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Natural Radioactivity Levels in Some Milk Samples Available in Local Markets in Babylon Governorate

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Abstract: 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±0.158-7.851±0.877 Bq/kg for ^{238}U for ^{232}Th from 0.821±0.273-3.067±0.569 Bq/kg and for ^{40}K from 266.558±5.018-485.740±7.197 Bq/kg. Also, it is found that the of radium equivalent activity and internal hazard index in milk samples ranged from 27.341-48.035 Bq/kg and from 0.07867-0.14720, respectively. But, the range of summation of the ingestion effective dose were varied from 4.861×10^{-5}-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.

Key words: Milk, natural radioactivity, Iraq market and gamma spectroscopy, internal hazard, measurements, Iraqi markets

INTRODUCTION

Radionuclides, both 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, like, 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 (Tawalbeh *et al*., 2012). Milk is an important vector of radionuclides that human beings may get from the environment through the mining activities (Anonymous, 1989). 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 (Anonymous, 1982). 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 research 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 in this study, 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 (Anonymous, 1999).

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

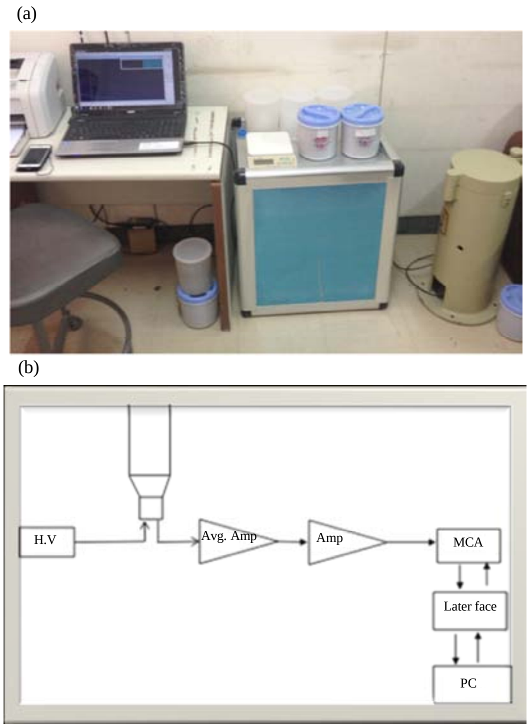

Fig. 1: a) Experimental set-up and b) Set-up block diagram

Table 1: Types and origins of the powdered milk samples

Sample codes	Sample name	Country of origin
M1	Golden cow's milk	Jordan
M2	Sponsor milk	Arab Emirates
M3	Surprisingly milk	Sultanate of Oman
M4	Anchor milk	New Zealand
M5	Pew milk	Ireland
M6	Diallak milk	Vietnam
M7	Aktal milk	France
M8	Lancy milk	Jordan
M9	Niro milk	Arab Emirates
M10	Nktalia milk	France

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) (Fig. 1).

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 a-ray 1-iCi 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 (Beretka and Matthew, 1985):

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

Where:

C	=	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 (Kumar *et al*., 2003)

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 Raeq has been introduced by E.I. Hamilton from the UK National Radiological Protection Board (NRPB) (Viruthagiri and Ponnarasi, 2011). 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 (Mohammed *et al*., 2015), 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 (Mohammed *et al*., 2015; Al-Masri *et al*., 2004; Alamoudi, 2013; Melquiades and Appoloni, 2002). Consequently, the following Radium equivalent activity (Ra_{eq}) of a sample in (Bq/kg) can be evaluated as (Melquiades and Appoloni, 2002; Hosseini *et al*., 2006; Anonymous, 2003, 1995):

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

Where:

A_{ra} = The specific activity of ^{226}Ra which is usually the same as that of ^{238}U (^{226}Ra) (Bq/kg)
A_{th} = The specific activity of ^{232}Th (Bq/kg)
A_K = The specific activity of ^{40}K (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 <370 Bq/kg (Al-Gazaly *et al*., 2014).

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 (H_{in}) is given by the following expression (Afshari *et al*., 2009):

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

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:

$$D = C \times I \times E \quad (4)$$

Where:

D = The annual effective dose (Sv/y)
C = The specific activity of radionuclides in the ingested sample (Bq/kg)
I = The annual intake of powdered milk (kg/y) which depends on a given age
E = 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 Anonymous (1995). The average consumption rate of milk for children in different age groups (age from 2-7, 7-12 years and 12-17 years) and adults (from 17 years) is 14 and 13 kg/year, respectively (Anonymous, 2003).

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±0.158-6.471±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±0.273-2.125±0.443 Bq/kg with an average value of 1.588±0.396 Bq/kg and from 266.558±5.018-483.595±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 27.341-48.035 Bq/kg with an average of 35.967 Bq/kg and from 0.078-0.147 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), (7-2), (12-17) years and for adults (age over 17 years) as shown in Table 5: due to the intake for the three different radionuclides has resulted to be: for ^{238}U (^{226}Ra): 0.337×10^{-4}, 0.435×10^{-4}, 0.185×10^{-5} and 0.548×10^{-4} Sv/y; for ^{232}Th 0.735×10^{-5}, 0.645×10^{-5}, 0.556×10^{-5} and 0.548×10^{-5} Sv/y; for ^{40}K 0.113×10^{-3}; 0.704×10^{-4}; 0.412×10^{-4} and 0.360×10^{-4} Sv/y (Table 4) have been found to be lower than the recommended reference limit by UNSCEAR: 412 Bq/kg (Kadhim, 2016) 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

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

		Specific activity		
Sample codes	Sample names	K-40	U-238	Th-232
M1	Golden cow's milk	319.780±6.200	1.108±0.350	1.800±0.499
M2	Sponsor milk	482.829±7.227	6.471±0.802	3.067±0.569
M3	Surprisingly milk	376.597±5.927	5.324±0.676	0.821±0.273
M4	Anchor milk	485.740±7.197	7.851±0.877	0.834±0.294
M5	Pew milk	466.497±6.813	1.740±0.399	0.875±0.291
M6	Diallak milk	266.558±5.018	4.782±0.644	2.125±0.443
M7	Aktal milk	372.640±6.090	0.274±0.158	1.362±0.364
M8	Lancy milk	306.744±5.935	1.796±0.435	1.347±0.389
M9	Niro milk	310.897±5.600	6.869±0.798	1.479±0.382
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

Sample codes	Sample name	Raeq (Bq/kg)	Internal hazard index
M1	Golden cow's milk	28.305	0.07942
M2	Sponsor milk	48.035	0.14720
M3	Surprisingly milk	35.496	0.11024
M4	Anchor milk	46.446	0.14664
M5	Pew milk	38.912	0.10977
M6	Diallak milk	28.355	0.08947
M7	Aktal milk	30.915	0.08421
M8	Lancy milk	27.341	0.07867
M9	Niro milk	32.923	0.10748
M10	Nktalia milk	42.939	0.12297
Average		35.967	0.10761
The global average		370.000	1.00000

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

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

Table 5: The conversion factor and annual intake (Kadhim, 2016)

	Conversion factor (Sv/Bq)			Annual intake
Types (years)	U-238	Th-232	K-40	(kg/years)
Powder milk Infants = 1	4.7×10^{-6}	4.6×10^{-6}	6.2×10^{-8}	22.4
Powder milk Infants = 1-2	9.6×10^{-7}	4.5×10^{-7}	4.2×10^{-8}	15.0
Powder milk Children 2-7	6.2×10^{-7}	3.5×10^{-7}	2.1×10^{-8}	14.0
Powder milk Children 7-12	8.0×10^{-7}	2.9×10^{-7}	1.3×10^{-8}	14.0
Powder milk Children 12-17	1.5×10^{-7}	2.5×10^{-7}	7.6×10^{-9}	14.0
Powder milk Adults 17	2.8×10^{-7}	2.3×10^{-7}	6.2×10^{-9}	13.0

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

Regions	^{226}Ra	^{232}Th	^{40}K	References
Egypt	0.44±0.23	---	134.7±12	Saleh *et al*. (2007)
Iran	---	---	17.3±3.3	Afshari *et al*. (2009)
Jordan	2.14	1.28	392	Ababneh *et al*. (2009)
Syria	----	---	435	Al-Masri *et al*. (2004)
Sadi Arabia	9.64	6.77	74.51	Alamoudi (2013)
Brazil	----	3.7	482±9.9	Melquiades and Appoloni (2002)
France	0.05±0.01	0.142±0.026	434.1±13	Hosseini *et al*. (2006)
Present work	3.881±0.563	1.588±0.396	387.187±6.299	

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 (Alamoudi, 2013). Also, it is found that the average annual effective dose from natural radioactivity turn out to be below the reference value of 1.0 m Sv/y recommended by ICRP for all ages (Anonymous, 1995).

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 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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