Estimating Radioactivity in Various Types of Milk using NaI (Tl) detector

تقيم النشاط الاشعاعي في انواع متغيره من الحليب باستخدام كاشف NaI(Tl)

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Abstract

In this research, uranium (^{238}U), thorium ,(^{232}Th) and potassium (^{40}K) specific activity in (Bq/kg) were measured in 10 different types of milk that are available in iraqi markets. The gamma spectrometry method with a NaI(Tl) detector has been used for radiometric measurements. Also we have calculated the radiation hazard indices (radium equivalent activity and internal hazard index) and ingestion effective dose in all samples.

It is found that the specific activity in milk samples were varied from (0.274±0.158) Bq/kg to (7.851±0.877) Bq/kg, for ^{238}U, for ^{232}Th from (0.821±0.273) Bq/kg to (3.067±0.569) Bq/kg and for ^{40}K from (266.558±5.018) Bq/kg to (485.740±7.197) Bq/kg. Also, it is found that the radium equivalent activity and internal hazard index in milk samples ranged from (27.341) Bq/kg to (48.035) Bq/kg and from (0.07867) to (0.14720) respectively. But the range of summation of the ingestion effective dose were varied from (4.861×10^{-5}) Sv/y to (1.541×10^{-4}) Sv/y. This study proves that the natural radioactivity, radiation hazard indices and ingestion effective dose were lower than the safe, except the some value of potassium.

Keywords: Milk; Natural radioactivity; Iraq market and gamma spectroscopy

الخلاصة:

في هذه الدراسة تم قياس النشاط النوعي لليورانيوم والثوريوم والبوتاسيوم في 10 نماذج من انواع مختلفة للحليب في الأسواق العراقية . استخدم اسلوب طيف كاما للكشف باستخدام كاشف ايوديد الصوديوم المطعم بالثاليوم ، حيث تم حساب معاملات الخطورة (مكافئ الراديوم ومعامل الخطورة الداخلي) والجرع المؤثرة لتناول الحليب في جميع النماذج.

وكانت نتائج النشاط النوعي لليورانيوم تتراوح بين 0.274±0.158 Bq/kg الى 7.851±0.877 Bq/kg ، الثوريوم بين 0.821±0.273 Bq/kg الى 3.067±0.569 Bq/kg اما البوتاسيوم 266.558±5.018 Bq/kg الى 485.740±7.197 Bq/kg. ووجد مكافئ الراديوم ومعامل الخطورة الداخلي في النماذج بمدى من 27.341 Bq/kg الى 48.035 Bq/kg 0.07867 الى 0.14720 على التوالي . اما الجرعة المؤثرة السنوية لتناول الحليب متغيرة بين 4.861×10^{-5} Sv/y الى 1.541×10^{-4} Sv/y. اثبتت هذه الدراسة ان النشاط الطبيعي ومعاملات الخطورة والجرع المؤثرة السنوية لتناول الحليب كانت منخفضه وضمن حدود الأمان ، ما عدى بعض قيم البوتاسيوم.

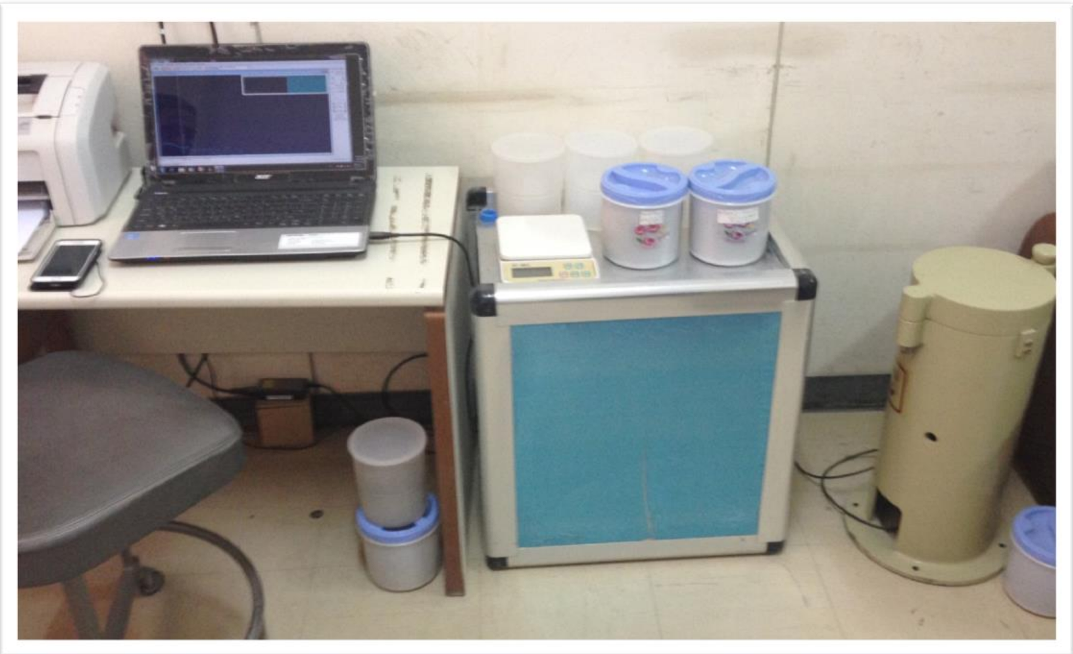
Introduction

Radionuclides of uranium-238, thorium-232, radium-226, and potassium-40 are enabled to enter human body mainly through inhalation of contaminated air and ingestion of contaminated water and food. Other pathways for radionuclide penetration into human body, for example through the skin, are irrelevant for the context of this study. According to their different physic-chemical properties, they can accumulate in different tissues and organs of the human body and, thus, representing a serious health issues for exposed workers and population all over the world. In particular, their assumption through daily intake of food can turn out to be quite relevant to be taken in consideration. For example, chemical uranium-238 turns out to be toxic primarily for lungs and kidneys, where it causes damages to the proximal tubule, besides the fact that when in the metal form, it has also been identified as a potential reproductive toxicant; thorium-232 affects lungs, liver and skeleton tissues; potassium accumulates in muscles and radium is retained primarily in bones, due to metabolic similarities with calcium. Therefore, depositions of large quantities of these radionuclides in particular organs will be able to affect the health condition, through the weakening the immune system, inducing various types of diseases, and finally contributing to increasing the mortality rate [1].

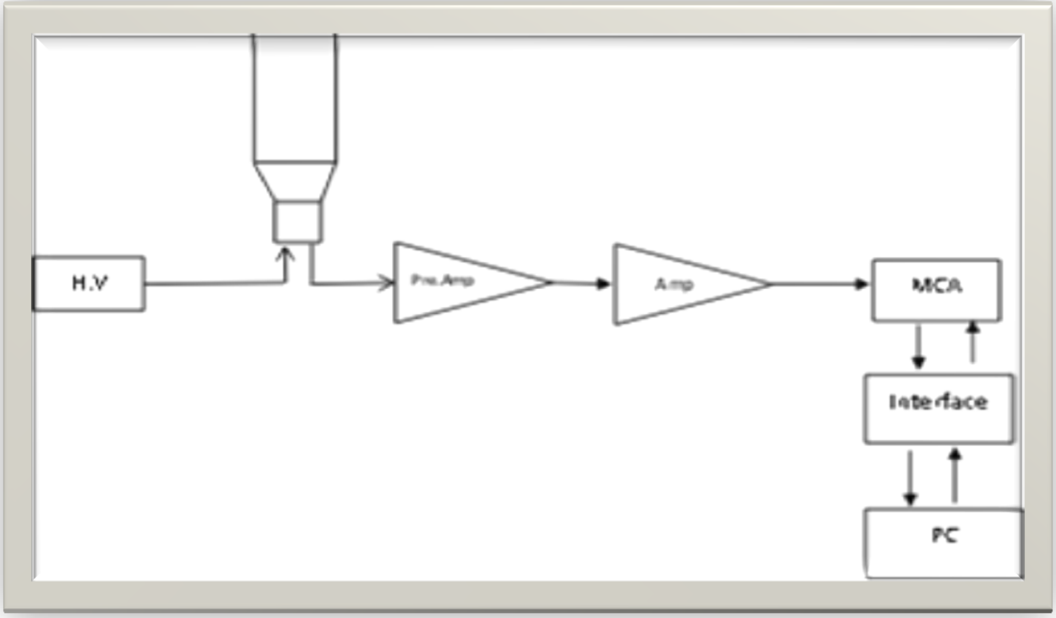
Milk is an important vector of radionuclides that human beings may get from the environment through the mining activities [2]. Also, milk is one of the important food for the human nutrition and contains all the macronutrients namely protein, carbohydrates, fat, vitamins (A, D and B groups) and trace elements particularly calcium, phosphate, magnesium, zinc and selenium [3]. In iraq no surveys of natural radioactivity in milk have been carried out and so far no baselines of concentration of natural and anthropogenic radioisotopes have been reported. Therefore, the monitoring of radioisotope activity concentrations will provide meaningful information that can contribute to the knowledge of population exposure and to the setting up of original baseline. This work has been undertaken with the purpose of measuring natural radioactivity to ^{238}U (^{226}Ra), ^{232}Th and ^{40}K in powdered milk samples that are available in the Iraqi markets. Also radium equivalent activity, internal hazard, the absorbed dose rates and annual effective dose for ^{238}U (^{226}Ra), ^{232}Th and ^{40}K to human beings in different ages have been calculated depending on standard world equations, so as to compare them with the recommended reference limits[4].

Materials and Methods

Ten samples of different types of powdered milk for infants have been collected from the local markets in Iraq to measure natural radioactivity. The type of milk samples are listed in Table (1), natural radioactivity levels have been measured using a gamma spectrometry facility, consisting of a gamma multichannel analyzer equipped with a (3"×3") NaI(Tl), figure (1):



(a)



(b)

Figure 1: (a) Experimental set-up, (b) set-up block diagram

Table (1): Types and origins of the powdered milk samples.

No.	Sample Code	Sample name	Country of Origin
1	M1	Golden cow's milk	Jordan
2	M2	Sponsor milk	Arab Emirates
3	M3	Surprisingly milk	Sultanate of Oman
4	M4	Anchor milk	New Zealand
5	M5	Pew milk	Ireland
6	M6	Diallak milk	Vietnam
7	M7	Aktal milk	France
8	M8	Lancy milk	Jordan
9	M9	Niro milk	Arab Emirates
10	M10	Nktalia milk	France

The theoretical part

The gamma spectra have been analyzed using the ORTEC Maestro-32 data acquisition and analysis system. The detector has a coaxial closed-facing geometry with the following specifications: the calculated resolution is 7.9% for energy of 661.66 keV of a ^{137}Cs standard source. Relative efficiency at 1.33 MeV for ^{60} Co was 2.2% and at 1.274 MeV for ^{22}Na was 2.4%. The detector has been shielded by a cylindrical lead shield in order to achieve the lowest background level. An energy calibration for this detector has been performed with a set of standard γ-ray 1-μCi active^{137}Cs, ^{60}Co, ^{54}Mn, and^{22}Na sources. In this study, the activity concentration of ^{40}K has been determined directly from the peak areas at 1460 keV. The Activity concentrations of ^{238}U (^{226}Ra) and^{232}Th have been calculated assuming secular equilibrium with their decay products. The gamma transition lines of ^{214}Bi (1765 keV) have been used to calculate the specific activity of the radioisotopes in the ^{238}U-series, while for the ^{232}Th-series the specific activities have been determined using gamma transition lines of ^{208}Tl (2614 keV). The counting time for each sample has been of about 18000 s. Since the counting rate is proportional to the amount of radioactivity contained in a sample, the Activity Concentration, or Specific Activity, A_x, can be evaluated in the following way [5].

$A_x = \frac{C - BG}{t\varepsilon\% MI\gamma}$.................(1)

Where C is the area under the photo-peaks, ε% the present of energy efficiency, Iγ the present of gamma-emission probability of radionuclide X under consideration, t the counting time, M the mass of sample and BG the background [6].

Radium Equivalent Activity

The most common Naturally Occurring Radioisotopes that can be found in foodstuff are represented by ^{232}Th, ^{40}K and ^{238}U (^{226}Ra). In order to assess the gamma radiation doses to human beings due to the (internal or external) exposure to them, it is necessary to evaluate how to take properly into account their specific radioactivity. To this purpose a suitable and practical index, the so-called radium equivalent activity index Ra_{eq} has been introduced by E. I. Hamilton from the UK National Radiological Protection Board (NRPB) [7]. Ra_{eq} has been defined on the basis of the preliminary estimation of the quantities of these radionuclides releasing the same gamma ray dose. From the extensive review of the investigations, performed in different countries, realized by a group of experts from the Organization for Economic Co-operation and Development (OECD)'s Nuclear Energy Agency [8] it turns out that a general agreement can be found on the statement that 370 Bq/kg (10 pCi/g) of ^{226}Ra, or 260 Bq/kg (7 pCi/g) of ^{232}Th or4810 Bq/kg (130 pCi/g) of ^{40}K provide the same gamma ray doses {15– 18}. Consequently, the following Radium Equivalent Activity (Raeq) of a sample in (Bq/kg) can be evaluated as {18-22}:

$Ra_{eq}=A_{Ra}+(1.43A_{Th}) + (0.077A_k)$ .... (2)

where A_{Ra} is the specific activity of ^{226}Ra, which is usually the same as that of ^{238}U (^{226}Ra), in (Bq/kg), A_{Th} is the specific activity of ^{232}Th, in (Bq/kg), and A_K the specific activity of ^{40}K, in (Bq/kg). This equation is based on the estimation that 10 Bq/kg of ^{226}Ra equal 7 Bq/kg of ^{232}Th and 130 Bq/kg of ^{40}K produce equal gamma dose. The maximum value of Ra_{eq} must be less than 370 Bq/kg [9].

Internal Hazard Index

In the international scientific community there is a general consensus that regarding the human internal exposure, due ingestion or inhalation, the introduction of a specific so-called Internal Hazard Index turns out to be very suitable to assess the radiation hazard. The Internal Hazard index (Hin) is given by the following expression [10]:

$H_{in}=\frac{A_{Ra}}{185}+\frac{A_{Th}}{259}+\frac{A_K}{4810} \leq 1$ …………………………………......... (3)

a consequence radiation hazard can be considered negligible if H_{in} turns out to be smaller than one.

Annual effective dose

The annual effective dose due to the intake of ^{238}U (^{226}Ra) , ^{232}Th and^{40}K in foods can be calculated using the formula from UNSCEAR (2000) [20]:

$D = C \times I \times E$……………………………………….................. (4)

Where D is the annual effective dose (Sv/y), C is the specific activity of radionuclides in the ingested sample (Bq/kg), I is the annual intake of powdered milk (kg/y) which depends on a given age and E is the ingested dose conversion factor for radionuclides (Sv/Bq). The conversion factor 'E' varies with both radioisotopes and the ages of the individuals and is determined by ICRP (1995). The average consumption rate of milk for children, in different age groups (age from 2 to 7 y, 7 to12 y and from 12 to 17 y), and adults (from 17 y) is 14kg/y and 13kg/y, respectively [11].

Results and Discussion

The measured specific activities of ^{238}U (^{226}Ra), ^{232}Th and ^{40}K detected in the samples of powdered milk under study including their uncertainty are summarized in Table 2. It can be noticed that the ^{238} U (^{226}Ra) activity concentrations detected in most of samples vary between (0.274±0.158) Bq/kg to (6.471±0.802) Bq/kg with an average value of (3.881±0.563) Bq/kg. For ^{232}Th and ^{40}K the measured specific activity ranged from(0.821±0.273) Bq/kg to (2.125±0.443) Bq/kg with an average value of (1.588± 0.396) Bq/kg and from(266.558±5.018) Bq/kg to (483.595±6.984) Bq/kg with an average value of (387.187± 6.299) Bq/kg, respectively. Also, the values of radiation hazard expressed by Ra_{eq} and H_{in} have been found to range from (27.341) to (48.035) Bq/kg with an average of (35.967) Bq/kg and from (0.078) to (0.147) with an average value of (0.107) respectively Table 3. The average value of the annual effective dose for children in the different age groups: (2-7)y, (7-2)y, (12-17)y and for adults (age over 17 y) as shown Table5: due to the intake for the three different radionuclides has resulted to be: for ^{238}U (^{226}Ra): 0.337×10^{-4} Sv/y,0.435×10^{-4}Sv/y, 0.185×10^{-5} Sv/y and 0.548×10^{-4} Sv/y; for ^{232} Th 0.735×10^{-5} Sv/y,0.645×10^{-5} Sv/y, 0.556×10^{-5} Sv/y and 0.548×10^{-5} Sv/y; for ^{40}K 0.113×10^{-3} Sv/y,0.704×10^{-4} Sv/y, 0.412×10^{-4} Sv/y and0.360×10^{-4} Sv/y Table 4.

Table (2): Specific activity (Bq/kg) in powder milk samples

No.	Sample Code	Sample Name	Specific activity		
			K-40	U-238	Th-232
1	M1	Golden cow's milk	319.780±6.200	1.108±0.350	1.800±0.499
2	M2	Sponsor milk	482.829±7.227	6.471±0.802	3.067±0.569
3	M3	Surprisingly milk	376.597±5.927	5.324±0.676	0.821±0.273
4	M4	Anchor milk	485.740±7.197	7.851±0.877	0.834±0.294
5	M5	Pew milk	466.497±6.813	1.740±0.399	0.875±0.291
6	M6	Diallak milk	266.558±5.018	4.782±0.644	2.125±0.443
7	M7	Aktal milk	372.640±6.090	0.274±0.158	1.362±0.364
8	M8	Lancy milk	306.744±5.935	1.796±0.435	1.347±0.389
9	M9	Niro milk	310.897±5.600	6.869±0.798	1.479±0.382
10	M10	Nktalia milk	483.595±6.984	2.599±0.491	2.170±0.462
Average			387.187±6.299	3.881±0.563	1.588±0.396
The global average			400	30	35

Table (3) :Radiation hazard in power milk samples

No.	Sample code	Sample name	Ra_{eq}(Bq/kg)	Internal hazard index
1	M1	Golden cow's milk	28.305	0.07942
2	M2	Sponsor milk	48.035	0.14720
3	M3	Surprisingly milk	35.496	0.11024
4	M4	Anchor milk	46.446	0.14664
5	M5	Pew milk	38.912	0.10977
6	M6	Diallak milk	28.355	0.08947
7	M7	Aktal milk	30.915	0.08421
8	M8	Lancy milk	27.341	0.07867
9	M9	Niro milk	32.923	0.10748
10	M10	Nktalia milk	42.939	0.12297
Average			35.967	0.10761
The global average			370	1

Table (4): Average annual effective dose ^{238}U (^{226}Ra),^{232}Th and ^{40}K for children in the age groups (2-7y), (12-17y) and adults (over17y) .

Sample code	Annual effective dose (Sv/y)			
	^{40}K	^{238}U	^{232}Th	Sum
Children (2-7) y	0.113×10^{-3}	0.337×10^{-4}	0.735×10^{-5}	1.541×10^{-4}
Children (7-12) y	0.704×10^{-4}	0.435×10^{-4}	0.645×10^{-5}	1.201×10^{-4}
Children (12-17) y	0.412×10^{-4}	0.815×10^{-5}	0.556×10^{-5}	4.861×10^{-5}
Adults(over 17y)	0.360×10^{-4}	0.163×10^{-4}	0.548×10^{-5}	5.778×10^{-5}

Table(5):the conversion factor and annual intake [12]

Type	Conversion factor(Sv/Bq)			Annual intake (kg/y)
	U-238	Th-232	K-40	
Powder milk (Infants ≤1Y)	4.7×10^{-6}	4.6×10^{-6}	6.2×10^{-8}	22.4
Powder milk (Infants ≤1-2Y)	9.6×10^{-7}	4.5×10^{-7}	4.2×10^{-8}	15
Powder milk (Children 2-7Y)	6.2×10^{-7}	3.5×10^{-7}	2.1×10^{-8}	14
Powder milk (Children 7-12Y)	8.0×10^{-7}	2.9×10^{-7}	1.3×10^{-8}	14
Powder milk (Children 12-17Y)	1.5×10^{-7}	2.5×10^{-7}	7.6×10^{-9}	14
Powder milk (Adults 17Y)	2.8×10^{-7}	2.3×10^{-7}	6.2×10^{-9}	13

These results have been compared with the outcomes of different investigations performed in other countries. The comparison is shown in Table (6).

Table (6): Comparison of the average specific activities (Bq/kg) of ^{238}U (^{226}Ra),^{232}Th and ^{40}K in powdered milk with data published in other countries.

Region	^{226}Ra	^{232}Th	^{40}K	Reference
Egypt	0.44±0.23	---	134.7±12	[13]
Iran	---	---	17.3±3.3	[14]
Jordan	2.14	1.28	392	[15]
Syria	----	---	435	[16]
Saadi Arabia	9.64	6.77	74.51	[17]
Brazil	----	3.7	482±9.9	[18]
France	0.05±0.01	0.142±0.026	434.1±13	[19]
Present work	3.881±0.563	1.588±0.396	387.187±6.299	

The results obtained show that for ^{238}U (^{226}Ra) the average measured specific activities and their average value appears to above the values reported in other countries; for ^{232}Th some values are greater and others lower. But for this two radioisotopes the results measured in Iraq turn out to be lower than the recommended reference limits by UNSCEAR (2000) [20]: 32 Bq/kg and 45 Bq/kg) respectively. For what it concerns the specific activity of ^{40}K the comparison with other outcomes shows the same behavior like for^{232}Th, but , nevertheless, the results from this study have been found to be lower than the recommended reference limit by UNSCEAR (2000): 412 Bq/kg [20] with the only exceptions of samples M2,M4,M5 and M10. Because potassium is a macronutrient indispensible to plants, this fact could be attributed, after a preliminary evaluation, to a local higher concentration of Potassium and hence, ^{40}K, in plants consumed by livestock in Iraq.

Radium equivalent activities and internal hazard index (H_{in}) values for all the samples are below the recommended value of 370 Bq/kg and one, respectively [17]. Also, it is found that the average annual effective dose from natural radioactivity turn out to be below the reference value of 1.0 mSv/y recommended by ICRP for all ages [21].

Conclusion

Natural radioactivity for the most available powdered and liquid milk brands consumed in Iraqi markets have been determined by using gamma spectroscopy. ^{238}U (^{226}Ra) and^{232}Th activities have been found below the prescribed international reference levels, while some samples have ^{40}K value which, however, have been found to be within the world wide ranges as reported in the international scientific literature.

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