Journal of Natural Sciences and Mathematics
Qassim University, Vol. 11, No. 1, pp 59-67 (January 2018/Rabi II 1439H.)

Gamma Ray Spectroscopic Analysis in infants' milk powder Consumed in Saudi Arabia, Qassim Province, Buraidah.

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Abstract. Infants' milk powder can be used in many countries because it contains vitamins, proteins and minerals, which are essential to grow infants. Therefore, the main objectives of this study were to measure the natural radionuclides concentrations (^{226}Ra, ^{232}Th, and ^{40}K). Gamma ray Spectroscopy, NaI(Tl) detector was used for measuring the radionuclides concentrations of ١٥ brands for new born till six months. The average radionuclides concentrations were ٢٫١±٠٫٠٤ BqKg^{-1}, ١٫٦±0.0٣ BqKg^{-1} and ١١١٫١±0.5 BqKg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K respectively. The average and range of the effective dose due to annual intake from infants' milk powder were estimated to be 75.1(19.8-109.5) μSvy^{-1}, 27.5(1.9-40.3) μSvy^{-1} and 177.4(57.8-247.2) μSvy^{-1} for ^{226}Ra, ^{232}Th and ^{40}K respectively. These values were lower than the recommended limit determined by ICRP-60, IAEA-152 and UNSCEAR. The comparison was obtained between our results and other published papers in different countries. The resulting data may serve as base-line levels of activity concentration in powdered milk in the area of study.

Keywords: Infants' Milk Powder; NaI(Tl); ^{226}Ra; Minerals; Trace elements; Ingestion Dose

1. Introduction

Human, animals or plants obtain dose from natural gamma ray radiation when they have been placed in the presence of gamma ray radiation (^{226}Ra, ^{232}Th and ^{40}K) [1-2]. The dose results from the natural radionuclides may transfer from plants or animals to human. The main source of human exposure to radionuclides is consumption food leading to ingestion radiation doses [3].

Breast milk is the optimal source of nutrition for infants because it provides all the essential nutrients for infant health. However, infant's milk powder is usually given for newborn if the breast milk was not enough. The infant's milk powder provides the energy, vitamins, proteins minerals and nutrients that are necessary for optimum growth [4]. In addition, the dairy products are main source of food not only for infant but also for other people as daily diet [5]. The average consumed weight from infant's milk powder per year is about 22.4 Kg [6]. Therefore, the accurate measurement of the radionuclides concentrations in milk is useful for radiation protection purposes. In general, the largest contributors to the ingestion dose resulting from milk and food is ^{40}K [6]. The dose causing cancer strongly depends on the amount of radionuclides presenting in the milk [2].

The present work aims to estimate the radionuclides concentrations (^{226}Ra, ^{232}Th and ^{40}K) in the milk powdered consumed by newborn (0-6 months) in Saudi Arabia, Qassim Province, Buridah using NaI(Tl) and to calculate the ingestion absorbed dose of radionuclides resulting from formula infant milk.

2. Materials and methods

2.1 radionuclides activity concentrations measurements in infants' milk powder

This study includes fifteen infants' milk powder samples for newborn (0-6 months). They were collected from Qassim local markets in Saudi Arabia during the year of the measurements. The brand, origin, density and expired date of samples were listed in Table 1. The powdered samples were prepared before analysis. The weight of the samples were between 105 gram and 185 gram. Next, the samples were placed in 250 mL polyethylene bottles. They were closed completely at least four weeks to ensure the secular equilibrium presenting between parents and daughters [7-10].

Table (1) the brand name, country of origin, production date and density of the selected study samples.

Sample code	Brand name	Origin	Age group (month)	Fat Per 100g	Expired date	Density (g/cm^3)
A	Biomil	Belgium	0-6	26.4	4-2018	0.45
B	Aptamil	Germany	0-6	24.7	17-9-2017	0.45
C	Bebelac	Netherland	0-6	24.5	8-2-2018	0.44
D	Nan	Swiss	0-6	27.7	6-2017	0.45
E	MaeilMam'ma	Korea	0-6	24.0	11-6-2017	0.45
F	Kabrita	Netherland	0-6	25.5	28-8-2017	0.49
G	Larilac	France	0-6	27.5	19-10-2017	0.45
H	Fabimilk	Netherland	0-6	29.0	17-11-2017	0.45
I	S-26 Gold	Ireland	0-6	29.0	29-9-2017	0.44
J	Premium care	France	0-6	26.5	22-9-2017	0.44
K	Lactonic gold	Spain	0-6	27.0	13-9-2017	0.45
L	Primalac premium	Swiss	0-6	27.0	10-10-2017	0.45
M	Novalac	France	0-6	25.5	16-11-2017	0.44
N	Blemil	Spain	0-6	25.0	18-11-2017	0.45
O	Wyeth Illume	Ireland	0-6	29.0	26-6-2017	0.38

NaI(Tl) detector 3×3 inch with a 1024-chanel computer analyzer was used to measure the natural radionuclides activity (count rate in the environmental samples). It was calibrated using known source such as ^{60}Co and ^{137}Cs point sources [10]. In order to calculate the radionuclide activity concentration (activity per unit mass) for each gamma-ray photo-peak rely on the secular equilibrium between parents and daughters in the samples, the equation1 was used [11].

$$A = \frac{[N_c - B] \times 100}{\varepsilon \times \eta \times m} \quad (1)$$

Where

N_c is the count rate (cps) for each infants' milk powder

B is the count rate (cps) for background radiation around the detector

ε is the abundance of the γ-peak in a radionuclide

m is the mass of sample

η is the measured efficiency for each gamma-ray peak observed for the same number of channels. The values of ε and η for each isotopes used to calculate the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were shown in table 2. The measuring time for gamma-ray spectra was 86400 s.

Equation 2 was used to obtain the Minimum Detectable Limit (MDL) of radionuclides activity concentration [12]. In order to calculate the MDL, an empty polystyrene container was used with same manner as the measured samples to count the background radiation. Any activity concentration of radioisotopes below MDL was neglected (table 2). The time measurements was 86400 s.

$$MDA = \frac{2.71 + 4.65\sqrt{B}}{\varepsilon \times \eta \times t} \quad (2)$$

2.2 Annual effective ingestion radionuclides dose

To calculate the annual effective ingestion radionuclides dose (D) resulting from the infants' milk powder, equation 2 was used [6,13-16].

$$D = AIE \quad (3)$$

Where:

D is the annual effective radionuclides dose (Svy^{-1})

A is the activity concentration of radionuclides in infants' milk powder sample ($BqKg^{-1}$)

I is the amount of milk powder taken in one year (Kgy^{-1}) and depends on a given age. Its value is 38 Kg/year for infant.

E is a conversion factor for ingestion radionuclides ($SvBq^{-1}$)

The conversion factor 'E' depends on both the radioisotopes and the baby age. Its value for infants between 0 and 12 months are 960, 450 and 42 $nSvBq^{-1}$ for ^{226}Ra, ^{232}Th, and ^{40}K respectively.

The total dose values for public should be lower than 1 mSv/year as recommended by UNSCEAR-2000 [2].

Table 2. Gamma rays and their related isotopes used to calculate the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K

		l efficiency for each ray peak (η) (%)	ce of the a ide (e)	ı detected)L)
^{214}Pb	351.90			0.14
^{214}Bi	609.30			0.20
^{214}Bi	1120.30			0.57
^{214}Bi	1764.5			0.30
^{228}Ac	911.10			0.30
^{212}Pb	238.60			0.11
^{40}K	1460.00			1.40

3. Results and discussion:

3.1 activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the infants' powdered milk samples present in table 3. The average and range obtained results of ^{226}Ra, ^{232}Th and ^{40}K were lower than the recommended reference level (50, 50 and 500 Bq/kg). It changed from 0.5 to 3.0 with the mean value 2.1 Bq/Kg for ^{226}Ra while it was between 1.1 and 2.4 and the average value was 1.6. The highest activity concentration resulting from 40K and its values ranged from 36.2 to154.9. Its mean value was 111.1 Bq/Kg. The highest activity concentration value was from 40K in the sample code K (Lactonic gold) from Spain while the lowest concentration of ^{40}K was found in the sample code A (Biomil) from Belgium. The relative contribution to dose due to ^{40}K was 62 %, followed by the contribution due to ^{226}Ra and ^{232}Th as 28.0 %, 10.0 % respectively as shown in figure 1. In order to test the correlations between ^{226}Ra and ^{232}Th and ^{226}Ra and ^{40}K, the obtained concentrations of naturally occurring radionuclides were plotted in the histogram Figure 2. It is noted that a good correlation between ^{226}Ra and ^{232}Th was observed with a correlation coefficient of 0.73, whereas a poor correlations between ^{226}Ra and ^{40}K. Table 4 shows the comparison between the present study and other published papers. The activity concentrations resulting from ^{226}Ra, ^{232}Th and ^{40}K in our results were lower than Malaysia and Nigeria [18,19]. When the comparison focuses on ^{226}Ra and ^{232}Th, this study was comparable with Jordan and other study in Saudi Arabia [6, 13]. The activity concentration of ^{40}K from this study was higher than the other study in Saudi Arabia and India [15, 20]. However, it was lower than the Jordan, Malaysia and Nigeria [6, 18, 19].

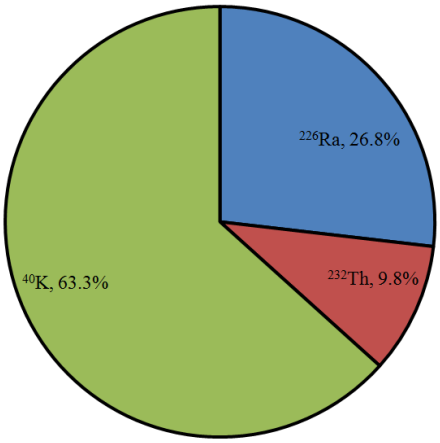

Fig 1: The relative contribution to ingestion dose due to ^{226}R, ^{232}Th and ^{40}K in infants' milk powder (newborn till 6 months) Consumed in Qassim province Saudi Arabia

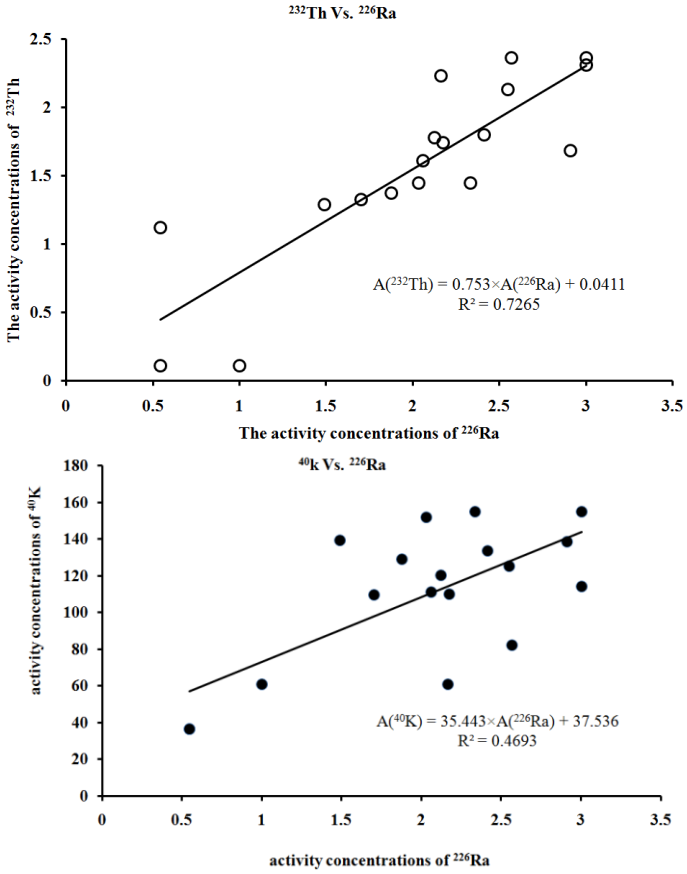

Fig 2: Correlations between (○) ^{226}Ra with ^{232}Th, (●) ^{226}Ra with 40K in infants' milk powder (newborn till 6 months) Consumed in Qassim province Saudi Arabia

3.2 Annual effective ingestion radionuclides dose

To estimate the annual effective ingestion radionuclides dose (D) in the powdered milk, equation 3 was used. The ingestion dose was ranged from 96.8 to 357.1 μSvy^{-1} with the mean value was 280.0 μSvy^{-1} obtained by the sum of contributions for ^{226}Ra, ^{232}Th and ^{40}K in the selected samples shown in Table (3). The comparison in the ingested dose between our study and other published results are shown in table (4). The average dose in this study was the lowest value except the average dose in Malaysia and Bulgaria [18, 21].

Table (3). Activity concentrations (Bq /Kg) of ^{226}Ra, ^{232}Th and ^{40}K with the Total annual effective ingestion radionuclides dose (μSvy^{-1}) obtained from infants' powdered milk sample

Sample code	Average activity concentration (Bq/Kg)			Total annual effective ingestion radionuclides dose (μSvy^{-1})
	^{226}Ra	^{232}Th	^{40}K	
A	1.0±0.002	BMDL	61.0±0.4	135.7
B	2.6±0.006	2.4±0.059	82.0±0.5	265.0
C	2.2±0.004	2.2±0.040	60.9±0.3	214.3
D	3.0±0.005	2.3±0.043	114.1±0.5	331.1
E	2.4±0.004	1.8±0.032	133.7±0.6	332.2
F	2.9±0.006	1.7±0.035	138.4±0.8	355.9
G	2.5±0.005	2.1±0.041	125.2±0.6	329.3
H	2.1±0.004	1.8±0.039	120.4±0.7	300.0
I	1.7±0.003	1.3±0.030	109.7±0.6	259.9
J	2.2±0.004	1.7±0.031	110.0±0.5	284.7
K	2.3±0.004	1.4±0.024	154.9±0.7	357.1
L	2.0±0.003	1.4±0.025	151.9±0.6	341.2
M	1.5±0.002	1.3±0.023	139.4±0.6	298.9
N	1.9±0.003	1.4±0.023	129.2±0.6	298.1
O	0.5±0.001	1.1±0.019	36.2±0.2	96.8
Average	2.1±0.004	1.6±0.031	111.1±0.5	280.0
Maximum	3.0±0.006	2.4±0.059	154.9±0.8	357.1
Minimum	0.5±0.001	0.1±0.003	36.2±0.2	96.8
IAEA [17]	-----	-----	540	-----

*BMDL=below minimum detected limit

Table 4: Comparison between the average radionuclides concentrations of ^{226}Ra, ^{232}Th, 40K, and Ingestion dose in this study with the published data for infants' milk powder.

Region	Average radionuclides concentrations (Bq/Kg)			ingestion dose (μSvy^{-1})	Reference
	^{226}Ra	^{232}Th	^{40}K	D	
Saudi Arabia	9.64	6.77	74.5	183.7	[15]
Kuwait	0.4	0.42	243.5	413	[14]
Malaysia	3.05	2.55	99.1	635.1	[18]
Saudi Arabia	0.46	0.35	234	410.0	[13]
Jordan	0.5	0.78	296.8	332.0	[6]
India	2.5	-----	34.35	-----	[20]
Nigeria	23.07	4.35	831.6	-----	[19]
Bulgaria	1.63	5.0	53.1	150.0	[21]
Saudi Arabia	2.1	1.6	111.1	280.0	Present study

Conclusion

Activity concentrations of naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) consumed by infant between 0 to six month in Saudi Arabia was determined using gamma ray Spectroscopy, NaI(Tl) detector. The average activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in all the brands of infants' powdered milk were 2.1(3.0-0.5), 1.6(2.4-0.1) and 111.1(154.9-36.2) $Bqkg^{-1}$ respectively. The radioactivity values were lower than the recommended limit determined by ICRP-60 and UNSCEAR. In addition, these values lie within the most values of the corresponding radionuclides as measured from different brands of milk around the world. The mean annual effective dose due to the ingestion of radionuclides in infants' powdered milk was estimated to be well below the ICRP recommendation.

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