

## RADIOLOGICAL IMPACT OF DRINKS INTAKES OF NATURALLY OCCURRING RADIONUCLIDES ON ADULTS OF CENTRAL ZONE OF MALAYSIA

(Impak Radiologi Radionuklid Tabii dalam Air Minuman ke atas Orang Dewasa di Kawasan Tengah Semenanjung Malaysia)

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### Abstract

Fifty three samples of different types of imported and locally produced drinks consumed in central zone of Malaysia were analyzed using gamma-ray spectrometry system. The measurement was conducted for 12 hours 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. The geometric means of daily intakes of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K were 0.05, 0.08 and 27.23 respectively. Also the values give annual committed effective doses of 0.8, 6.5 and 61.53  $\mu\text{Sv yr}^{-1}$  for <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K, respectively for population in central zone of Malaysia. The net radiological impact of these radionuclides is 68.83  $\mu\text{Sv yr}^{-1}$ . This value gives cancer risk factor of  $1.72 \times 10^{-7}$ . Also the value of net radiological impact gives loss of life expectancy of 0.43 days only. Whereas ICRP cancer risk factor for general public is  $2.5 \times 10^{-3}$  and total risk involve from the all natural radiation sources based on global average annual radiation dose of 2.4 mSv  $\text{yr}^{-1}$  is  $6.0 \times 10^{-3}$ . The estimated cancer risk shows that probability of increase of cancer risk from daily Malaysian drinks is only a minor fraction of ICRP values. Therefore the drink samples investigated here does not pose any significant health hazard and is considered radiologically safe for human consumption.

**Keywords:** <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K, naturally occurring radionuclides (NORM), drinks, dietary intake, radiation dose, cancer risk

### Abstrak

Lima puluh tiga sampel daripada pelbagai jenis minuman tempatan import yang terdapat di zon tengah Malaysia dianalisis menggunakan sistem spektroskopi sinar gama. Pengukuran ini dijalankan selama 12 jam menggunakan gamma spektrometer Canberra jenis-p germanium berketulian tinggi (HPGe) dengan resolusi kecekapan 30% relatif sebanyak 1.8 keV di 1.33 MeV. Pengesan dihubungkan ke komputer dengan MCA kad (Accuspec B) dan Genie-2000 Analisis perisian Canberra Industries, Amerika Syarikat. Cara geometri pengambilan harian <sup>238</sup>U, <sup>232</sup>Th dan <sup>40</sup>K 0.05, 0.08 dan 27.23 masing-masing. Nilai ini juga memberikan dos komited berkesan tahunan 0.8, 6.5 dan 61.53  $\mu\text{Sv yr}^{-1}$  untuk <sup>238</sup>U, <sup>232</sup>Th dan <sup>40</sup>K, masing-masing untuk penduduk di zon tengah Malaysia. Impak bersih radiologi dari radionuklid ini adalah 68.83  $\mu\text{Sv yr}^{-1}$ . Nilai ini memberikan faktor risiko kanser  $1.72 \times 10^{-7}$ . Juga nilai impak radiologi bersih memberikan kerugian jangka hayat 0.43 hari sahaja. Manakala faktor risiko kanser ICRP bagi orang awam ialah  $2.5 \times 10^{-3}$  dan risiko jumlah yang terlibat dari semua sumber radiasi semulajadi berdasarkan purata dos sinaran tahunan global 2.4 mSv  $\text{yr}^{-1}$  adalah  $6.0 \times 10^{-3}$ . Anggaran risiko kanser menunjukkan kebarangkalian peningkatan risiko kanser dari minuman harian di Malaysia hanyalah sebahagian kecil nilai ICRP. Oleh itu, sampel minuman yang dikaji di sini tidak menimbulkan sebarang bahaya kesihatan yang ketara dan dianggap selamat secara radiologi untuk penggunaan manusia

**Kata kunci:** <sup>238</sup>U, <sup>232</sup>Th dan <sup>40</sup>K, radionuklida semulajadi (NORM), minuman, ambilan diet, dos radiasi, risiko kanser

### Introduction

$^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  are three long-lived naturally occurring radionuclides present in the earth crust. Radionuclides such as  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  that occur naturally in soil are incorporated metabolically into plants and ultimately find their way into food and water [1]. Generally, there are two sources of environmental radionuclide, natural (mainly from the  $^{238}\text{U}$ ,  $^{232}\text{Th}$  series) and artificial (i.e  $^{137}\text{Cs}$ ) 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 (e.g 1986 Chernobyl Disaster and 2011 Fukushima Earthquake). Ionizing Radiation is dangerous to the health, especially the charged particles and the high energy photon [2]. The main natural radioactive sources of ionizing radiation are the long lived  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and their decay series and the  $^{40}\text{K}$ . The radiological hazard can be the consequence 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,  $^{238}\text{U}$  accumulated in human lungs and kidney,  $^{232}\text{Th}$  in lungs, liver and skeleton tissues and  $^{40}\text{K}$  in muscles. 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 increase in mortality rate. The environmental radionuclides present the most risk to human health, so it is important for us to understand the transport, fate and effects of radionuclides moving through the drinks and food. There are many types of drink in Malaysia. The main component of daily serving are such as water, tea, coffee, and chocolate drink. Generally, central zone of Malaysia consumes a large amount of different type of drinks as shown in Table1 [3]. Since naturally occurring radioactive materials (NORM) are present in all type of drinks commodities, the levels in some type of 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. Thus, the main objectives of this research are to quantify the presence of natural radionuclides in some important drinks consumed in central zone of Malaysia, and to determine impact of these radionuclides on human in normal and natural background conditions. Finally, to estimate the health detriment in terms of cancer risk.

Table 1. Drinks consumption statistics for central zone of Malaysia.

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

<sup>1</sup>Estimated mean intake, <sup>2</sup> Standard error of mean,

Source: Ministry of Health Malaysia (MHO) and Food Safety Quality. 2006.

## Materials and Methods

### Materials

Fifty three types of drinks were taken as the sample. All samples used were both local and imported drinks and were obtained from local markets. The name of the samples and their number are as follows: drinking water (3), mineral water (3), tea (6), coffee (3), chocolate drink(3), herbs drink (3), carbonated drink (3), fruit juices(4), soybean drink (3), energy drink (3), frish 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 in to allow reaching the secular equilibrium of  $^{232}\text{Th}$  and  $^{238}\text{U}$  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 weightless the bottle without and with samples.

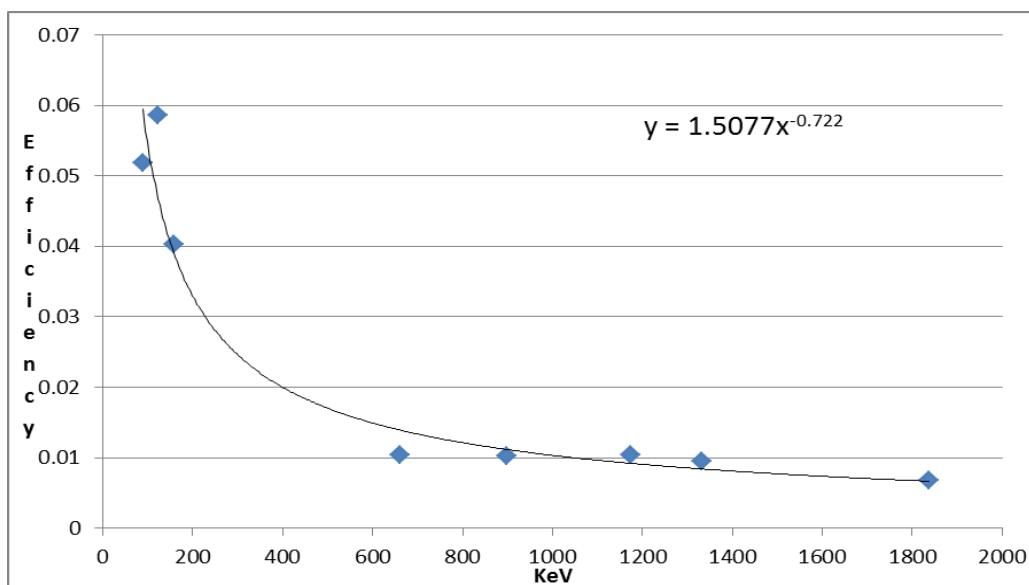


Figure 1. Efficiency graph for standard water on 12/04/2011 for software PCA.

### Natural radioactivity measurement

The measurement was conducted for 12 hours 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  $^{241}\text{Am}$ ,  $^{109}\text{Cd}$ ,  $^{57}\text{Co}$ ,  $^{123}\text{Te}$ ,  $^{51}\text{Cr}$ ,  $^{113}\text{Sn}$ ,  $^{85}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{88}\text{Y}$ ,  $^{60}\text{Co}$ , 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. Figure1 shows the Efficiency graph for standard water. The radionuclides were identified according to their individual photopeak, which are 609 keV ( $^{214}\text{Bi}$ ) and 351.9 keV( $^{214}\text{Pb}$ ) for  $^{238}\text{U}$ , 238 keV ( $^{212}\text{Pb}$ ), 583.191, 510.80 keV ( $^{208}\text{Tl}$ ) and 911keV( $^{228}\text{Ac}$ ) for  $^{232}\text{Th}$ . And 1460 keV for  $^{40}\text{K}$ . The activity of  $^{226}\text{Ra}$  during the equilibrium was assumed to be the same as its parent,  $^{238}\text{U}$ . The specific activity for each radionuclide was calculated using this equation [4]:

$$A_S = \frac{(C_s - C_b)}{t E_\gamma P_\gamma M_s} \quad (1)$$

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,  $E_\gamma$  and  $P_\gamma$  are detection efficiency and emission probability of  $\gamma$ -ray,  $t$  is counting time and  $M_s$  is the mass of the sample in kg.

### Results and Discussion

Contribution of radioactivity due to  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  to internal radiation doses on adult population in Malaysia was estimated using their published mass fractions in daily drinks. Geometric mean (GM) on these estimated daily intakes in terms of elemental concentration and activity are reported in Table 2. Radioactivities levels of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in daily drinks are calculated by using conversion rate for uranium = 0.64 Bq/kg, thorium = 1.02 Bq/kg and potassium = 340.38 Bq/kg. Detailed discussion on the estimated radiation doses due to ingestion these radionuclides are given below.

#### Uranium-238

The estimated GM activity is 0.05 Bq/d. The Annual Committed effective dose (ACED) is calculated using the observed annual intake of  $^{238}\text{U}$  activity of 18.25 Bq and ICRP dose coefficient of  $4.4 \times 10^{-8}$  Sv/Bq for adult members of the public [5]. It is estimated as  $0.8 \mu\text{Sv yr}^{-1}$ . Details are listed in Table 3.

Table 2. Daily intakes of uranium, thorium and potassium concentrations and their activities from the Malaysian diet

Analysis parameter	Uranium		Thorium		Potassium	
	Conc. (kg/d)	Activity (Bq/d)	Conc. (kg/d)	Activity (Bq/d)	Conc. (k g/d)	Activity (Bq/d)
Geometric mean (GM)	0.08	0.05	0.08	0.08	0.08	27.23

Table 3. Annual committed effective dose of adult population of Malaysian from intake of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$

Radionuclide	Daily intake (Bq)	Annual intake (Bq)	Dose coefficient (Sv/Bq)	Annual committed dose ( $\mu\text{Sv}$ )
$^{238}\text{U}$	0.05	18.25	$4.4 \times 10^{-8}$	0.8
$^{232}\text{Th}$	0.08	29.78	$2.2 \times 10^{-7}$	6.5
$^{40}\text{K}$	27.23	$99.39 \times 10^4$	$6.2 \times 10^{-9}$	61.53
Total contribution				68.83

Comparison with other countries of the world was also made by calculating committed effective doses of their populations using published estimates for  $^{238}\text{U}$  intake and presented in Table 4. The table shows that population of UK receives lowest annual radiation dose  $0.01 \mu\text{Sv yr}^{-1}$  [6]. While population of China receives highest annual radiation dose  $1 \mu\text{Sv yr}^{-1}$  [7]. Whereas population of Malaysia receives second highest annual radiation dose  $0.8 \mu\text{Sv yr}^{-1}$ . The radiation doses due to ingestion of  $^{238}\text{U}$  activity determined in the present work ( $0.8 \mu\text{Sv yr}^{-1}$ ) lie well within the observed range (i.e.  $0.01$ -  $1 \mu\text{Sv yr}^{-1}$ ) of other countries listed in Table 4.

Table 4. Annual committed effective dose of adult population of world from intake of <sup>238</sup>U.

Radionuclides	Dose (μSv yr <sup>-1</sup> )	Reference
UK	0.01	UNSCEAR (2000)
Philippine	0.11	Akhter et al. (2007)
India	0.11	Akhter et al. (2007)
Japan	0.13	Akhter et al. (2007)
World average	0.25	UNSCEAR (2000)
USA	0.26	UNSCEAR (2000)
S.Korea	0.75	Akhter et al. (2007)
Malaysia	0.8	Present study
China	1.00	Akhter et al. (2007)

### Thorium-232

The estimated GM activity is 0.08 Bq/d. The ACED due to the ingestion of <sup>232</sup>Th was estimated using the observed annual intake of 29.78 Bq and ICRP dose coefficient of  $2.2 \times 10^{-7}$  Sv/Bq (Table 3) for adult members of the public [5]. It is 0.08 μSv yr<sup>-1</sup>. ACED of adult population of other countries are also calculated using the available data in literature and listed in Table 5. The table shows that population of Philippine receives lowest radiation dose 0.05 μSv yr<sup>-1</sup> while population of Malaysia receives highest dose 6.5 μSv yr<sup>-1</sup>. The second and third highest are Bangladesh and China countries. The radiation doses due to ingestion of <sup>232</sup>Th activity determined in the present work is larger than radiation doses due to ingestion of <sup>232</sup>Th activity of other countries. This is due to high concentration of <sup>228</sup>Ra in Malaysian soil compared to the concentration of <sup>228</sup>Ra in others countries soil [8].

Table 5. Annual committed effective dose of adult population of world from intake of <sup>232</sup>Th.

Radionuclides	Dose (μSv yr <sup>-1</sup> )	Reference
Philippine	0.05	Akhter et al. (2007)
Japan	0.10	Akhter et al. (2007)
India	0.21	Akhter et al. (2007)
USA	0.33	Akhter et al. (2007)
S.Korea	0.33	Akhter et al. (2007)
World average	0.37	UNSCEAR (2000)
Pakistan	0.8	Akhter et al. (2000)
China	1.02	Akhter et al. (2007)
Bangladesh	1.15	Akhter et al. (2007)
Malaysia	6.5	Present study

### Potassium-40

The estimated GM activity is 27.23 Bq/d. The ACED due to the ingestion of <sup>40</sup>K was estimated using the observed annual intake of  $99.39 \times 10^4$  Bq and ICRP dose coefficient of  $6.2 \times 10^{-9}$  Sv/Bq for general public [5] as presented in Table 3. The ACED is calculated to be 61.53 μSv. Comparison was also made with the available Potassium ingestion data of other countries of the world, by calculating committed effective doses of various populations and presented in Table 6. The results shows that population of Malaysia receives lowest annual dose (61.53 μSv) while population of China receives 164 μSv annual dose [9]. While Iran receives high est annual dose (262 μSv). The

total estimated value of annual effective dose from  $^{40}\text{K}$  (61.53  $\mu\text{Sv}$ ) is less than annual effective dose world reference value which is 178  $\mu\text{Sv}$  [10].

Table 6. Annual committed effective dose of adult population of world from intake of  $^{40}\text{K}$ .

Radionuclides	Dose ( $\mu\text{Sv yr}^{-1}$ )	Reference
Malaysia	61.53	Present study
Indonesia	74	Akhter et al. (2007)
Philippine	74	Akhter et al. (2007)
India	106	Akhter et al. (2007)
Japan	128	Akhter et al. (2007)
Turkey	160	Akhter et al. (2007)
China	164	Quan et al. (2008)
World average	178	UNSCEAR (2000)
USA	178	UNSCEAR (2000)
Pakistan	178.75	Akhter et al.(2007)
S.Korea	192	Akhter et al. (2007)
Canada	197	IAEA (1992 )
UK	215	UNSCEAR (2000)
Spain	242	IAEA (1992 )
Iran	262	IAEA (1992 )

Cumulative radiological impact of these three naturally occurring radionuclides i.e.  $^{238}\text{U}$  (0.8 $\mu\text{Sv}$ ),  $^{232}\text{Th}$  (6.5  $\mu\text{Sv}$ ) and  $^{40}\text{K}$  (61.53  $\mu\text{Sv}$ ) in terms of annual committed effective dose is 68.83  $\mu\text{Sv yr}^{-1}$ , in which  $^{40}\text{K}$  has major contribution. The net radiological impact of drinks from these radionuclides is significantly less than the ICRP dose limit of 223.29  $\mu\text{Sv yr}^{-1}$ .

### Cancer risk

The risk incurred by a population is estimated by assuming linear dose-effect relationship with no threshold as per ICRP practice. For low doses ICRP fatal cancer risk factor is 0.05  $\text{Sv}^{-1}$  [11]. The risk factor states the probability of a person dying of cancer increases by 5% for a total dose of 1 Sv received during his lifetime. The estimated cumulative annual dose of 68.83  $\mu\text{Sv}$  is used to estimate cancer risk for an adult person using the following relationship [7].

$$\text{Risk} = \text{Dose (Sv)} \times \text{risk factor (Sv}^{-1}\text{)} \quad (2)$$

The estimated cumulative annual dose of 68.83  $\mu\text{Sv}$  to Malaysian adults is used and life-time (50 yr) exposure is estimated as 3.44  $\mu\text{Sv}$ , which gives a risk factor of  $1.72 \times 10^{-7}$ . This value is a negligible fraction of the total risk involve ( $6.0 \times 10^{-3}$ ) from all natural radiation sources based on global average annual radiation dose of 2.4 mSv /yr to man. The estimated values are significantly less than ICRP cancer risk factor of  $2.5 \times 10^{-3}$  based on annual dose limit of 1 mSv for general public, which gives annual death probability of  $10^{-5}$ , i.e. 1 in 100, 000 [12]. The risk of cancer from measured radiation is also compared with other kinds of health risks using Cohen and Lee estimates [7]. The authors estimated an average loss of 15 days from occupational exposure of 3 mSv /yr from age 18 to 65. Based on their estimates the calculated health risk from the measured average radiation dose 0.068 mSv /yr from present study is only 0.43days .

It is therefore concluded that 0.068 mSv/yr annual radiation dose attributable to drinks intake of three naturally occurring radionuclides  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in Malaysia would not pose any significant radiological impact on health and cancer risk to population . The drinks are considered safe for human consumption.

### **Conclusion**

The annual committed effective doses to Malaysian population from the drinks intake of naturally occurring radionuclides  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  have been determined and cancer risk is estimated. These estimated committed effective doses are 0.8, 6.5 and 61.53  $\mu\text{Sv yr}^{-1}$  respectively. The net radiological impact of these radionuclides is 68.83  $\mu\text{Sv yr}^{-1}$ . This gives a cancer risk factor of  $1.72 \times 10^{-7}$  and loss of life expectancy of 0.43 days only. The estimated risk has no significant health hazard and Malaysian drinks is radiologically safe, as per international standards.

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