Natural Radioactivity and Heavy Metals in Milk Consumed in Saudi Arabia and Population Dose Rate Estimates

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Abstract
This paper represents an important part of the Saudi Food and Drug Authority plane to reach its aims regarding the safety and effectiveness of food for humans.The results of radioactivity analysis carried out for ^{40}K,^{232}Th and^{226}Ra in powdered infant's milk used in Saudi Arabia (Jeddah city).The main detected activity corresponding to^{40}K was within the range reported in different parts of the world with average activity of 234.18±1.9BqKg^{-1}, while the average activities of^{226}Ra,^{232}Th were 0.46 Bqkg^{-1}, and 0.35 Bqkg^{-1}, respectively, although the activity of some samples were below the detection limit. 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 infants were estimated to be 410μSv for infant ≤1Y and157 μSv for infants (1-2Y) ,which are lower than allowed value (1mSv). The heavy metals analyses were done by atomic absorption spectrophotometer. The geometric mean of Fe, Zn, Mn,Cu and Pb in the samples of powdered milk was found to be 3.033 ,2.91, 0.031 ,0.182 and 0.034 mg/ kg respectively, where as the daily intake was computed to be 0.186, 0.179, 0.002 ,0.001 and 0.002 mg / day, respectively. The results showed that the intake of heavy metals through the ingestion of milk did not exceed the limit of one as proposed by US-IPA. This study could be useful as a baseline data for radiation and heavy metals exposure to infant's milk and their impact on infant's health**[**J. H. Al-Zahrani. **Natural Radioactivity and Heavy Metal s in Milk Consumed in Saudi Arabia and Population Dose Rate Estimates.** Life Sci J 2012;9(2):651-656]. (ISSN:1097-8135). http://www.lifesciencesite.com. 98

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1-Introduction:

Milk is an important vector of radio nuclides and heavy metals to man that may get into the environment from the mining activities .The radio nuclides and heavy metals enter the human body mainly by two routes namely : inhalation and ingestion, Licata *et al.,(* 2004).Also milk is one of the important food for human nutrition and contains all the macronutrients namely protein, carbohydrates, fat, vitamins (A,D and B groups) and trace elements particularly calcium, phosphate, magnesium, zinc and selenium, Abollino *et al.,(*1998); Buldini *et al.,(* 2002). Milk is the main basic foodstuffs for the infants especially for infants less than one year because they generally consume more milk on apery body weight basis than adults. So, the assessment of radioactivity and heavy metals levels in the powdered infant's milk and the associated doses are of crucial importance for controlling the radiation levels and necessary in establishing rules and regulations relating to radiation protection ,Quindos *et al*.,(1994) . It is also important to understand the behaviour of natural radio nuclides and heavy metals in the environment because such informations can be used as the associated parameter values for radiological assessment (Vera et al. (2003). In addition Potassium is an essential constituent of cellular tissue,^{40} K is one of the most important natural radio nuclides . Also, the heavy metals such as Fe ,Zn,Cu , Mn and Pb are essential at very low concentrations for the survival of all forms of life .

There is increasing world wide concern about quality of powdered milk , studies in recent years have been indicated,Shukla(1994) 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 and Borut (2011) ; Soma *et al (*2011) .

In Saudi Arabia no surveys of radioactivity in powdered infant's milk have been carried out and no baselines of concentration of natural and anthropogenic radioisotopes have been reported. Therefore, the establishment of radio-isotope concentrations will prove meaningful information that can contribute to knowledge of population exposure and to the setting up of original baseline, IAEA (1989).

The aim of this study was to investigate the concentration of some long-lived radio nuclides (^{226}Ra, ^{232}Th and ^{40}K) and the concentration of some heavy metals such as Fe , Zn , Cu , Mn and Pb in the powdered infant's milk . In addition estimation of the annual internal dose from the intake of natural isotopes and heavy metals . These measurements can be useful as baseline values for the estimation of the internal radiation and heavy metals doses .

2 . Materials and Methods:

Twenty five samples of different types of powdered infant's milk were collected from the local markets in Saudi Arabia (Jeddah City) between 2010-2012 . The

Type of samples are listed in Table (1). The powdered samples were 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 infant's 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 $1.2x10^{-5}$at1332.5KevCo-60and an energy resolution (FWHM)of7.5%for662kev.samples were accounted 10 hours, the activity concentration of Pb^{214} (352Kev) and Bi^{214} (609 Kev,1120Kev) were chosen to provide an estimate of ^{226}Ra ,while that of the daughter radionuclidesTi^{208} (2651Kev)Pb^{212} (239Kev) Ac^{228} (911Kev) were chosen as indicator of ^{232}Th,^{40} K was directly measured using its single photo peak at 1460 KeV emitter ,jibiri *et al.*,(2007). The activity *concentra*tion (AE,i) in Bq kg^{-1}, for a radionuclide i with a detected photo peak at energy E,was obtained from the following equation, Noorddin (1999) :

AE;i (Bq kg^{-1}) = NE;i / e_E t cd M ....(1)

Where:

NE;i *= is the net peak-area of the radionuclide i at energy E* , e_E *is the detector energy-dependent efficiency at energy E , t is the counting live time in sec, cd is the gamma-ray yield per disintegration of the nuclide i for its transition at energy E , M is the mass of the sample.*

Heavy metals were analysed using an atomic absorption spectrophotometer (A Analyst 700) reagents blank determinations were used to correct the instrument readings. Also, after every 4 samples readings standards were run to make sure that the obtained results were within ranges.

3. Results and Discussions

3.1. Radioactivity analysis of infant's milk

The measured activity concentration of ^{226}R, ^{232}Th and ^{40}K detected in the samples of powdered infant's milk under study including their uncertainty are summarized in Table (1).It can be noticed that ^{40}K was detected in most of samples and varied between 210.21±.31 $Bqkg^{-1}$ to 257.51±3.33 $Bqkg^{-1}$ with an average value of 234.18 ±1.9 $Bqkg^{-1}$.The measured concentration ranged from 0.25±0.03 $Bqkg^{-1}$ to 0.85±.12 $Bqkg^{-1}$ and from 0.09±.02 Bqkg to 0.76±.12 $Bqkg^{-1}$ for^{226}Ra and ^{232}Th respectively. On the other hand, the highest values of ^{40}K were detected with activities 250.8±2.55 , 257.51±3.33 and 252.3±275 $Bqkg^{-1}$in samples No. (12, 14 and 20) respectively. The lowest concentration of ^{40}K was found 210.20±.31$Bqkg^{-1}$ in sample No.(24) . While the lowest concentrations of ^{226}Ra and ^{232}Th were 0.25±.03 $Bqkg^{-1}$ and 0.09±0.02 $Bqkg^{-1}$ in samples No.(1) and No.(15), respectively.The ^{226} Ra,^{232} Th,^{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 (2). It is important to remark that the ^{226}Ra ^{232}Th and ^{40}K activities levels determined in the present study are similar to those of powdered milk consumed in other countries, Melquiades *et al.*, (2002); Al-Marsi *etal.*,(2004); Hosseni *et al.,(*2006) ;Ibrahim *et al.* ,(2007);Zaid *et al*.,(2010)

Table(1) :activity concentrations of ^{226}Ra,^{232}Th And ^{40}K in powered infant's milk (Bqkg

Table (2): Comparison of the average concentrations of

No.	Samples	226Ra	232Th	40K
1	Semilac Gain	0.25±0.03	-----	-----
2	Eptamil	0.55±0.10	0.29+0.02	210.7±2.26
3	Bi0mil	0.32±.02	0.76±.12	222.3±0.47
4	Iasomil	0.68±.08	--------	-------
5	Babelac	0.61±.07	---------	-------
6	Hiap	0.30±.03	---------	--------
7	Expret	0.44±.0.03	0.64±.06	216.9±0.9
8	Novalac	0.49±.04	-------	-------
9	Maial mam	0.28±.02	0.45±.05	-------
10	Meloppa	0.33±.02	0.23±.07	-------
11	Fabimilk	0.85±.12	0.62±.04	------
12	Saha	………	………	250.8±2.55
13	Smilac total	--------	0.30±.09	-------
14	Gain kids	---------	0.65±.14	257.51±3.33
15	France	---------	0.09±0.02	229.9±2.12
16	Blemil plus	--------	0.45±0.04	226.1±2.37
17	Eptajenuer	---------	--------	248.7±3.31
18	Gain plus	-------	0.48±0.06	247.3±2.73
19	Brame care	--------	0.39±0.02	219.1±3.05
20	Nan	-------	0.41±0.07	252.3±2.57
21	Novalac	-------	--------	243.2±0.41
22	Ronagrow	-------	-------	219.9±0.88
23	Larilac	--------	0.73±0.03	227.2±2.76
24	Soupermail	--------	0.56±.03	210.21±0.31
25	Promil	--------	0.22±0.07	231.5±0. 61
	Average	0.46	0.35	234±1.9

Table (2): Comparison of the average concentrations of ^{226}Ra,^{232}Th and ^{40}Kin powered infant's milk with those published data in powder milk(Bqkg^{-1})

Region	^{226}Ra	^{232}Th	^{40}K	Reference
Present Work	0.25 – 0.85	0.09 - 0.76	210 – 257	present Work
Iran/France	0.05	0.142	434	Hosseni *et al.,* (2006)
Jordan	0.5 --2.14	0.78--1.28	349 —392	Zaid *et al.,*(2010)
Newzealand	0.149--0.186	0.147 –.166	594-605	Hosseni *et al.,* (2006)
France	0.05±.011	0.142±.026	434.1	Hosseni *et al, ,*(2006)
Brazil	----	1.7 – 3.7	489	Melquiades *et al.,* (2002)
Egypt	------	-----	222.11	Ibrahim *et al.* (2007)
Syria	-----	----	129- -435	Al-Marsi *et al.,*(2004)

3.2. Internal dose of the radio nuclides from ingested milk

Radiation doses to population from intake of radio nuclides in foods can be calculated from the Formula reported in Reference (UNSCEAR 2000) :

D = C A R --------- (2)

Where :

D *is the effective dose by ingestion of the radionuclide (Sv Y^{-1}),**A**is the activity concentration of the radionuclides in the sample (Bqkg^{-1}) ,* ***C*** *is the internal dose conversion factor by ingestion of the radionuclides (Sv Bq^{-1}) ,**R** is the annual intake of milk (Kg Y^{-1}) which depends on a given age* (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 infant ≤ 1Y and infant 1-2Y were 22.4KgY^{-1} and 15KgY^{-1} ,UNSCEEAR(1993).

Table 3:The dose conversion factor of ^{40}K, ^{226}Ra, ^{232}Th for the age infants (≤1Y)and (1-2Y

Dose conversion factors (Sv Bq-1)			
	^{40}K	^{226}Ra	^{232}Th
Infants ≤1Y	6.2×10^{-8}	4.7×10^{-6}	4.6×10^{-6}
Infants (1-2Y)	4.2×10^{-8}	9.6×10^{-7}	4.5×10^{-7}

For the calculation, the recommended conversion factors ICRP (1996) in Table (3) with Table (1) were used . The results listed in Table (4) showed that the average annual doses received from the intake of ^{40}K,^{232}Th, ^{226}Ra due to the ingestion of the powdered infant's milk were 410 µSv Y^{-1} and 157 µSv Y^{-1} for the ages ≤ 1Y and 1-2 year respectively .Also ^{40}K gives the largest contribution to the total average annual effective dose due to the high consumption rate of milk in the first year, after the age of one year the baby starts to have more solid foodstuff than milk .These results for all ages of infants are within the typical world wide range of annual dose (200–800 mSv) due to the ingestion of all natural radiation sources UNCEAR(2000) .

Table 4: Annual radionuclide intake and effective ingestion dose due to the intake of ^{226} Ra , ^{232}Th and^{40}K in powdered infant's milk

Radionuclide		In*take* *(BqY^{-1})*		Ingestion dose (µSv Y^{-1})	
		Infants		***Infants***	
		≤1Y	1-2 Y	≤ 1Y	1-2Y
^{40}K	Minimum	4709	3153	291.94	132.43
	Maximum	5768	3882	357.63	163.07
	Average	5246	3512	325	148
^{226}Ra	Minimum	5.6	4.0	26.32	3.6
	Maximum	19	13	89.49	12.24
	Average	10	7.0	48	7.0
^{232}Th	Minimum	2.0	1.35	9.27	0.61
	Maximum	17	11	78.31	5.13
	Average	8.0	5.0	36	2.0
Total Average				410	157

3.3. Heavy metals analysis of infant's milk

The range of Fe, Zn, Mn ,Cu and Pb in the milk samples were 2.125 - 3.971 mg/kg , 2.04 – 3.943mg/kg ,0.005 - 0.063 mg/kg , 0.019 – 0.336 mg/kg and 0.012 – 0.098 mg/kg respectively, Table (5). The geometric mean concentration of Fe , Zn , Mn , Cu and Pb was found to be 3.033 ,2.91 ,0.031 ,0.182 and 0.034 mg/kg respectively .The mean of each heavy metal was compared with acceptable limits recorded by International Dairy Federation ,IDF(1979) in which the limits were given as 0.37 ,3.28 , 0.025 , 0.1 and 0.049 mg/kg for Fe , Zn , Mn , Cu and Pb respectively. It was appeared that all heavy metals values reported in this study were within the accepted limits, except the value of iron , which was found exceeded the permissible limit, but it is similar to average value 4.02 – 3.94 mg/kg in milk reported by Jelena *et al.,(*2007) and the value 4.91 mg /l represented by Soma *et al.,(*2011).The infants need iron unusually o rapid growth. In addition, the institute of medicine has estimated that growing infant needs to gain iron of 0.7 mg per day, IMDR (2001).

3.4. Daily intake of heavy metals and hazard quotient

Risk from heavy metals intake through ingestion may be characterized using a hazard quotient (HQ).This is the ratio of the average daily dose (ADD ;milligrams per kilogram of body weight per day) of a chemical to a reference dose (Rf Do; milligrams per kilograms per day)defined as the maximum tolerable daily intake of specific metal that dose not result in any deleterious health effects:

$$\mathbf{HQ = ADD / RfDo} \quad \text{--------(3)}$$

The HQ was estimated for the heavy metals by the intake of the milk , if HQ > 1.0 ,then the ADD of a particular metal exceeds the RfDo ,indicating that there is a potential risk associated with that metal .

The average daily dose(ADD) was calculated by dividing the intake by the average body weight of infants 8.5 Kg and 14.6 Kg for infants ≤ 1Y and infants (1-2Y) respectively ,Dang et al .,(1996).The daily intake was estimated taking into account

Table 5 . Concentration of heavy metals in infant's milk

Samples	Concentration of heavy metals (mg/kg)					Sampels	Concentration of heavy metals (mg/kg)				
	Fe	***Mn***	***Zn***	***Cu***	***Pb***		***Fe***	***Mn***	***Zn***	***Cu***	***Pb***
Semilac Gain	3.271	0.022	2.142	0.132	0.025	**Gain kids**	3.124	0.037	3.153	0.300	0.018
Eptamil	3.367	0.026	3.549	0.213	0.098	**France**	3.057	0.026	2.593	0.146	0.026
Bi0mil	3.614	0.015	2.348	0.202	0.045	**Blemil plus**	3.478	0.042	3.046	0.215	0.038
Iasomil	3.271	0.037	3.362	0.243	0.027	**Eptajenuer**	3.001	0.009	3.182	0.135	0.021
Babelac	3.1.22	0.031	2.611	0.122	0.021	**Gain plus**	3.345	0.005	3.943	0.221	0.097
Hiap	2.535	0.008	3.139	0.162	0.023	**Brame care**	3.971	0.032	3.087	0.143	0.016
Expret	3.208	0.063	4.343	0.120	0.015	**Nan**	3.036	0.026	4.085	0.205	0.042
Novalac	3.114	0.052	3.849	0.232	0.019	**Novalac**	2.858	0.025	2.195	0.335	0.059
Maial mam	3.421	0.012	2.457	0.202	0.024	**Ronagrow**	3.593	0.054	2.041	0.146	0.043
Meloppa	3.225	0.046	3.449	0.143	0.032	**Larilac**	2.125	0.018	3.278	0.019	0.012
Fabimilk	2.223	0.036	2.510	0.119	0.015	**Soupermail**	2.146	0.058	2.127	0.152	0.061
Ssaha	3.118	0.041	2.321	0.336	0.016	**Promil**	2.374	0.021	2.846	0.141	0.024
Smilactotal	2.231	0.035	3.081	0.155	0.041						
Geomean	Fe 3.033	Mn 0.031	Zn 2.91	Cu 0.182	Pb 0.034						

the geometric mean concentration of the metals in milk and the average milk consumption per day for infants≤1y and infants (1-2Y) respectively UNSCEEAR(1993).The RfDo of all the heavy metals except Pb were considered from US-EPA(2003). The RfDo of Pb was taken from WHO(1993).

Estimated exposure and hazard quotient due to intake of infant's milk are given in Table (6). Results showed that the daily intake of Fe ,Mn , Zn, Cu and Pb was 0.186,0.002,0.011,0.021,0.0002 mg/day respectively. These results are normally significantly lower than the recommended desirable levels of , 3.0 – 5.0 mg/kg and 0.5 – 1.0 mg/kg for Zn and Cu respectively, FAO/WHO (1992).The hazarded quotients(HQ) of the heavy metals suggest that the heavy metals in the infant's milk does not pose any apparent threat to the infants, where the HQs of all the considered heavy metals were below the value of (1) as suggested by US-EPA .The HQ ranges from 0.0017(Mn) to 0.07 (Zn).

Table 6 Intake and hazard quotient of heavy metals milk due to ingestion of infant's

Heavy metals	[a]RfDo (mg/kg body weight/day	Geomean (mg/kg)	[b]Intake (mg /day)	[b]ADD (mg/kg body weight/day)	HQ (hazard Quotient)
Fe	7.0×10^{-1}	3.033	0.186	0.022	0.031
Mn	1.4×10^{-1}	0.031	0.002	0.0002	.0017
Cu	4.0×10^{-2}	0.182	0.011	0.0013	0.033
Zn	3.0×10^{-1}	2.91	0.179	0.021	0.07
Pb	3.5×10^{-3}	0.034	0.002	0.0003	0.007

[a]RfDo (Reference oral dose) , [b]Intake and [b]ADD (for infants age ≤1Y)

Conclusions

Natural radioactivity such as ^{226}Ra ,^{232}Th ,and ^{40}K radio nuclides were determined for most available powdered infant's milk consumed in Saudi Arabia. The main gamma activity arises from ^{40} K which was a detected value to be within the world wide ranges as reported in other regions around the Natural radioactivity such as^{226}Ra ,^{232}Th ,and ^{40}K radio nuclides were determined for most available powdered infant's milk consumed in Saudi Arabia. The main gamma activity arises from ^{40} K which was a detected value to be within the world wide ranges as reported in other regions around the world. ^{226}Ra and ^{232}Th activities were below the detection limits. In addition the annual effective internal dose due to the intake of powdered infant's milk was calculated, the main contributor was ^{40}K at the first year due to high consumption rate of milk concentration of heavy metals in infant's milk will provide baseline data and there is a require for intensive of the sampling for quantification of the result. The contribution to the dietary intake of heavy metals was lower than Provisional Tolerable Daily Intake reported by the Joint, FAO/WHO (1992).The largest contributors to the dose received from ingestion of milk in general was due to natural radio nuclides, particularly ^{40}K which was an essential constituent of cellular tissue.^{40} K is one of the most important natural radio nuclides. Regular monitoring of these radio nuclides and the metals in the milk and in other food to prevent excessive build up of the metals in the food chain.

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