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Measurement of Natural Radioactivity Levels for Some Milk Products Samples Available in Yemen by Gamma Spectrometry

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Abstract – The aim of this study is to know how much the some imported and produced milk products are contaminated with naturally occuring and man- made radionuclide as Cesium. Four samples of baby food and Seven samples of cheese and cream 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 6.14, 23.33 and 184.65 Bq/kg respectively in a baby food samples, and 0.469, 0.368 and 21.7 Bq/kg, in cheese and cream. Some important coifficient were also determined such as the Internal hazard index (H_{in}). 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 baby food samples are higher than the international values, but it is lower for K^{40} in all samples. The results also showed that the internal hazard coifficient (H_{in}) in all samples lower than one (< 1) , then don't consider any risks on the human's health.

Keywords – Radionuclide Concentrations, Milk Products , Radiation dose, Internal hazard index , 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 K^{40}. 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)[1] . 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)[2]. Milk is one of the important food for human nutrition and contains all the macro nutrients (Alamoudi Zain et al. 2013) [3]. 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) [4] . Many countries and several international organizations have determined the concentrations of natural radionuclides in milk products and the annual intake and , hence, the radiation dose due to its consumption have been evaluated (R. Sasaki , and T. Tsugo., et al 1959; A. Aarkrog, et al 1980; Beresford, N.A. et al 2000; Abdulfatah and Abdualmajed1995; Marmuleva et al 2003; H. Mukallati, A. Al-Hamwi et al 2004; Alzuhairi and Al-shami et al 2005; and T.Hosseini et al 2006;) ,[5, 6, 7, 8, 9, 10, 11, 12] . The present work is the first study of the radioactive content of 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) [11]. So, the main objectives of this study are to identify and determine natural radionuclide activity concentrations in milk products 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) [13]. 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 products samples such as baby food, cheese and cream, which used in Yemen, and to estimate the Internal hazard index (H_{in}).

II. Materials And Methods

2.1 Sampling and Sample Preparation

Eleven samples of different types of 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), and (3).

An ampty marrnilli beakers was also counted under the same conditions to determine the background.

2.2 Radioactivity determination:

The activity concentrations of U238 ,Th232, Ra226, and K40, 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 Co60 [Shaw.G.; 2007] ,[14]. A mixed radionuclide gamma-ray reference standard consists of a solution in 4M HCl of the ten radionuclides which are Am241 (60KeV), Cd109 (88 KeV), Co57 (122 KeV), Ce139 (166 KeV), Hg203 (279 KeV), Sn113 (392 KeV), Sr 85 (514 KeV), Cs137 (662KeV), Co60 (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 that illustrated in the Fig.1, The multichannel analyzer (MCA), ORTEC type with 8062 channel was used for the measurements in this research .

Fig.1: The Photogragh Picture for High Pure Germanium Detector(HPGe) which used to Analyse the Samples .

And estimated some of Exposure Risks such as the Internal hazard index (H_{in}) .

2.2.1 Specific Activity:

Each Sample was measured for (12 hrs). Genie2000 was used for the acquisition and analysis, and diagnosed radionuclides U238,Th232, U235, and K40 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 Ra226 (U238) in the samples. Energy lines (338.3 keV, 911.1 keV, and 2614.7 keV) were used to calculate the radioactivity of Th232 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) AEi , of a nuclide i and for a peak at energy Ei, is given by (Curie L et al 1968) [15] ,

$$A_{Ei} = \frac{N_{Ei}}{\varepsilon_E \times t \times \gamma_d \times M_s} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(1)$$

where N_{Ei} is the net peak area of a peak at energy E, ε_E the detection efficiency at energy E, t the counting live-time, γ_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 least specific radioactivity of radionuclides can be measured. Curie equation used to express the detection limit (UNSCEAR- 2000) [4] ,

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

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) [2] ,

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

Where; B .G: is the background counts in the region of interest or of the counting system, t : is the counting time (sec.),

ε :is the counting efficiency at the specifice energy.

2.2.4 The internal Hazard Index(Hazard Coefficient H_{in}):

In addition to the external hazard, radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter products is quantified by the internal hazard index H_{in} which is given by the equation (4) [16].

$$\mathbf{H_{in}} = \frac{\mathbf{C_{Ra}}}{\mathbf{185}} + \frac{\mathbf{C_{Th}}}{\mathbf{259}} + \frac{\mathbf{C_K}}{\mathbf{4810}} \leq \mathbf{1} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(4)$$

Where :

C_{Ra}, C_{Th} and C_K represents concentration of radium, thorium and potasium in the sample.

It is assumed that 185 Bq/kg of Ra226, 259 Bq/kg of Th232 and 4810 Bq/kg of K40 produce the same gamma-ray dose rate . If the maximum concentration of radium is half that of the normal acceptable limit, then Hin will be less than 1.0 [17]. For the safe of consume a foodstuffs of population, Hin should be less than unity (<1).

III. Results And Discussions

3.1 Activity concentrations of analyzed radionuclide's in baby food samples

Measured activity concentrations of **Th^{232}, Ra^{226} and K^{40}** in the baby food samples under study including their uncertainty are reported in Table 1. The activity concentrations of, **Th^{232}** vary from 0 Bq/Kg in (s.no. 9),which is less than the detection limit (<MDA)to 9.21 Bq/Kg in (s.no.10) with aaverage value 6.14 Bq/Kg , **Ra^{226}** concentrations vary from 13.03 Bq/Kg in (s.no.8) , 35.08 Bq/Kg in (s.no.9) with an average value 23.33 Bq/Kg.). Table (1) shows that the main gamma detected activity arises from **K^{40}** was detected in all samples and varied between 134.56 Bq/Kg (s.no.9) to 238.3 Bq/Kg in (s.no.10) with an average value of 184.65 Bq / kg , this variation in the activity concentration is not significantly fairly large from one brand of baby food samble to another. Figure 2 shows comparison between the mean activity concentration of **Th^{232}, Ra^{226} and K^{40}** in baby food 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 baby food samples around the world were presented in Table (2) . 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 Syrian (**H. Mukallati, A. Al-Hamwi,.. et al 2004**) [10] and Iranian(**T.Hosseini et al 2006**) [12] and lower than the values in baby food from some parts of the world as shown in Table (2) . As can be seen from this table, the **K^{40}** activities are, on an average, lower than the other study by almost 5% approximately.

Table(1): The Specific Activity in (Bq/kg) of baby food samples available in local market

No.	Sample Lab No	Specific Activity in (Bq/kg)			
		Cs^{137}	Ra^{226}	Th^{232}	K^{40}
8	S-8	<MDA	13.03	7.22	144.70
9	S-9	<MDA	35.08	<MDA	134.56
10	S-10	<MDA	25.17	9.21	238.02
11	S-11	<MDA	20.02	8.11	221.3
Average concentration		–	23.33	6.14	184.65
Range		–	13.03-35.08	0-6.14	134.56-238.02

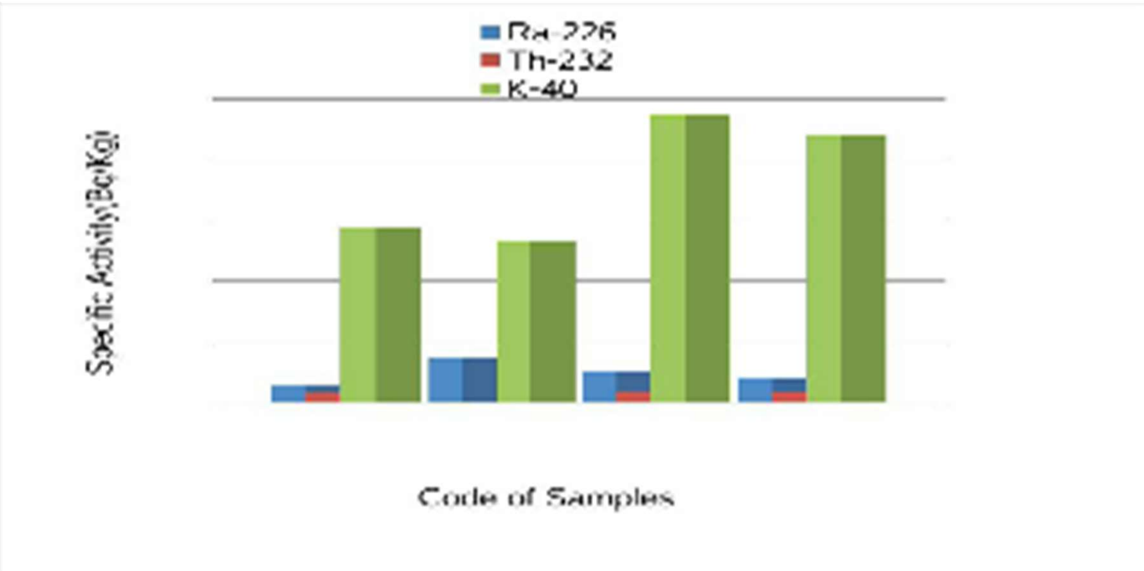

Fig.2: Levels of Specific Activity radionuclides in baby food samples (in Bq/kg)

Table 2: A comparison of specific activity values in(Bq/kg) of baby food samples in this study with studies from other countries.

No	Name of Sample	Specific Activity in (Bq/kg)			the origin for sample	Reference
		Th^{232}	Ra^{226}	K^{40}		
1	Baby Food	0-9.21 (6.14)	13.03-35.08 23.33	134.56-238.02 184.65	Yemen	This work
2	Baby Food	<0.023	0.14±0.02	42.4±0.8	Belgium	T.Hosseini et al 2006 [12]
3	Baby Milk- Bebelac-1	-	-	197.38	Holland	Abdulfatah and Abdualmajed et al 1995; [8]
4	Baby Food-Cerelac(Wheat&honey)	-	-	103.97	-	Abdulfatah and Abdualmajed et al 1995 [8]

5	**Baby Milk-Similac**	**-**	**-**	**225.73**	**-**	**Abdulfatah and Abdualmajed et al 1995 [8]**
6	**Baby Food**	**4**	**3**	**-**	**Syria**	**H. Mukallati, A. Al-Hamwi,.. et al 2004 [10]**

3.1.2 Internal Hazard Index(H_{in})

The calculated values of the results of Hin for all samples are lower than unity (<1). The values of Hin for the studied baby food samples range from 0.128 to 0.222. So they don't consider any risk on human's health.See table.3, and fig.3.,

Table(3): Internal hazard index Hin for the baby food samples

No.	1	2	3	4	Average
Sample code	S-8	S-9	S-10	S-11	
H_{in}	0.128	0.216	0.222	0.19	1.89

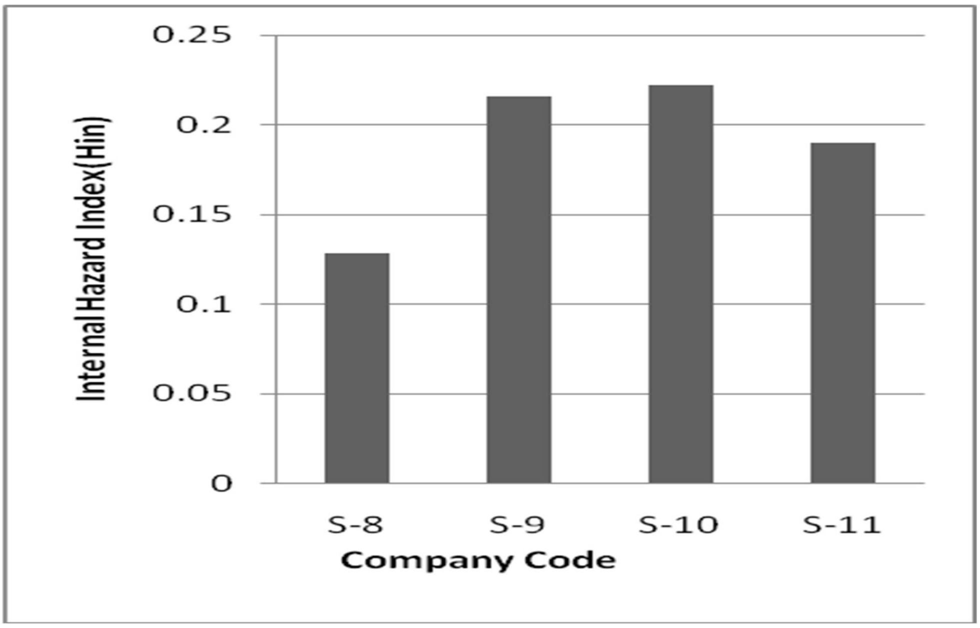

Fig.3: Internal hazard index Hin of baby food samples

3.2 Activity concentrations of analyzed radionuclide's in cheese and cream samples:

Measured activity concentrations of Th232, Ra226 and K40 in the cheese and cream samples under study including their uncertainty are reported in Table 4 . The activity concentrations of ,Th232 vary from 0.17 Bq/Kg in (s.no.16) to 1.2 Bq/Kg in (s.no.12) with an average value 0.469 Bq/Kg , Ra226 concentrations vary from 0 Bq/Kg in (s.no.17), it is less than the detection limit (<MDA), to 0.92 Bq/Kg in (s.no.15) with an average value 0.368 Bq/Kg. Table (4) shows that the main gamma detected activity arises from K40 was detected in all samples and varied between 9.5 Bq/Kg (s.no.17) to 31.1 Bq/Kg in (s.no.14) with an average value of 21.7 Bq / kg , this variation in the activity concentration is not significantly large from one brand of sample to another. Figure 4 shows comparison between the mean activity concentration of Ra226 ,Th232 and K40 in cheese and cream under investigation. The Ra226 ,Th232 and, K40 activities measured in the present work were comparable with completion of others activated values of cheese and cream around the world were presented in Table (5) . The results obtained show that the measured activity concentrations of Th232 in all samples and the average value appears much above the recommended values of other countries , except the values of Iranian (Hosseini et al 2006) (12) . The measured activity concentrations values for Ra226 and K40 and their average values in these samples are found to be higher than the values in cheese and cream samples from some parts of the world as shown in Table (5) .

Table(4) : The Specific Activity in (Bq/kg) of commercial cheese and cream samples

No	Sample Lab No	Specific Activity in (Bq/kg)			
		Cs^{137}	Ra^{226}	Th^{232}	K^{40}
1	S-12	<MDA	0.39	1.2	20.02
2	S-13	<MDA	0.53	0.47	27.12
3	S-14	<MDA	0.027	0.25	31.1
4	S-15	<MDA	0.92	0.27	29.43
5	S-16	<MDA	0.11	0.17	22.03
6	S-17	<MDA	<MDA	0.4	9.5
7	S-18	<MDA	0.6	0.52	12.5
Average concentration		_	0.368	0.469	21.7
Range		_	0 - 0.92	0.17 - 1.2	9.5 - 31.1

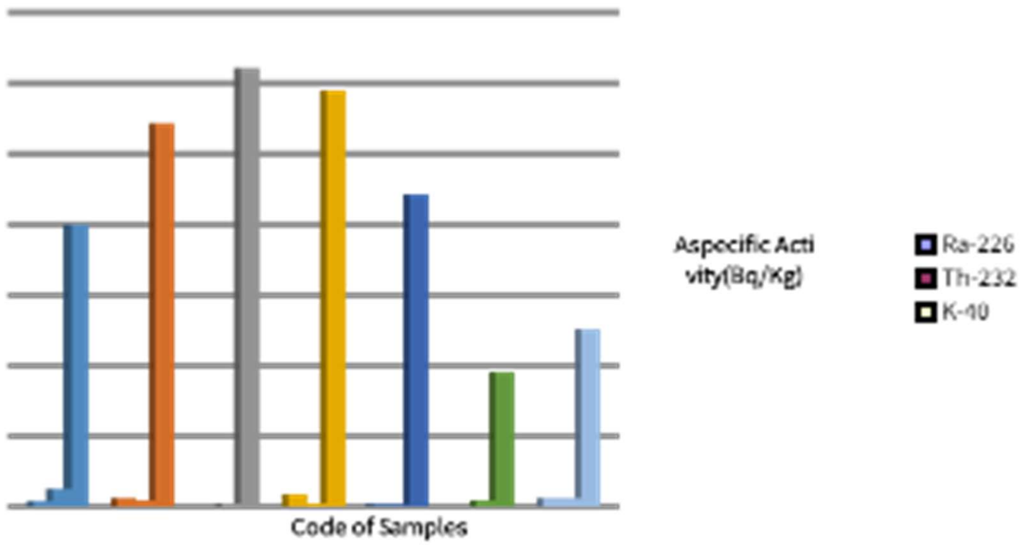

Fig. 4: Levels of Specific Activity radionuclides in cheese and cream samples(in Bq/kg).

Table 5: represents a comparison specific activity values in(Bq/kg) of cheese and cream samples in this study with studies from other countries.

No	Name of Sample	Specific Activity in (Bq/kg)				the origin of sample	Reference
		Cs^{137}	Th^{232}	Ra^{226}	K^{40}		
1	Cheese	4.3	–	–	-	France	IAEA 295, Viena. et al (1989).[18]
2	Cheese	–	–	–	17	Westrn 8	..
						Europe	

3	**Butter Cream**	**0.18□0.02**	**0.83□0.15**	**0.298□0.08**	**6.4□0.3**	**Netherlnd**	**T.Hosseini et al 2006**(12)
4	**Cheese**	**<0.2**	**<0.7**	**<0.4**	**25**	**Australia**	**T.Hosseini et al 2006**(12)
5	**Cheese**	**8**	_	_	_	**Swedish**	**T.Hosseini et al 2006**(12)
6	**Cheese**	**0.08 Bq**	_	_	_	**Denmark**	**A. Aarkrog,.., et al 1980** (6)
7	**Cheese**	**-**	_	**0.03**	_	**-**	**Beresford, N.A,... et al 2000** (7)
8	**butter**	**8.03**	_	_	_	**Russia**	**Marmuleva,..et al 2003**:(9)
9	**Cheese**	_	**0.652**	**0.495**	**25.35**	**Yemen**	**This work**
10	**Cream**	_	**0.252**	**0.363**	**14.68**	**Yemen**	**This work**

3.2.1. Internal Hazard Index (H_{in})

The calculated values of the results of Hin for all cheese and cream samples are lower than unity. The calculated values of Hin for the studied samples range from 0.004 to 0.012 Once again, all these values are less than unity(<1) . See table.6 ,and Fig.(5).

Table 6 : Internal hazard index Hin , for the cheese and cream samples

No.	1	2	3	4	5	6	7	
Sample code	S-12	S-13	S-14	S-15	S-16	S-17	S-18	Average
H_{in}	0.0109	0.0102	0.008	0.012	0.006	0.004	0.0078	0.0084

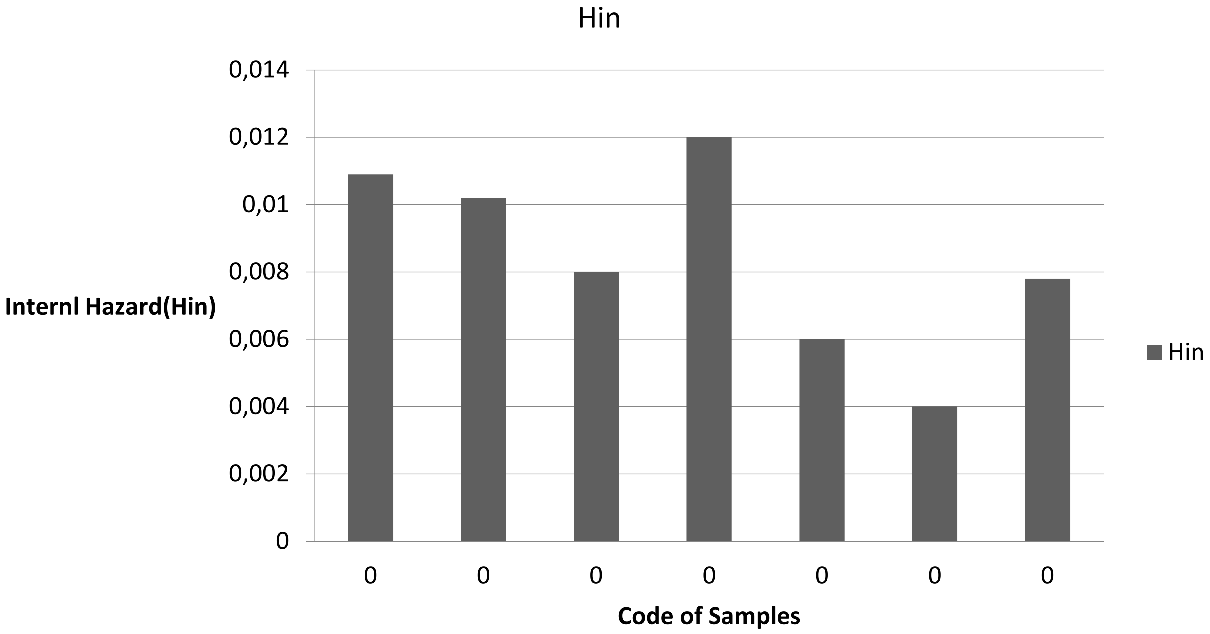

Fig.5: Internal hazard index (Hin) for cheese and cream Samples.

IV. CONCLUSIONS

In this study, the radionuclides present in some milk products samples consumed in Yemen such as Th232, Ra226 and K40 are identified.

The results show that the average concentrations of the radionuclides in the different baby food samples are 6.14 Bq/kg, 23.33Bq/kg and 184.65 Bq/kg for Th232, Ra226, and K40 respectively. Results of the average activity concentration of Th232 higher than the international values ,but it is lower for Ra226 ,and K40 . In regard to the above results, we conclude that the baby food consumed as food important for Yemeni babies generally have low specific activities. For cheese and cream samples, the average value of the concentrations for Th232, Ra226, and K40 have been found to lie within the average 0.469 Bq/kg, 0.368 Bq/kg and 21.7 Bq/kg, respectively. Then the activity concentrations of Th232, Ra226 and K40 in cheese and cream samples are higher than results studied in other countries. But were lower than the world reference values as has been reported in ICRP(2005) report [19] .

Also, the results showed that the averages hazard coefficient Hin are 0.189 and 0.0084 in baby food , cheese and cream samples. All samples show lower than unity(< 1), then they don't consider any risks on the human's health.

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

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REFERENCES

[1] Report on a World Health Organism (WHO),Health Hazards and Radiocaesium from the following Chernobyl nuclear accident, meeting, aschloss Reisensborg , Copenhagen ,Germany(1987) .

[2] Michael Poschl,- Radionuclide concentrations in food and the environment-, puplishing by Taylor & Francis Group, LLC, © (2007).

[3] Alamoudi Zain, Assessment of Natural Radionuclides in Powdered milk Consumed in Saudi Arabia and Estemated of the Corresponding annual Effective Dose , Journal of Americn Science.267 - 273 .(ISSN: 1545 - 1003).9(6) , 2013. (http: //www.americanscienc.org. 31).

[4] United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), Sources and effects of ionizing radiation, vol.1, New York, united Nations publication, (2000).

[5] Sasaki. R., Tango. T, Koyama. S., and Omomo. Y.,, Radioactive Contamination of Milk and Milk Products in Japan,, Japan, Department of Agricultural Chimistry, University of Tokyo, (1959), published online : 29 July 2014 ., by Journal of the agricultural chemical Society of Japan.To link to this article : https://doi.org/10.1080/03758397.1960.10857636 .

[6] A. Aarkrog, J. Lippert, and H. Dahlgaard,, Environmental Radioactivity in the Faroes in 1980,, Risø National Laboratory, DK-4000 Roskilde, Denmark,(1980).

[7] Beresford, N.A., Mayes, R.W., Cooke, A.I., Barnett, C.L., Howard, B.J. Stuart Lamb, C., and Naylor, G.P.L., The importance of source-dependent bioavailability in determining the transfer of ingested radionuclides to ruminant-derived food products, Environ. Sci. Technol., 34, 4455,(2000).

[8] A.F. Abdul-fattah& S. Abdul-Majed, Cs^{137}, and K^{40} Radioactivity Levels in Foodstuffs in Geddah and Riyadh Local Markets,, The Fourth Saudi Engineering Conference, Nov.. Volume V Jeddah & Saudi Arabia, (1995).

[9] Marmuleva, N. I.; Barinov, E. Ya; Petukhov, V. L. Radionuclides accumulation in milk and its products , JPhy4.107..827M ,(2003).

[10] H. Mukallati, A. Al-Hamwi, H. Khalili, M. Hassan H. Assaf, Y. Amin and A. Nashawati- , Atomic Energy Commission of Syria, (2004).

[11] A. Al-shamy , M H. AlZuhairy ,-Determination of radionuclides for some Imported Foodstuffs in Republic of Yemen - ,Abhath Al-Yarmouk. U, Basic Sci&Eng. Vol.14, NO.1,PP,85-92,© Jordan (2005).

[12] T. Hosseini, A.A. Fathivand, H. Barati, M. Karimi, Assessment of radionuclides in imported foodstuffs in Iran, Iran. J. Radiat. Res.; 43: 149-153. (2006).

[13] Environmental Radiation DATA (2001) United States Environmental Protection Agency Office of Radiation and Indoor Air, Environmental Radiation DATA. Report 106, April-June.

[14] Shaw.G ; Environmental Terrestrial in Radioactivity, First edition, (2007).

[15] Curie L., A., Analyses Chemestry, 40(3)- 578, (1968).

[16] Abdulazize Bazohier , Determination of Natural Radioactivity Concentrations in Soil and Rocks in Yemen and Analysis by nuclear methods , D.Sc. degree in Nuclear Physics at Baghdad University , (2000).

[17] Conceição, F. T., Bonotto, D. M. and Jiménez-Rueda J. R.,, Radionuclides Supply by Sugar Cane Crops at Corumbatai River Basin, , Brazil,, São Paulo, Brazil. Available at e-mail: ftomazini@sorocaba.unesp.br.

[18] IAEA 295, Measurements of radionuclides in food and the environment – International Atomic Energy Agency: a Guidebook. Viena. (1989).

[19] Risk values and dose limits for ICRP60 and NCRP116, (2005). available at: http://www.tenorom.com/ncrp1 icrp60.htm.Accessed, 2018.