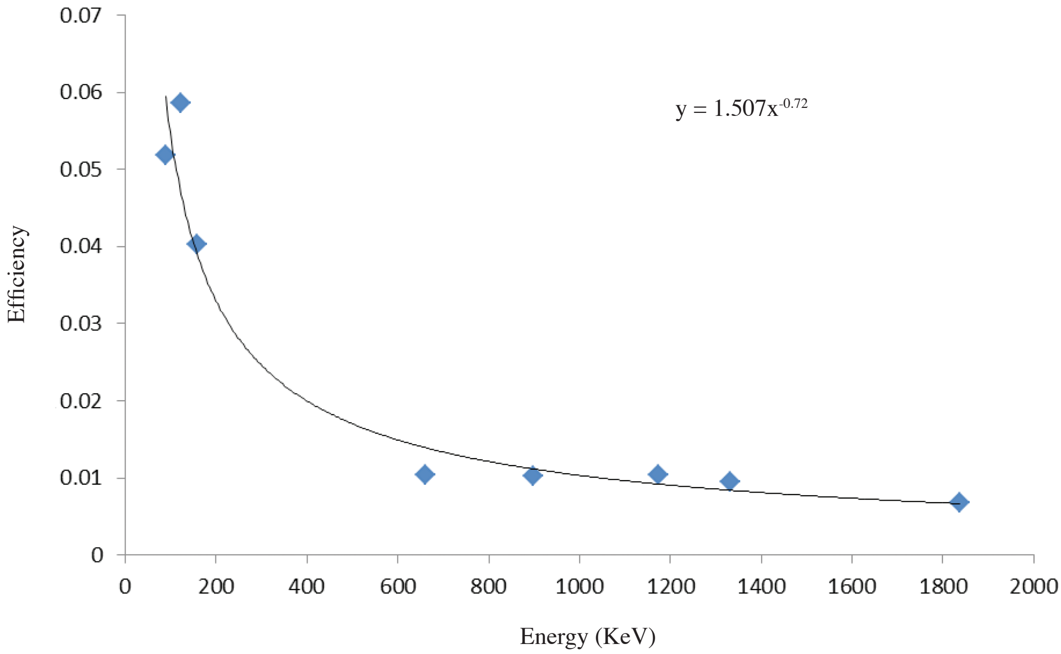

FIGURE 1. Efficiency graph for standard water on 12/04/2011 for software PCA

TABLE 2. Measured concentration activity of ^{238}U, ^{232}Th and ^{40}K in drinks types (Bq/kg)

Drinks type (n)	^{238}U	^{232}Th	^{40}K
Drinking water (3)	$1.3 \pm 8\times10^{-4}$	$5.75 \pm 8\times10^{-4}$	$6.18 \pm 6\times10^{-4}$
Mineral water (3)	$0.37\pm7\times 10^{-4}$	$0.87 \pm 9\times10^{-4}$	$11.7 \pm 7\times10^{-4}$
Isotonic drink (4)	$0.57\pm 7\times10^{-4}$	$0.10 \pm 8\times10^{-4}$	$3.64 \pm 6\times10^{-4}$
Tea (6)	$0.50 \pm 8\times10^{-4}$	$0.22 \pm 9\times10^{-4}$	$0.11 \pm 6\times10^{-4}$
Coffee / local (3)	$0.60 \pm 9\times10^{-4}$	$0.42 \pm 9\times10^{-4}$	$0.32 \pm 8\times10^{-4}$
Chocolate drinks (3)	$0.54 \pm 7\times10^{-4}$	$1.31\pm 9\times10^{-4}$	$5.93 \pm 7\times10^{-4}$
Herbs drinks (3)	$0.73 \pm 7\times10^{-4}$	$1.03 \pm 8\times10^{-4}$	$27.66 \pm 7\times10^{-4}$
Carbonated drinks (3)	$0.25 \pm 8\times10^{-4}$	$1.06 \pm 8\times10^{-4}$	$2.35 \pm 6\times10^{-4}$
Fruit juices (4)	$0.60 \pm 7\times10^{-4}$	$2.72 \pm 8\times10^{-4}$	$6.79 \pm 6\times10^{-4}$
Soybean drinks (3)	$0.97 \pm 7\times10^{-4}$	$0.92 \pm 8\times10^{-4}$	$26.07 \pm 7\times10^{-4}$
Energy drinks (3)	$0.55 \pm 9\times10^{-4}$	$0.4 \pm 9\times10^{-4}$	$5.07 \pm 7\times10^{-4}$
Fresh milk (3)	$0.95 \pm 8\times10^{-4}$	$1.85 \pm 9\times10^{-4}$	$50.12 \pm 8\times10^{-4}$
Sweetened condensed milk (3)	$1.05 \pm 8\times10^{-4}$	$1.22 \pm 8\times10^{-4}$	$92.44 \pm 1\times10^{-3}$
Yoghurt (3)	$0.57 \pm 8\times10^{-4}$	$1.47 \pm 8\times10^{-4}$	$51.92 \pm 7\times10^{-4}$
Wine (3)	$0.57\pm 7\times10^{-4}$	$0.87 \pm 8\times10^{-4}$	$11.83 \pm 7\times10^{-4}$
Water melon (3)	$0.90 \pm 1\times10^{-3}$	$0.75\pm 7\times10^{-4}$	$6.18 \pm 6\times10^{-4}$

The concentration of ^{238}U ranged from ($0.25 \pm 8\times10^{-4}$ to $1.30 \pm 8\times10^{-4}$ Bq/kg). The highest concentration of ^{238}U was found in drinking water, while the lowest concentration was found in carbonated drink. Studies reported by Quan et al. (2008) showed that the average concentration of ^{238}U in drinks was 0.02 Bq/kg, which is lower than the average concentration of ^{238}U in this study. They also showed that the concentration of ^{238}U were lower in fresh milk and wine compared with drinking water. The same results were found in this study which the mean concentration were $0.95 \pm 8\times10^{-4}$, $0.57 \pm 7\times10^{-4}$ and $1.3 \pm 8\times10^{-4}$, respectively. ^{238}U concentration is comparable with the results of other researchers and lies within the acceptable value, which is less than 20 Bq/kg for safe consumption as was reported in (UNSCEAR 2000).

The concentration of ^{232}Th ranged from $0.1 \pm 8\times10^{-4}$ to $5.75 \pm 8\times10^{-4}$ Bq/kg. The results indicated that the maximal uptake of ^{232}Th occurred mainly in drinking water and the minimal in isotonic drink. The result also showed that the measured concentration value of ^{232}Th content of fresh milk is 1.85 ± 8×10^{-4}, higher than the concentration value of ^{232}Th in fresh milk previously reported. For comparison, the concentration activity of ^{232}Th in fresh milk reported by Quan et al. (2008) and Laili et al. (2006) were found to be 0.02 and 0.5 Bq/kg, respectively. It was also observed that the average concentrations of ^{232}Th were higher than the average concentrations of ^{238}U. This is due to high concentration of ^{228}Ra in Malaysian soil compared with the concentration ^{226}Ra (Omar 1993). In general, the measured concentration values of ^{232}Th lies within the acceptable values as was reported by UNSCEAR (2000). Potassium is a very soluble element and an essential element in metabolism of living organism. The concentration of ^{40}K ranged from 0.1 ± 6×10^{-4} to 92.44 ± 1×10^{-3} Bq/kg. The highest activity concentration of ^{40}K was found in sweetened condensed milk (92.44 ± 1×10^{-3} Bq/kg), followed by Yoghurt (51.92 ± 8×10^{-4} Bq/kg) and fresh milk (50.12 ± 8×10^{-4} Bq/kg). The lowest concentration was found in tea with a value of 0.1 ± 6×10^{-4} Bq/kg. The levels detected in this study were slightly less than those published by UNSCEAR (2000), that ranges from 170 to 430 Bq/kg.

RADIUM EQUIVALENT INDEX (Ra_{eq})

Table 3 illustrates the calculated values of Ra_{eq} which ranged between 0.79 and 9.67 Bq/kg. Of all the 53 samples measured in this study, sweetened condensed milk has the highest concentrations of Ra_{eq} 9.67 Bq/kg, whereas carbonated drink showed the lowest concentration 0.79 Bq/kg. The average radioactivity concentration of Ra_{eq} was found to be 4.14 Bq/kg. All the values of Ra_{eq} were within the safety limits recommended by UNSCEAR (2000).

TABLE 3. The radium equivalent and hazard index of the studied samples

Drinks type	Ra_{eq} (Bq/kg)	HI
Drinking water	9.1	0.03
Mineral water	1.8	0.01
Isotonic drink	1.67	0.003
Tea	0.82	0.002
Coffee / local	1.23	0.003
Chocolate drinks	3.31	0.01
Herbs drinks	4.33	0.01
Carbonated drinks	0.79	0.005
Fruit juices	1.98	0.01
Soybean drinks	4.36	0.014
Energy drinks	1.51	0.004
Fresh milk	6.17	0.02
Sweetened condensed milk	9.67	0.03
Yoghurt	5.38	0.02
Wine	2.3	0.007
Water melon	2.45	0.007

HAZARD INDEX (HI)

As shown in Table 3, the HI ranged between 0.002 and 0.03. It is obviously clear that the values of HI were much lesser than the safe value, which is one (Tufail et al. 2000).

CONCLUSION

The results showed that the concentration of natural radionuclides of ^{238}U , ^{232}Th and ^{40}K in samples of different types of imported and locally produced drinks consumed in the central zone of Malaysia were relatively lower than the world recommended limits. The calculated radium equivalent and hazard index (HI) was also not exceeding the maximum suggested values. Thus, the drinks consumed in central zone of Malaysia do not pose any radiological complication.

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