International Letters of Natural Sciences *Online: 2015-11-03*
ISSN: 2300-9675, Vol. 48, pp 49-49
doi:10.56431/p-nay9g9

Contamination and Radionuclides Concentration in Imported Canned Foodstuffs in Baghdad Markets

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Keywords: Contamination and Radionuclides Concentration; Imported Canned Foodstuffs; Baghdad Markets; ^{40}K, ^{137}Cs, ^{214}Bi (^{238}U series) and ^{228}Ac (^{232}Th series)

ABSTRACT. This work was concerned with study the contamination and concentration of natural and man-made radioactivity ^{40}K, ^{137}Cs, ^{214}Bi (^{238}U series) and ^{228}Ac (^{232}Th series) in 19 different imported foodstuffs canned samples (Mushrooms, Black Olives, Broad Beans, Peaches, Sweet Corn, Tomato Past, Green Peas, Chick Peas, Baby milk 1,2,3 and Full milk powder) , were collected from locally markets in different regions in Baghdad city. Samples were analyzed by high-purity germanium (HPGe) detector gamma spectrometer systems from Canberra, with high resolution (2.0 keV at 1332 keV) and relative efficiency of ~30% for ^{137}Cs (661.7 keV) and provide (Software Genie 2000 from Canberra). From the data obtained, the activity concentrations (Bq/kg or Bq/l) for ^{40}K, ^{137}Cs, ^{214}Bi and ^{228}Ac in above foodstuff samples were measured. It was found that these values lie within the worldwide range for radioactivity in food. The mean value of radium equivalent activity (Ra_{eq}) in all samples it was below the threshold value of 370 Bq/kg. The all value of indoor and outdoor annual effective dose equivalent in samples it was lower than the world average values (0.45 *mSv/y* for indoor and 0.07 *mSv/y* for outdoor).

1. INTRODUCTION

Radioactive contamination, also called radiological contamination, is the deposition of, or presence of radioactive substances on surfaces or within solids, liquids or gases (including the human body), where their presence is unintended or undesirable [1].

Contamination may affect a person, a place, an animal, or an object such as clothing. Following an atmospheric nuclear weapon discharge or a nuclear reactor contamination breach, the air, soil, people, plants, and animals in the vicinity will become contamination by nuclear fuel and fission products[2].

Radioactive contamination can enter the body through ingestion, inhalation, absorption, or injection. For this reason, it is important to use personal protective equipment when working with radioactive materials. Radioactive contamination may also be ingested as the result of eating contaminated plants and animals or drinking contaminated water or milk from exposed animals[3]. According to their different physic-chemical properties, they can accumulate in different tissues and organs of the human body and, thus, representing a serious health issues for exposed workers and population all over the world. In particular, their assumption through daily intake of food can turn out to be quite relevant to be taken in consideration. For example, chemical ^{238}U turns out to be toxic primarily for lungs and kidneys, where it causes damages to the proximal tubule, besides the fact that when in the metal form, it has also been identified as a potential reproductive toxicant [4]; ^{232}Th affects lungs, liver and skeleton tissues; potassium accumulates in muscles [5] and radium is retained primarily in bones, due to metabolic similarities with calcium. Therefore, depositions of large quantities of these radionuclides in particular organs will be able to affect the health condition, through the weakening the immune system, inducing various types of diseases, and finally contributing to increasing the mortality rate [6].

Food can be contaminated with radioactive materials as a result of an unclear emergency. The surface of food like fruits and vegetables or animal feed can become radioactive by deposit of radioactive materials falling on it from the air or through rain water. Under such circumstances, the large leafy areas of leafy vegetables can effectively intercept the radioactive contaminants that

deposited from the atmosphere, making them more susceptible to radioactive contamination. On the other hand, when lactating cows graze on contaminated herbage of the early fallout, the radionuclides will soon transfer and appear in their milk. Therefore, in general, foods consumed fresh, such as milk, leafy vegetables, and fruit, are initially more likely to be contaminated after a nuclear accident. Depending on the agricultural practices in the area of contamination and the stage of growing or harvest season at the time of the accident, grains, root corps, other produce, and animal-derived food products may become contaminated at later stage[7]. Radionuclides travel through the environment along the same pathways as other materials. They travel through the air, in water (both groundwater and surface water), and through the food chain. Radionuclides may enter the human body by ingestion (eating or drinking), by inhalation, or through the skin[8]. Fig. 1 describes the cycle of radionuclides incorporation in the soil until it's finally absorption for the man. Greatest contamination is achieved when cows graze during fallout periods ingesting pasture grass and soil [9]. Even if they are kept indoors, contamination of milk may occur by inhalation of radionuclides or ingesting them in drinking water and contaminated feed. Milk from goat and sheep, should be checked periodically over a longer period because their grazing habits [10].

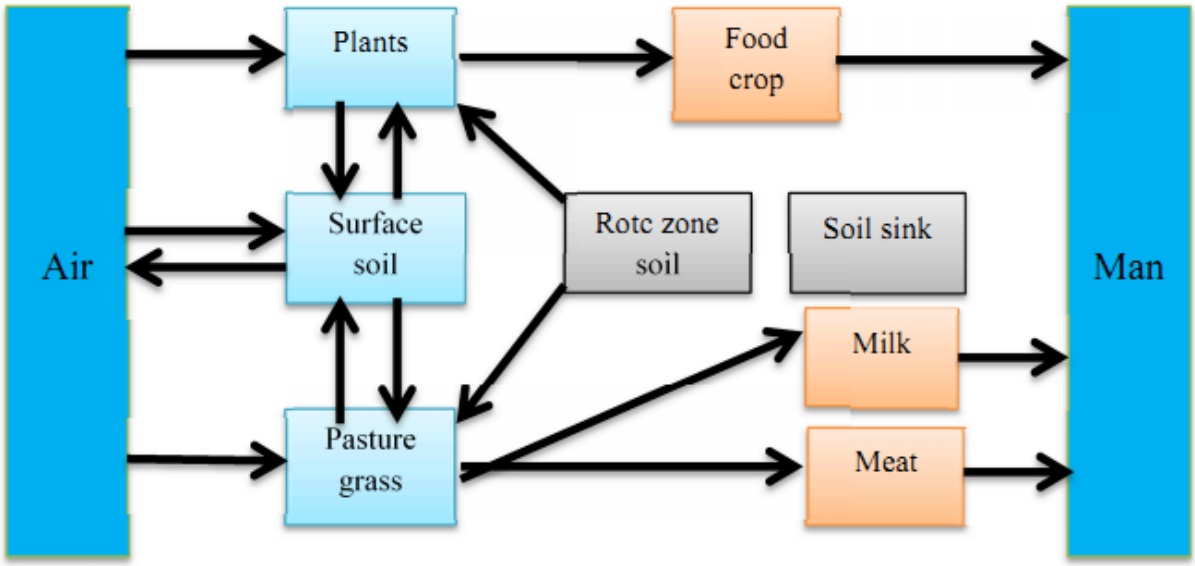

Fig.1: A typical set of pathways through the environment from a radiation source exposed to the air

The most important radionuclides associated to internal radiation exposure (due to ingestion of contaminated water and food) and the contamination of the environment, are ^{89}Sr, ^{90}Sr, ^{131}I, ^{134}Cs, ^{137}Cs, ^{238}Pu, ^{239}Pu, ^{241}Am, ^{242}Cm and tritium. With regard to milk, radionuclides of specific interest are ^{89}Sr, ^{90}Sr, ^{131}I, ^{134}Cs and ^{137}Cs [11]. As they decay at different rates, the doses from these elements and other radionuclides, are delivered in different manners. The aim of present study are assessing the specific activities and examines some of the radiation hazard indices of these naturally and man-made radionuclides (^{40}K, ^{137}Cs, ^{214}Bi and ^{228}Ac) in 19 samples of imported foodstuffs canned from locally markets in Baghdad city, Iraq using γ- ray spectrometry.

2-THEORY CONCEPTS

2-1 Calculation of Radiation Hazard Indices

It is justifiable to exploit as many as possible of the known radiation health hazard indices to achieve a safe conclusion on the health status of an exposed person or environment. To represent the activity levels of ^{40}K, ^{226}Ra and ^{232}Th and by a single quantity, which takes into account the radiation hazards associated with each component, Radium equivalent (Ra_{eq}) is a common index used to compare the specific activities of materials containing ^{40}K, ^{226}Ra, and ^{232}Th by a single quantity, which takes into account the radiation hazards associated with them[12].The activity index provides a useful guideline in regulating the safety standard dwellings. The radium equivalent activity represents a weighted sum of activities of the above mentioned natural radionuclides and is based on the estimation that 13Bq/kg of ^{40}K, 1Bq/kg of ^{226}Ra and 0.7Bq/kg of ^{232}Th, and produce

the same radiation dose rates. The Radium Equivalent activity (Ra_{eq}) which is defined mathematically by Eq.(1) [13].

$$Ra_{eq} = C_{Ra} + 1.43\ C_{Th} + 0.077\ C_K \quad (1)$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively in Bq/kg[14].

2-2 Calculation of Dose in Air, Annual Effective Dose and the Corresponding External and Internal Indices

The total air absorbed dose rate (D) due to the mean specific activity concentrations of ^{238}U, ^{232}Th and ^{40}K (Bq/kg) was calculated using the following Eq.[15].

$$D\ (nGy/h) = 0.427C_{Ra} + 0.662C_{Th} + 0.043C_K \quad (2)$$

The annual effective dose equivalent (AEDE) received outdoor by a member is calculated from the absorbed dose rate by applying dose conversion factor of 0.7Sv/Gy and the occupancy factor for outdoor and indoor was 0.2 (5/24) and 0.8 (19/24) respectively. AEDE is determined using the following Eqs.[16]:

$$\text{AEDE (Outdoor)}\ (mSv/y) = AD\ (nGy/h) \times 8760\ h \times 0.7\ Sv/Gy \times 0.2 \times 10^{-6} \quad (3)$$

$$\text{AEDE (Indoor)}\ (mSv/y) = AD\ (nGy/h) \times 8760\ h \times 0.7\ Sv/Gy \times 0.8 \times 10^{-6} \quad (4)$$

The value for the annual effective dose rate in (*mSv/y*) adequately falls within the permissible dose equivalent limit of 1mSv/year[17]. The external hazards index (H_{ex}) and the internal index (H_{in}) was also determined using the following Equations[18

$$H_{ex} = [C_{Ra}/370(Bq/kg)] + [C_{Th}/259(Bq/kg)] + [C_K/4810(Bq/kg)] \quad (5)$$

$$H_{in} = [C_{Ra}/180(Bq/kg)] + [C_{Th}/259(Bq/kg)] + [C_K/4810(Bq/kg)] \quad (6)$$

Another radiation hazard index called-representative level index ($I_\gamma r$), is defined as follows[19]:

$$I_\gamma r = C_{Ra}/150 + C_{Th}/100 + C_K/1500 \quad (7)$$

This gamma index is used to correlate the annual dose rate due to the excess external gamma radiation caused by superficial materials. It is a screening tool for identifying materials that might become health of health concern when used for construction[16].

3- MATERIALS AND METHODS

In this study, samples were collected from the imported foodstuffs canned and available in locally markets of different regions in Baghdad city, Iraq. The samples are coded and stored in the laboratory for a month to get the case of balance and an appropriate degree heat. Table 1 as shown imported foodstuffs canned samples and the cursor in front each sample the production and expiry date and the origin of sample.

Table 1: Imported canned foodstuffs samples

Sample No	Sample	Origin	Prod. and Expiry date
C 01	Mushrooms in brine	China	9/2014 – 2/2016
C 02	Black Olives in brine	Spain	3/2015 – 3/2016
C 03	Broad Beans	Jordan	5/2015– 5/2016
C 04	Peaches in glucose fructose syrup	Greece	1/2015–12/2017
C 05	Sweet Corn	P.R.C	4/2015 – 4/2017
C 06	Tomato Past	Turkey	5/2015 - 5/2017
C 07	Green Peas	China	2/2015 - 8/2016
C 08	Chick Peas	Italy	4/2015 – 4/2017
C 09	Baby milk/1 (Francelait)	France	2/2015 - 7/2016
C 10	Baby milk/1 (Novalac)	Germany	6/2014 - 12/2015
C 11	Baby milk/1 (Biomil)	Belgium	2/2014 - 11/2015
C 12	Baby milk/2 (Dielac)	Vietnam	7/2014 - 7/2016
C 13	Baby milk/2 (Nactalia)	France	7/2013 - 12/2015
C 14	Baby milk/2 (Quigoz)	France	7/2013 - 12/2015
C 15	Baby milk/3 (NIDO/3)	UAE	6/2014 - 9/2015
C 16	Full milk powder(Almudhish)	Oman	6/2014 - 6/2016
C 17	Full milk powder (Dielac)	Vietnam	5/2014 - 5/2016
C 18	Full milk powder (Landoz)	Malaysia	4/2014 - 3/2016
C 19	Full milk powder (7 Cows)	Austria	6/2014 - 4/2016

The measurements were carried out using gamma spectrometry systems from Canberra, equipped with high purity P-type coaxial germanium detector (HPGe) with high resolution (2.0 keV at 1332 keV) and relative efficiency of ~30% for ^{137}Cs (661.7 keV). The detector were surrounded with an 11.5 cm thick lead shield in order to reduce the background and by a 0.3 cm copper layer to attenuate x-rays emitted by the lead shield. The detector were connected to standard electronics and the spectra were accumulated in 8K MCA (integrated data processor 1510 with S100 MCA band a desktop inspector from Canberra), Fig.2. The detector was energy calibrated using a multi gamma standard source prepared in the same geometry (Marinelli beaker) as the analyzed samples. The efficiency calibration was performed and the curves were obtained by fitting the experimental efficiencies for each sample density. Efficiency curves were corrected for attenuation and absorption. The linearity of the detectors was checked using mixed source from Canberra For quality assurance, certified reference materials foodstuffs were used in the same counting geometry as the samples. The background spectra were frequently measured under the same conditions as the samples measurements and were used to correct the calculated sample activities. Due to the low activity concentrations in the measured samples especially for natural and man-made radionuclides such as (^{40}K, ^{137}Cs, ^{214}Bi and ^{228}Ac have energy 1460.8, 661.7, 609.3 and 969.1 keV, respectively). To ensure good statistical significance the average counting time identified 3600 second for each sample. The spectra were analyzed off-line using Genie 2000 software from Canberra including peak search, nuclide identification, and activity and uncertainty calculation modules. Activity concentrations were expressed in Bq/kg dry or wet weight depending on the sample type. The uncertainty reported is the combined uncertainty calculated using error propagation law and at 95% confidence level, based on the relative standard uncertainties of the sample mass, the net peak area, the full energy peak efficiency, the half-life of the radionuclide of interest and the emission probability.

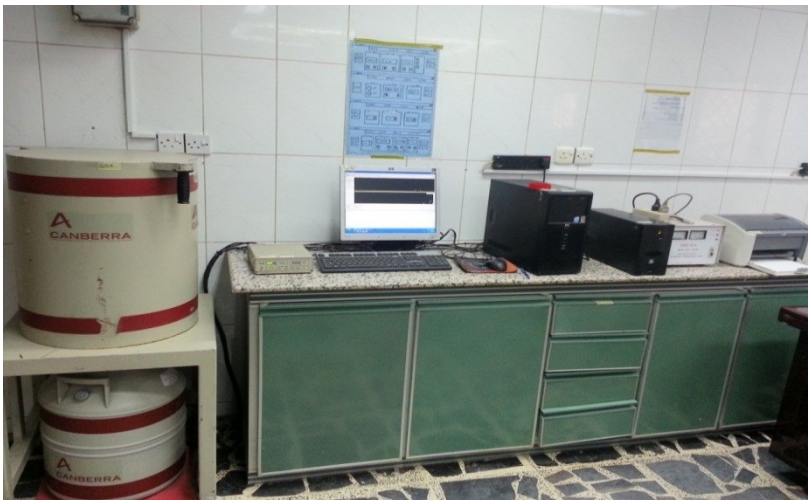

Fig.2: Photo of the HPGe detector and the system of data acquisition

4- RESULTS AND DISCUSSION

In order to complete the monitoring program, some of the locally and imported milk samples have been analyzed. Table 2 shows the results of the activity concentration in Bq/kg of ^{40}K, ^{137}Cs, ^{214}Bi (^{238}U series) and ^{228}Ac (^{232}Th series) in studied samples. In Mushrooms in brine sample were found 180.93, B.D.L., 0.33 and B.D.L., respectively. In Black Olives in brine sample were found 220.38, B.D.L., 0.12 and B.D.L., respectively. In Broad Beans sample were found 222.28, B.D.L., 0.21 and 0.32, respectively. In Peaches in glucose fructose syrup sample were found 160.66, B.D.L., 0.10 and 0.1640, respectively, In Sweet Corn sample were found 229.24, B.D.L., B.D.L. and 0.1093, respectively. In Tomato Past sample were found 227.65, 0.031, 0.21 and 0.21, respectively. In Green Peas sample were found 222.91, 0.020, 0.18 and B.D.L., respectively. In Chick Peas sample were found 214.56, B.D.L., 1.30 and B.D.L., respectively. In Baby milk/1samples were found (80.304-101.48), (0.003- 0.079), (0.08-3.83) and (B.D.L.- 0.51),

respectively. In Baby milk/2 samples were found (92.54-172.27), (B.D.L.- 0.39), (0.39-1.14) and (B.D.L. - 0.027), respectively. In Baby milk/3 sample were found 125.27, 0.063, B.D.L. and B.D.L., respectively. In Full milk powder samples were found (208.3- 255.0), (0.071-0.098), (0.068-0.57) and (B.D.L.- 0.136), respectively, as shown in Fig. 3and 4, these radioactivity concentration values obtained in this study are below the world average values[13].The results obtained for the radium equivalent activity was ranged in Mushrooms in brine, Black Olives in brine, Broad Beans, Peaches in glucose fructose syrup, Sweet Corn, Tomato Past, Green Peas, Chick Peas, Baby milk/1, Baby milk/2, Baby milk/3 and Full milk powder 7.51-19.70 Bq/kg, as shown Fig.5, which is well below the threshold value of 370 Bq/kg [13]. Also, The range values of total air absorbed dose rate (D) in above samples it was 3.705-10.99 *nGy/h*, as shown Fig.6, all values were found to be below the limit (1.0 *mSv/y*) recommended by the International Commission on Radiological Protection. The present range values of outdoor annual effective dose equivalent in samples it was 0.00451 - 0.0134 *mSv/y*. For indoor annual effective dose equivalent it was 0.0181 - 0.0538 *mSv/y*, as shown Figs. 7 and 8, all values of indoor and outdoor annual effective dose equivalent lower than the world average values (0.45 *mSv/y* for indoor and 0.07 *mSv/y* for outdoor)[20].The range values of external hazard indices in samples 0.018- 0.053. For internal hazard indices it was 0.018 - 0.053. For the another radiation hazard index called-representative level index ($I_\gamma r$) were found 0.057- 0.17, as shown Figs.9,10 and 11, All values of hazard indices are less than the world permissible value of unity[20].

Table 2:Concentration of radionuclides and the hazard indices in imported foodstuffs canned samples

Sample No	^{40}K Bq/kg	^{137}Cs Bq/kg	^{214}Bi Bq/kg	^{228}Ac Bq/kg	$^{226}Ra_{eq}$ Bq/kg	D *nGy/h*	AEDE Outdoor *mSv/y*	AEDE Indoor *mSv/y*	H_{ex}	H_{in}	$I_\gamma r$
C 01	180.9325	-	0.3359	-	14.26	7.923	0.0097	0.0388	0.0385	0.0394	0.122
C 02	220.3855	-	0.1233	0.3284	17.56	9.747	0.0119	0.0478	0.0476	0.0478	0.151
C 03	222.2809	-	0.2125	0.3280	17.79	9.865	0.0120	0.0484	0.0480	0.0486	0.153
C 04	160.6627	-	0.1062	0.1640	12.71	7.062	0.0086	0.0346	0.0343	0.0346	0.109
C 05	229.2437	-	-	0.1093	17.81	9.930	0.0121	0.0487	0.0481	0.0481	0.154
C 06	227.6510	0.0318	0.2125	0.2186	18.05	10.024	0.0123	0.0491	0.0487	0.0493	0.155
C 07	222.9192	0.020	0.1853	-	17.35	9.665	0.0118	0.0474	0.0468	0.0474	0.150
C 08	214.5638	-	1.3054	-	17.82	9.783	0.0120	0.0479	0.0481	0.0518	0.151
C 09	94.113	0.079	3.8318	-	11.07	5.683	0.0069	0.0278	0.029	0.040	0.088
C 10	80.304	0.0318	0.0822	0.328	10.66	3.705	0.00451	0.0181	0.018	0.018	0.057
C 11	101.48	0.0038	0.2673	0.5193	8.82	4.821	0.0058	0.0236	0.024	0.024	0.074
C 12	172.27	0.3977	1.1446	-	14.40	7.896	0.0096	0.0386	0.039	0.042	0.122
C 13	165.19	0.0159	0.5621	0.0273	13.32	7.361	0.0089	0.0360	0.036	0.037	0.114
C 14	92.547	-	0.383	-	7.51	4.143	0.0050	0.0203	0.020	0.021	0.064
C 15	125.27	0.063	-	-	9.64	5.386	0.0065	0.0263	0.026	0.026	0.083
C 16	231.9	0.071	0.574	0.027	18.47	10.21	0.0124	0.0500	0.05	0.051	0.15
C 17	255.0	0.098	0.068	-	19.70	10.99	0.0134	0.0538	0.053	0.053	0.17
C 18	208.3	0.079	0.30	-	16.34	9.08	0.0110	0.0444	0.044	0.044	0.14
C 19	239.4	0.071	0.089	0.136	18.72	10.42	0.0127	0.0510	0.05	0.05	0.16

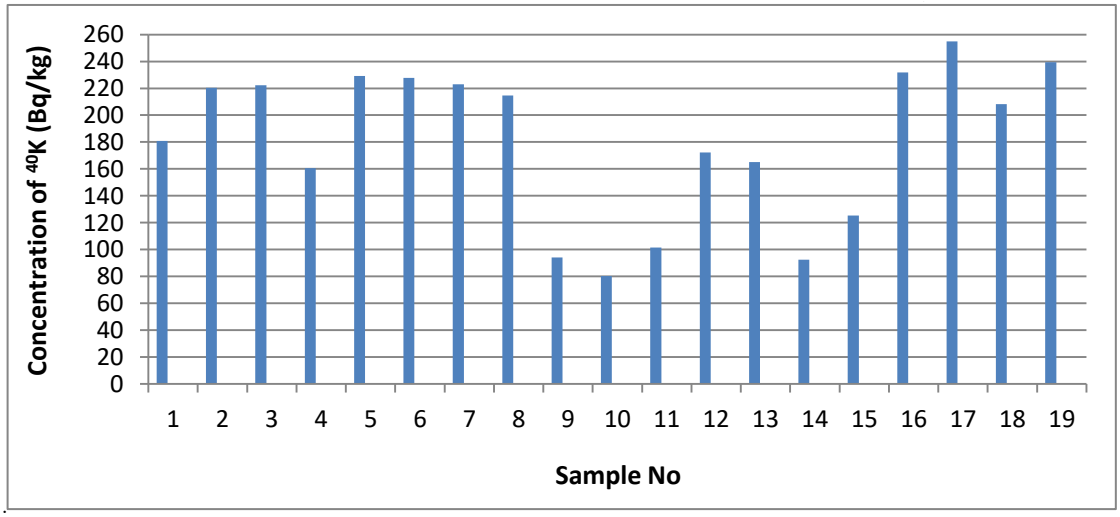

Fig. 3: Concentration of ^{40}K in imported foodstuffs canned samples

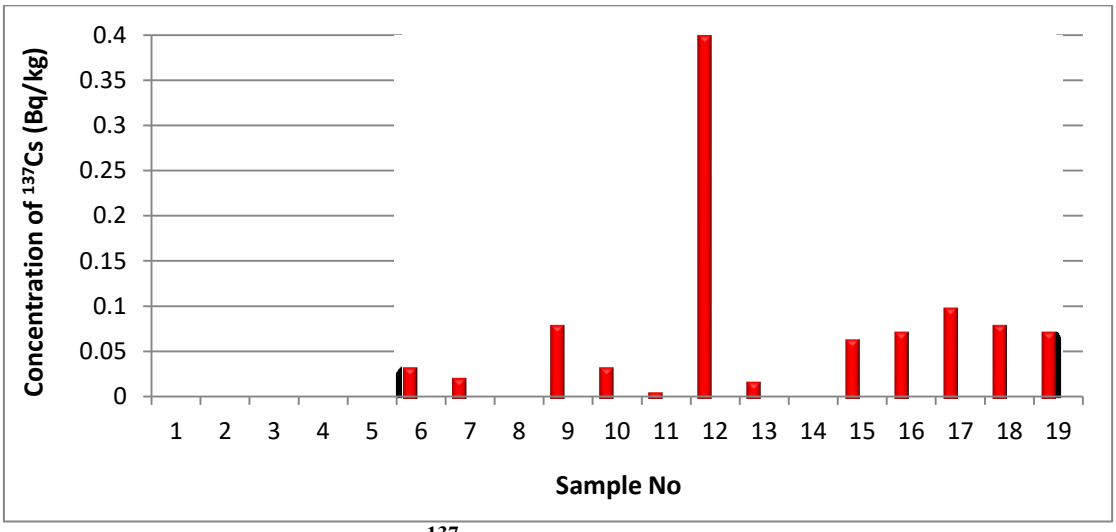

Fig. 4: Concentration of ^{137}Cs in imported foodstuffs canned samples

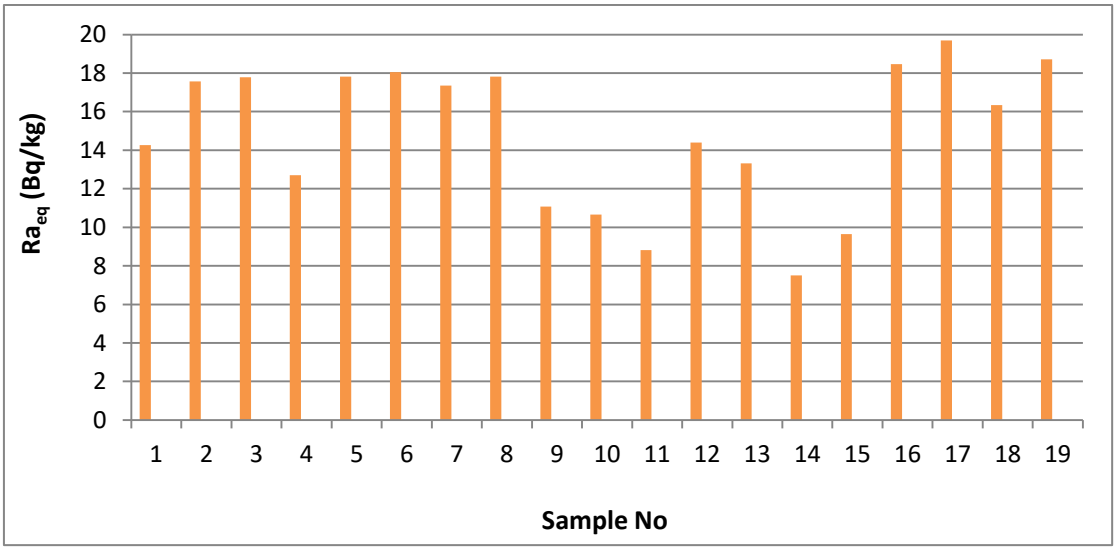

Fig. 5: Ra_{eq} in imported foodstuffs canned samples

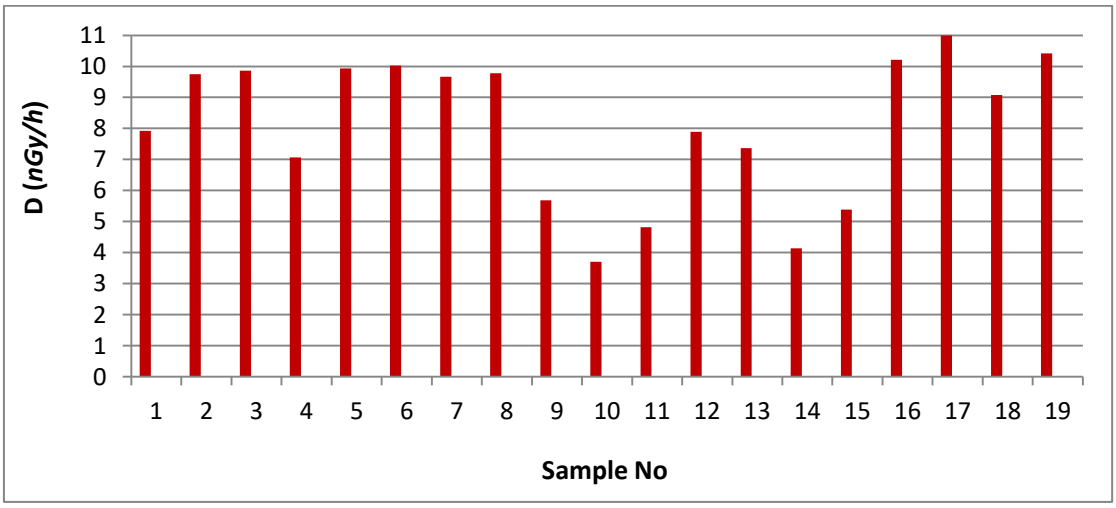

Fig. 6: D in imported foodstuffs canned samples

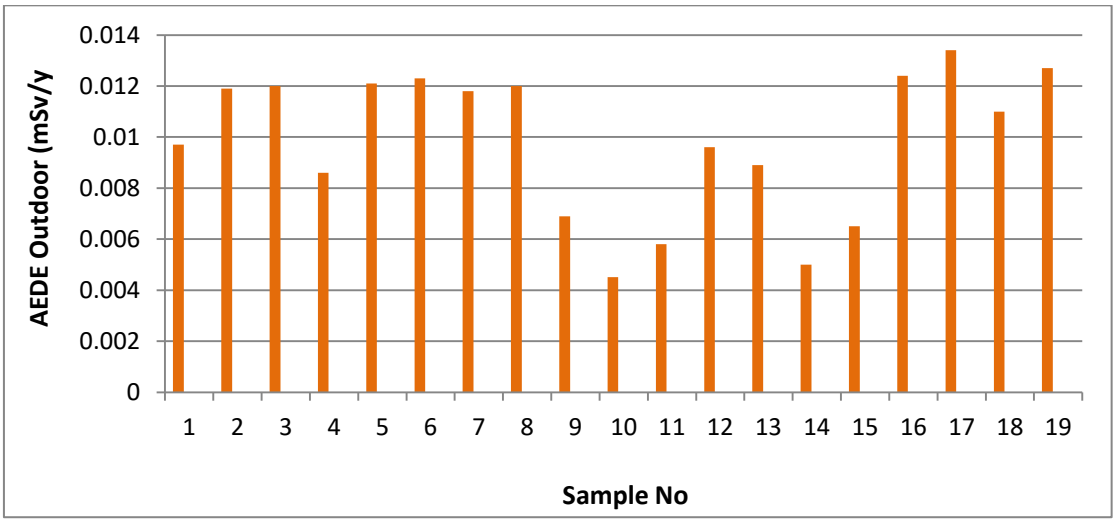

Fig. 7: AEDE Outdoor in imported foodstuffs canned samples

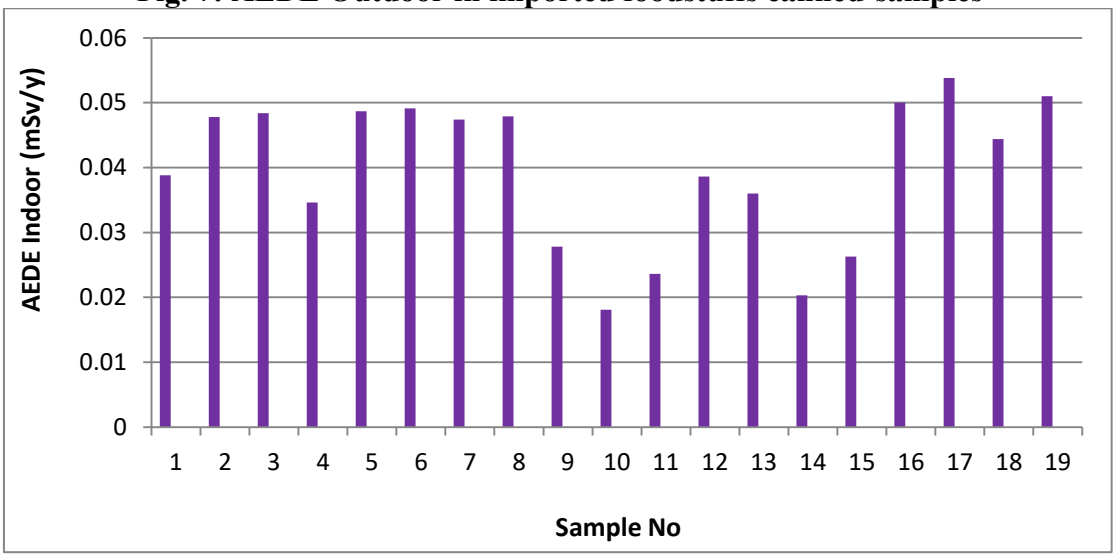

Fig. 8: AEDE Indoor in imported foodstuffs canned samples

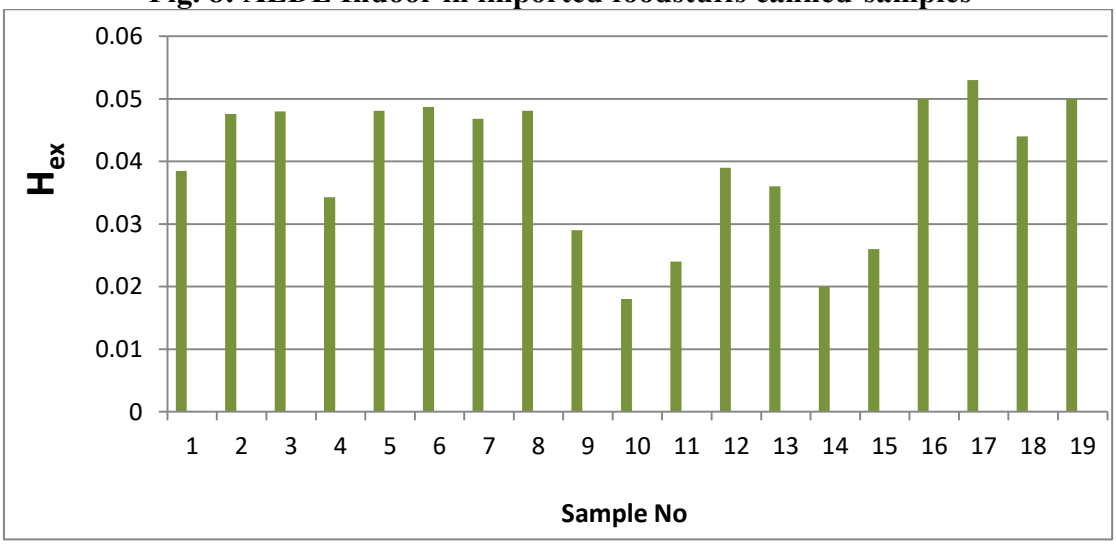

Fig. 9: H_{ex} in imported foodstuffs canned samples

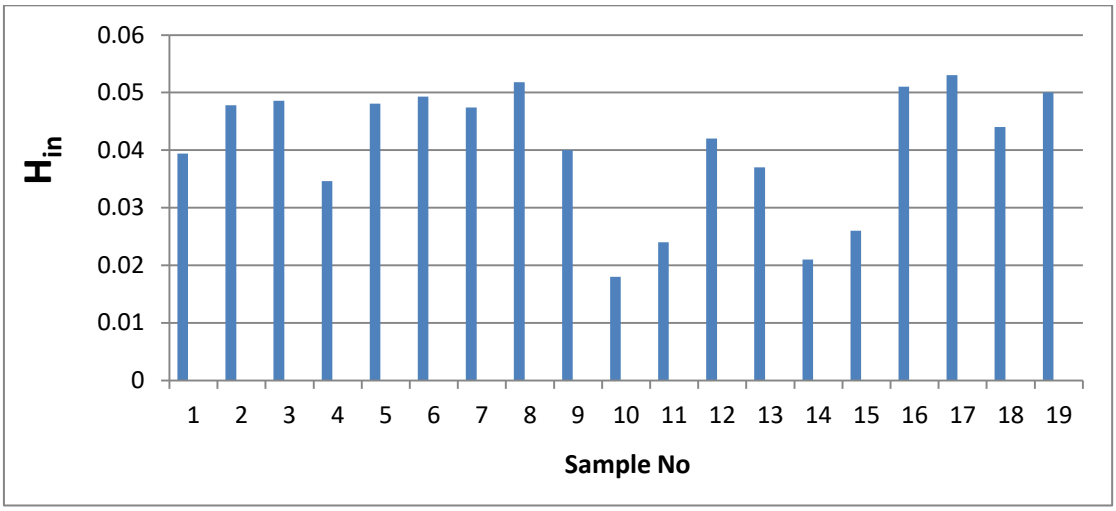

Fig. 10: H_{in} in imported foodstuffs canned samples

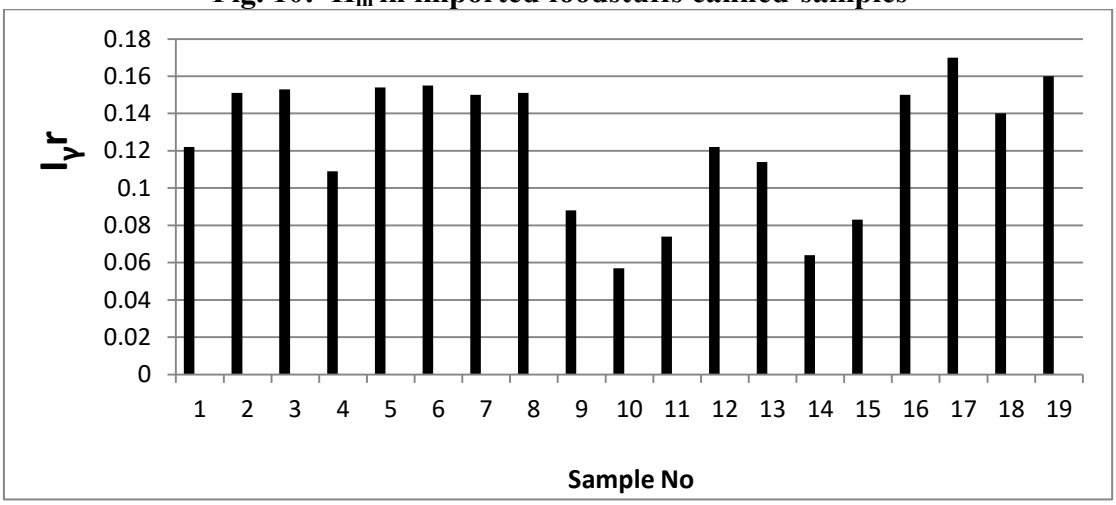

Fig. 11: Iγr in imported foodstuffs canned samples

5- CONCLUSIONS

1-The natural and man-made ^{40}K, ^{137}Cs, ^{214}Bi and ^{228}Ac could be identified in foodstuffs canned samples spectra. ^{226}Ra and ^{232}Th activity was calculated from ^{214}Bi and ^{228}Ac data, respectively, assuming the secular equilibrium between these samples.

2-The values of activity found to ^{40}K, ^{137}Cs, ^{214}Bi and ^{228}Ac in foodstuffs canned samples are lower than the world average allowed.

3-The values of activity found to ^{137}Cs was lower. At the opposite, the ^{40}K activity was highest. As the potassium is rough uniformly distributed in the body, follows intake in foods, and its concentration in the body is under homeostatic control [13], it is less dangerous for human health than ^{137}Cs.

4-The values for the (Ra_{eq}) were found to be within the world average allowed maximum value of 370 Bq/kg. This study could be useful as a baseline data for radionuclide concentration and radium equivalent activity.

In general terms, it can be concluded that the implemented technique presented good results when compared with other literature data. Also it can be concluded that imported foodstuffs canned in Baghdad Markets here analyzed are safe for human consumption because their radioactivity levels are lower than the maximum permitted levels.

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