Assessment of Natural radionuclides in Powdered milk Consumed in Saudi Arabia and Estimates of the Corresponding annual Effective Dose

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Abstract: This paper presents the activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th radionuclides measured in 20 brands of powdered milk samples collected from local markets of Saudi Arabia .The main detected activity corresponding to^{40}K with average activity of 74.51 BqKg^{-1}, while the average activities of ^{226}Ra,^{232}Th were 9.64 Bqkg^{-1}, and 6.77 Bqkg^{-1}, respectively. The determination of the radiation dose due to the consumption of these brands of milk are the main objectives of this program. The total average effective dose due to annual intake of ^{226}Ra, ^{232}Th and ^{40}K from the ingestion of the powdered milk for children(age 2-7y ,7-12y and 12-17y) were estimated to be 183.74 µSvy^{-1} 198.34 µSvy^{-1} and 234.15 µSvy^{-1} and for adults (▯ 17Y) is 29.41 µSvy^{-1},which these values are lower than the ICRP recommended limit of 1.0 mSv y^{-1} for all ages . Results are compared with those of different countries worldwide. The resulting data may serve as base-line levels of activity concentration in powdered milk in the area of study.

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Key word: Natural Radioactivity, Milk, Heavy Metals, Ingestion dose, Hazard quotient.

1. Introduction

The terrestrial or primordial radionuclides have sufficiently longer half-lives , so that they survived since their creation and keep decaying to attain the stable state and producing ionizing radiation in various degrees UNSCEAR, 1993) . Gamma radiation from these radionuclides represents the main external source of irradiation of the human body (Tzortzis *et al* .2003, UNSCEAR 2000). Doses by ingestion are due mainly to ^{40}K and to the ^{238}U and ^{232}Th series radionuclides present in food and drinking water. Milk is one of the important food for human nutrition and contains all the macronutrients (Abollino *et al*. 1998). Also, milk and milk products are main constituents of human daily diet (Davies *et al.* 1986, Buldini *et al*. 2002). 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 (Shukla1994; Melquiades *et al* 2001 ; Melquiades and Appoloni 2002 ; Al-Masri *et al* 2004 ; Navarrte *et al* 2007 ; Desmani *et al* 2009 ; Zaid *et al* 2010 ; Shanti *et al* 2010 ; Marko & Borut 2011 ; Soma *et al* 2011and .Alzahrani , 2012) .) .

The present work is the first study of the radioactive content of the powdered milk consumed by children and adults in Saudi Arabia, where, as, the powdered milk for infants were studied before (Alzahrani , 2012) . So, the main objectives of this study are to identify and determine natural radionuclide activity concentrations in powdered milk samples available in Saudi Arabia 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.

2. Materials and methods

Twenty samples of different types of powdered milk were collected during the year of the experiment from the local markets in Saudi Arabia (Jeddah City) . The brand of samples are listed in Table (1). The powdered samples were prepared before analysis, 48-53 g of milk powder was dried in an oven at 80^{o} C for 6 h to a constant weight and then stored in tight plastic containers for four weeks to allow radioactive equilibrium to be reached between parents and their daughter radio nuclides (Ibrahim and Pimpl 1994) . Detection and measurements of the radio nuclides in the powdered milk samples were carried out by gamma ray spectrometer using a NaI (TI) detector

3x3 inch with a 1024-chanel computer analyzer. The detector has a peak efficiency of $1.2x10^{-5}$ at 1332.5Kev Co-60and an energy resolution (FWHM)of 7.5% for 662kev. samples were accounted 15 hours, the activity concentration of ^{214}Pb (352Kev) and ^{214}Bi (609 Kev,1120Kev) were chosen to provide an estimate of ^{226}Ra ,while that of the daughter radionuclides ^{208}Ti (2651Kev) ^{212}Pb (239Kev) ^{228}Ac (911Kev) were chosen as indicator of ^{232}Th . ^{40}K was directly measured using its single photo peak at 1460 Kev emitter . An empty beaker was also counted under the same conditions to determine the background.The activity concentrations for the natural radionuclides in the measured samples were computed by : (Noorddin 1999, El-Taher., 2011)

$A_s = C_a /\ \ P_r M_s$ (Bq/kg) …. (1)

where C_a is the net counting rate of -ray (counts per second), is the detector efficiency of the specific -ray, P_r is the absolute transition probability of -decay and M_s the mass of the sample (kg). The calibration of the counting system was checked by standard samples having the same geometry.

2.1 Calculation of annual effective dose

The annual effective dose due to the intake of natural radionuclides in foods can be calculated from the Formula given by (Alam *et al*.1999 , UNSCEAR 2000):

D = C I E . . ….. (2)

Where : D is the annual effective dose (Sv y^{-1}) to an individual due to the ingestion of radionuclides , C the activity concentration of radionuclides in the ingested sample (Bq Kg^{-1}), I the annual intake of powdered milk (Kg y^{-1}) which depends on a given age (ICRP 1996) and E the ingested dose conversion factor for radionuclides (Sv Bq^{-1}).

The conversion factor 'E' varies with both radioisotopes and the age of the individual are listed in Table (2) (ICRP 1996). Annual effective ingestion dose due to milk consumption strongly depends on the milk consumption . In our study the average mass of the milk consumed by the children (age from 2 to 7 y, 7to12y and from 12 to 17 y) and adults (age from 17 y and above) of 14Kgy^{-1} and 13Kgy^{-1}, respectively, (UNSCEEAR,1993 ; Zaid *et al* 2009) . For the calculation, the total dose is obtained by using the sum of contributions for the radioisotopes in the samples Table (1) with the recommended conversion factors ICRP (1996) in Table (2) .

Table (1). Activity concentrations (Bq /Kg) of different radionuclides present in powdered milk sample

Code No.	Samples	^{226}Ra	^{232}Th	^{40}K
M1	Klim (full)	10.06±0.44	9.54±0.59	82.41±7.09
M2	Velor	8.95±0.38	5.52±0.34	57.91±4.98
M3	Nido	6.69±0.29	3.72±0.24	95.91±8.25
M4	Wadi Fatma	19.53±0.84	4.61±0.28	82.41±7.09
M5	Saudia	6.94±0.35	4.06±0.26	56,18±4.83
M6	Alkhair	7.51±0.39	1.59±0.09	58.45±5.03
M7	Aalia	17.72±0.88	10.25±0.62	46.23±3.98
M8	Baity	15.23±0.76	7.77±0.48	69.72±5.99
M9	Luna	5.59±0.26	6.27±0.38	98.42±8.47
M10	Anlene	10.32±0.51	6.91±0.42	107.43±9.24
M11	Klim(low fat)	14.18±0.71	8.49±0..53	102.80±8.84
M12	Altaie	5.31±023	5.16±0.31	75.98±6.54
M13	Puck	6.43±0.41	3.19±0.20	54.86±4.72
M14	Green Farms	3.26±0.16	3.45±0.21	29.46±2.53
M15	Freshco	7.29±0.28	8.82±0.54	91.57±7.88
M16	Rainbow	8.39±0.37	4.24±0.26	49.38±4.25
M17	Anchor	8.22±0.42	6.46±0.39	39.59±3.41
M18	Two Cows	11.12±0.48	10.11±0.62	93.10±8.01
M19	Orlanda	8.57±0.44	13.57±0.83	52.13±4.48
M20	Regilait	11.64±0.51	11.72±0.72	146.33±12.58
	Average	9.64	6.77	74.51

Table(2). Dose conversion factors of ^{226}Ra ,^{232}Th and ^{40}K for the age groups children (2-7y ,7-12y,12-17y) and adults □ 17 y

Dose conversion factors (Sv Bq^{-1})			
	^{226}Ra	^{232}Th	^{40}K
Children 2-7 Y	6.2×10^{-7}	3.5×10^{-7}	2.1×10^{-8}
Children 7-12Y	8.0×10^{-7}	2.9×10^{-7}	1.3×10^{-8}
Children 12-17Y	1.5×10^{-6}	2.5×10^{-7}	7.6×10^{-9}
Adults 17 Y	2.8×10^{-7}	2.3×10^{-7}	6.2×10^{-9}

3. Results and Discussions

3.1 Activity concentrations of analyzed radionuclide's in milk samples

Measured activity concentrations of ^{226}Ra ^{232}Th and ^{40}K in the powdered milk samples under study including their uncertainty are reported in Table 1. The activity concentrations of, ^{226}Ra vary from 3.26±0.16 Bq/Kg in (s.no. 14) to 17.72±0.88 Bq/Kg in (s.no.7) with an average value 9.64 Bq/Kg , ^{232}Th concentrations vary from 1.59±0.09 Bq/Kg in (s.no.6) to 13.57±0.83 Bq/Kg in (s.no.19) with an average value 6.77 Bq/Kg.

Table (1) shows that the main gamma detected activity arises from ^{40}K was detected in all samples and varied between 29.46±2.53 Bq/Kg (s.no.14) to 146.33±12.58 Bq/Kg in (s.no.20) with an average value of 74.51 Bq / kg , this variation in the activity concentration is not significantly large from one brand of milk to another. Figure 1 shows comparison between the mean activity concentration of ^{226}Ra ,^{232}Th and ^{40}K in powdered milk under investigation. The ^{226}Ra ,^{232}Th and ^{40}K 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 ^{226}Ra and ^{232}Th in all samples and their average values appears much above the recommended values of other countries . The measured activity concentrations values for ^{40}K and the average value in these samples are found to be higher than the values of Indian(Shanthi *et al* 2010) and Egypt(Ibrahim *et al* 2007) and 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 ^{40}K activities are, on an average, lower than the other study by almost 20 % .

Table (3). Comparison of the average concentrations of ^{226}Ra,^{232}Th and ^{40}K with those published data in powdered milk ($Bqkg^{-1}$)

Region	^{226}Ra	^{232}Th	^{40}K	Reference
Saudi Arabia	**3.26 – 17.72 (9.64)**	**1.59 – 13.57 (6.77)**	29.46– 146.33 **(74.51)**	present Work
Iran/France	0.05	0.142	434	Hussein *et al.*, (2006)
Brazil	----	1.7 – 3.7	489	Melquiades *et al*, (2002)
Indian	2.5	1.02	34.35	Shanthi et al 2010
Egypt Elexandria	0.44	-----	47. 25	Ibrahim *et al* . (2007)
Syria	-----	----	129- -435	Al-Marsi *et al*.,(2004)
Jordan	0.5 --2.14	0.78--1.28	349 —392	Zaid *et al*.,(2010)

3.2 Annual effective dose for different age groups

As shown in Table (3), the results of the calculated age-dependent annual effective dose indicate that :-

(1). The annual effective dose due to the intake of ^{226}Ra varies from the minimum value 28.29 µSv y^{-1} to the maximum value 169.56 µSv y^{-1} with an average 83.74 µSv y^{-1} , from 36.51 µSv y^{-1} to 218.78 µSv y^{-1} with an average 108.05 µSv y^{-1} and from 68.46 µSvy^{-1} to 410.46 µSvy^{-1} with an average of 202.93 µSvy^{-1} for children (age 2–7 y) ,(age 7-12 y) and (age 12-17y) respectively. For adults (age 17 y) the minimum annual effective value is 1.19 µSv y^{-1} and the maximum value is 7.11 µSv y^{-1} with an average 3.51 µSv y^{-1} .

(2). The annual effective dose due to the intake of ^{232}Th varies from 3.32 µSv y^{-1} to 66.49 µSv y^{-1} , from 2.75 µSv y^{-1} to 55.94 µSv y^{-1} and from 5.57 µSv y^{-1} to 47.49 µSv y^{-1} for children age groups of 2-7y , 7-12y and 12-17y , respectively ,with an average values 77.91 , 64.55 and 23.19 µSv y^{-1} , respectively.

The annual effective for adults varies from the minimum value 4.75 µSv y^{-1} to the maximum value 40.57 µSv y^{-1} with an average value of and 19.81 µSv y^{-1}

(3). The annual effective dose due to the intake of ^{40}K has been estimated ranging from 8 .66 to 43.02 µSv y^{-1} for children (age2-7y) , 5.36 to 26.63 µSv y^{-1} for (age 7-12 y) 3.13 to 15.58 µSv y^{-1} for (age 12-17y) , with averages of 22.19 , 13.74 and 8 .03 µSv y^{-1}, respectively. While, for adults the annual effective dose varies from 2.37 to 11.97 µSv y^{-1} with an average 6.09 µSv y^{-1} . It appears that the average annual dose from^{40}K is below the reference value of 1.0 mSvy^{-1} recommended by ICRP in all ages.

It can also be seen that average annual effective dose of ^{226}Ra is higher than the other two natural radionuclides of ^{232}Th and ^{40}K against children age groups . ^{226}Ra is a highly radiotoxic radionuclide,when humans ingest radium, 20% is absorbed into the blood stream , the absorbed radium is initially distributed to soft tissues and bones, but its retention is mainly in growing bones (El Arabi 2006). A bar diagram comparison of annual effective dose (µSv y^{-1}) for all four age groups in this study is presented in Fig.2 .

(4) . The total average annual doses received from the intake of ^{226}Ra ^{232}Th and ^{40}K due to the ingestion of the powdered milk were (in µSvy^{-1}): 183.74 , 186.34 , and 234.15 µSv y^{-1} for children , ages (2-7y, 7-12y and 12-17y) , respectively. For adults the total average annual effective is 29.41 µSv y^{-1} (Table 4). This indicates that children have a higher risk factor compared with adults. These results for all age groups are within the typical worldwide range of annual dose (200– 800 mSv) due to the ingestion of all natural radiation sources reported by UNSCEAR 2000.

Also the results showed that ^{226}Ra gives the largest contribution to the total average annual effective dose due to the intake of the powdered milk ; 46% , 58 % and 87 % for children , ages , 2-7y ,7-12y and 12-17y , respectively and the lowest contribution 7 % for adults . For the natural radionuclide ^{232}Th, the results showed that it is the second largest contributor to the total annual effective dose contributing to 42%, 35 %, 10 % of the total annual effective dose for all the three above said age children groups, respectively and gives the largest contribution 67 % for adults . The contribution due to the intake of ^{40}K to the total annual effective dose is 12%, 17% and 3 % for the children (age 2-7y, 7-12y and 12-17y) , respectively and 21 % for adults. Fig. 3 shows percentage contribution to activity concentration by age groups.

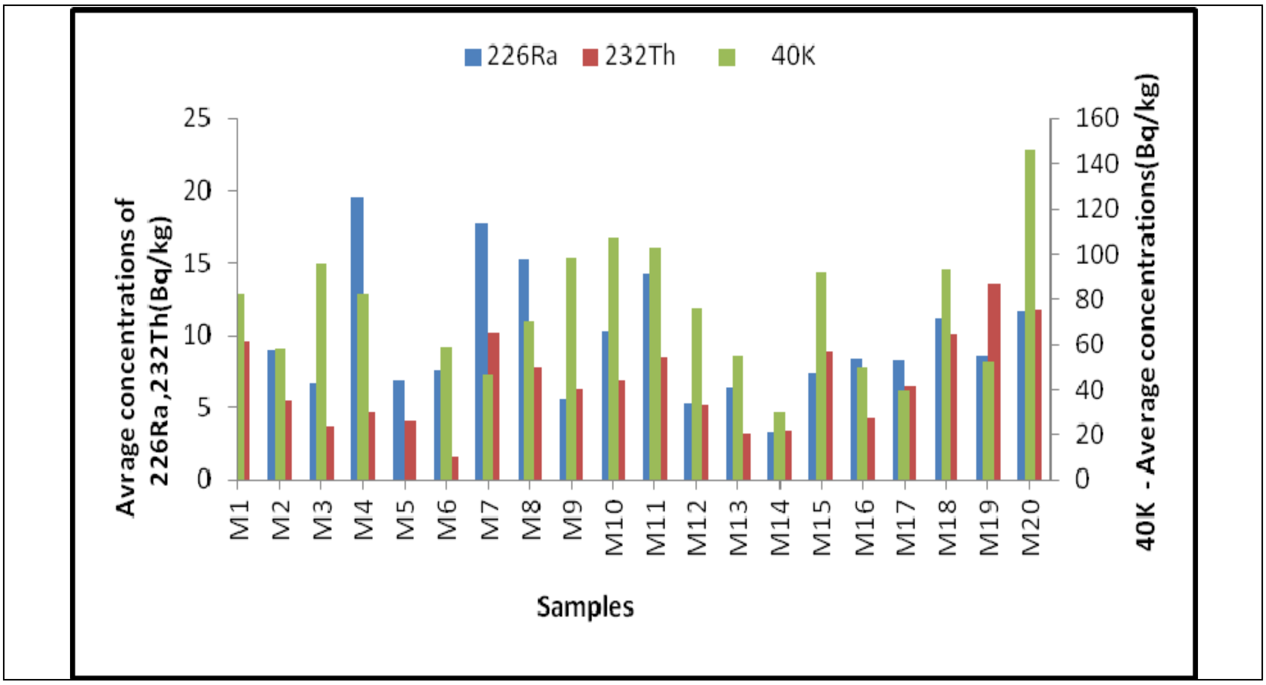

Fig.1 Comparison between the mean activity concentrations of ^{226}Ra ,^{232}Th and ^{40}K in powdered milk in Saudi Arabia

Table 4: Annual effective dose (μSv y^{-1}) for different age groups

Radionuclide		**Effective dose (μSv/ y)**			
		Children			***Adults***
		2-7 y	**7-12 y**	**12-17 y**	**17 y**
^{226}Ra	Minimum	28.29	36.51	68.46	1.19
	Maximum	169.56	218.78	410.21	7.11
	Average	83.74	108.05	202.93	3.51
^{232}Th	Minimum	3.32	2.75	5.57	4.75
	Maximum	66.49	55.94	47.49	40.57
	Average	77.91	64.55	23.19	19.81
$^{40}K^{22}$	Minimum	8.66	5.36	3.13	2.37
	Maximum	43.02	26.63	15..58	11.97
	Average	22.19	13.74	8.03	6.09
Total Average		183.74	186.34	234.15	29.41

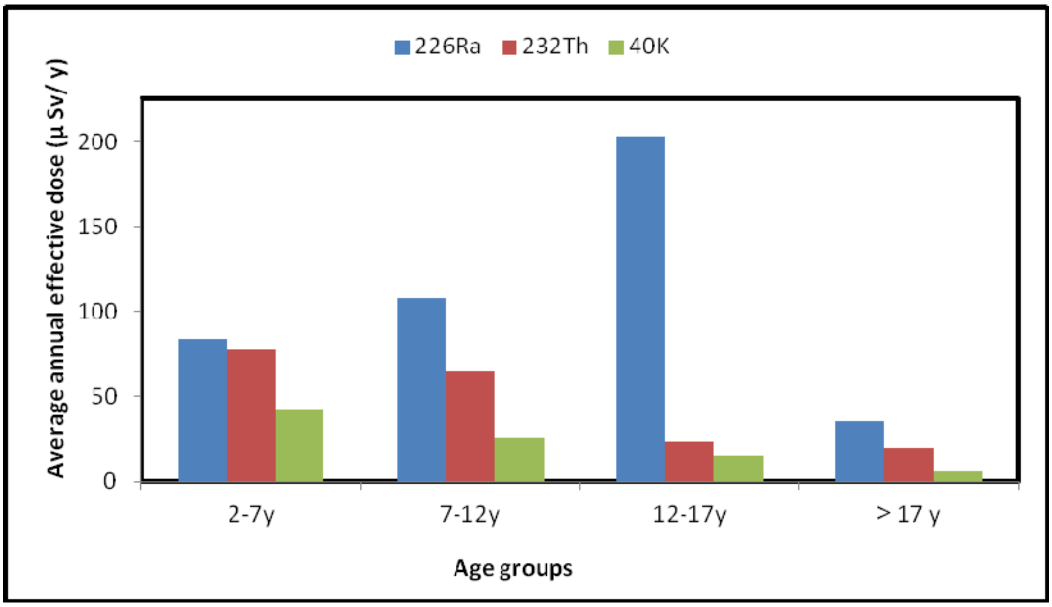

Fig. 2 Comparison between average annual effective dose rate (μSvy^{-1}) of natural radioactivity in powdered milk consumed by different age groups in Saudi Arabia

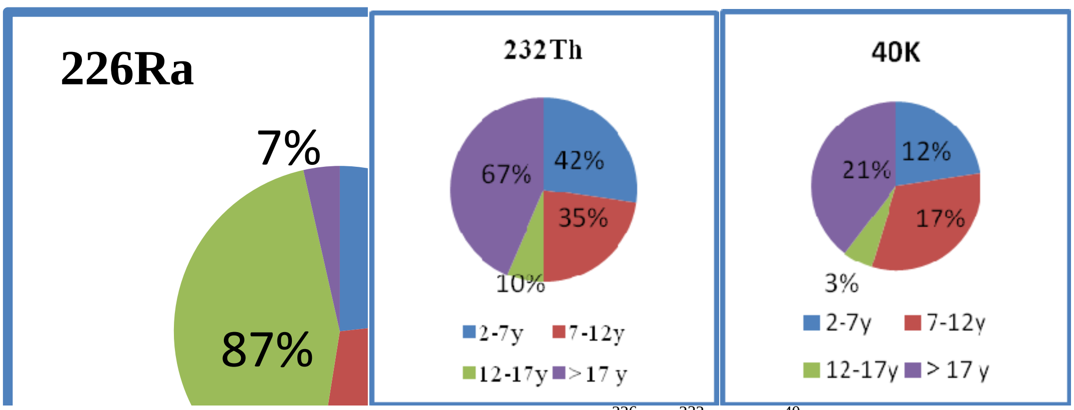

Fig.(3) Percentage contribution to activity concentration ^{226}Ra, ^{232}Th and ^{40}K by age groups

4. Conclusions

In this study, the radionuclides present in powdered milk consumed in Saudi Arabia such as ^{40}K, ^{226}Ra and ^{228}Ra are identified with averages of 9.64 Bq/Kg , 6.77 Bq/Kg and 74.51 Bq/Kg , respectively. The values of ^{226}Ra and ^{232}Th concentrations are found to be greater than those reported of other countries in the literature ,where ^{40}K concentrations are lower the other study .So , we can deduce that the radionuclides level in the studied samples of powdered milk in Saudi Arabia for children and adults is obviously below the maximum level permitted by IAEA , so , these kinds of milk can be normally consumed.

Total averages annual effective dose due to the ingestion of all three natural radionuclides for different age groups of children (2–7 y, 7–12 y, 12–17 y) and adults (_ 17 y) are found to be within the average annual ingestion radiation dose due to natural sources .The total average annual effective dose received by children being 2.2 , 2.1 and 2.9 times higher than those received by adults, which are far below the WHO recommended limit and ICRP for radiological safety .

The data generated in this study will provide base-line radiometric values for natural and artificial radioactivity in milk and help to develop future guidelines in Saudi Arabia for radiological protection for the population.

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