



Assessment of ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K concentrations and annual effective dose due to the consumption of Vietnamese fresh milk

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Abstract

Vietnam has little data on radionuclide concentrations in milk, despite steadily increasing domestic consumption. Eight milk brands were investigated by gamma-spectrometry, and the resulting ingestion dose was calculated. The ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K concentrations varied from 0.60 ± 0.19 to 1.21 ± 0.24 , 1.45 ± 0.18 to 2.45 ± 0.24 , below detection limit to 0.13 ± 0.06 , and 341 ± 6 to 387 ± 7 Bq/kg (dry w.t). The total average Annual Effective Dose for all age groups were similar for all brands, and concentrations are far less than the WHO guidance level. All brands are safe for consumption.

Keywords Radionuclide concentration · Fresh milk · Annual effective dose · Risk factor · Infants · Children · Adults

Introduction

The humans are often exposed to natural and artificial radionuclides, which can enter the human body through breathing, eating, and drinking. In the food chain, milk is one of the fundamental foodstuffs for humans, especially for infants, children and old people. In addition, milk is sensitive to be contaminated because some radionuclides such as ^{137}Cs , ^{40}K are easily transferred to the milk through the grass-cow-milk pathway [1]. Therefore, evaluating and monitoring the level of radionuclides in milk (fresh and powdered) play an important role in estimating the annual effective dose for the population.

The radionuclides in milk have received much attention from researchers in many parts of the world. In Nigeria, a report showed unusually high average activity concentrations of ^{40}K compared to other reported values, reaching 831.6 ± 53.8 Bq/kg [2]. For powdered milk in Brazil, Melquiades & Appoloni [3] indicated that the activity of ^{40}K (475 ± 12 for polly and 489 ± 13 for cativa) was usual for powdered milk, while the observed ^{137}Cs and ^{232}Th activity concentrations were comparatively small. The average

activity concentrations of ^{40}K were also monitored in fresh and powdered milk in Tehran-Iran [4]. Observed mean activity concentrations of radionuclides in imported infant powdered milk in Malaysia of ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K were 2.6 ± 2.3 , 3.1 ± 1.8 , 0.3 ± 0.2 , and 99.1 ± 69.4 (Bq/kg), respectively [5]. Amongst these, the infant powdered milk from the Philippines had the lowest level of radioactivity, and the brand from Singapore showed the highest one [5]. In addition, the research also determined the mean annual effective dose due to consumption of powdered milk in Malaysia and found it to be 635 and 111 $\mu\text{Sv}/\text{year}$ for infants ≤ 1 and infants from 1 to 2 years old respectively [5]. In general, the radioactivity of fresh and powdered milk in many countries has been investigated and the effective dose has been calculated for various parts of the population, especially for infants. These investigations have shown that the level of radioactivity and effective dose can change an order of magnitude depending on the source of origin.

In Vietnam, the demand for milk is on the rise year by year. Most of the fresh milk brands consumed in Vietnam are domestic products. Although Vietnam has no nuclear power plant, the country is somewhat affected by the Fukushima nuclear disaster in 2011 (transfer by fallout and rain-fall to surface soil) [6, 7]. In addition, there are many natural radioactive sources, such as mines, along the mainland of Vietnam, which contain natural, and long half-life artificial radionuclides such as ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K . Thus, the radionuclides may transfer from soil to plant (grass) and from grass and air to cow's milk. These reasons could

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potentially lead to high artificial radionuclide activity concentrations in milk products produced by cows in Vietnam. These pathways are well described by many, such as the transfer of radionuclides from soil to plant (grass) [8–11] and from grass and air to cow's milk [12–14]. However, so far there is no research nor monitoring on the radionuclides in milk consumed in Vietnam. This study deals with the investigation of the radioactive levels in the local fresh milks in Vietnam. The research focuses on the determination of radioactivity level in fresh milk using a high-resolution HPGe detector. The results of activity concentration measurements will be used to calculate the average annual effective doses for different age groups. The data reported here can be used to establish the baseline for natural and artificial radioactivity in milk as well as comparison for new data, when there is some event involving nuclear plants or exploration activities in radioactive bearing mines in Vietnam.

Materials and methods

Eight brands of the fresh milk consumed commonly in Vietnam were bought from local prestigious markets and were used to investigate their radionuclide contents, including ^{232}Th , ^{226}Ra , ^{137}Cs and ^{40}K . All these milk brands were produced from local dairy cow's farms in Vietnam and were collected between 2018 and 2019. These brands are, namely: Vinamilk (VN); TH True Milk (TH); Moc Chau (MC); Da Lat Milk (DL); Dutch Lady (DuL); Vinamilk Organic (VO); Ba Vi (BV); and Nutifood (NT).

About three kilograms of each type of fresh milk (wet w.t) were taken for analysis. The fresh milk in the form of liquid was converted to powder and dried at 90 °C in a clean non-stick container to avoid radionuclide loss, contamination and to ensure that the moisture was completely removed. The obtained samples were weighted and packed in a plastic cylindrical beaker and sealed to prevent the escape of radon. The samples were left for at least 28 days to obtain the secular equilibrium between ^{226}Ra and its daughters (mostly ^{214}Bi and ^{214}Pb) [15]. The weight of milk after drying (dry w.t) is shown in Table 1.

After equilibrium was reached, activity concentration measurements were performed using a high-resolution HPGe detector (Canberra-GC5019) with 30 % relative efficiency. The analysis was performed using Genie-2000 software. The detector's energy resolution is 1.9 keV at 1.33 MeV of ^{60}Co gamma-ray peak. To reduce the surrounding natural background radiation at the laboratory, the detector is shielded by a 15 cm thick lead cylinder.

The activity concentration of each sample was determined based on their respective gamma lines: 609.3 keV, 1120.3 keV and 1764.5 keV from ^{214}Bi were used to determine the activity concentration of ^{226}Ra , while that of

Table 1 The information of studied samples

No	Brands	Wet w.t (g)	Dry w.t (g)	Concentration ratio
1	VN	3000	369.6	8.1
2	TH	3000	359.0	8.4
3	MC	3000	369.3	8.1
4	DL	3000	377.4	7.9
5	DuL	3000	380.7	7.9
6	VO	3000	390.9	7.7
7	BV	3000	352.1	8.5
8	NT	3000	372.8	8.0
Mean \pm SD				8.1 \pm 0.3

^{232}Th were determined from the gamma lines of 911.2 keV and 969.0 keV from ^{228}Ac and 583.0 keV and 2614.4 keV from ^{208}Tl [15–17]. The radioactive equilibrium in the ^{232}Th decay chain might not be present all the way to ^{232}Th , just ^{228}Ra , but similarly to the current literature, the values will be reported as ^{232}Th . For ^{40}K , its activity concentration was determined from its 1461 keV gamma line and 662 keV was used for ^{137}Cs . The samples were counted for over 50 h to avoid the statistical counting error. The gamma spectrometer was calibrated using IAEA reference materials RGU, RGTH and RGK [15]. The quality control tests were carried out based on standard reference materials (IAEA-321 and IAEA-414, IAEA reference materials produced by the International Atomic Energy Agency). The results agreed well with the values of IAEA-321 and IAEA-414 samples (except for ^{232}Th because of low activity) for ^{40}K , ^{137}Cs , ^{238}U and ^{40}K , ^{137}Cs in 492 ± 25 , 3.02 ± 0.10 , 1.36 ± 0.21 and 71.9 ± 2.1 , 544 ± 29 Bq/kg and the reference values 480 ± 22 , 3.09 ± 0.07 , 1.40 ± 0.36 and 72.6 ± 1.6 , 552 ± 17 Bq/kg respectively. The self-gamma absorption difference resulting from the difference in density of the studied samples and standard ones were corrected for following the method described by Debertain [18] and Jodlowski [19]. To lessen the photoeffect absorption in the sample, all of the gamma lines used are higher than 500 keV. The detection limits for ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K were 0.56, 0.32, 0.11 and 2.1 Bq/kg, respectively.

The activity concentrations of ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K is calculated by the formula (1).

$$A_{sp} = \frac{N_{sp} \times M_{st} \times A_{st} \times C_i}{N_{st} \times M_{sp}} \quad (1)$$

where N_{sp} , M_{sp} , and N_{st} , M_{st} are the net intensity, mass of the measured sample and of standard sample respectively; A_{st} is activity concentration of standard sample; C_i is the corrected factor for the differences between the densities of the sample and standard sample.

Table 2 Dose conversion factors for different age groups

Age groups	Dose conversion factors ($\mu\text{Sv/Bq}$)			
	^{232}Th	^{226}Ra	^{137}Cs	^{40}K
Infants	5.7	0.96	0.011	0.042
Children	3.9	0.8	0.0098	0.013
Adults	0.69	0.28	0.013	0.0062

Table 3 Activity concentrations of ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K in eight fresh milk brands in Vietnam

Brands	Activity concentrations of fresh milk (Bq/kg dry w.t)			
	^{232}Th	^{226}Ra	^{137}Cs	^{40}K
VN	0.87 ± 0.16	1.84 ± 0.24	0.13 ± 0.06	365 ± 7
TH	0.64 ± 0.17	1.84 ± 0.17	BDL	381 ± 7
MC	0.99 ± 0.16	2.45 ± 0.24	BDL	387 ± 7
DL	1.18 ± 0.16	2.05 ± 0.16	BDL	386 ± 7
DuL	0.79 ± 0.24	2.04 ± 0.24	BDL	374 ± 7
VO	1.05 ± 0.23	2.39 ± 0.23	BDL	375 ± 7
BV	0.60 ± 0.19	1.86 ± 0.17	BDL	384 ± 7
NT	1.21 ± 0.24	1.45 ± 0.18	BDL	341 ± 6
Min	0.60 ± 0.19	1.45 ± 0.18	BDL	341 ± 6
Max	1.21 ± 0.24	2.45 ± 0.24	0.13 ± 0.06	387 ± 7
Mean \pm SD	0.91 ± 0.23	1.98 ± 0.36		371 ± 17

BDL below detection limit

The annual effective dose D ($\mu\text{Sv/year}$) of radionuclides to individuals due to the consumption of milk is calculated based on the following equation below [20].

$$D = A \times I \times E \quad (2)$$

where A is the activity concentration of radionuclides in milk (Bq/kg); E is the dose conversion coefficient for the radionuclides due to ingestion ($\mu\text{Sv/Bq}$) (Table 2) [21]; I is the annual intake of milk (kg/year) which depends on the age groups. In this study, the annual ingestion dose for three age groups was investigated, including infants; children; and adults. The average annual intakes of fresh milk are 14.8 kg, 13.6 kg and 13.0 kg (dry w.t) for infants, children, and adults respectively [22].

Results and discussions

The activity concentration of ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K in eight selected brands of fresh milk are presented in Table 3. The results showed that smallest activity concentration belongs to the artificial radionuclide, ^{137}Cs , 0.13 ± 0.06 (Bq/kg) was found in VN milk and all other milk samples had activity concentrations below the detection limit (BDL). All results for this radionuclide are well below the guideline

level [23]. The low ^{137}Cs activity concentration in milk samples could be explained by low ^{137}Cs activity concentration in soil which was reported to be 3.82 Bq/kg (average) in surface soil (0–20 cm from surface) [24] before the Fukushima accident; the low ^{137}Cs activity concentration in water; the reduction due to the transfer processes from soil to grass, and from grass and water to cow milk. More recent data, albeit local data confirms the low ^{137}Cs activity concentration in soil (0.17–5.28 Bq/kg at Luong My Farm in Hoa Binh Province) [25]. The major radionuclide is ^{40}K in all brands, it shows the highest value among other radionuclides with a low standard deviation. The MC milk sample shows the highest activity concentration with 387 ± 7 Bq/kg of ^{40}K and the highest activity concentration of ^{226}Ra with 2.45 ± 0.24 Bq/kg. The lowest activity concentration of ^{40}K and ^{226}Ra are found in NT milk samples with 341 ± 6 and 1.45 ± 0.18 Bq/kg respectively. The relatively high activity concentration of ^{40}K in milk samples could be justified because potassium is a highly mobile in the environment [26–28], it is one of the major radionuclide elements in soil, and it naturally poses a part of potassium, which is a major nutrient for plants, animals and humans [10, 11]. These reasons lead to high ^{40}K activity concentration in comparison with other radionuclides due to the transfer process from soil to grass and from grass and water to cow milk. The highest and lowest activity concentrations of ^{232}Th are observed in NT and BV samples with 1.21 ± 0.24 and 0.60 ± 0.19 Bq/kg, respectively. The activity concentration of ^{226}Ra is two times greater than that of ^{232}Th . In general, ^{40}K is the dominant radionuclide in the studied samples. All of the radionuclides (^{232}Th , ^{226}Ra , ^{137}Cs and ^{40}K) are similar in value for the eight milk brands and are far less than the guidance level reported by the WHO [29].

The activity of concentrations of ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K in fresh milk found by this study are compared with reports by other authors from different countries in Table 4. Therein, for the natural radionuclides, countries such as Saudi Arabia, Singapore, Indian, Malaysia, New Zealand, Thailand, Spain show higher results of ^{232}Th , ^{226}Ra activity concentrations, but lower ^{40}K activity concentration compared to those in Vietnam. Other countries such as Jordan, Israel and Iran reported lower values of the ^{226}Ra , ^{232}Th activity concentrations, but higher value of ^{40}K activity concentrations compared to those in Vietnam. On the other hand, there are two countries having values of activity concentrations higher than those in Vietnam with 23.1, 4.35, 832 (Bq/kg) for ^{226}Ra , ^{232}Th , ^{40}K respectively in Nigeria and 1.6–3.7, 5.1–11.2, 475–489 (Bq/kg) for ^{232}Th , ^{137}Cs , and ^{40}K respectively in Brazil. By contrast, Australia has activity concentrations lower than those in Vietnam. For artificial radionuclides, ^{137}Cs activity concentration in the world is generally higher than that in Vietnam, except for Australia and Israel with 0.11 and BDL-0.08 (Bq/kg) respectively

Table 4 Activity concentration of ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K in milk from previous investigations in different countries

Country	Activity concentration (Bq/kg dry w.t)				Reference
	^{232}Th	^{226}Ra	^{137}Cs	^{40}K	
Vietnam	0.60–1.21	1.45–2.45	BDL-0.13	341–387	Present study
Australia	1.04	1.51	0.11	47	[5]
Bangladesh	15.0	26.7	–	494	[30]
Brazil	1.6–3.7	–	5.1–11.2	475–489	[1]
Egypt	0.60	0.91	0.42	477	[31]
India	1.48	2.5	–	34	[32]
India (Karnataka)	–	0.03	0.06	71.9	[33]
India (Vizag)	0.25–2.8	1.10–2.70	0.03–0.90	3–26	[34]
Iran	0.094–0.166	0.05–0.186	–	434–610	[35]
Israel	BDL-0.8	–	BDL-0.08	54–472	[36]
Jordan	BDL-1.28	BDL-2.14	BDL-1.5	297–393	[1]
Korea	–	–	0.023–0.024	18–48	[37]
Malaysia	0.31–8.57	1.36–7.06	–	40–254	[5]
Malaysia	BDL-1.86	1.09–3.62	–	124–213	[38]
Malaysia	0.20–2.83	0.20–2.93	0.10–0.95	212–508	[39]
New Zealand	0.31–1.37	1.89–2.49	0.63–0.73	52–78	[5]
Nigeria	4.35	23.1	–	832	[7]
Saudi Arabia	6.77	9.64	–	75	[40]
Saudi Arabia	0.09–0.76	0.25–0.85	–	210–257	[41]
Saudi Arabia	0.52	0.49	–	402	[42]
Singapore	8.57	7.06	–	235	[5]
Spain	2.22	3.86	0.17	104	[5]
Thailand	1.83–2.42	2.37–4.95	0.16–0.23	94–245	[5]
Tunisia	BDL	BDL	2.26	160	[43]

BDL below detection limit

[5, 33]. The variations of activity concentrations of ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K in powdered milk in the world may be affected by the type of milk, which is related to the animal of origin, different studied breeds, and the pollution in the local environment that affects the concentration of ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K as well.

The calculated annual effective doses (AED) due to the ingestion of ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K in Vietnamese milk for different age groups are presented in Table 5. The results show that the annual effective dose due to the intake of radionuclides in this study (except for ^{137}Cs) by infants is much higher than that of children and adults, especially the adults. For the ^{137}Cs , the AEDs by infants, children and adults are almost the same. This result is similar to that of Ababneh and coworkers [1]. The research results also indicate that ^{40}K has the highest contribution to the total annual effective dose due to the ingestion of fresh milk: 66.9% for infants, 48.4% for children and 66.1% for adults. Since K is in homeostasis in the body, it might be argued, that the resulting dose from ^{40}K should be disregarded from the excess dose. ^{232}Th gives the second-highest contribution to the total effective dose: 22.9% for infants; 35.7% for children and 18.2% for adults. On the other hand, the contribution of ^{137}Cs to the total

Table 5 Annual effective dose (AED) for different age groups

Radionuclides	Value	Age groups ($\mu\text{Sv/y}$)		
		Infants	Children	Adults
^{232}Th	Maximum	103.3	64.8	10.9
	Minimum	47.9	30.0	5.1
	Average	77.9	48.9	8.3
	SD	± 21.3	± 13.3	± 2.3
^{226}Ra	Maximum	35.8	27.4	9.1
	Minimum	20.9	15.9	5.3
	Average	28.5	21.7	7.3
	SD	± 5.1	± 3.9	± 1.3
^{137}Cs	Maximum	0.021	0.172	0.022
	Minimum	0	0	0
	Average	0.009	0.007	0.009
	SD	± 0.008	± 0.006	± 0.008
^{40}K	Maximum	246.6	70.0	31.8
	Minimum	214.9	61.0	27.8
	Average	227.4	66.3	30.2
	SD	± 10.8	± 3.1	± 1.4
Total average AED, μSv		340.0	136.9	45.7

effective dose is insignificant for all age groups and accounts for the smallest proportions among the four radionuclides.

The total average annual effective doses due to the intake of ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K from Vietnamese fresh milk are 340.0, 136.9 and 45.7 μSv for infants, children, and adults in respectively (Table 5). This shows that infants have the highest risk factor compared to other age groups. The risk factor for infants is 2.4 and 7.4 times higher than that for children and adults, respectively. This is similar to the results reported by Ababneh et al. [1] for the intake of milk in Jordan. In general, the total average annual effective dose results for the three age groups in this study are within the typical worldwide range of annual effective dose (200–800 μSv) due to the ingestion of natural radiation sources [20]

Conclusions

A systematic study of radioactivity concentration in Vietnamese fresh milk is presented for the first time. Activity concentrations of radionuclides (^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K) in selected eight fresh milk brands consumed in Vietnam have been determined by a HPGe detector. The annual effective dose due to the ingestion of fresh milk in Vietnam was also calculated. Based on the analysis of results, the following conclusions can be drawn:

The artificial of ^{137}Cs has a very minor presence, while ^{40}K is present in all brands in two order of magnitude higher concentration than the other observed radionuclides. The activity concentrations of the four radionuclides in each fresh milk brand was quite similar. In general, the level of radioactivity in Vietnamese fresh milk brands was found to be similar to the worldwide level reported in the literature.

The ^{40}K has the highest contribution to the total AED due to the ingestion of Vietnamese fresh milk, followed by ^{232}Th , ^{226}Ra , and ^{137}Cs . The AED due to the intake of radionuclides in this study (except for ^{137}Cs) for infants is much higher than that for children and adults. For ^{137}Cs , the AEDs by infants, children and adults are almost the same. The ^{40}K contribution to the total annual effective dose was 66.9 % for infants, 48.4 % for children and 66.1 % for adults, due to K being in homeostasis in the body it might be argued, that the AED is less by this amount.

The total average AEDs due to the intake of ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K from Vietnamese fresh milk for infants have the highest risk factor compared to other age groups.

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