

Measurement of Natural Radionuclides Levels and their Annual Effective Doses in Different Types of Powdered Milk Consumed by Infants in Jordan

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ABSTRACT

In this study, fourteen samples of different types of imported and locally produced powdered milk consumed by infants in Jordan were analyzed using gamma-ray spectrometry system equipped with a High Purity Germanium (HPGe) detector. The activity concentrations of the natural radionuclides ²³⁸U, ²³²Th and ⁴⁰K present in the studied samples were measured. The measurements indicated that the radioactivity concentrations for: ²³⁸U ranged from 0.23±0.10 Bq.kg⁻¹ to 2.03±0.74 Bq.kg⁻¹ with an average of 1.1±0.3 Bq.kg⁻¹, for ²³²Th ranged from 0.12±0.08 Bq.kg⁻¹ to 0.68±0.29 Bq.kg⁻¹ with an average of 0.42±0.13 Bq.kg⁻¹ and for ⁴⁰K ranged from below detection limit to 479±27.1 Bq.kg⁻¹ with an average of 346±19.6 Bq.kg⁻¹. The annual effective dose rate that infants may experience as a result of the consumption of powdered milk (the study samples) and the contents of the above-mentioned radioactive elements were approximately 268 μSv per year. This total is way less than the permissible value of 1 mSv⁻¹ for public. The obtained results were compared with the international accepted values, and found to be within the acceptable limits. Therefore, according to the findings of this study, the investigated powdered milk samples does not pose any significant health hazard and is considered radiologically safe for infants consumption in Jordan.

Keywords: ²³⁸U, ²³²Th and ⁴⁰K, Naturally Occurring Radionuclides, Powdered Milk, Infants, Radioactivity Concentrations, Annual effective Dose (AED).

INTRODUCTION

The sources of ionizing radiation are found everywhere in the environment of the human being. They include Naturally Occurring Radioactive Materials (NORM) contained in the earth, building materials, air, food, water and cosmic rays¹. NORM include cosmogenic radionuclides such as ¹⁴C and ³H, primordial radionuclides, including the series of ²³⁸U, ²³⁵U, ²³²Th, and some independent nuclides such as ⁴⁰K². External dose from primordial radionuclides due to gamma radiation depends mainly on the geological and geographical

environmental conditions, and appear at different levels in the soils of each region in the world^{3, 4}. Basically, radionuclides in water and food are from natural sources. This natural radioactivity can be transferred from rocks and mineral present in the soil to plants. When animal eat the plant, this radionuclides can be transferred and remained in different organs of their bodies. Accordingly, throughout of chain food, plants and animals that will become food for people. Thus, provide a pathway for radionuclides to move from environment to people who live in places surrounded by NORM⁵. The world is naturally radioactive and around 90% of human exposure arises from natural sources^{6,7,8}. Therefore, assessment of the gamma radiation dose from natural sources is

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important as it is the biggest contributor to the external dose of the world population⁹. The ingested radionuclides could be concentrated in certain organs of the body. For example, ^{238}U is accumulated in human lungs and kidney, ^{232}Th in lungs, liver and skeleton tissues, and ^{40}K in muscles¹⁰. 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 rate¹¹.

The major foodstuffs contributing to the radiation ingestion dose will be those in which the radionuclides have large transfer rates or those consumed in large quantities. Such as milk (fresh/ powdered). Milk and its products are important components of diet in many countries, especially in the underdeveloped countries; people in these countries tend to use this product extensively in the diets of their children¹². Milk is consumed in significant amount, particularly by infants < 2 years old, since its minerals and proteins are essential for their growth. Apart from this, infant formula is produced from purified cow's milk and other protein sources. Contamination of milk will be greatest when cows are grazing during the fallout period. But even when cows are kept indoors, contamination of milk may occur by inhalation of radionuclides or ingestion of radionuclides in drinking water and contaminated feed. Since the first year of life is a very sensitive period in the development of human nervous, reproductive, digestive, and immune systems, detail study on the composition of baby foods (especially concentration of radionuclides) is crucial to ensure the safety and suitability of these foods¹³.

Among the types of food that are commonly consumed worldwide is milk powder. Hence, studies on the radioactivity of milk powder were performed in various regions across the globe¹⁴⁻²¹. In Jordan, several studies were performed to measure the level of radioactivity in environmental samples²²⁻²⁵. These studies mostly concentrated on measuring the radioactivity from soil and

radon in limited regions of the country. A thorough literature search reveals that there are insufficient data on the subject and a small number of studies on radionuclides content of food consumed in Jordan^{26,27}. The lack of information about the level of radioactivity in the powdered milk has persuaded one to initiate such studies^{12,28}. The levels in some type of powdered milk consumed by infants in Jordan need to be established in order to forecast any possible radiological risk associated with the consumption of the powdered milk. Thus, the main objectives of this research are: **i**) to quantify the presence of natural radionuclides in some powdered milk consumed by infants in Jordan, and **ii**) to estimate the average annual effective dose equivalent for ingestion of NORMS in milk powder samples. The collected data also can be used as baseline data on powdered milk radioactivity in Jordan for future reference.

MATERIALS AND METHODS

Materials

Six types of different brands of infant's powdered milk were taken as the samples. All samples used were both local and imported powdered milk and were purchased from local markets. The name of the powdered milk samples and their numbers are as follows: Jordanian samples (3), Ireland samples (3), France samples (2), Switzerland samples (3), Holland samples (2) and Dubai samples (1). This makes the total number of samples collected as fourteen. Table 1 lists the country of origin, Brand names, code No., age group consumers and production date of the powdered milk samples used in the present study.

Sample Preparation

The samples were prepared for the natural radioactivity concentration measurements. Each powdered sample was weighted (500 g) and sealed in 500 ml marinelli beakers. The samples were kept at room temperature (25 °C) for at least 30 days, before counting, to allow reaching the secular equilibrium of radium isotopes coming from

natural radioactive series (^{238}U and ^{232}Th) with their respective decay products, in which the activities of all radionuclide within each series are nearly equal.

Natural radioactivity measurement

The measurements were conducted at Physics Department/ Yarmouk University, Irbid-Jordan, using a Canberra p-type high purity germanium (HPGe) gamma spectrometer with 25% relative efficiency and resolution of 1.9 keV at 1.33 MeV of ^{60}Co gamma ray peak. The detector was connected to a computer with MCA card (Accuspec B) and Genie-2000 Analysis software of Canberra Industries, USA. A100-mm thick lead bricks shielded the detector from the background radiation from the radionuclides in the environment and cosmic rays. The calibrations of the energy and relative efficiencies for the detector were carried out using a multi-gamma ray reference standard source (MGS-5, Canberra, USA), which emits gamma rays in the range of 60 -1461 keV. The samples were counted for 86000 s to reduce the statistical counting error and quality control tests were performed using standard reference materials (IAEA-321 Powdered milk). An empty beaker was also counted under the same conditions to determine the background. The activity concentrations of each sample for the radionuclides of interest were determined from their respective gamma lines or gamma lines emitted by their progenies. The gamma lines used were 609 keV for ^{238}U (^{214}Bi), with emission percentage of 44.6, 238 keV for ^{232}Th (^{212}Pb), with emission percentage of 43.5 and 1461 keV for ^{40}K with emission percentage of 10.66

Determination of radioactivity concentration:

The activity concentration, A_i measured in Bq.kg^{-1} , of a radionuclide of interest in each sample was evaluated; ^{238}U concentration was calculated from ^{214}Bi (energy 609.3 keV, yield = 44.6%), ^{232}Th concentration was calculated from ^{212}Pb (energy 238.6 keV, yield = 43.5%) and ^{40}K concentration was calculated from its own gamma photopeaks (energy 1461, yield = 10.7%). using the

following equation^{29,30}.

$$A_i = \frac{C_i}{\varepsilon(E) m p_\gamma(E,i)} \quad (1)$$

where C_i is the net count rate of nuclide i , $\varepsilon(E)$ is the detector efficiency at energy E , m is the mass of the sample in kilograms, and $p_\gamma(E,i)$ is the emission probability of radionuclide i at energy E .

Calculation of Average “Annual effective Dose” (AED):

The average annual effective dose equivalent for ingestion of NORMS in milk powder samples were evaluated using the following expression³¹:

$$AED \left(\frac{\text{mSv}}{\text{a}} \right) = \sum_i I (DCF)_i A_i \quad (2)$$

where I is the consumption rate from intake of NORMS in milk powders, A_i is the specific activity of radionuclide i in the milk samples and $(DCF)_i$ is the dose conversion factor of radionuclide i for certain group age. For infants, $(DCF)_U = 960 \text{ nSv/Bq}$, $(DCF)_{Th} = 450 \text{ nSv/Bq}$ and $(DCF)_K = 42 \text{ nSv/Bq}$ ³².

RESULTS AND DISCUSSION

The measured activity concentration of ^{238}U , (^{226}Ra), ^{232}Th , and ^{40}K in Bqkg^{-1} present in each brand are displayed in Table 2. For the powdered milk it is shown that ^{238}U and ^{232}Th were detected in all samples and displayed relatively low activity concentrations ranged from 0.64 ± 0.58 to $1.64 \pm 0.63 \text{ Bqkg}^{-1}$ and 0.14 ± 0.09 to $0.55 \pm 0.34 \text{ Bqkg}^{-1}$, respectively. On The other hand, ^{40}K was measured above the detection limit in eleven brands and varied between $197 \pm 13.12 \text{ Bqkg}^{-1}$ and $479 \pm 27.1 \text{ Bqkg}^{-1}$. This variation in the activity concentrations is not significantly large from one brand to another. The average values of the activity concentration of the ^{238}U (^{226}Ra), ^{232}Th , and ^{40}K in Bqkg^{-1} are displayed in Table 2. The average concentrations of ^{238}U , ^{232}Th , and ^{40}K were $1.1 \pm$

0.3·0.42 ± 0.13 and 346 ± 20 Bq/kg, respectively. Figure (1) shows that the highest average concentration of ^{238}U was found in Holland samples, while the lowest average concentration was found in Dubai samples. ^{238}U average concentration for all samples ($1.1 \pm 0.3 \text{ Bqkg}^{-1}$) is comparable with the world average and lies within the acceptable value, which is less than 35 Bqkg^{-1} for infant's safe consumption¹¹. The average value concentration of ^{232}Th was $0.42 \pm 0.13 \text{ Bqkg}^{-1}$. The results indicate that the maximum concentration of ^{232}Th occurred mainly in Ireland samples (0.55 Bqkg^{-1}) and the minimum in Holland samples (0.14 Bqkg^{-1}). It was also observed that the average concentration of ^{238}U in Jordanian milk samples was higher than the average concentration of ^{232}Th . This is due to high concentration of ^{226}Ra in Jordanian soil compared to the concentration of ^{228}Ra ^{22,24}. In general the measured average concentration value of ^{232}Th ($0.42 \pm 0.13 \text{ Bqkg}^{-1}$) lies within the acceptable values as was reported by UNSCEAR (United Nation Scientific Committee on the Effects of Atomic Radiation) which is 30 Bqkg^{-1} ¹¹. Potassium is a very soluble in water and an essential element in metabolism of living organism. The highest activity concentration of ^{40}K was found in Ireland samples-Pro3 ($479 \pm 27.1 \text{ Bqkg}^{-1}$), followed by Jordanian samples-Fo3 ($387 \pm 21.7 \text{ Bqkg}^{-1}$) and Holland samples-Be2 ($360 \pm 20.3 \text{ Bqkg}^{-1}$). The lowest concentration was found in Switzerland samples with a value of $197 \pm 13.9 \text{ Bqkg}^{-1}$. ^{40}K grand average concentration in the studied samples was ($346 \pm 20 \text{ Bqkg}^{-1}$). This average is less than the internationally recommended upper limit (400 Bqkg^{-1}) for the safe use¹¹.

The contribution of ^{238}U , ^{232}Th and ^{40}K into the annual effective dose (*AED*) was calculated using equation (2). Figure (2) shows the results of these calculations. *AED* due ^{40}K was the largest, $247 \mu\text{Svy}^{-1}$, while *AED* from ^{232}Th was the smallest, $3.1 \mu\text{Svy}^{-1}$ and *AED* from ^{238}U was $18.12 \mu\text{Svy}^{-1}$. However, the total of *AED* was $268 \mu\text{Svy}^{-1}$. This total is way less than the permissible value of 1 mSvy^{-1} for public^{33,34}.

The results reported here are compared with those obtained by other studies worldwide (Table3). As can be seen from the table, the ^{40}K activities are, on an average lower by almost 25%¹⁴⁻²¹ of the global value. As for ^{232}U (^{226}Ra) and ^{232}Th (^{228}Ra), the data available in the literature are not enough to allow for an exhaustive comparison because in most cases these isotopes were not detected¹⁴⁻¹⁸

Analysis of Variance

Table 4 shows the descriptive statics of activity concentration of ^{238}U , ^{232}Th , and ^{40}K of different countries. The variation in the levels of radionuclides activity concentrations were mainly due to the geological formation of the country of origin. Milk powder imported from Ireland showed high concentration of radionuclide as the country consists of granitic rock and sediments and experienced many volcanic activities¹⁴. The milk powder imported from Switzerland show low level of radionuclide concentration as the country has a clean and healthy environment which also being far from any battle fields and facilities where nuclear explosions took place¹⁵. The radiation dose could vary also due to differential bioaccumulation of radionuclide by organisms, contamination of food through processing and preparation techniques and feeding habits by a particular population^{16,31}.

CONCLUSION

In this research, 14 samples of different types of imported and locally produced powdered infant milk in central zone of Jordan were examined and evaluated. The gamma-ray spectrometry system equipped with a high Purity germanium (HPGe) detector was used in this study. The results have shown that the grand average activity concentrations of natural radionuclides of ^{238}U , ^{232}Th , and ^{40}K ($1.1 \pm 0.3 \text{ Bqkg}^{-1}$, $0.42 \pm 0.13 \text{ Bqkg}^{-1}$, $346 \pm 20 \text{ Bqkg}^{-1}$ respectively) in samples of different types of imported and locally produced powdered milk consumed by infants in Jordan are relatively lower than the world recommended

limits (35 Bqkg⁻¹, 30 Bqkg⁻¹ and 400 Bqkg⁻¹ for ²³⁸U, ²³²Th and ⁴⁰K, respectively)¹¹. The calculated total average annual effective dose (AED = 68.3 μSv.y⁻¹) from these natural radioactive nuclides (²³⁸U, ²³²Th and ⁴⁰K) due to the ingestion of powdered milk by infant (less than two years) were found to be less than the WHO recommended limit and ICRP for radiological safety^{33,34}. As far as we concerned, this study is the first in the country for the estimation of AED from the radionuclide concentration in infant's milk powder. The data reported here might be

useful to establish a baseline and help to develop future guidelines in the country for radiological protection.

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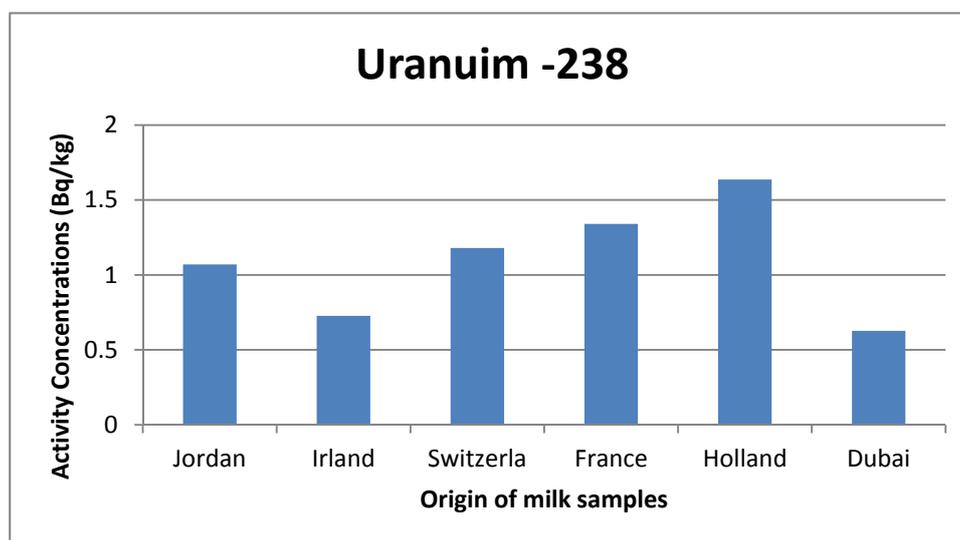
Table 1. The country of origin, Brand names, code No., age group and production date of the powdered milk samples used in the present study.

Origin	Brand name	Code No.	Age group (months)	Production date(dd/mm/yy)
Jordan	Sahha	Sa ₁	0-6	20/8/2017
	Sahha	Sa ₂	6-12	23/7/2017
	Formelac	FO ₃	12-24	18/7/2017
Ireland	S-26 Promil	Pro ₁	0-6	21/6/2017
	Similac Gain	Si ₂	6-12	21/11/2016
	S-26 Progress	Pro ₃	12-24	18/7/2017
Switzerland	Nan	Na ₁	0-6	/6/2016
	Nan	Na ₂	6-12	22/4/2017
	Primalac	Pri ₃	12-24	2/2/2016
France	Nidina	Ni ₁	0-6	/6/2017
		Ni ₂	6-12	/6/2017
Holland	Bebelac	Be ₁	0-6	28/3/2017
		Be ₂	6-12	18/4/2017
Dubai- U.A.E	Nido	Nid ₃	12-24	/9/2017

Table 2. Origin, code no (CN) of samples, activity concentrations and their averages for ^{238}U , ^{232}Th and ^{40}K in powdered milk samples (Bq/kg) used in the present study.

Origin	CN	Activity Concentration, Bq/kg		
		^{232}U	^{238}Th	^{40}K
Jordan	Sa1	1.34 ± 0.57	0.42 ± 0.26	BDL
	Sa2	1.41 ± 0.62	0.37 ± 0.29	333 ± 18.89
	Fo3	0.46 ± 0.31	0.4 ± 0.29	387 ± 21.7
	Average	1.1	0.4	360
Ireland	Pro1	1.43 ± 0.75	0.46 ± 0.35	BDL
	Si2	0.52 ± 0.2	0.68 ± 0.29	445 ± 24.7
	Pro3	0.23 ± 0.1	0.51 ± 0.39	479 ± 27.1
	Average	0.73	0.55	462
Switzerland	Na1	2.03 ± 0.74	0.45 ± 0.2	197 ± 13.12
	Na2	0.61 ± 0.2	0.41 ± 0.29	327 ± 18.60
	Pri3	0.9 ± 0.58	0.53 ± 0.28	BDL
	Average	1.2	0.47	262
France	Ni1	1.26 ± 0.75	0.57 ± 0.45	295 ± 17.4
	Ni2	1.42 ± 0.64	0.37 ± 0.29	333 ± 18.85
	Average	1.34	0.47	317
Holland	Be1	1.41 ± 0.64	0.15 ± 0.09	307 ± 17.6
	Be2	1.86 ± 0.62	0.12 ± 0.08	360 ± 20.3
	Average	1.64	0.14	334
Dubai	Nid3	0.64 ± 0.58	0.5 ± 0.27	327 ± 18.3

BDL: Below Detection Limit.

**Figure 1: Average activity concentrations of U-238 in milk samples from different country (an example from Anova outcome)**

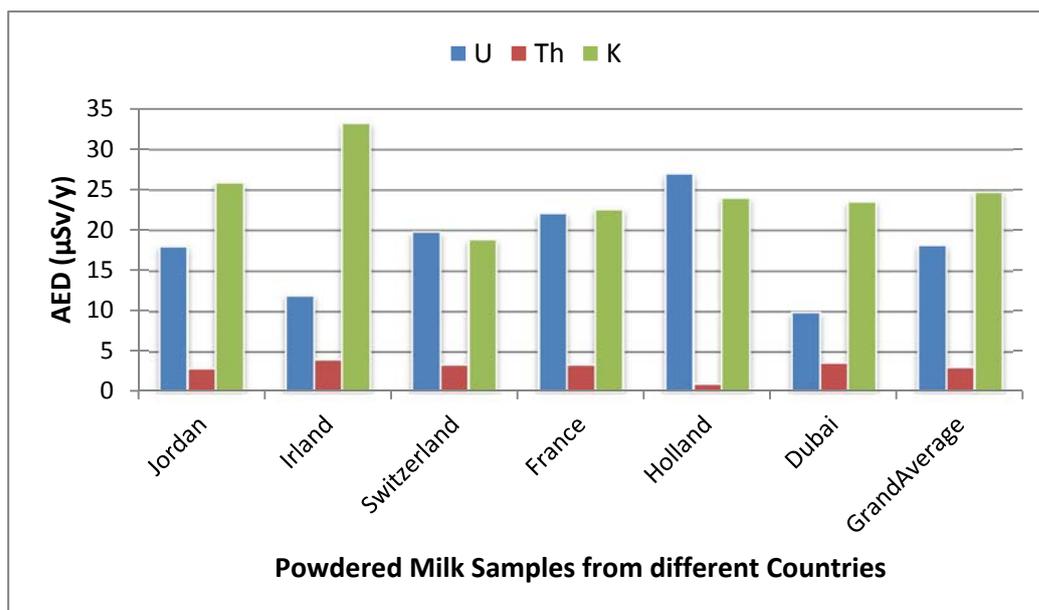


Figure 2: Shows the annual effective dose of U-238, Th-232 and K-40 due to ingestion of milk samples from different countries. Values of ⁴⁰K must be multiplied by 10.

Table 3. Measured average concentration activity of ²³⁸U, ²³²Th and ⁴⁰K in powdered milk samples (Bq/kg); in compared with the literature.

Origin of sample manufactured (number of sample)	Activity Concentration, Bq/kg			Reference
	²³² U	²³⁸ Th	⁴⁰ K	
This study	1.1 ± 0.3	0.42 ± 0.13	346 ± 19.6	
Jordan	1.24 ± 0.77	1.11 ± 0.20	348.30 ± 26.03	12
Malaysia	-	0.82 ± 0.08	238.71 ± 1.04	14
Iraq	3.90 ± 1.97	1.96 ± 1.40	177.91 ± 13.33	15
Brazil	-	-	464.96 ± 34.68	17
Venezuela	-	-	401.71 ± 13.45	17
Nigeria	-	BDL	39.60 ± 12.6	18
Saudi Arabia	-	0.29 ± 0.02	210.70 ± 2.26	19
Germany	-	0.09 ± 0.03	610.00 ± 18.30	20
France	-	0.14 ± 0.03	434.10 ± 13.00	20
New Zealand	-	0.15 ± 0.04	605.550 ± 12.10	20
Iran	BDL	BDL	254.78 ± 3.38	21
Spain	0.35 ± 0.02	BDL	241.16 ± 3.38	21
World average	35	30	400	11

BDL: Below Detection Limit.

Table 4. One way Anova output of ^{238}U activity concentrations in milk samples from different countries

Anova: Single Factor for Th-232

SUMMARY						
Groups	Count	Sum	Average	Variance		
Jordan	3	1.19	0.396667	0.000633		
Irland2	3	1.65	0.55	0.0133		
Switzerland	3	1.39	0.463333	0.003733		
France	2	1.41	0.47	0.01		
Holland	2	0.41	0.136667	0.000233		
Dubai	1	1.92	0.64	0		

ANOVA						
Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	0.442161	5	0.088432	19.01768	2.5E-05	3.105875
Within Groups	0.0558	12	0.00465			
Total	0.497961	17				

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قياس "مستويات النويدات المشعة الطبيعية" والجرعات الفعالة الخاصة بها في أنواع مختلفة من الحليب المجفف المستهلك من قبل الأطفال الرضع في الأردن

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ملخص

تم في هذا البحث تحليل اربع عشرة عينة من انواع مختلفة من الحليب المجفف المستورد والمنتج محليا والتي يستهلكها الرضع في الاردن باستخدام نظام قياس طيف اشعة جاما المجهز بكاشف جرمانيوم عالي النقاوة . تم قياس تراكيز النشاط من النويدات المشعة الطبيعية الموجودة في العينات التي تم دراستها ، كما تم حساب معدل الجرعة الفعالة السنوية. لقد بينت القياسات ان متوسط تراكيز اليورانيوم-238 والثوريوم-232 والبوتاسيوم-40 هي كالتالي: 1.1 ± 0.3 ، 0.42 ± 0.13 و 343 ± 20 بيكريل/كغم على التوالي. وكان معدل الجرعة الفعالة السنوية التي يمكن ان يتعرض لها الأطفال الرضع نتيجة استهلاك حليب البودرة (عينات الدراسة) وما تحويه من العناصر الطبيعية المشعة سالفة الذكر حوالي 268 ميكروسيبرت في السنة. ان هذه النتائج متواضعة جدا مقارنة مع المعايير الدولية المتفق عليها. ولقد قورنت نتائج هذه الدراسة مع نتائج دراسات مشابهة في مناطق أخرى من العالم وتبين انها تقع ضمن معدلات تلك الدراسات. لذلك فان عينات الحليب المجفف التي تم التحقق منها في هذه الدراسة لا تشكل اي خطر صحي وتعتبر آمنة اشعاعيا لاستهلاك الرضع في الاردن.

الكلمات الدالة: النويدات المشعة التي تحدث بشكل طبيعي ، الحليب المجفف في الاردن، الرضع، تركيز ^{238}U ، ^{232}Th ، ^{40}K النشاط الاشعاعي، معدل الجرعة الفعالة السنوية..

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