International Journal of Progressive Sciences and Technologies (IJPSAT)
ISSN: 2509-0119.

http://ijpsat.ijsht-journals.org
Vol. 23 No. 2 November 2020, pp. 461-469

Assessment of Natural Radionuclides in Some Powdered milk Samples Consumed in Yemen and Estimated of the Corresponding annual Effective Dose

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Abstract – The aim of this study is to know how much the some imported and produced milk are contaminated with naturally occuring and man- made radionuclide as Cesium. Seven samples of powdered milk samples were collected. They are the most consummed in Yemen. These Samples were prepared in the laboratory, sieved through a fine mesh, well sealed in standard Marinelli beakers (30ml) for about four weeks in order to establish secular equilibrium between Th^{232} and Ra^{226} with their progeny. Gamma ray spectroscopy is used to analyze the samples and the concentrations of radioisotopes were determined in Bq/kg by using hyper-purity germanium (HPGe) detector Canberra p-type (model) 707 with a relative efficiency of 30% and energy resolution is of 1.85 keV.
the average concentrations of Th^{232} , Ra^{226} and K^{40} were found to be concentrations of 5.37 , 31.29 and 96.84 Bq/ kg respectively in powdered milk samples. Also, the total average annual doses received from the intake of K^{40}, Th^{232} ,Ra^{226}, and due to the ingestion of the powdered milk were measured (in μSv): 970.575, 660.81and 315.50 for infants, children and adults, respectively. These results indicate that infants have a higher risk factor compared with other age groups. Finally, the measured activity concentrations for these radionuclide were compared with the reported data obtained from similar foodstuffs consumed in other countries and with International references value. The value of the average activity concentration of the radionuclides Th^{232} , Ra^{226} measured in powdered milk samples are higher than the international values, but it is lower for K^{40} in all samples.

Keywords – Radionuclide Concentrations, Milk, Radiation dose, Annual Effective Dose, high-purity germanium (HPGe) detector.

I. Introduction

It is inevitable that all populations have been subject to radiation exposure from external and internal sources almost every day of their lives. The greatest contribution of irradiation to the human body comes from natural sources, which are emitted from the members of the natural decay series as well as from the radioisotope of potassium K40. In addition to the natural sources, the environment has been considerably affected from man-made sources such as those produced artificially through nuclear reactions, thermo nuclear weapons testing and the Chernobyl power station accidentin1986(WHO, 1987). Agricultural products are usually contaminated either directly or indirectly with natural and artificial radionuclides. Therefore, consumption of such products increases the internal radiation dose to the humans and contributes a large portion to the mean annual effective dose (Michael Poschl,2007). Milk is one of the important food for human nutrition and contains all the macro nutrients (Alamoudi Zain et al. 2013). Also, milk and milk products are main constituents of human daily diet. So, the accurate measurement of the activity concentration of naturally occurring radionuclides in milk is useful for determining human population exposure to ionizing radiation by ingestion and domestic uses because the doses from these pathways are strongly related to the amount of radionulcides present. Internal exposure of humans to high levels of radium for a long time may produce bone and sinus cancers (UNSCEAR-2000) . Many countries and several international organizations have determined the concentrations of natural radionuclides in milk and the annual intake and , hence, the radiation dose due to its consumption have been evaluated (Nasriev and Ahmedovo1987; Abdulfatah and Abdualmajed1995; Marmuleva et al 2003; R .Hewamanna et al 1999; Melquiades et al 2001; Alzuhairi and Al-shami et al 2005; T.Hosseini et al 2006; Shanti etal 2010; and Alamoudi Zain et al 2013.) . The present work is the first study of the radioactive content of the powdered milk, and milk products consumed by all age groups in Yemen , where, as, one kind from the powdered milk for adult were studied before (Alzuhiri ,and Alshami, 2005). So, the main objectives of this study are to identify and determine natural radionuclide activity concentrations in powdered milk samples available in Yemen local markets. Data obtained here can provide an opportunity to verify any impact from the ingestion of radionuclides in the people who consume milk, also may be relevant for designing local rules and regulations for radiation protection and safety purposes. Among different kinds of foodstuffs milk, and milk products is a reliable indicator of the general population intake of certain radionuclides, since milk is consumed by a large segment of the population, and contains several biologically significant radionuclides (E R DATA, U.S-2000). By these considerations, the aim of the present study has been to determine the activity concentration of Th^{232}, Ra^{226}, and K^{40}, in different milk samples used in Yemen, and to estimate its effective ingestion dose.

II. MATERIALS AND METHODS

2.1 Sampling and Sample Preparation

Seven samples of different types of powdered milk , and milk products were collected during the year of the experiment .The samples have been collected from Yemeni markets. Every sample's weight 250g for solid samples or its capillaries The samples have been put alone in marnilli peakers. They were put in a suitable cans about four weeks in order to establish secular equilibrium between Ra^{226} , Th^{232} with their progeny before measuring. Those samples are represented in tables (1). An ampty marrnilli beakers was also counted under the same conditions to determine the background.

2.2 Radioactivity determination:

The activity concentrations of U^{238} ,Th^{232}, Ra^{226}, and K^{40}, in the samples analyzed, as derived from Windows Analysis . The samples were measured using a closed end-coaxial Canberra p-type (model 707) high-purity germanium (HPGe) detector with a relative efficiency of 30%. The germanium crystal is located inside the lead shield. Energy resolution of 1.85KeV, full width at half maximum (FWHM) for the 1332KeV gamma line of $Co^{60.}$,[G.Shaw., et al 2007]. A mixed radionuclide gamma-ray reference standard consists of a solution in 4M HCl of the ten radionuclides which are Am^{241} (60KeV), Cd^{109} (88 KeV), Co^{57} (122 KeV), Ce^{139} (166 KeV), Hg^{203} (279 KeV), Sn^{113} (392 KeV), Sr^{85} (514 KeV), Cs^{137} (662KeV), Co^{60} (1173 and 1333KeV), Y^{88} (898 and 1836 KeV) of activities 1131, 644, 577, 738, 1886, 2054, 3838, 2496, 3377, 3382, 6288 and 6651-rays/s.g, respectively as in the reference certificates number QCY48 issued by GE Healthcare limited is used to calibrate HPGe detector. And calculated some of Exposure Risk such as, , annual effective dose AED (μSv/yr) ,

2.2.1 Specific Activity:

Each Sample was measured for (12 hrs). Genie2000 was used for the acquisition and analysis, and diagnosed radionuclides U^{238},Th^{232}, U^{235}, and K^{40} by choosing an overlapping energies. Energy lines (351.9 keV, 609.4 keV, 1120.1 keV, and 1764.4 keV) were used to calculate the radioactivity of Ra^{226} (U^{238}) in the samples. Energy lines (338.3 keV, 911.1 keV, and 2614.7 keV) were used to calculate the radioactivity of Th^{232} in the samples. Energy line 1460.8 keV it used to calculate the radioactivity of potassium in the samples. Ampty marrnilli beakers was counted under the same conditions to determine the background. Unit Bq/Kg, was used for measurments specific activity (in Bq/kg) A_{Ei} , of a nuclide i and for a peak at energy E_i, is given by (Curie L et al 1968.).

$$A_{Ei} = \frac{N_{Ei}}{\varepsilon_E \times t \times \gamma_d \times M_s} \qquad \text{.............. (1)}$$

where N_{Ei} is the net peak area of a peak at energy E, $\square_E$ the detection efficiency at energy E, t the counting live-time, $\square_d$ the intensity per decay(%) of photon emission at the energy E, and M_s the mass in kg of the measured sample. If there is more than one peak in the energy range of analysis, then the peak activities are averaged and the result is the weighted average nuclide activity. The software calculate the net counts in a particular photopeak , the background subtracted net counts ,and calculate of the A_{Ei}. Also, the specific radioactivity(Bq/kg) was calculated by dividing the radioactivity over the sample volume for the powder samples.

2.2.2 Detection Limits (D.L)

Detection limits is the leas. specific radioactivity of radionuclides can be measured Curie equation used to express the detection limit (UNSCEAR- 2000).

$$\mathbf{D.L} = (\mathbf{2.77} + \mathbf{3.29})\frac{\text{Specfic activity}}{\text{Net area}} \quad \text{........(2)}$$

Where; Net area: is the net peak area.

2.2.3 The minimum detectable activity (MDA):

The minimum detectable activity (MDA) is defined as the smallest concentration of a radionuclide that can be determined reliably with a giving detection system. For low-level radioactivity measurements, it is essential to reduce the background count and increase detection efficiency. Counting efficiency can be improved by using a larger amount of source sample and a larger detector volume, and by improving the source-detector geometry.

The MDA are given by the following equation (Michael et al 2007),

$$\mathbf{MDA} = (\mathbf{2.71} + \mathbf{4.65})\frac{\sqrt{\mathbf{B.G}}}{\mathbf{t.\varepsilon}} \qquad \mathbf{(3)}$$

Where; B .G: is the background counts in the region of interest or of the counting system, t : is the counting time (sec.), and ε :is the counting efficiency at the specifice energy(%).

2.2.3 Annual ingestion dose for different age groups AED (μSv/yr)

The annual effective dose (D) to individuals due to the ingestion of the radionuclides in the powdered milk is estimated using the following equation (Zaid Q. Ababneh,... et al 2009).

$$D = AIE \qquad . (4)$$

where :

D is in (Sv /y), A is the activity concentration of radionuclides in milk (Bq/ kg), E is the dose conversion factors for the radio nuclides (Sv/Bq), and I is the annual intake of milk (kg/y), which depends strongly on a given age group. The calculation of the annual ingestion dose has been done for different age groups: infants, children and adults. Corresponding conversion factors are taken for those of 1–2, 3–12 and (< 17 y) and listed in Table 2. The average annual intakes of the powdered milk, used in the dose calculations, are taken as those of fresh milk consumption divided by a concentration ratio. This yields for the annual intake (I) the values of about 15, 14 and 13 kg/y for infants, children and adults, respectively (Zaid Q. Ababneh,..et al 2009).

Table(1): The Specific Activity in (Bq/kg) of powder milk samples available in yemen.

No	Sample Lab No.	Specific Activity in (Bq/kg)			
		Cs^{137}	Ra^{226}	Th^{232}	K^{40}
1	S-1	<MDA	16.73	8.27	170.32
2	S -2	<MDA	35.9	3.50	140.77
3	S -3	<MDA	<MDA	8.40	7.86
4	S -4	<MDA	30.25	4.24	14.35
5	S -5	<MDA	37.94	9.13	273.01
6	S -6	<MDA	47.63	1.14	48.45
7	S -7	<MDA	51.18	2.90	23.10

Average concentration	**--**	**31.29**	**5.37**	**96.84**
Range		**0-51.2**	**1.14 -9.13**	**7.86- 273.01**

Table 2:The dose conversion factors of K40, Ra226, Th232 (Ra228) and Cs137 for the age groups: infants (1–2 y), children (3–12 y) and adults (>17 y) [ICRP - 1996].

	Dose conversion factors (mSv/ Bq)			
Age Group	K^{40}	Cs^{137}	Ra^{226}	Th^{232} (Ra^{228})
Infants (1–2 y)	42	11	960	5700
Children (3–12 y)	13	9.8	800	3900
Adults (< **17**y)	6.2	13	280	690

III. Results and Discussions

3.3.1 Activity concentrations of analyzed radionuclide's in milk samples:

Measured activity concentratins of Th^{232}, Ra^{226} and K^{40} in the powdered milk samples under study including their uncertainty are reported in Table 1. The activity concentrations of, Th232 vary from 1.14 Bq/Kg in (s.no. 6) to 9.13 Bq/Kg in (s.no.5) with an average value 5.37 Bq/Kg , Ra^{226} concentrations vary from 16.73 Bq/Kg in (s.no.1) , 51.18 Bq/Kg in (s.no.7) with an average value 31.29 Bq/Kg. although, (s.no.3) is less than the detection limit (<MDA) Table (1), shows that the main gamma detected activity arises from K^{40} was detected in all samples and varied between 7.86 Bq/Kg (s.no.3) to 273.01 Bq/Kg in (s.no.5) with an average value of 96.84 Bq / kg , this variation in the activity concentration is significantly fairly large from one brand of milk to another. Figure.1, shows comparison between the mean activity concentration of Th^{232}, Ra^{226} and K^{40} in powdered milk under investigation. The Th^{232}, Ra^{226} and K^{40} activities measured in the present work were comparable with completion of others activated values of milk samples around the world were presented in Table (3) . The results obtained show that the measured activity concentrations of Th^{232}, Ra^{226} and K^{40} in all samples and their average values appears much above the recommended values of other countries . The measured activity concentrations values for K^{40} and the average value in these samples are found to be higher than the values of Jordanian(Zaid Q. Ababneh,.. et. al 2009) only , but lower than the values in powdered milk from some parts of the world as shown in Table (3) . As can be seen from this table, the K^{40} activities are, on an average, lower than the other study by almost 40% approximately.

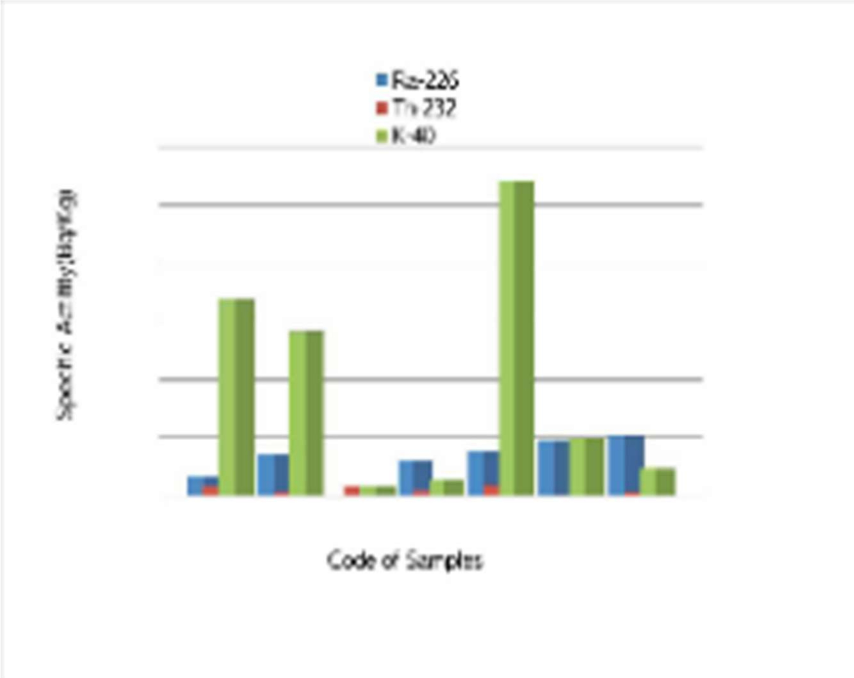

Fig.1: Comparison of Specific Activity radionuclides in milk samples in **Yemen.**

Table 3 : represents a comparison specific activity values in(Bq/kg)

of milk samples in this study with studies from other countries.

No.	country	Specific Activity in (Bq/kg)				Reference
		Cs^{137}	Th^{232}	Ra^{226}	K^{40}	
1	Yemen	<MDA	1.14 - 9.13 (5.37)	<MDA-51.2 (31.29)	7.86- 273.01 (96.84)	present work
2	Iyran	--	5.4±0.2	2.9±0.1	778.4±23	T.Hosseini et al 2006
3	Nigeria	--	4.35±2.06	23.07±7.7	831.66±54.8	O. A. Osibote, et al 1999
4	Brazil	--	-	-	482 ± 37	Melquiades et al 2001;
5	uzbekistan	--	4.8×10^3	4.4×10^2	12.95×10^3	Nasriev and Ahmedovo et al 1987;

6	**Iraq**	**--**	**-**	**-**	**404.822**	**R. M. Yousif, et al**
7	**New zealand**	**--**	**0.166±0.03**	**0.186±0.03**	**549.0±16.5**	**T.Hosseini et al 2006**
8	**Jordan**	**--**	**0.78–1.28**	**0.50–2.14**	**40.8+0.8**	**Zaid Q. Ababneh,.. et. al2009**

3.2 Annual ingestion dose for different age groups :

Table.4 ,shows the calculated annual effective doses due to the intake of K^{40}, Th^{232}, Ra^{226} in the reconstituted powdered milk for different age groups. The results showed that the average annual effective dose due to the intake of the natural radionuclide K^{40} by infants is 61.007μSv, which is much higher than the average affective doses of 17.25 and 7.80μSv corresponding to those for children and adults. Also the results showed that the contribution due to the intake of K^{40} to the total annual effective dose due to the intake of the powdered milk; 6.29% for infants, 2.61 % for children and 2.47 % for adults.

For the natural radionuclide Th^{232}, the results showed that it is the largest contributor to the total annual effective dose contributing to 61.1%, 44.36% and 47.3 % of the total for adults, children and infants, respecti-vely. Also the results showed that the contribution due to the intake of the Ra^{226} total annual effective dose is 46.42, 53.03 and 36.43 % for the infants, children and adults, respectively.

The total average annual doses received from the intake of K^{40}, Th^{23} ,Ra^{226}, and due to the ingestion of the powdered milk were (in μSv): 970.575, 660.81and 315.50 for infants, children and adults, respectively (Table 4). This indicates that infants have a higher risk factor compared with other age groups.

From all radionuclidies, the cumulative annual effective dose received by infants is _ 1.5 and 3.1 higher than that received by children and adults, respectively.

Table4: Age-dependent annual effective dose(μSv) due to the intake of K40, Ra226, and Th232from powdered milk.

Radionuclides	**Annual effective dose (μSv)**			
		Infants	**Children**	**Adults**
K^{40}	**Minimum**	**4.951**	**1.431**	**0.628**
	Maximum	**171.996**	**49.688**	**22.004**
	Average	**61.10**	**17.251**	**7.8**
Ra^{226}	**Minimum**	**240.912**	**90.376**	**30.897**

	Maximum	**736.992**	**573.216**	**193.575**
	Average	**450.552**	**350.432**	**114.93**
Th^{232}	**Minimum**	**97.470**	**62.244**	**10.226**
	Maximum	**780.615**	**498.498**	**81.896**
	Average	**459.013**	**293.124**	**192.768**
Total average		**970.576**	**660.807**	**315.503**

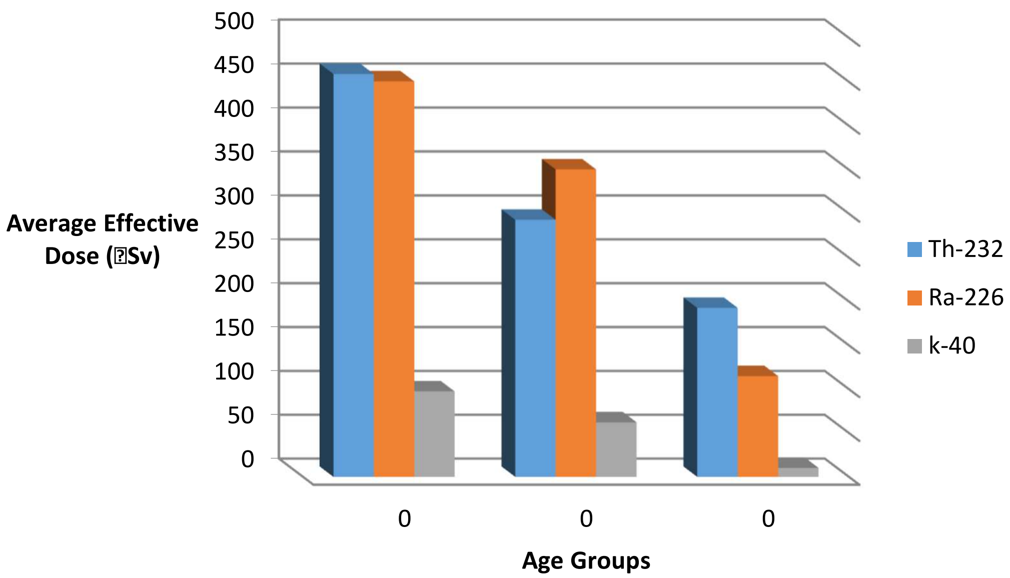

Fig.2: Average the annual effective dose (µSv) in Age Groups due to the intake of K40, Ra226, and Th232 from powdered milk

These results for all age groups are within the typical worldwide range of annual dose (200–800µSv) due to the ingestion of all natural radiation sources reported by UNSCEAR 2000[l]. Figure 2, represents average values due to annual effective dose (µSv) in age groups due to the intake of K^{40}, Ra^{226}, and Th^{232} from powdered milk samples.

IV. CONCLUSIONS

In this study, the radionuclides present in powdered milk consumed in Yemen such as Th^{232}, Ra^{226} and K^{40} are identified with averages of 5.37 Bq/Kg , 31.29 Bq/Kg and 96.84 Bq/Kg , respectively. The values of Th^{232} and Ra^{226} concentrations are found to be greater than those reported of other countries in the literature ,where K^{40} concentrations are lower the other study .So , we can deduce that the radionuclides level in the studied samples of powdered milk in Yemen for children and adults is obviously below the maximum level permitted by IAEA , so , these kinds of milk can be normally consumed. The total average annual doses received from the intake of K^{40}, Th^{23} ,Ra^{226}, and due to the ingestion of the powdered milk were (in μSv): 970.575, 660.81 and 315.50 for infants, children and adults, respectively . The total average annual effective dose received by infants being 1.5, and 3.1 times higher than that received by children and adults, respectively. Which are far below the WHO recommended limit . Also, all athese results were lower than the world reference values as has been reported in ICRP(2005) report.

The data generated in this study will provide baseline radiometric values for natural and artificial radioactivity in milk and help to develop future guidelines in Yemen for radiological protection for the population.

ACKNOWLEDGEMENTS

I express my gratitude to Dr.Taher Mohammed, Dr.Abdualhameed Othman and Dr.Sahar Ahmed for her fruitful discussions and advise.

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