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Measurement of Radioactivity in Common Powered Milk Consumed in Sana'a, Yemen and Estimation of the Associated Annual Effective Dose

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Abstract

This study focuses on measuring the radioactivity in milk, given its critical importance and sensitivity as a fundamental component of human nutrition, particularly due to its vital role as a primary food source for children during the early stages of life. The radionuclides of ^{232}Th, ^{226}Ra and ^{40}K were measured in 16 brands of powdered milk consumed in Yemen, imported from various regions worldwide. The measurements were conducted using gamma-ray spectroscopy with an HPGe detector, equipped with a specially designed shield. The range of ^{226}Ra from (0.42±0.08 to 1.86±0.30 with an average of 0.77±0.12) Bq/kg, the range of ^{232}Th from (0.21±0.05 to 0.40±0.07 with an average of 0.30±0.05) Bq/kg, and the range of ^{40}K (from 140.69±3.90 to 195.96±5.29 with an average of 169.06±4.63) Bq/kg, respectively, for infant group ($\leq$ 1 year). The range of ^{226}Ra from (0.14±0.03 to 7.21±1.34 with an average of 1.88±0.54) Bq/kg, the range of ^{232}Th from (0.25±0.04 to 1.22±0.34 with an average of 0.68±0.15) Bq/kg, and the range of ^{40}K (from 141.52±3.90 to 534.39±13.54 with an average of 320.36±8.35) Bq/kg, respectively, for individuals above 2 years, respectively. The total average annual effective dose due to ingestion of radionuclides was calculated for three age groups and found to be (214.50, 123.27, and 41 μSv/year) for infant group ($\leq$ 1 year), children (2-17 years), and adults (>17 years), respectively. Overall, these values are lower than the WHO/FAO and ICRP recommended limit of (1.0 mSv/year) for all age groups, indicating that the radioactivity levels in the tested powdered milk are within acceptable safety limits.

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1. INTRODUCTION

Radioactivity is a natural phenomenon that has been present since the dawn of time. It occurs when unstable atomic nuclei decay, releasing radiation in the process. Radioactivity is present in a variety of sources, including food, water, rocks, soil, and cosmic rays, but it can also be artificially generated through nuclear reactions [1].

Radionuclides, both natural and artificial, can enter the human body through inhalation of dust or by consuming contaminated food and water, resulting in internal radiation exposure. Studies have shown that approximately one-eighth of the total effective dose from natural sources comes from food consumption [2]. Also fertilizers used in agricultural soils are a contributing factor to the transfer of radiation into the food chain. A study conducted by researcher Abdul-Rahim, A. K., et al. measured the levels of radiation and heavy metals in fertilizers. The findings revealed that the radiation levels were close to the recommended guideline limits, highlighting the importance of monitoring the use of agricultural fertilizers to ensure the safety of soil and crops [3]. This is influenced by the concentration of radionuclides in the food and the amount consumed. As a result, monitoring radioactivity levels in food is essential for protecting public health. In response, international organizations such as the IAEA, ICRP, UNSCEAR, FAO, and WHO have committed to establishing guidelines for radioactivity levels in food [4].

Milk is one of the most crucial food products to be tested for radioactivity levels, as it plays a key role in human nutrition, particularly for infants and children, and assessing radioactivity levels in milk is a critical part of this process [5]. Numerous global studies have been conducted on foodstuffs to evaluate radiation levels and prevent unnecessary exposure [6]. In Yemen, although there is some research on radioactivity in food, data on powdered milk is limited [7]. A study conducted by Wail Al-Sharabi in 2016 analyzed 7 powdered milk samples [8]. In our research, reflecting the extensive variety of imported brands available in the market. This increase allows for a more comprehensive examination of the quality and safety of milk products consumed by consumers today. Powdered milk is a primary food source for many infants in Yemen, with most of it being imported. Therefore, verifying radioactivity levels and assessing the potential radiation dose from its consumption is essential [9, 10]. This study investigates the radioactivity levels in milk, emphasizing its critical importance as a fundamental component of human nutrition, particularly due to its essential role as a primary food source for children during their early stages of life. The research aims to evaluate the activity concentrations of the radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in powdered milk brands commonly consumed in Yemen, which are imported from various regions worldwide. Additionally, the study seeks to estimate the annual effective dose resulting from the ingestion of these radionuclides for different age groups, including infants, children, and adults, to assess compliance with international safety standards.

2. MATERIALS AND METHODS

Sixteen powdered milk samples collected from the Yemen Standardization and Metrology Organization and various markets in Sana'a city were used in this study. The samples were categorized into two different age groups. The first age group included 5 samples with assigned codes ranging from S_1 to S_5, representing milk for infants under 1 year of age. The second age group included 11 samples with assigned codes ranging from S_6 to S_{16}, representing milk intended for individuals aged 2 years and above. All powdered milk samples were imported from 12 different countries, as shown in Table 1.

A 200 ml portion of each sample was weighed and stored for 30 days to allow secular equilibrium to be established between ^{226}Ra, ^{232}Th, ^{238}U, and their decay products. The measurements were conducted in the gamma laboratory at the National Atomic Energy Commission (NATEC) in Yemen using the Gamma-Ray Spectrometry Technique with an HPGe detector having 35% relative efficiency and an energy resolution of 1.80 keV at 1332 keV of ^{60}Co. The detector was shielded in a chamber made of lead (8 cm in thickness) to reduce background radioactivity. The spectra of the samples were analyzed using Genie 2000 software (Canberra). The detector was calibrated using a gamma multinuclide standard sample (R8/31/38QCT48) containing ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{137}Cs, ^{88}Y and ^{60}Co. Absolute efficiency was achieved, as shown in Figures 1 and 2. Each sample was counted for 172,800 seconds to minimize statistical counting errors and ensure sufficient counts for the various gamma-ray peaks. The background spectra were counted under the same conditions and for the same duration using an empty geometry.

Table 1. Samples Codes and brands collected in the study.

S. Code	Country	Brands
S_1	**UAE**	Reina
S_2	**France**	Franclait
S_3	**Belaraus**	AnDo
S_4	**Denmark**	Regalac
S_5	**New Zealand**	Golden
S_6	**Irlanda**	Glenor
S_7	**Malaysia**	Latif
S_8	**New Zealand**	Gardo
S_9	**Switzerland**	Nido
S_{10}	**New Zealand**	NoNo
S_{11}	**Ukraine**	Veselynivskiy
S_{12}	**Turkey**	Montana
S_{13}	**European Union**	Al-Jawker
S_{14}	**Germany**	Arla
S_{15}	**New Zealand**	SLG
S_{16}	**New Zealand**	Divalley

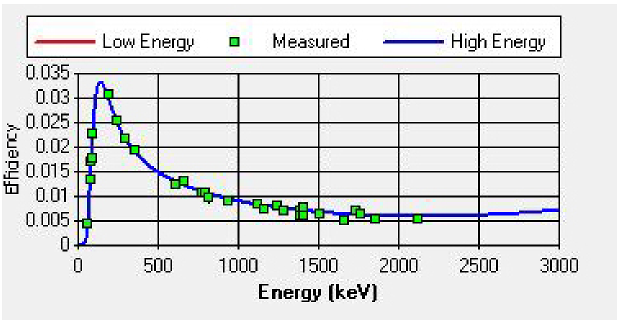

Figure 1. Efficiency calibration curve for HPGe detector.

The activities of ^{226}Ra, ^{232}Th and ^{40}K were determined through their progeny energy peaks, specifically ^{214}Pb (351,9 keV) and ^{214}Bi (609.3 kev) for ^{226}Ra, and ^{228}Ac (911 keV), ^{208}Tl (583.1 keV) and ^{212}Pb (238,6 keV) for ^{232}Th. The nuclide-specific activity, based on the peak at radionuclide energy, is given by [11, 12]:

$$A = \frac{N}{\epsilon_\gamma . t . \gamma_d . m} \quad (1)$$

Where ***A*** is the specific activity (Bq/kg) of the nuclide, ***N*** is the net peak area for the peak, ϵ_γ is the detector efficiency, ***t*** is the counting live-time (in seconds), $\gamma_{\mathbf{d}}$ is the gamma emission probability, and mmm is the sample's

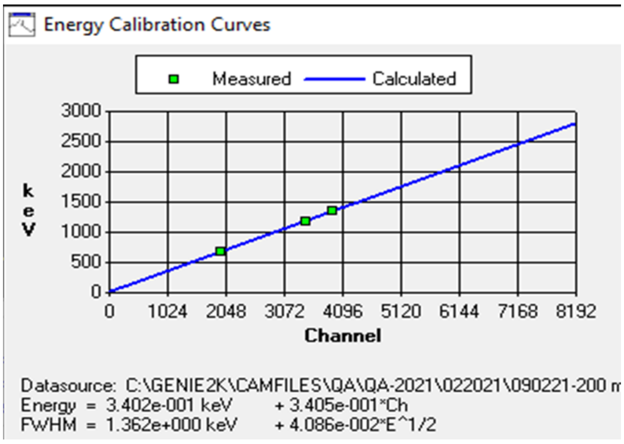

Figure 2. Energy calibration curve for HPGe detector

Table 2. Dose conversion factors for different age groups[12, 13]

Age groups	Dose conversion factors (μSv/Bq) ^{226}Ra	^{232}Th	^{40}K
Infants	0.96	5.7	0.042
Children	0.8	3.9	0.013
Adults	0.28	0.69	0.0062

mass (in kg). The annual effective dose **D** (μSv/year) of radionuclides resulting from milk consumption is determined using the equation provided below [1].

$$D = A \times I \times E \quad (2)$$

Where **A** represents the activity concentration of radionuclides in milk (Bq/kg), while **E** stands for the dose conversion coefficient for the radionuclides due to ingestion (μSv/Bq) see Table (1). Additionally, I denotes the annual intake of milk (kg/year), which varies according to the age groups. In this investigation, the annual ingestion dose was examined for three distinct age groups: infants ($\leq$ 1 year), children (2–17 years), and adults (> 17 years). The average annual intakes of powdered milk were (22.4, 14.0, and 13.0 kg/year) for infants, children, and adults, respectively [13–16]. The annual effective dose for both children and adults was calculated based on the activity concentration for the age group older than two years from Table 3, as both groups consume the same brands. However, the dose conversion factors from Table 2 differ for each group, as well as the consumption rates for each group.

3. RESULTS AND DISCUSSIONS

3.1. Radioactivity in powdered milk Samples

The activity concentrations of the collected powdered milk samples in (Bq kg^{-1}) of ^{226}Ra, ^{232}Th, and ^{40}K are listed in Table 3. The activity concentrations for infants ($\leq$1 year) were as follows: ^{226}Ra ranged from (0.42±0.08 to 1.86±0.30 Bq kg^{-1}) with a mean value of (0.77±0.12

Table 3. Activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in powdered milk samples (Bq/kg).

S. Code	Country	Age group	^{226}Ra	^{232}Th	^{40}K
S_1	UAE	$\leq$ 1 year	1.86±0.30	0.21±0.05	195.86±5.32
S_2	France		0.47±0.07	0.40±0.07	164.54±4.52
S_3	Belaraus		0.59±0.07	0.39±0.05	140.69±3.90
S_4	Denmark		0.42±0.08	0.25±0.05	148.26±4.11
S_5	New Zealand		0.53±0.09	0.27±0.05	195.96±5.29
Min.			0.42±0.08	0.21±0.05	140.69±3.90
Max.			1.86±0.30	0.40±0.07	195.96±5.29
Mean±SD			0.77±0.12	0.30±0.05	169.06±4.63
S_6	Irlanda	Above 2 years	UD[1]	0.76±0.24	386.63±9.84
S_7	Malaysia		0.44±0.09	1.06±0.22	275.28±7.28
S_8	New-Zealand		6.14±0.96	0.34±0.06	251.77±6.70
S_9	Switzer-land		1.95±0.34	0.33±0.05	351.36±9.26
S_{10}	New-Zealand		0.18±0.04	0.45±0.13	353.91±9.25
S_{11}	Ukraine		0.26±0.05	0.65±0.15	534.39±13.54
S_{12}	Turkey		0.14±0.03	0.25±0.04	243.81±6.39
S_{13}	European Union		1.09±0.08	0.44±0.08	268.41±7.18
S_{14}	Germany		7.21±1.34	0.94±0.19	460.46±11.66
S_{15}	New Zealand		5.08±3.97	1.01±0.15	141.52±3.90
S_{16}	New Zealand		UD	1.22±0.34	256.43±6.84
Min.			0.14±0.03	0.25±0.04	141.52±3.90
Max.			7.21±1.34	1.22±0.34	534.39±13.54
Mean±SD			1.88±0.54	0.68±0.15	320.36±8.35

[1]**UD Undetected.**

Bq kg^{-1}). The activity concentration of ^{232}Th ranged from (0.21±0.05 to 0.40±0.07 Bq kg^{-1}) with a mean value of (0.30±0.05 Bq kg^{-1}). The activity concentration of ^{40}K ranged from (140.69±3.90 to 195.96±5.29 Bq kg^{-1}) with a mean value of (169.06±4.63 Bq kg^{-1}). The activity concentrations for the above 2 years age group were as follows: ^{226}Ra ranged from (0.14±0.03 to 7.21±1.34 Bq kg^{-1}) with a mean value of (1.88±0.54 Bq kg^{-1}). The activity concentration of ^{232}Th ranged from (0.25±0.04 to 1.22±0.34 Bq kg^{-1}) with a mean value of (0.68±0.15 Bq kg^{-1}). The activity concentration of ^{40}K ranged from (141.52±3.90 to 534.39±13.54 Bq kg^{-1}) with a mean value of (320.36±8.35 Bq kg^{-1}). Figures 2, 3, and 4 show the specific activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in (Bq kg^{-1}), respectively, for all samples considered in this research. The highest activity concentrations of radionuclides in milk samples were as follows: for ^{226}Ra (1.86±0.30 Bq kg^{-1} and 7.21±1.34 Bq kg^{-1}) in S_1 (UAE) and S_{14} (Germany) for infants and above 2 years, respectively. Since the passage of radionuclides to organisms occurs through the food chain, the relatively existing concentration of ^{226}Ra detected in Germany and UAE milk could be attributed, in part, to the radium content of the environments, which is transferred to the milk through the grass–cow–milk pathway [12]. In the global marketplace, many industrial facilities rely on importing raw materials from diverse supply chains worldwide. It is highly likely that the final manufacturing process of these products incorporates raw materials sourced from various regions. Consequently, the detected radioactivity levels in the investigated formula could be attributed to the presence of raw materials imported from different parts of the world [12]. The activity concentrations of ^{226}Ra in powdered milk found in this study were compared with those reported by other authors from different countries. Table 4 shows that the mean activity of ^{226}Ra in powdered milk available to the Yemeni population is higher than in Malaysia [12], Nigeria [17], New Zealand [12], and Vietnam [18], but significantly lower than the reported data from Egypt [14], Iran [19], Jordan [7], Saudi Arabia [20], and Tunisia [21]. The highest activity concentrations for ^{232}Th (0.40±0.07 Bq kg^{-1} and 1.22±0.34 Bq kg^{-1}) were detected in S_2 (France) and S_{16} (New Zealand) for infants and above 2 years, respectively. The imported infant milk brand from New Zealand exhibits the highest ^{232}Th activity, supporting the notion that the raw materials used in processing infant formula are typically sourced from various global supply chains. However, this is influenced by the global economy, which necessitates the use of internationally imported materials. Additionally, the soil–plant ecosystem and specific food intake by animals, such as cows (e.g., silage, hay), can affect the radioactivity levels in infant formula or milk [12]. Furthermore, food processing and preparation methods play a significant role in redistributing radionuclides, which may contribute to the observed radioactivity levels in the infant milk brand under investigation [13]. Notably, several previous reports have linked infant deaths and illnesses to unsafe manufacturing practices in the production of infant formula [21]. Additionally, other brands exhibit similarly low levels of ^{232}Th activity, which may be attributed to the limited mobility of this radionuclide within environmental compartments, resulting in a low transfer through the soil–plant/grass–cow–milk pathway [22]. As shown in Table 4, the ^{232}Th levels in powdered milk available to the Yemeni population are higher than in Egypt [14], Malaysia [12], Nigeria [17], New Zealand [12], and Vietnam [18][18], but significantly lower than the reported data from Iran [19], Jordan [7], Saudi Arabia [20], and Tunisia [21]. The results of the investigation reveal that the activity concentration of ^{40}K in the powdered infant milk exhibits relatively high values compared to other radionuclides. The activity of ^{40}K (140.69±3.90 Bq kg^{-1} and 534.39±13.54 Bq kg^{-1}) was highest in S_3 (Belarus) and S_{11} (Ukraine) for infants and above 2 years, respectively. ^{40}K was the predominant radionuclide across all samples, exhibiting the highest concentrations with minimal variation. The S_{11} (Ukraine) sample had the highest ^{40}K activity (534.39±13.54 Bq kg^{-1}) across all samples for all age groups. This observation is expected, as potassium is an essential element for body metabolism and is widely distributed throughout the environment. Consequently, it can easily transfer through the food chain. The relatively high activity concentration of ^{40}K in the milk samples can be attributed to the high environmental mobility of potassium, moreover, the relatively high levels of radionuclides in Ukraine may be attributed to previous radiation incidents, such as the Chernobyl disaster. This

Table 4. Comparison of the activity concentrations (Bq/kg) of ^{226}Ra, ^{232}Th ,and ^{40}K in powdered milk with different countries.

Country	Age group	^{226}Ra	^{232}Th	^{40}K	Reference
Yemen	≤ 1 year	0.42-1.86	0.21-0.40	140.69-195.96	Present study
Bangladesh		6.33-12.52	4.81-12.28	40.81-359.86	[23]
Malaysia		1.36-7.06	0.31-8.57	40-254	[12]
Tunisia		-	-	130-181	[21]
Jordan	Aabove 2 years	0.50-2.14	0.78-1.28	296.78-392.94	[7]
Egypt		0.91	0.60	477	[14]
Iran		0.05-0.18	0.094-0.16	434-610	[19]
New Zealand		1.89-2.49	0.31-1.37	52-78	[12]
Saudi Arabia		0.09-0.76	0.25-0.85	210-257	[20]
Nigeria		23.1	4.35	832	[17]
Vietnam		1.45-2.45	0.60-1.21	341-387	[18]
Yemen		0.14-7.21	0.25-1.22	141.52-534.39	Present study

- Not reported.

event led to a significant increase in radiation levels, particularly radioactive iodine-131, which directly impacted milk and plants [17]. Potassium is a major radionuclide component in soil and is naturally present as an essential nutrient for humans, animals, and plants [18]. With respect to countries, the milk samples from Nigeria [17] show the highest mean activity, followed by those from Iran [19], Egypt [14], Jordan [7], and Vietnam [18], while samples from New Zealand [12], Malaysia [12], Saudi Arabia [20], and Tunisia [21] show the least mean values. To assess the additional radiation exposure to humans resulting from environmental pollution, particularly due to nuclear accidents such as Chernobyl and Fukushima, it is crucial to investigate the deposition of radionuclides on soil and grass and their subsequent transfer to infant milk through the soil–grass–cow–milk pathway [12].

These factors contribute to a higher concentration of ^{40}Kactivity compared to other samples.

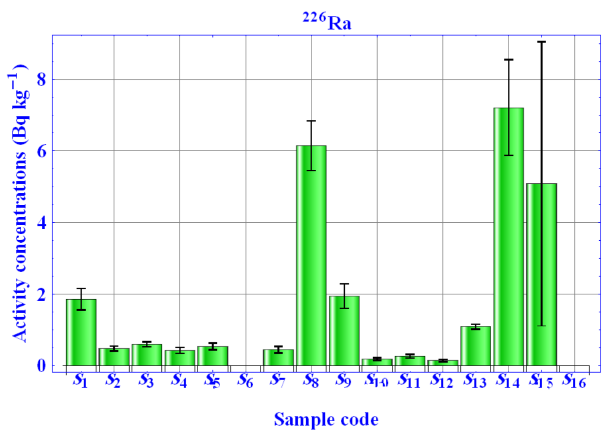

Figure 3. Distribution of activity concentrations of ^{226}Ra of powdered milk samples.

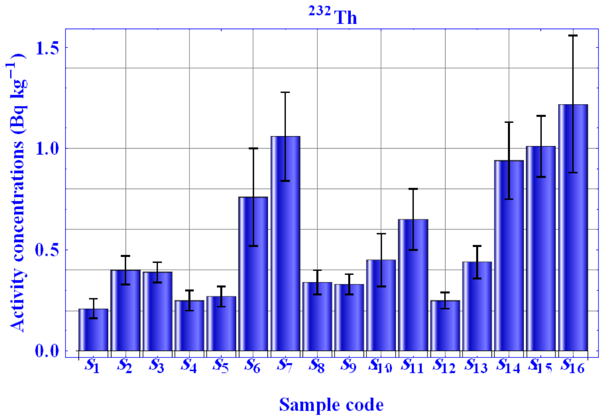

Figure 4. Distribution of activity concentrations of ^{232}Th of powdered milk samples.

Table 5. Annual effective dose for different age groups (μSv/y).

Radionuclides		Age groups		
		Infants	Children	Adults
^{226}Ra	Min.	9.032	1.57	0.51
	Max	39.99	80.75	26.24
	Avg.	16.64	27.99	9.10
^{232}Th	Min.	26.81	13.65	1.88
	Max	51.07	66.61	10.94
	Avg.	38.81	36.98	6.08
^{40}K	Min.	132.36	25.76	11.34
	Max	184.36	97.26	43.07
	Avg.	159.05	58.31	25.82
Total Minimums		168.20	40.97	14.16
Total Maximums		275.42	244.62	80.25
Total average		214.50	123.27	41
The WHO recommended limit		1000		

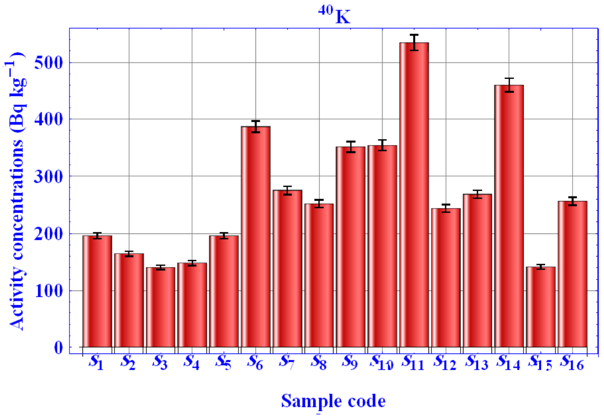

Figure 5. Distribution of activity concentrations of ^{40}K of powdered milk samples.

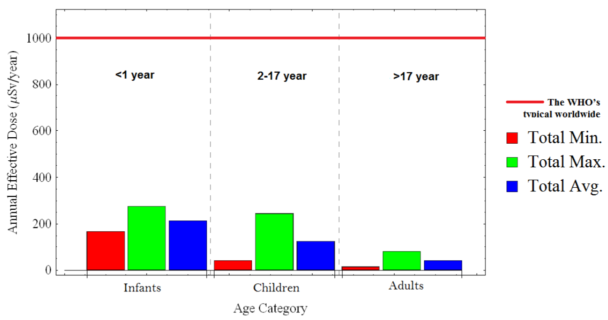

Figure 6. Distribution of annual effective dose for different age groups.

3.2. Annual Effective Dose

Calculating the annual effective dose from milk ingestion provides valuable insights into the potential radiation exposure from this dietary source. By calculating this dose, we can assess the risk of radiation exposure from consuming milk, especially in regions where radionuclides may be present in the environment of the imported samples. This calculation helps determine whether the population's exposure remains within safe limits and provides valuable insights for establishing food safety guidelines and public health recommendations [24–26].

The annual effective doses of ^{226}Ra, ^{232}Th, and ^{40}K in powdered milk were calculated in this study and are presented in Table 4. The average annual effective doses for ^{226}Ra, ^{232}Th, and ^{40}K were **16.64, 38.81, and 159.05 μSv/y** for infants, 27.99, 36.98, and 58.31 μSv/y for children, and 9.10, 6.08, and 25.82 μSv/y for adults, respectively, as shown in Table 5 and Figure 5.

The research findings indicate that ^{40}K contributes the highest percentage to the total annual effective dose resulting from the ingestion of powdered milk, accounting for 74.15% in infants, 47.30% in children, and 62.97% in adults. However, the ^{40}K dose obtained in this study is used solely for comparative purposes. This is because potassium is an essential element that is strictly regulated through homeostatic mechanisms within the body and is not influenced by variations in environmental levels. As a result, the dose from ^{40}K within the body remains constant. ^{226}Ra contributes to the total effective dose by 7.75% for infants, 22.71% for children, and 22.20% for adults. ^{232}Th contributes 18.10% to the total effective dose for infants, 29.99% for children, and 14.83% for adults, as shown in Figures 6, 7, and 8. The total average annual effective doses due to the intake of ^{226}Ra, ^{232}Th, and ^{40}K from powdered milk were as follows: 214.50, 123.27, and 41 μSv/year for infants, children, and adults, respectively, as listed in Table 5. This indicates that infants face the highest risk compared to other age groups because powdered milk is their primary food source, consumed in large quantities during the early stages of life. In general, the total average annual effective dose results for the three age groups in this study were within the typical global limits recommended by the WHO for annual effective dose, which is 1 mSv/year due to the ingestion of natural radiation sources [26–28].

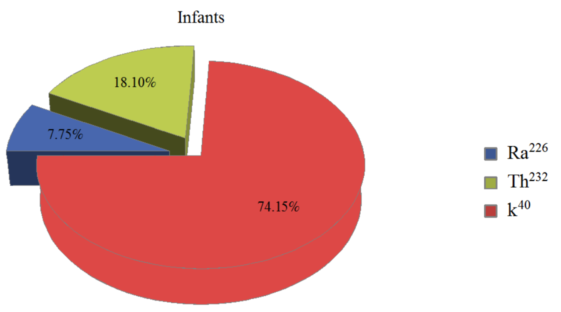

Figure 7. Histogram of the total annual effective dose for infants and contribution of ^{226}Ra, ^{232}Th and ^{40}k.

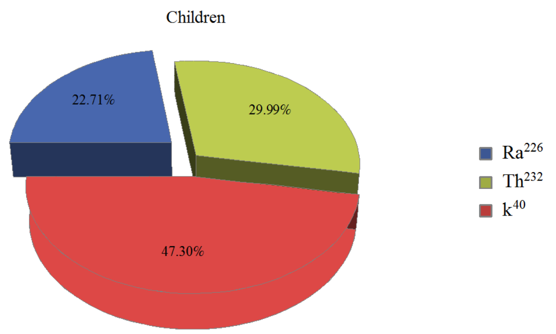

Figure 8. Histogram of the total annual effective dose for children and contribution of ^{226}Ra,^{232}Th and ^{40}k

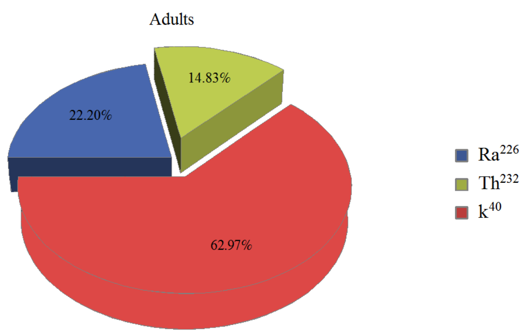

Figure 9. Histogram of the total annual effective dose for adults and contribution of ^{226}Ra,^{232}Th and ^{40}k.

4. CONCLUSIONS

For the first time, a systematic investigation of the radioactivity concentration in milk consumed in Yemen has been conducted. Using an HPGe detector, the activity concentrations of radionuclides ^{226}Ra, ^{232}Th, and ^{40}K were measured in 16 powdered milk samples. Additionally, the annual effective dose (AED) from milk ingestion in Yemen was calculated. ^{40}K contributes the most to the total AED, accounting for 74.15% in infants, 47.30% in children, and 62.97% in adults. The contribution of ^{232}Th to the total effective dose is 18.10% for infants, 29.99% for children, and 14.83% for adults, while ^{226}Ra contributes 7.75% for infants, 22.71% for children, and 22.20% for adults. In summary, the total average annual effective doses (AED) from the intake of ^{232}Th, ^{226}Ra, and ^{40}K in powdered milk are 214.50, 123.27, and 41 μSv/year for infants, children, and adults, respectively. To conclude, the total average annual effective dose for the three age groups is within the permissible limits set by the WHO (1 mSv/year). This study provides essential data on natural radioactivity in milk samples, which can support the formulation of radiological protection guidelines for the country's population.

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