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Original Article

Measurement of radioactivity levels in powdered milk consumed in Algeria and estimation of annual effective doses

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ABSTRACT

The monitoring of radioactivity in food, especially in milk, has become a top priority in many countries. This paper presents the activity concentrations of ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs radionuclides measured in twenty-two samples of the powdered milk available in the Algerian market, sourced from different regions worldwide. The measurements were conducted using an HPGe detector by employing the gamma spectrometry method. The results show that the specific activity of ^{40}K, ^{226}Ra and ^{232}Th in infant powdered milk samples varied from (133.57±1.75 to 195.95±2.56 Bq/kg), (1.35±0.03 to 2.70±0.07 Bq/kg) and (1.34±0.03 to 1.63±0.06 Bq/kg), respectively. In adult powdered milk samples, the activity concentration of ^{40}K, ^{226}Ra ^{232}Th and ^{137}Cs were varied from (110.52±1.49 to 687.89±8.94 Bq/kg), (1.38±0.04 to 2.59±0.10 Bq/kg), (1.25±0.03 to 2.63±0.09 Bq/kg) and (3.60±0.07 to 7.78±0.11 Bq/kg), respectively. The obtained results were subsequently compared with data from various studies conducted globally. The estimated annual ingestion dose resulting from the consumption of powdered milk was found to be 583.80 µSv/y infants (≤ 1y), and 56.85 µSv/y for adults (≥ 17y). These values indicate that powdered milk in Algeria does not have a significant radiological impact on the population, as they are below the dose limit recommended by the World Health Organization.

1. Introduction

Monitoring radioactivity has become a common practice in many countries, especially regarding food products and the environment. This approach plays a crucial role in safeguarding public health by regulating human exposure to radiation [1].

The primary sources of internal radiation exposure in humans through food ingestion are the naturally occurring radionuclides, particularly ^{40}K, and the radionuclides from the ^{238}U and ^{232}Th series, which are extensively present in the environment [2]. In addition to natural radionuclides, various artificial radioactive elements, such as ^{137}Cs, have the ability to spread through the food chain via food consumption. These elements have been introduced into our environment as a result of numerous nuclear weapons experiments and various incidents involving nuclear reactors [3,4]. One of the primary food items that necessitate examination for its radioactivity levels is milk. It is an essential component of human dietary needs, particularly for infants under 1-year-old and children. Furthermore, milk acts as a vector of radioactive contamination in the food chain since it can be contaminated by radionuclides like ^{137}Cs. These radionuclides can be transferred to milk from the grass ingested by the cows, which is laden with radioactive isotopes [5].

In Algeria, although milk is considered as a fundamental component of the diet, a considerable portion, especially powdered milk, is imported, primarily from Europe. That's

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why Algerian regulations require strict monitoring of radioactive contamination in this product.

The purpose of this study is to perform a qualitative and quantitative analysis of the radionuclides found in powdered milk consumed in Algeria, and subsequently estimate the annual ingestion dose for various age groups in the population resulting from this consumption.

Materials and Methods

2.1. Samples conditioning

Twenty-two samples of powdered milk, imported from various countries and consumed in Algeria, were chosen for analysis. The origins and types of the samples are detailed in Table 1. A quantity of 250 g of each sample was packed in a marinelli beaker (Fig 1), and sealed for a duration of four weeks to achieve the state of radioactivity equilibrium between the parent and daughter radionuclides.

Fig 1. Samples conditioning in Marinelli Beakers

Table 1. Sample codes and countries of origin for analyzed powdered milk samples.

	Sample code	Origin		Sample code	Origin
Adult milk powder	**Sample 01**	Netherlands	Infant milk powder	**Sample 14**	France
	Sample 02	Ireland		**Sample 15**	Denmark
	Sample 03	Argentina			
	Sample 04	Holland		**Sample 16**	France
	Sample 05	Ukraine			
	Sample 06	Lithuania		**Sample 17**	Germany
	Sample 07	Belgium		**Sample 18**	Switzerland
	Sample 08	Poland		**Sample 19**	France
	Sample 09	Canada		**Sample 20**	France
	Sample 10	France		**Sample 21**	Switzerland
	Sample 11	New Zealand			
	Sample 12	Uruguay		**Sample 22**	France
	Sample 13	USA			

2.2. Gamma Spectrometric Analysis

The powdered milk samples were analyzed using a high-resolution gamma-ray spectrometry system (Fig 2) that incorporated a coaxial hyper-pure germanium detector (Canberra model GC3018). This detector has an energy resolution of 1.8 keV at the 1.33 MeV γ-ray line of ^{60}Co and an efficiency of 30%. The system also included a preamplifier (model 2002CSL) positioned in close proximity to the detector, an amplifier (model 2024A), and a 16 k digital multichannel analyzer. In order to maintain a low background environment, the detector was enclosed within a 10 cm steel shield coated internally with a 1 mm copper layer.

The energy calibration is performed using a multi-nuclides gamma-ray reference source (^{241}Am, ^{137}Cs and ^{60}Co) that covered a wide energy range from 59.5 to 1332.5 keV, while the efficiency calibration was done using a standard source that has the same matrix and same geometry as the measuring samples; it is powder milk contaminated with a radioactive source of 152Eu, with an activity of 326 Bq in 1993, emitting gamma rays in the energy range of 121–1408 keV.

Fig 2. Gamma spectrometry system

By employing this standard source, the counting efficiency curve of the HPGe detector was drawn and is calculated using the following equation:

$$\xi = \frac{N}{A * I\gamma * tc} \quad (1)$$

Where N is the net area at the interest peak energy, corrected for background counts, A represents the source activity in Becquerel with decay time correction; tc stands for the collection time in seconds and I_γ signifies the intensity of gamma ray.

Figure 3 and 4 show respectively the energy calibration line and the curve of the efficiency calibration.

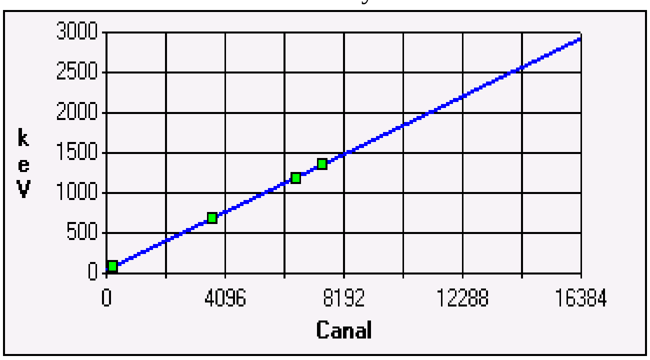

Fig 3. Energy calibration line.

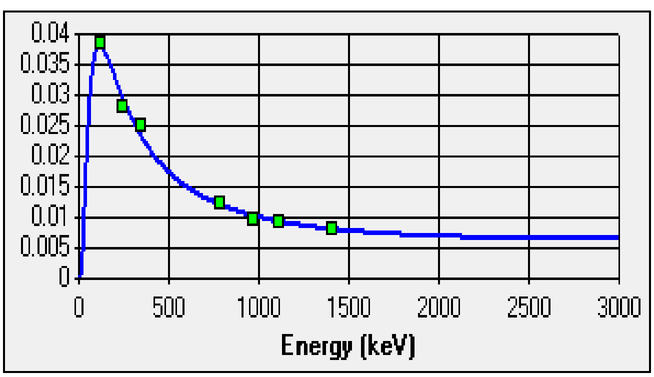

Fig 4. Efficiency calibration curve.

The samples were placed above the detector for 14400 s to minimize statistical counting errors. The background was also measured for the empty container with the same counting time and subtracted from the samples spectra. The figure below illustrates an example of spectra from an adult milk powder sample obtained using the Genie 2000 software.

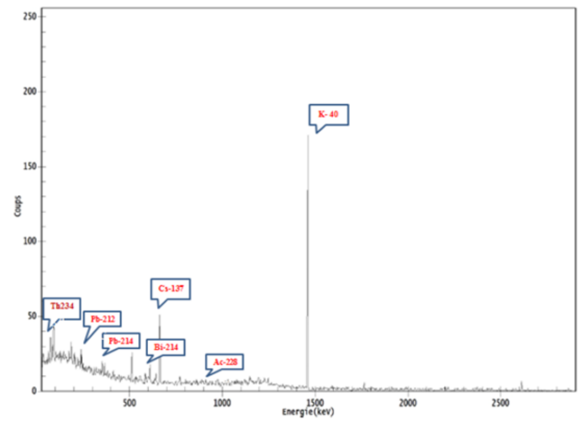

Fig 5. Gamma ray spectrum of Adult milk Powder sample (Sample 05: Ukraine).

2.3. Activity calculation:

The activity concentration of radionuclides in milk samples, expressed in Bq/kg, was determined using the following equation **[6]**:

$$A = \frac{N}{\xi * I\gamma * tc * m} \quad (2)$$

The uncertainty related to the activity concentration is determined through error propagation, as described by the following relationship **[7]**:

$$\delta A = A\sqrt{\left(\frac{\sigma\xi}{\xi}\right)^2 + \left(\frac{\sigma N}{N}\right)^2 + \left(\frac{\sigma I\gamma}{I\gamma}\right)^2 + \left(\frac{\sigma m}{m}\right)^2 + \left(\frac{\sigma tc}{tc}\right)^2} \quad (3)$$

Where *N* represents the net peak counts after background subtraction; ξ, corresponds to the detector counting efficiency at the specific gamma ray energy; tc, is the measured time in seconds; *I*, represents the intensity of γ-ray; while m indicates the weight of the sample in kilograms.

The activity concentration of ^{226}Ra and ^{232}Th was assessed by relying on their short-lived progenies, utilizing their distinct and notably strong gamma spectral lines **[10]**. This approach was necessary due to the limited intensity of gamma rays emitted by ^{226}Ra and ^{232}Th, making them undetectable by conventional gamma-ray spectrometry [8,9].

The activity concentrations of ^{226}Ra and ^{232}Th were then calculated using equations 4 and 5 respectively:

$$A_{226Ra}(Bq/kg) = \frac{A_{214Bi} + A_{214Pb}}{2} \quad (4)$$

$$A_{232Th}(Bq/kg) = \frac{A_{228Ac} + A_{212Pb} + A_{208Tl}}{3} \quad (5)$$

On the other hand, the activity concentrations of ^{40}K and ^{137}Cs were directly assessed using their individual gamma lines at 1460.8 keV and 661.6 keV, respectively.

2.4. Estimation of annual effective dose

To calculate the annual effective dose (D) resulting from the ingestion of radionuclides through the consumption of powdered milk, we used the following equation [11]:

$$D = AIE \tag{6}$$

Where D is the annual effective dose in (Sv/y), A is the activity concentrations of the interested radionuclides in the sample (Bq/kg), I is the annual powdered milk intake (kg/y), which depends on the human age (for infants ≤1 year is 22.4 kg/y and adult >17 year is 13 kg/y) [12] and E is the dose conversion factors for the radionuclides (Sv/Bq).The conversion factor E varies depending on both the radioisotopes and the ages of the individuals (Table 2).

Table 2: Dose conversion factors of radionuclides for infants (≤1) and adults (>17 y) [13].

Age group	**Dose conversion factors (Sv/Bq)**			
	^{40}K	**^{226}Ra**	**^{232}Th**	**^{137}Cs**
Infants (≤ 1y)	$6.2*10^{-8}$	$4.7 * 10^{-6}$	$4.6 * 10^{-6}$	$2.1*10^{-8}$
Adults (> 17y)	$6.2*10^{-9}$	$2.8 * 10^{-7}$	$2.3 * 10^{-7}$	$1.3*10^{-8}$

The annual effective dose resulting from the consumption of radionuclides found in powdered milk are tabulated in Table 5.

2. Results and Discussion

3.1. Activity Concentration:

The activity concentrations of radionuclides measured in the powdered milk samples are given in Table 3. The table indicates that the activity concentrations of ^{40}K in all samples were higher than for ^{226}Ra and ^{232}Th. This can be attributed to the natural abundance of potassium-40 in the environment. Its widespread presence in the Earth's crust and soil allows it to be easily incorporated into plants and subsequently transferred through the food chain, making it an integral part of the biological processes within ecosystems [14].

The measurement results also reveal that the specific activities of ^{40}K, ^{226}Ra, and ^{232}Th exhibit variation across different samples. In infant powdered milk samples, the activity concentrations varied from 133.57 ± 1.75 (Germany) to 195.95 ± 2.56 Bq/kg (France) for ^{40}K, from 1.35 ± 0.03 to 2.70 ± 0.07 Bq/kg for ^{226}Ra, and from 1.34 ± 0.03 to 1.63 ± 0.06 Bq/kg for ^{232}Th, respectively.

In adult powdered milk samples, the activity concentrations of ^{40}K, ^{226}Ra, and ^{232}Th varied from 110.52 ± 1.49 (New Zealand) to 687.89 ± 8.94 Bq/kg (USA), from 1.38 ± 0.04 to 2.59 ± 0.10 Bq/kg, and from 1.25 ± 0.03 to 2.63 ± 0.09 Bq/kg, respectively.

This variation can be attributed to diverse factors including background levels of natural radioactivity in various regions of the world, the transfer of radionuclides from soil to plants/grass, the geological characteristics of local dairy farming areas, climate and agricultural practices in various farming areas, the intake of feedstuffs by different animals (cow in this case), and the raw materials involved in processing [15] .

Besides natural radioactivity, the presence of artificial radioactivity (^{137}Cs) was also detected in two samples of adult milk powder from Poland (3.60 ± 0.07 Bq/kg) and Ukraine (7.78 ± 0.11 Bq/kg). However, the measured activities are below regulatory limits [16]. The occurrence of this radionuclide in powdered milk is a result of its dispersion in the environment due to nuclear activities or accidents [17].

Table 3: Activity concentrations of different radionuclides found in milk powder samples.

Samples code	**Activity concentration (Bq/kg)**			
	^{40}K	**^{226}Ra**	**^{232}Th**	**^{137}Cs**
Sample 01	552.28±7.18	2.59±0.10	2.63±0.09	BDL
Sample 02	685.18±8.90	2.46±0.09	1.63±0.05	BDL
Sample 03	466.84±6.07	1.38±0.04	1.46±0.04	BDL
Sample 04	568.21±7.38	2.26±0.07	1.71±0.06	BDL
Sample 05	558.72±7.26	2.56±0.08	2.04±0.08	7.78±0.11

Sample 06	597.37±7.76	1.93±0.07	1.49±0.05	BDL
Sample 07	595.67±7.74	2.32±0.08	1.91±0.07	BDL
Sample 08	619.40±8.05	1.74±0.06	1.50±0.04	3.60±0.07
Sample 09	632.29±8.22	2.34±0.09	1.57±0.05	BDL
Sample 10	668.90±8.69	2.13±0.07	1.79±0.06	BDL
Sample 11	110.52±1.49	1.62±0.06	1.25±0.03	BDL
Sample 12	383.10±4.98	1.78±0.05	1.56±0.04	BDL
Sample 13	687.89±8.94	1.83±0.08	1.43±0.03	BDL
Sample 14	179.00±2.34	1.75±0.06	1.52±0.05	BDL
Sample 15	165.10±2.16	1.29±0.04	1.37±0.04	BDL
Sample 16	182.05±2.38	2.46±0.08	1.63±0.06	BDL
Sample 17	133.57±1.75	2.97±0.09	1.51±0.04	BDL
Sample 18	171.88±2.28	1.35±0.03	1.44±0.05	BDL
Sample 19	174.93±2.29	1.83±0.07	1.47±0.06	BDL
Sample 20	141.37±1.85	1.49±0.05	1.46±0.04	BDL
Sample 21	144.08±1.89	1.71±0.06	1.34±0.03	BDL
Sample 22	195.95±2.56	2.70±0.07	1.50±0.05	BDL

BDL: below detection limit.

Table 4: Comparison of activity concentration of radionuclides detected in powdered milk with worldwide values.

Country	**Activity Concentration (Bq/kg)**				**Reference**
	^{226}Ra	**^{232}Th**	**^{40}K**	**^{137}Cs**	
Thailand^{I}	3.20±0.51	1.24±0.10	231.20±25.50	4.90±0.74	[18]
Saudia Arabia^{I}	0.46±0.05	0.35±0.08	234.18±1.90	*N/A*	[19]
Malaysia^{I}	3.05±0.45	2.55±0.64	99.10±5.58	0.27±0.19	[20]
Bangladesh^{I}	5.40±0.40	5.70±0.50	334.40±12.00	N/A	[21]
Iraq^{I}	3.88±0.56	1.58 ±0.39	387.18±6.29	N/A	[22]
Egypt^{I}	0.91 ± 0.20	0.60 ± 0.11	478,0 ± 25,00	0.42 ±0.11	[23]
Algeria^{I}	N/A	N/A	106.70±5.44	N/A	[24]
Iraq^{A}	*BDL*	*BDL*	290.67±14.97	N/A	[25]
Brazil^{A}	*BDL*	*0.50*	489.52±13.00	2.54	[3]
Iran^{A}	*0.11*±0.02	*0.13*±0.02	549.65±14.97	1.43±0.04	[26]
Jordan^{A}	*1.24±0.77*	*N/A*	348.30±7.76	0.52+0.39	[5]
Malaysia^{A}	*1.97 ±0.32*	*1.07±0.24*	307.74 ± 7.20	N/A	[27]
Saudia Arabia^{A}	*9.64*±0.45	*6.77*±0.40	74.51±6.40	N/A	[28]
India^{A}	2.50±1.20	N/A	34.35±5.20	N/A	[29]
Algeria^{I}	1.95±0.06	1.47±0.05	165.5±2.20	BDL	Present work
Algeria^{A}	2.07±0.07	1.69±0.05	548.17±7.12	5.69±0.09	Present work

BDL: below detection limit, N/A represents data not available, I: Infant milk powder, A: Adult milk powder.

The findings presented in this study were compared to the results obtained in other studies worldwide, which focused on the activities of radionuclides in powdered milk. These comparative values are summarized in Table 4. As shown in Table 4, our study found that the average activity concentrations of ^{40}K in powdered milk for infants were higher than those in Malaysia but lower than levels detected in powdered milk from several other regions. Regarding adult samples, the average concentration was higher than what previous research had reported, except for Iran. Concerning the activity values of ^{226}Ra and ^{232}Th, as they are compared to published data from various countries, it is evident that the majority of these values are within the range of the published data. In the case of ^{137}Cs, the average radioactivity concentrations in this study are relatively elevated when compared to levels found in other nations. The results indicate that the differences in the levels of radioactivity in milk can be explained by differences in their original surroundings and/or the raw materials used in their production [25].

3.2. The Annual Effective Dose for milk ingestion

Table 5 illustrate the annual effective dose calculations resulting from the ingestion of radionuclides in powdered milk for different age groups.

Table 5: Annual effective dose resulting from the ingestion of radionuclides.

Samples Code		Annual effective dose (µSv/y)			
		^{40}K	^{226}Ra	^{232}Th	^{137}Cs
Powder milk (Adults >17Y)	1	44.51	9.44	7.86	BDL
	2	55.23	8.96	4.88	BDL
	3	37.63	5.03	4.36	BDL
	4	45.80	8.24	5.14	BDL
	5	45.80	9.34	6.12	1.3
	6	48.15	7.03	4.46	BDL
	7	48.01	8.46	5.72	BDL
	8	49.92	6.35	4.49	0.61
	9	50.96	8.53	4.69	BDL
	10	53.91	7.78	5.36	BDL
	11	8.91	5.90	3.75	BDL
	12	30.88	6.49	4.67	BDL
	13	55.44	6.69	4.28	BDL
	Average	**44.24**	**7.55**	**5.06**	**0.95**
Powder milk (Infants ≤1Y)	14	248,61	183,98	156,07	BDL
	15	229,30	135,63	140,72	BDL
	16	252,84	259,22	168,03	BDL
	17	185,51	312,69	155,57	BDL
	18	238,72	142,64	148,34	BDL
	19	242,96	169,01	151,08	BDL
	20	196,34	156,87	150,10	BDL
	21	200,11	180,10	139,10	BDL
	22	272,15	284,24	154,35	BDL
	Average	**248,61**	**183,98**	**156,07**	**BDL**

The results showed that 40K is the primary factor influencing the internal radiation dose experienced by the population through the consumption of powdered milk. Moreover, the average annual effective dose resulting from 40K intake in infants is significantly higher in comparison to that in adults. This difference can be attributed to the fact that milk acts as the primary nutritional source for infants in their initial six months of life, leading to a higher rate of consumption [19]. In the case of 137Cs, the dose resulting from the consumption of this radionuclide is negligible, indicating that it does not present a potential health hazard. The findings of this study demonstrate that the annual effective doses resulting from the consumption of radionuclides in powdered milk are all below the recommended limit of 1 mSv/y established by the Joint Food and Agriculture Organization of the United Nations/World Health Organization (FAO/ WHO) [30] and the International Commission on Radiological Protection (ICRP) [31].

4. Conclusion

The activity concentrations of radionuclides were measured in 22 samples of powdered milk imported from different countries and marketed in Algeria using High-resolution Gamma-ray spectrometry with H5PGe detector.

The results show that the average activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in infant powdered milk samples were 165.32±2.16, 1.95±0.06 and 1.47±0.05 Bq/kg, respectively.

In adult powdered milk samples, the activity concentration of ^{40}K, ^{226}Ra ^{232}Th and ^{137}Cs were 548.18±7.12, 2.07±0.07, 1.69±0.05 and 5.69±0.09 Bq/kg, respectively.

The annual effective dose resulting from the consumption of these powdered milk samples by infants and adults was also evaluated. The overall result indicates that the radionuclide levels in the studied samples of powdered milk for children and adults are clearly below the recommended limits issued by FAO/WHO. In general terms, it can be concluded that the powdered milk consumed in Algeria is radiologically secure and is unlikely to pose substantial health risks. However, it is advisable to maintain continuous monitoring of the radiological safety of milk consumed by the population in Algeria.

Conflict of Interest

The authors declare that they have no conflict of interest

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