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Gamma Spectroscopic Measurements of Some Imported Foodstuffs in Egypt

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Abstract

All imported foodstuff coming into Egypt is analyzed for radioactivity level through Radiation Detection Project. In this research, a study of long-lived gamma emitting radionuclides was performed in some imported foodstuffs and feed additives especially those imported from different countries that affected by nuclear accidents as Russia and Japan (Chernobyl and Fukushima accidents).The samples are fish, soya bean, wheat, yellow corn, lentils, dry peas, yellow butter and feed additives. These samples have been tested by gamma spectrometry method using HPGe detector. The specific activity and the annual effective dose of U-238, Th-232, K-40, Cs-137 and Cs-134 in all samples were determined. It was found that the specific activity varied from 0.293 Bq/kg to 6.971 Bq/kg (average of 1.875 Bq/kg) for U-238, from 0.158 Bq/kg to 10.395 Bq/kg (average of 2.73 Bq/kg) for Th-232, from 11.6 Bq/kg to 660.66 Bq/kg (average of 168.7 Bq/kg) for K-40, from 0.225 Bq/kg to 4.26 Bq/kg (average of 0.797 Bq/kg) for Cs-137 and from 0.345 Bq/kg to 1.25 Bq/kg for Cs-134 in food samples. Relative to feed additives the radionuclide Ra-226, U-235, Pa-234m, Th-234, Pb-214, Pb-212 and K-40 were found with different wide range. Also the annual effective dose due to ingestion was calculated. The results were compared with international recommended values and were found to be below the Maximum Permissible Limits.

Keywords: Cesium-137,U-238,Th-232, Effective dose, Food chain, Feed additives.

1. Introduction

The broad use of nuclear energy, nuclear weapon, nuclear accidents, burning of coal production, processing of phosphate minerals and disposal of radioactive waste contribute to unequal distribution of radioactivity on the planet. It is necessary to play a special attention to systematic radioactive hygienic control of foodstuffs of vegetable, mineral and animal origin [1].

Natural and anthropogenic radionuclides are found in terrestrial and aquatic food chains. Natural radionuclides include isotopes of potassium ^{40}K, uranium ^{238}U and its decay series, and thorium ^{232}Th, and its decay series. These naturally occurring radioactive materials (NORM) are long-lived (in the order of 10^{10} year) and are typically present in environmental samples [2]. Anthropogenic radionuclides can be released to the environment either controlled (regulated discharges) or uncontrolled (accidents) as (^{137}Cs and ^{134}Cs). It was estimated that 9 x 10^{16} Bq of the cesium isotope ^{137}Cs, were released to the environment from the Chernobyl accident [3]. Also, the amount of radioactive contaminants released directly into the marine environment was estimated to be 3 -- 27 PBq for ^{137}Cs from Fukushima nuclear accident [4].

Phosphate-based products derived from the wet acid digestion of phosphate rock enhance several radionuclides within the final product and waste/by-products that are subsequently considered to be Naturally Occurring Radioactive Materials (NORM).Such products as monocalcium phosphate (MCP), dicalcium phosphate d (DCP), and tricalcium phosphate (TCP), are important in modern livestock production, and mostly used as food additives for cattle, pig and poultry diets as well as a human diet supplement [5]. During the preparation of MCP (monocalcium phosphate) or DCP (dicalcium phosphate), the secular equilibrium of U with its daughters is disrupted

due to digestion with acid. Some daughters are follow the DCP production as ($^{234}Pa_m$ and ^{214}Pb) and others are eliminated together with sludge such as (^{232}Th and ^{209}Po) [6].

The food chain can be contaminated with nuclear fission isotope as (^{137}Cs and ^{134}Cs) or natural radioactivity as Uranium and Thorium and their daughters. Food additives prepared from phosphate rocks contain elevated quantities of ^{238}U and its decay chain daughters (~1000Bq/kg) [5]. The ingestion of chicken previously fed with food containing some dicalcium phosphate as an additive increases the exposure of human to radiation risk.

The aim of the present work is to quantify the presence of long-lived gamma emitters in foodstuffs and feed additives imported to Egypt from different countries especially that affected by the nuclear accidents as Russia and Japan. Also to estimate annual effective doses to consumers via the ingestion of chicken meat fed with food containing certain amounts of DCP and TCP.

2. Materials and methods

2.1. Samples Collection

Food stuff samples and feed additives samples have been collected from Alex, Port Said and Damietta marine ports which were imported from different countries. Brief description of the collected samples and its origin are given in table 1.

Table.1: Types and origin of imported food stuffs samples in this study

Sample	Sample name	Origin
1	Sardine fish	Japan
2	Mackerel fish	
3	Mackerel fish	
4	Lizard fish	
5	sardine fish	
6	Haring fish	Russia
7	Haring fish	
8	Wheat	
9	Wheat	
10	Yellow corn	Ukraine
11	Yellow butter	
12	Soya bean	
13	Dry peas	
14	Soya bean	
15	Yellow corn	
16	Yellow corn	
17	Soya bean	
18	wheat	
19	wheat	
20	Sardine fish	Norway
21	Sardine fish	
22	Mackerel fish	
23	Mackerel fish	
24	Mackerel fish	
25	Sabary fish	Yemen
26	Sardine fish	
27	Mackerel fish	
28	Free Saba fish	Vietnam
29	Lentils	Turkey
30	yellow corn	
31	yellow corn	
32DCP	Feed additives	
33DCP	Feed additives	
34MCP	Feed additives	
35MCP	Feed additives	

2.2. Samples Preparation

The edible part of the samples were put in plastic jars (300 ml volume) weighed, sealed and analyzed for gamma emitting radionuclides after keeping inside for 30 days allowing secular equilibrium to be attained between ^{238}U, ^{232}Th and their daughters before starting the measurements.

2.3. Detecting System Setup:

The radioactivity measurements were performed by a high-resolution gamma spectroscopic system employing a high purity germanium crystal (HPGe) coupled with multichannel analyzer (TENNELEC) 8192 channels. It is a p-type co-axial detector.

Both of energy and efficiency calibration were performed using standard sources of ^{283}U (R G U.1) and ^{232}Th (R G Th.1) from IAEA/RL/148 Vienna, 1987 with activity 4.9298 $Bq.g^{-1}$ for ^{283}U and 3.247 $Bq.g^{-1}$ for ^{232}Th. Each standard source was counted in the same plastic jars (300 ml volume). The efficiency calculation was made for all gamma lines energy; the results were used to plot the relation between photo peak efficiency and gamma line energy (KeV).

2.4. Counting procedure

Radioactivity concentration of each sample and the back ground were measured for about 20 hours. Since the detection system gives only the count rate that is proportional to the amount of radioactivity in the samples, the radioactivity concentration in the environmental samples was obtained as follow:

$$A = \frac{\text{net cps}}{I \times E \times m} \quad \ldots\ldots\ldots\ldots (1)$$

Where net cps (net count per sec) = cps (sample) – cps (background), A is activity concentration in Bq/kg, I = intensity of the gamma line in a radionuclide, E is measured efficiency for each gamma line observed and m is mass of sample in kg.

Under the assumption that secular equilibrium has been attained between ^{232}Th and ^{238}U and their daughter products, the concentration of ^{232}Th was determined from the average concentrations of ^{212}Pb (238 keV), ^{212}Bi (727.2 KeV) and ^{228}Ac (911 keV) in the samples, and that of ^{238}U was determined from the average concentrations of the ^{214}Pb (352 keV) and ^{214}Bi (609 keV) [7]. For non-equilibrium, $^{234}Pa_m$ activity was determined from emission line at 1001.0 KeV. ^{234}Th activity was determined through the emission line at 92.9 KeV. The ^{235}U was determined from its emission line at143.3KeV, 163.4KeV and 205.3 KeV and also can be determined from emission line at 186 KeV after subtraction of the concentration of ^{226}Ra.A direct measurement of ^{40}K and ^{137}Cs concentrations was determined from 1460 keV and 662 keV gamma energy respectively.

3. Results and discussion

Table (2) summarizes the measured concentrations in $Bq.kg^{-1}$ of ^{238}U, ^{232}Th, ^{40}K, ^{137}Cs and ^{134}Cs in in different types of imported food under study. The results indicated that the main contribution to the background gamma-radiation in different foodstuff is the radiation from the natural radioactive series notably ^{238}U and ^{232}Th in addition to ^{40}K.

From the table, it is noticed that, ^{238}U was detected in all samples except six samples with a minimum value of 0.293 $Bq.kg^{-1}$ for sample.29 imported from Turkey and a maximum value of 6.971 $Bq.kg^{-1}$ for sample.25 imported from Yemen, For ^{232}Th, it was detected in all samples except one sample .25 imported from Yemen with a minimum value of 0.158 $Bq.kg^{-1}$ for sample.2 imported from Japan and a maximum value of 10.395$Bq.kg^{-1}$ for sample.6 imported from Russia .^{40}K was detected in all samples except one sample. 25 imported from Yemen, with a minimum value of 11.6 $Bq.kg^{-1}$ for sample.30 imported from Turkey and a maximum value of 660.66 $Bq.kg^{-1}$ for sample.12 imported from Ukraine. For ^{137}Cs, it was detected in 15 samples only with a minimum value of 0.225 $Bq.kg^{-1}$ for sample.27 imported from Yemen and a maximum value of 4.26 $Bq.kg^{-1}$ for sample No.1 imported from Japan (average of 0.797 $Bq.kg^{-1}$). Also, ^{134}Cs was detected in two samples only (No 1, 2) with values 1.25 and 0.319 $Bq.kg^{-1}$ which are imported from Japan.

The highest specific activity of ^{238}U for sample.25 imported from Yemen may be due to the extraction of uranium from the water which is related to the high concentration of ^{238}U in beach sediment of Yemen [8].Also, it is noticed that the sample.6 showed the highest activity concentration for ^{232}Th. These values are still lower than recommended limit (33 and 45 Bq/kg) for ^{238}U and ^{232}Th, respectively, reported by UNSCEAR (2008) [9].As for ^{238}U, its detection in most samples does not necessarily imply its absence in others. In fact, the infrequency of ^{238}U detection in food samples was reported by various authors [7].

The short lived isotope ^{134}Cs ($T_{1/2}$ = 2 years) was detected in samples imported from Japan that affected by recent nuclear accident (Fukushima). While^{137}Cs was detected in samples imported from Russia, Japan and Ukraine (Chernobyl and Fukushima). This means that these isotopes, are still being up taken by vegetation [4]

The reason of increasing a specific activity of ^{40}K (only in soya bean samples) over the world wide average may be due to plant fertilizers that rich phosphate used to grow the soya bean [10]. The samples .12, 14 and 17 showed highest activity concentration for 40K, which are much higher

than the world wide average (412 Bq.kg^{-1}) reported by UNSCEAR (2008) [9].

Table.2: The activity concentrations in (Bq.kg^{-1}) in the collected samples

Sample	U-238	Th-232	K-40	Cs-137	Cs-134
1	6.45	4.347	183.59	4.26	1.25
2	-	0.158	121.14	1.71	0.345
3	4.176	7.238	135.69	0.811	-
4	0.73	2.469	79.06	0.53	-
5	0.615	1.95	125.75	0.262	-
6	2.581	10.395	139.7	-	-
7	1.96	1.4789	95.98	-	-
8	0.618	3.266	149.04	3.244	-
9	1.117	3.462	129.27	-	-
10	4.6459	0.2175	98.82	-	-
11	-	2.732	24.16	-	-
12	5.564	2.844	660.66	2.483	-
13	1.5305	1.915	275	2.109	-
14	1.222	5.676	660.66	3.77	-
15	3.109	1.553	122.12	2.93	-
16	1.183	2.392	144.185	0.332	-
17	-	4.644	631.57	0.803	-
18	1.017	4.851	160.98	-	-
19	1.26	1.865	149.38	-	-
20	0.776	1.74	115.95	0.693	-
21	-	2.138	100.8	0.553	-
22	-	0.489	80.76	-	-
23	0.907	2.832	106.35	-	-
24	3.569	3.202	95.19	-	-
25	6.971	-	-	-	-
26	2.984	4.37	119.6	-	-
27	3.616	3.193	117.85	0.225	-
28	0.819	0.588	38.01	-	-
29	0.293	0.337	303.75	-	-
30	-	1.538	11.6	-	-
31	0.428	0.794	56.09	-	-

Table 3 shows the radioactivity concentrations of, ^{234}Th, ^{234m}Pa, ^{214}Pb, ^{214}Bi, ^{212}Pb ^{212}Bi ^{228}Ac, ^{208}Tl, ^{40}K, ^{235}U, and ^{226}Ra in the imported feed additives. The results showed that ^{235}U is found in three samples out of four feed additives samples with minimum value of 29.28 Bq.kg^{-1} for sample 35MCP and maximum value of 72.07 Bq.Kg^{-1} for sample 32DCP. The results showed the highest activity concentrations of ^{134}Pa$_{m,}$ the first daughter of ^{238}U with minimum value of 1044.9 Bq.Kg^{-1} for sample 35MCP and maximum value of 2784 Bq.Kg^{-1} for sample 32DCP. ^{134}Pa$_m$ has biological half-life 50 days .So; it has higher risk factor due to its higher energetic β-emitter. The daughters of U cannot be used as a measure of its concentration because its equilibrium is interrupted during digestion with acid (HCl or H_2SO_4) to form dicalcium phosphate DCP as final product [11].

Also ^{40}K was detected in three samples out of four with minimum value of 5.02 Bq.kg^{-1} for sample 32DCP and maximum value of 23.81 Bq.Kg^{-1} for sample 35MCP. Sample 33DCP showed very low activity, this may be due to much purification for human consumption. Different radioactivity concentrations in DCP samples are related to the following two reasons: (i) the geological characteristics of the phosphate rock and (ii) the industrial manufacturing processes used for its production [5].

Table.3: The activity concentrations (Bq.kg^{-1}) in the imported feed additives

Sample No.	^{226}Ra	^{235}U	^{234}Th	^{234m}Pa	^{214}Pb	^{214}Bi	^{212}Pb	^{212}Bi	^{228}AC	^{208}Tl	^{40}K
	58	72.0	672	278	30.6	24.4	45.3	40.2	10.7	12.7	5.02
33DC	-	-	2.28	-	1.41	0.93	0.33	-	-	0.48	-

34MCP	6.3	33.0	224.	113	5.98	6.29	436.	462	88.1	164	11.3
35MCP	4.5	29.2	220.	104	5.8	4.17	398	420	84.3	145	23.8

3.1. Annual effective dose

In this study, annual effective dose were calculated for adults eating imported foods containing ^{238}U, ^{232}Th, ^{40}K, ^{137}Cs and ^{134}Cs using Eq.2 as following **[12]:**

$$D=E\times A\times I \quad (2)$$

Where, D is the annual effective dose $Sv.y^{-1}$, A is the activity concentration for the radionuclide $Bq.kg^{-1}$, E is the dose conversion factor for the radionuclide in $Sv.Bq^{-1}$ (ICRP, 2012) **[13]** and I is mass of food eaten per year in $kg.y^{-1}$.Table 4 shows the dose conversion factors ($nSv.Bq^{-1}$) for the adult (ICRP, 2012) **[13].**

Table 4: Dose conversion factors ($nSv.Bq^{-1}$) for the adult

Age	^{238}U	^{232}Th	^{40}K	^{137}Cs	^{134}Cs	^{226}Ra	^{235}U	^{234m}Pa	^{234}Th	^{214}Pb	^{212}Pb
adult	44	220	6.2	13	19	280	46	0.51	3.4	0.14	5.9

Table 5 summarizes the annual effective doses in $\mu Sv.y^{-1}$ of ^{238}U, ^{232}Th, ^{40}K, ^{137}Cs, ^{134}Cs and their total values. The annual effective dose has been determined from Eq. (2) and the annual consumption of Egyptians according to FAOSTAT 2011**[14]**. The calculated annual effective dose showed the lowest value $0.067\mu Sv.y^{-1}$ (for dry peas) and the highest value $249.32\ \mu Sv.y^{-1}$ (for wheat) imported from different countries (Japan and Russia). The total annual effective dose due to ingestion of all imported food found equal $502.72\ \mu Sv.y^{-1}$ which approximately equal $0.5\ mSv.y^{-1}$ that still is less than those reported of ($1\ mSv.y^{-1}$)**[9].**

Table 5: The calculated annual effective dose in (µSv.y-1) for adults due to consumption of imported food containing 238U, 232Th, 40K, 137Cs and 134Cs

Mean sample	(kg/capita/yr) *	^{238}U	^{232}Th	^{40}K	^{137}Cs	^{134}Cs	Total
Fish	22.1	2.151	13.96	19.205	0.106	0.0268	35.451
yellow corn	63.3	4.447	17.78	187.34	0.447	-	210.01
Wheat	146.1	6.448	108.03	133.30	1.54	-	249.32
Yellow butter	2.9	-	1.743	2.188	-	-	3.931
soya beans	1.1	0.1095	1.062	1.058	0.034	-	2.263
Dry peas	0.1	0.0067	0.042	0.015	0.0027	-	0.067
Lentils	5.2	0.067	0.386	1.225	-	-	1.678

* According to FAOSTAT 2011 **[14]**

3.2. Annual effective dose due the ingestion of chicken meat fed with contaminated food:

The annual effective dose due to ingestion of chicken meat can be calculated from the concentration of radioactive material in chicken tissue which is proportional to the quantity of radioactive material taken through the diet.

The quantity of radioactive material intake by chicken (A) can be calculated according to Eq.3 as following:

$$A = C \times F \times d \times w \quad (3)$$

Where, C is the concentration of radioactive material in feed additive in Bq.kg^{-1}, F is the fraction of radionuclide absorbed by chicken tissue **[15],** d is the percentage of feed additive added to chicken diet equal 5% **[5]** and w is the weight of total diet consumed by the chicken till its weight reaches 1.5 kg .At the end of the production cycle (45 days), the chicken weight reaches 3.5 kg (according to poultry farming).

The annual effective dose (D) due to ingestion of chicken meat can be calculated according to Eq.2 as following:

$$D=E\times A\times I \quad (2)$$

Where A is the calculated concentration of radioactive material in chicken tissue in Bq.kg^{-1}, E is the dose conversion factor for each radionuclide in Sv.Bq^{-1} (ICRP, 2012) **[13]** and I is mass of chicken eaten per year in kg.y^{-1} (according to Egyptian habits).

Table 6 shows the calculated annual effective dose of consumption of chicken meat in µSv.y^{-}1.The annual effective dose was calculated due to the effective radionuclides ^{226}Ra, ^{235}U, ^{234m}Pa, ^{234}Th, ^{214}Pb and ^{212}Pb. The total value of the annual effective dose for two MCP samples (34 MCP and 35MCP) is ranging from (0.0003 to 0.9) µSv.y^{-1}for ^{214}Pb and ^{212}Pb radionuclides respectively. Also for DCP sample (32DCP) the total value of the annual effective dose is ranging from (0.0002 to 76.14) µSv.y^{-1} for ^{40}K and ^{226}Ra radionuclides respectively**.** But 33DCP sample shows very low radioactive concentration because it is used to human consumption. The summation of the total value of the annual effective dose for all radionuclides is ranging from (1.64 to 76.79) µSv.y^{-1}for MCP and DCP respectively. This may increase the annual effective dose to human due to food consumption.

Table.6: The calculated annual effective dose due to consumption of chicken meat in µSv.y^{-1}.

isotope	F	32DCP		34MCP		35MCP	
		C	D	C	D	C	D
^{226}Ra	0.2	582.7	76.14	6.39	0.83	4.56	0.60
^{235}U	0.05	72.07	0.39	33.09	0.18	29.28	0.16
^{234m}Pa	0.05	2784.7	0.17	1132	0.07	1044.9	0.06
^{234}Th	0.0005	672	0.0027	224.7	0.0009	220.8	0.0009
^{214}Pb	0.15	30.69	0.0015	5.98	0.0003	5.8	0.0003
^{212}Pb	0.15	45.33	0.094	436.7	0.902	398	0.822
^{40}K	1	5.02	0.0002	11.39	0.002	23..81	0.002
			Total=76.79		Total=1.98		Total=1.64

Where:

D- The annual effective dose Sv.y^{-1}

C- The concentration of radioactive material in feed additive in Bq.kg^{-1}.

F- The fraction of radionuclide absorbed by chicken tissue.

4. Conclusions

A study of long-lived gamma emitting radionuclides was performed in some imported foodstuffs and feed additives imported from different countries especially those affected by nuclear accidents as Russia and Japan (Chernobyl and Fukushima accidents).The samples were fish, soya bean, wheat, yellow corn, lentils, dry peas, yellow butter and feed additives. It is concluded that:

The main contribution to the background gamma-radiation in samples is the radiation from the natural radioactive series notably ^{238}U and ^{232}Th in addition to ^{40}K. This means that the presence of natural radionuclides in different foodstuff samples was expected specifically.

The short lived isotope ^{134}Cs was detected in samples imported from Japan that affected by recent nuclear

accident (Fukushima). While ^{137}Cs was detected in samples imported from Russia, Japan and Ukraine (Chernobyl and Fukushima). This means that these isotopes are still being up taken by vegetation.

The concentrations of the naturally occurring 238U, 235U and 232Th decay series in DCP and MCP samples indicated that the radioactive equilibrium in the phosphate rock was disrupted during industrial chemical processing.

The total annual effective dose due to ingestion of all imported food is found to be (0.5 $mSv.y^{-1}$) which was found to be below the Maximum Permissible Limits (1 $mSv.y^{-1}$). The total annual effective dose due to feed additives (MCP and DCP) for all radionuclides is ranging from 1.64 to 76.79 ($\mu Sv.y^{-1}$) respectively. This may increase the annual effective dose to human due to food consumption.

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