Chiang Mai J. Sci. 2019; 46(4) : 778-786
http://epg.science.cmu.ac.th/ejournal/
Contributed Paper

Assessment of Activity Concentrations and Their Associated Radiological Health Risks in Commercial Infant Formulas in Thailand

Worawat Poltabtim [a] and Kiadtisak Saenboonruang*[a]

[a] Department of Applied Radiation and Isotopes, Faculty of Science, Kasetsart University, Bangkok, 10900 Thailand.

*Author for correspondence; e-mail: fscikssa@ku.ac.th, kiadtisak.s@ku.th

Received: 28 January 2019
Revised: 7 March 2019
Accepted: 13 March 2019

Abstract

This work assessed the activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs, their corresponding annual effective doses, and their radiological health risks associated with the consumption of infant formulas in Thailand. The results showed that the average (± standard deviation) activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs radionuclides were 3.20±0.51 Bq kg^{-1}, 1.24±0.10 Bq kg^{-1}, 213.20±25.50 Bq kg^{-1}, and 4.90±0.74 Bq kg^{-1}, respectively, leading to an average annual effective dose for infants aged ≤1 year and 1–2 years of 762.82±91.34 μSv year^{-1} and 189.60±22.47 μSv year^{-1}, respectively, and radiological health risks of (5.63±0.66)×10^{-6} and (3.77±0.44)×10^{-6}, respectively. The overall results suggested that infant formulas in Thailand posed no significant effective dose and radiological health risks, implying safe consumption for infants.

Keywords: powdered milk, radioactivity, annual effective dose, risk, gamma spectroscopy

1. Introduction

As parenting culture and styles continue to change from breastfeeding to bottle feeding due to economic and cultural needs [1], infant formulas have become one of the important sources of nutrients and energy for babies and infants aged under 12 months. The change could be seen in a report released by the U.S. Centers for Disease Control and Prevention (CDC), which showed that only 47% and 25% of infants born in 2014 were exclusively breastfeed through the first 3 and 6 months, respectively. In addition, approximately 28% and 34% of infants were supplemented with infant formulas before 3 and 6 months, respectively [2].

Most infant formulas are made from cow's milk that has been processed to satisfy the basic nutritional and caloric needs of infants, while additional nutrients such as O-mega 3, 6, 9 fatty acids, nucleotides, choline, and taurine, are also added to some infant formulas in order to enhance infant development [3]. Although the importance and benefits of infant formulas are recognized in today's parenting cultures, possible radiological risks associated with natural and artificial radionuclides (^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs) in infant formulas have raised concerns to consumers as evidence of elevating radioactivity levels has been found in grass and plants used to feed cows due to increases in the use of fertilizer and residues from mining [4–6], which could be transferred to the milk before it is processed into infant formulas [7, 8]. These radionuclides, once in an infant's body, could potentially affect and damage cells of important organs, causing possible abnormality in the infant's health and development [9].

In previous reports, levels of activity concentrations in infant formulas varied depending on regions and dates of manufacture. For example, Priharti et al. showed that the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in infant formulas obtained from Malaysian markets were 1.87±0.17 Bq kg^{-1}, 0.89±0.13 Bq kg^{-1}, and 213.00±2.09 Bq kg^{-1}, respectively, which led to the annual effective doses for infants aged ≤ 1 year and 1–2 years of 0.46 and 0.21 mSv year^{-1}, respectively [10]. Jemii and Alharbi also reported that activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in samples purchased from Saudi Arabian markets for infants aged ≤ 1 year and 1–2 years were 0.52±0.09 Bq kg^{-1}, 0.51±0.06 Bq kg^{-1}, and 371.98±3.68 Bq kg^{-1}, leading to the annual effective doses of 0.62 and 0.24 mSv year^{-1}, respectively [11]. The report also showed the total risks of 7.91×10^{-6} and 5.32×10^{-6} for infants aged ≤ 1 year and 1-2 years, respectively, indicating no radiological risk to infants from the consumption of the investigated formulas as the risks were within the recommended limit of 1×10^{-6}–1×10^{-4}. For activity concentrations arising from the artificial radionuclide, ^{137}Cs, Pietrzak-Fiecko et al. reported that powdered cow milk from four regions of Poland had the average ^{137}Cs activity concentrations of 1.90 Bq kg^{-1} in addition to natural radionuclides, which could potentially increase radiological health risks from powdered milk consumption [12].

Despite the wide availability of data on activity concentrations in infant formulas, the same information is considered inadequate for Thai consumers as no official reports have yet been released to the public, posing uncertainty regarding purchaser safety and trust. As a result, the current work investigated and reported both natural and artificial activity concentrations from ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in nine different brands of commercial infant formulas available in Thailand using an HPGe gamma-ray spectrometer. Values of the measured activity concentrations were then used to calculate the annual effective doses and total radiological health risks associated with the consumption of the investigated infant formulas. This report is the first scientific report of radiation hazard indices from infant formula consumption in Thailand and could be used to raise the safety awareness of purchasers as well as to develop safety guidelines and recommendations.

2. MATERIALS AND METHODS

2.1 Sampling and Sample Preparation

This work investigated the activity concentrations of nine different commercial infant formulas purchased from local grocery stores in Bangkok, Thailand. These nine brands were also widely available through the country. Details of locations/dates of manufacture, and recommended age groups are shown in Table 1.

All samples were oven-dried at 70 °C for 24 h until constant weights were achieved and 3 sets of each sample weighing 200 g were transferred to separate 500 mL Marinelli-type beakers, which were sealed with masking tape to avoid radon escaping. The sealed samples were then stored for 4 weeks to let ^{226}Ra, ^{232}Th, and their progenies reach secular equilibriums prior to gamma spectroscopy [13].

2.2 Determination of Activity Concentrations

The activity concentration of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs were determined using a reverse

Table 1. Sample codes, origins, dates of manufacture, and recommended age groups of the investigated infant formulas.

Sample	Manufacturing countries	Manufacturing dates	Recommended age group
S1	Singapore	27/5/2016	<1 year
S2	Thailand	28/11/2017	<1 year
S3	Thailand	20/12/2017	<1 year
S4	Singapore	19/5/2017	0.5–3 years
S5	Thailand	16/12/2017	0.5–3 years
S6	Singapore	14/8/2017	>1 year
S7	Thailand	16/8/2017	>1 year
S8	Thailand	14/6/2017	>1 year
S9	Thailand	16/8/2017	>3 year

electrode, closed-end, coaxial HPGe gamma spectrometer (Canberra; Model GR2519; Serial number 12946022; 51.7 mm crystal diameter; 58.5 mm length), having a relative efficiency of 25% and an energy resolution of 1.9 keV-FWHM at the 1.33 MeV peak of ^{60}Co. The HPGe gamma detector was kept inside a 12 cm-thick cylindrical lead shield with a fixed bottom and a movable cover in order to reduce external and background radiation. The detector was connected to a full-featured 16k channel integrated multichannel analyser (MCA; Canberra; Model DSA1000). Spectra of gamma rays emitted from the samples were analysed using the Genie 2000 (3.2) software, in which the calibration of the energy and efficiency was performed using a mixed gamma standard-type EG-ML developed by Eckert & Ziegler (California, USA) with isotopes of ^{109}Cd, ^{57}Co, ^{123m}Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y, and ^{60}Co. All measurements were performed with a 60,000 s counting time period.

The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in the samples were calculated using Eq.1 [11]:

$$A = \frac{N_{net}}{m \times t \times P_\gamma \times \epsilon_\gamma} \qquad (1)$$

where A, N_{net}, m, t, P_γ, and ε_γ are the activity concentration (Bq kg^{-1}), the net count of gamma rays at the respective energy per second (cps), the mass of the sample (kg), the collection time (s), the probability of the transition of the interested radionuclide at the respective gamma energy, and the efficiency of the gamma spectrometer at the respective gamma energy, respectively. The value of ε_γ for a respective gamma energy was calculated using a polynomial fitting method described in the report of Asaduzzaman et al. and the calibrated results are shown in Figure 1 [14]. Transition isotopes for each radionuclides and their corresponding P_γ were described in Saenboonruang et al. [15]. It should be noted that the minimum detectable activity (MDA) levels for this work were calculated as 1.36 Bq kg^{-1}, 0.75 Bq kg^{-1}, 7.01 Bq kg^{-1}, and 0.57 Bq kg^{-1} for ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs, respectively [16].

2.3 Determination of Annual Effective Dose

The annual effective dose from ingestion of the investigated infant formulas was calculated using Eq. 2 [11]:

$$H = AIF \qquad (2)$$

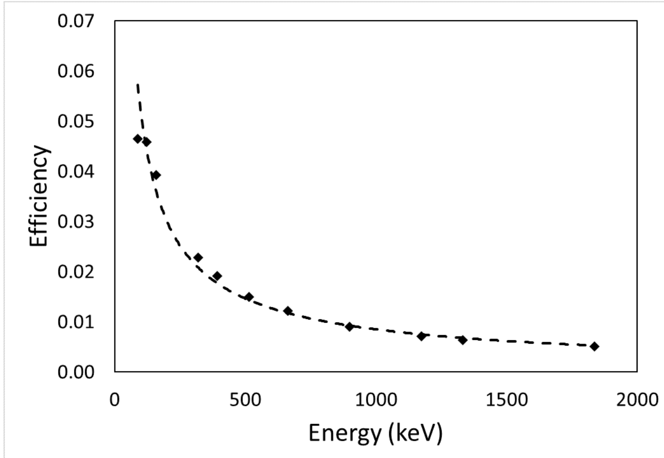

Figure 1. Efficiencies of the gamma spectrometer at respective gamma energies.

where H, A, I, and F were the annual effective dose (Sv year^{-1}), the activity concentrations of the interested radionuclides in the samples (Bq kg^{-1}), the ingestion rates of infants (kg year^{-1}), and the dose conversion factors (Sv Bq^{-1}), respectively. Values of I and F varied depending on the age group of the infants and are shown in Table 2.

Table 2. Annual intake of infants and dose conversion factors for each age group.

Age group	Ingestion rate [16] (kg year^{-1})	Dose conversion factor [17] (Sv Bq^{-1})			
		^{226}Ra	^{232}Th	^{40}K	^{137}Cs
≤1 year	22.4	4.7×10^{-6}	4.6×10^{-6}	6.2×10^{-8}	2.1×10^{-8}
1-2 year	15	9.6×10^{-7}	4.5×10^{-7}	4.7×10^{-8}	1.2×10^{-8}

2.4 Determination of Radiological Health Risk

The radiological risk associated with infant formula consumption was calculated using Eq. 3 [11]:

$$\text{Risk} = D_{int} \times SF \times t \quad (3)$$

where D_{int}, SF, and t are the average daily intake of the infant formula for each age group (pCi day^{-1}), the slope risk factor or the morbidity risk of each radionuclide (risk pCi^{-1}), and the exposure duration (days), respectively. The value of D_{int} was calculated using the relationship shown in Eq. 4:

$$D_{int} = \frac{A \times I}{365} \quad (4)$$

The values of SF for ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs used in this work were 5.14×10^{-10} risk pCi^{-1}, 1.33×10^{-10} risk pCi^{-1}, 3.43×10^{-11} risk pCi^{-1}, and 3.74×10^{-11} risk pCi^{-1}, respectively, while the value of t was 365 days.

2.5 Statistical Analysis

The statistical analysis of this experiment was performed using IBM Statistical Package for the Social Sciences (SPSS) v.24.0 software. The reported values were expressed as mean±standard deviation and the analysis of variance (ANOVA) was used to determine statistical significance at $p<0.05$.

3. Results and Discussion

The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs and their corresponding total activity concentrations for all samples are shown in Table 3, which indicates that the activity concentrations of ^{40}K in all samples were higher than for ^{226}Ra, ^{232}Th, and ^{137}Cs ($p<0.05$) with the values varying from 180.66 to 253.63 Bq kg^{-1} and the average value being 213.20 Bq kg^{-1}. The activity concentrations of ^{226}Ra, ^{232}Th, and ^{137}Cs were much smaller, ranging from 2.54 to 3.82 Bq kg^{-1}, 1.05 to 1.42 Bq kg^{-1}, and 4.07 to 6.53 Bq kg^{-1}, respectively, while the average values were 3.20 Bq kg^{-1}, 1.24 Bq kg^{-1}, and 4.90 Bq kg^{-1}, respectively. The highest values of activity concentrations in ^{40}K was expected as K is an essential element for the grasses and plants used to feed cows and also for the body metabolism of cows, leading to a high accumulation of K in infant formulas derived from cow milk [11, 13, 15]. On the other hand, the lower activity concentrations of ^{226}Ra, ^{232}Th, and ^{137}Cs in all samples could have been due to the fact that these radionuclides are not used metabolically by grasses and cows; hence, they did not accumulate in large quantities, but rather were transported with other essential elements that are chemically similar. For example, ^{226}Ra could be transported along with Ca and thus accumulate in the grass and cows.

The ^{226}Ra and ^{232}Th activity concentrations in this work were in the same order of magnitude as the reports from Malaysia, Saudi Arabia, and Brazil (Table 4). However, the values were smaller than those from Nigeria. For ^{40}K, the value in the present study was similar to that from Malaysia but smaller than the values from Saudi Arabia, Nigeria, Brazil, and Venezuela (though still in the same order of magnitude). In terms of ^{137}Cs, the present value was in between the values obtained from Poland, Venezuela and Brazil, respectively (also in the same order of magnitude). Hence, the overall results of the activity concentrations found in this work were in agreement with other reports, suggesting that the radionuclide concentrations in infant formulas throughout the world have similar characteristics with slight deviations, probably due to differences in environmental backgrounds and farming cultures.

Table 3. Individual activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs, and the total activity concentration for all infant formula samples. Values are shown as mean±standard deviation of the mean.

Sample	Activity concentration (Bq kg^{-1})				Total activity concentration (Bq kg^{-1})
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	
S1	3.62±0.30	1.28±0.60	208.92±10.21	4.71±0.34	218.53±11.45
S2	2.66±0.57	1.05±0.51	180.66±13.76	4.76±0.33	189.13±15.17
S3	2.68±0.52	1.18±0.47	181.97±11.88	6.53±0.13	192.36±13.00
S4	3.49±0.46	1.24±0.47	245.82±16.37	4.32±0.49	254.87±17.79
S5	2.90±0.42	1.32±0.55	195.40±12.32	4.85±0.03	204.47±13.32
S6	3.79±0.56	1.28±0.44	216.33±10.99	4.70±0.45	226.10±12.44
S7	3.26±0.85	1.22±0.56	224.25±9.61	4.07±0.29	232.80±11.31
S8	2.54±0.67	1.18±0.70	211.80±8.86	4.54±0.42	220.06±10.65
S9	3.82±0.64	1.42±0.46	253.63±8.27	5.58±0.21	264.45±9.58
Average	3.20±0.51	1.24±0.10	213.20±25.50	4.90±0.74	222.53±11.45

Table 5 shows the annual intake and the annual effective dose due to the radionuclides of interest with infants in the ≤1-year-old

Table 4. Comparison of activity concentrations between the present work and literature values. Values are shown as mean±standard deviation of the mean.

Country	Activity concentration (Bq kg^{-1})				Reference
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	
Thailand	3.20±0.51	1.24±0.10	213.20±25.50	4.90±0.74	Present work
Malaysia	1.87±0.17	0.89±0.13	213±2.09	N/A[a]	Priharti et al. [10]
Malaysia	3.05±1.84	2.55±2.48	99.1±69.5	0.27±0.19	Uwatse et al. [19]
Saudi Arabia	0.52±0.087	0.51±0.062	371.98±3.68	N/A	Jemii and Alharbi [11]
Nigeria	19.3±7.2	12.1±4.8	468.0±72.7	N/A	Agbalagba et al. [20]
Brazil	N/A	1.7-3.7	489±13	7.0-11.2	Melquiades and Appoloni [21]
Venezuela	N/A	N/A	329±20	1.43±0.4	Labrecque et al. [22]
Poland	N/A	N/A	N/A	1.9	Pietrzak-Fiecko and Smoczynski [12]
Poland	N/A	N/A	N/A	1.39±0.26	Pietrzak-Fiecko and Smoczynski [23]

[a] N/A represents data not available.

Table 5. Annual intakes and annual effective doses for ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs concentrations for all infant formula samples and age groups. Values are shown as mean±standard deviation of the mean.

Sample	Annual intake (kBq year^{-1})		Annual effective dose (μSv year^{-1})	
	≤1 year	1-2 year	≤1 year	1-2 year
S1	4.89±0.26	3.28±0.17	805.45±105.86	193.26±14.74
S2	4.24±0.34	2.84±0.23	641.29±131.67	160.04±20.33
S3	4.31±0.29	2.89±0.20	659.83±119.50	162.40±18.12
S4	5.71±0.40	3.82±0.27	838.37±119.93	214.26±20.19
S5	4.58±0.30	3.07±0.20	714.88±118.31	174.62±17.56
S6	5.06±0.28	3.39±0.19	833.51±119.76	200.36±18.07
S7	5.21±0.25	3.49±0.17	782.12±160.47	197.20±22.13
S8	4.92±0.24	3.30±0.16	685.20±154.33	178.78±19.93
S9	5.92±0.21	3.97±0.14	904.74±125.85	225.47±17.52
Average	4.89±0.58	3.34±0.39	762.82±91.34	189.60±22.47

group clearly having a higher average annual intake than the 1–2-year-old group ($p<0.05$) due to the higher ingestion rates and higher dose conversion factors in the younger age group. When comparing the annual effective doses in the work with the ones reported by Jemii and Alhabi [11], the results showed that both reports had the values in the same order of magnitude (762.82 to 620 μSv year^{-1} in the <1-year-old group and 189.60 to 240 μSv year^{-1} in the 1–2-year-old group). These average annual effective doses were less than the recommended limit of 1 mSv year^{-1} issued by the Joint Food and Agriculture Organization of the United Nations/World Health Organization (FAO/WHO) and the International Commission on Radiological Protection (ICRP) [24, 25]. Furthermore, percentage contributions to the average annual effective dose shown in Figure 2a indicate that ^{226}Ra contributed the

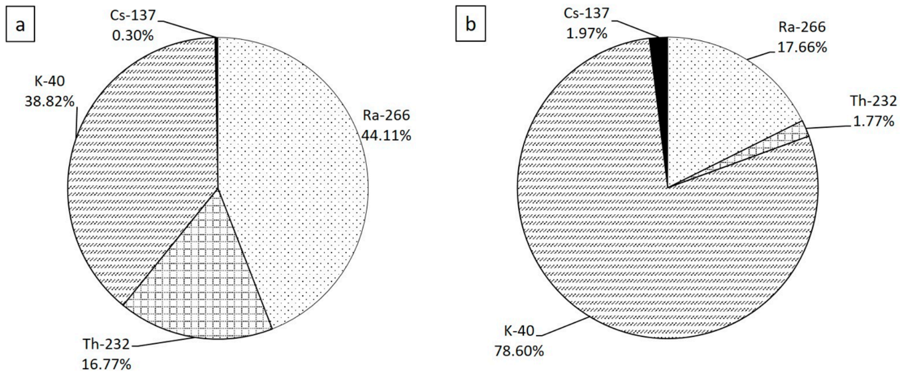

Figure 2. Contributions of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in infant formulas to (a) annual effective dose and (b) total radiological health risk.

highest percentage (44.11%) among other radionuclides, of which, ^{40}K, ^{232}Th, and ^{137}Cs contributed 38.82%, 16.77%, and 0.30%, respectively. The largest contribution from ^{226}Ra was mainly due to its dose conversion rates being the highest in both age groups, despite having a small average activity concentration of only 3.20 Bq kg^{-1} compared with 213.20 Bq kg^{-1} in ^{40}K.

Table 6 shows the daily intake and radiological health risks from the consumption of infant formulas, which indicates that the average risk for infants in the ≤1-year-old group had a higher risk value (5.63×10^{-6}) than infants in the 1–2-year-old group (3.77×10^{-6}) ($p<0.05$), mainly due to the higher ingestion rates in younger infants. The radiological health risks in this work were within the recommended

Table 6. Daily intakes and total health risks for ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs concentrations for all infant formula samples and age groups. Values are shown as mean±standard deviation of the mean.

Sample	Daily intake (pCi day^{-1})		Risk (×10^{-6})	
	≤1 year	1-2 year	≤1 year	1-2 year
S1	362.12±18.99	242.49±12.49	5.67±0.35	3.80±0.24
S2	313.38±25.28	209.82±16.83	4.77±0.51	3.19±0.34
S3	318.75±21.48	213.45±14.41	4.85±0.45	3.25±0.30
S4	422.31±29.26	282.80±19.74	6.38±0.53	4.27±0.36
S5	338.80±22.19	226.87±14.79	5.17±0.43	3.46±0.29
S6	374.65±20.75	250.88±13.80	5.88±0.45	3.93±0.30
S7	385.74±18.67	258.31±12.55	5.86±0.52	3.92±0.35
S8	364.64±17.60	244.18±11.80	5.38±0.46	3.60±0.31
S9	438.21±15.63	293.44±10.63	6.69±0.41	4.48±0.28
Average	368.73±42.63	246.92±28.55	5.63±0.66	3.77±0.44

excess lifetime risks issued by the United States Environmental Protection Agency (US EPA), which are between 1×10^{-6} and 1×10^{-4}, implying that the consumption of infant formulas was radiologically safe in both age groups [26]. In addition, Figure 2b clearly shows that ^{40}K has the highest contribution to the risks (78.60%), mostly due to the much larger activity concentrations of ^{40}K. These results were in good agreement with previously reported results in Malaysia and Saudi Arabia [10, 11].

4. Conclusions

Radioactivity due to ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in infant formulas could potentially harm infants where there are higher activity concentrations, annual effective doses, and radiological health risks than the recommendations issued by responsible agencies/organizations. To ensure the safety of infants consuming infant formulas, this work measured the activity concentrations in nine different brands of infant formulas that were available in Thailand. The results indicated that ^{40}K had the highest activity concentrations in all samples and contributed the most to the radiological health risks. Furthermore, the levels of activity concentrations found in this work were in agreement with other literature values, while the annual effective doses and the associated health risks were within the recommended limits issued by FAO/WHO and US EPA, implying safe consumption of infant formulas in Thailand. The outcomes of this work not only strengthen trust and safety levels of purchasers, especially parents, but also provide the first scientific report of radioactivity of both natural and artificial radionuclides in infant formulas in Thailand. We encourage other researchers who are also interested in food safety, especially in infants, to further conduct research in other foods or in other regions so that more information can be made available for future reference.

Acknowlegements

The authors acknowledge financial support from the Faculty of Science, Kasetsart University, Bangkok, Thailand under the Undergraduate Research Matching Fund (URMF).

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